University of Groningen

Casimir force from complex systems

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DOI:
[10.33612/diss.178363820](https://doi.org/10.33612/diss.178363820)

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2021

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):
Babamahdi, Z. (2021). *Casimir force from complex systems*. [Thesis fully internal (DIV), University of Groningen]. University of Groningen. <https://doi.org/10.33612/diss.178363820>

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Casimir force from complex systems

PhD thesis

to obtain the degree of PhD at the
University of Groningen
on the authority of the
Rector Magnificus Prof. C. Wijmenga
and in accordance with
the decision by the College of Deans.

This thesis will be defended in public on

Friday 10 September 2021 at 12.45 hours

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To the memory of all the innocent students among the passengers of the flight PS752, whose loving lives, their bright future and the chance of ever finishing their study were brutally taken away.

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Chapter 1

Introduction

1.1. Adhesion surface forces

The tendency of two dissimilar surfaces to cling to one another is called the adhesion force, which can be the consequence of different physical and chemical phenomena. The corresponding interatomic and intermolecular surface forces are listed as dispersion forces [1–3], electrostatic forces [4], chemical forces [5], capillary forces [6], entropic repulsion forces [7], etc., depending on the regime in which they operate. The knowledge of each individual contribution of the different surface forces is substantially required to provide better understanding of their effect. The various types of surface interactions play a decisive role depending on several factors like surface morphology, distance between bodies, size of the objects, temperature, humidity, and surface hydrophobicity and hydrophilicity. Surface forces are important in situations when objects come in close proximity or into contact. Generally, they directly influence or depend on phenomena such as dispersion, agglomeration, adhesion, coating, and polishing, which all play a decisive role in materials processing. Surface forces are particularly important in the study and design of micro/nanoelectromechanical systems (MEMS/NEMS), since the surface to volume ratio increases with decreasing system size [8–10].

Dispersion forces, which originate from nature and are known as van der Waals or Casimir forces, could become dominant when the bodies are separated by distances smaller than, typically, 100 nm. They play an important role in nanoscience and nanotechnology including MEMS/NEMS devices [11–14].

Dispersion forces stem from quantum and thermal fluctuations of electric currents inside of the interacting media and in the gap separating them. An overview of the Casimir force and how it can be measured, is given in the following.

1.2. Casimir force

The Casimir force is associated with topological constraints on quantum fields. In quantum mechanics, two reflecting plates facing each other in a vacuum attract each other even if there is no charge involved. This startling phenomenon is known as the Casimir effect [3], named after the Dutch physicist Hendrik B. Casimir, who discovered it in 1948. This is a quantum-mechanical effect that arises because even a perfect vacuum is not entirely empty but filled with unavoidable fluctuations of the electromagnetic (EM) field, imposed by the Heisenberg uncertainty principle, and imbuing the vacuum with a non-zero energy. The Casimir force is the most famous mechanical effect of vacuum fluctuations, and it is the retarded extension of the London/van der Waals force between two objects, formally acting at a larger range (>10 nm). The reflecting plates provide boundaries in between which only some EM fluctuations can exist compared with those in the unrestricted vacuum outside the plates [15]. Therefore, the net result is that there is a force, which tends to push the two plates towards each other (see Figure 1-1). Hence, the Casimir effect appears to be the macroscopic manifestation of the quantization of the EM field between two objects, since it arises from the quantum EM fluctuations that always occur in a vacuum. In fact, the mirror attraction is attributed to EM vacuum fluctuations, which are different in between and outside of the mirrors due to presence of ideally reflecting boundaries. Inside the mirror cavity, only the EM field modes that have nodes on the mirrors are allowed, while outside all the EM field modes can exist. Therefore, the radiation pressure from outside will be larger than that from inside of the cavity resulting in an attractive force pushing the plates together [12,13,15,16].

The Casimir force is relatively long-range in comparison to atomic forces, but only becomes measurable at sub-micrometer length scales. In the mesoscopic regime (~ 100 nm), it is quite substantial, and as a result it has been measured with increasing precision during the past decades. H. B Casimir derived this force as a consequence of the change in (infinite) zero-point energy (ZPE) of the EM field due to the presence of the plates. The original

Figure 1-1. A schematic of what is termed as the quantization of the electromagnetic field. On average the external pressure is greater than the internal pressure. Both mirrors are mutually attracted to each other by the Casimir force.

equation of the Casimir force F_c for perfect reflecting mirrors has the form [3]:

$$F_c = -\frac{c\hbar\pi^2 A}{240L^4}. \quad (1-1)$$

The force is dependent on the distance L between the plates and the plate area A , and it is always attractive in the ideal world. Furthermore, it is dependent on the speed of light c , with the role of converting the electromagnetic wavelength into frequency, and the Planck constant $\hbar$ with the role to convert the frequency into energy. These parameters are the principal constants of quantum mechanics and special relativity. However, this form of interaction as described by Equation (1-1) only holds under the assumption of perfectly reflecting plates. The attraction is attributed to perturbation of the quantum mechanical fluctuations of the EM fields in vacuum. The average of the EM fields with respect to these fluctuations is zero, but the standard deviation (the uncertainty) is not. This fact gives rise to a non-zero electromagnetic energy in vacuum called zero-point energy (ZPE). This name refers to the fact that it persists even at zero $T = 0$ K, and each EM mode (photon) with frequency ω carries a ZPE of $\hbar\omega/2$. The parallel plates impose boundary conditions on the EM fields between them: at the position of the plates the values of the fields must be zero. This ensures that only modes with certain frequency are allowed to exist inside the cavity formed by the plates, while no such restriction applies to the modes outside the cavity [12,13]. This gives rise to the net attractive force between the plates as expressed by Equation (1-1) above. The more realistic calculation between dielectric surfaces was presented in

1952 by E. Lifshitz [17]. Within this theory (see also Chapter 2) the van der Waals and Casimir forces can be considered the short- and long-range limits respectively of the same force.

1.3. Measuring the Casimir force

The Casimir force is of fundamental interest as it is taken as evidence for the existence of the EM fluctuations associated with the quantum vacuum [3]. Progress in understanding the fundamental Casimir force has been put forward when in 2000, almost 40 years after Lifshitz and co-workers, Böstom and Sernelius introduced their ideas of how to treat materials permittivities in the zero-frequency limit [18]. This fundamental quantum mechanical force is nowadays posing a limitation on how to miniaturize very small devices, and it becomes significantly important in some aspects of emerging micro and nanotechnology, as well as is a probe of new forces beyond the standard model. Therefore, the Casimir effect is not only of fundamental importance in understanding the concepts of quantum vacuum and matter-field interactions, but also because it becomes considerably strong at nanoscale separations affecting the motion of parts in nano-devices leading to strong attention of experimental physicists.

The first attempt to directly measure the Casimir force was performed in the 1950s by Marcus Sparnaay [19]. Two parallel metal plates were brought in contact after each measurement to prevent any unintentional electrostatic contribution from becoming too large. Despite of this, the experimental uncertainty was so large that the resulting Casimir force was neither significantly different from zero nor did it contradict Equation (1.1). Later, in the 1970s, an experiment involving metal test bodies was similarly inconclusive [20]. It was not until 1997 when Steve Lamoreaux performed the first high accuracy measurements of the Casimir force using a torsion pendulum [21]. A major source of uncertainty in, e.g., Sparnaay's experiment was the inability of keeping the two plates parallel at nanometer to micrometer separations. A solution for this problem is to use the plane-sphere setup, in which the radius of the sphere is much larger than the typical separation distance. In this case, the curvature of the sphere can be considered gentle enough so that each surface segment dA of the curved body – in this case a sphere – is approximated by a parallel plate configuration, and the contribution of each segment is added to the total force. This approximation is called “*proximity force approximation*” (PFA), see Figure 1-2. This approximation allows to

Figure 1-2. A schematic of the application of the proximity force approximation in defining the set-up of the sphere-plate geometry with a sphere of Radius R above a flat plate for calculation of Casimir force.

Figure 1-3. The SEM image of a probe made by attaching a micro-size Borosilicate sphere at the end of a tipless cantilever. The probe is then coated with 60 nm gold.

calculate the Casimir force by integrating the Casimir pressure between opposing infinitesimal surface area elements, as if they were parallel plates, over the area that the sphere and the plate expose to one another [22].

The sphere-plate geometry is the configuration in which the most precise Casimir force measurements are currently performed [23]. A Scanning Electron Microscopy (SEM) image of the micro-size sphere attached at the end of a tipless cantilever is shown in Figure 1-3.

One can distinguish between static and dynamic setups to measure the Casimir force. An example of a static setup is the technique called contact mode force measurement with atomic force microscopy (AFM), which

Figure 1-4. (a) Schematic of established ways to measure the force in sphere-plate geometry. The force (F) is balanced by a spring constant (k). (b) AFM experiment set-up. (c) Torsional rod realization [36].

was introduced soon after Lamoreaux's experiment [24]. In a static AFM force measurement, a micro-size sphere is rigidly attached to a cantilever. This sphere is brought into proximity to a plate (substrate). The resulting surface force deflects the cantilever by a certain amount. (See Figure 1-4b). From this deflection and the Hooke's law the total surface force can be calculated. This surface force is not equal to the Casimir force: due to charge accumulation there is always a nonzero electrostatic contribution which must be compensated with a reverse bias, even if both surfaces are grounded. In a dynamic setup, one of the surfaces remains immobile whereas the other one moves towards the immobile surface (but does not make contact) performing continuously oscillations around its pivot point (See Figure 1-4a; the same applies in using dynamic force measurement by AFM). In this type of setup, the frequency of rotation ω_r is shifted as compared to the system's natural frequency ω_0 due to the surface interactions. The frequency ω_r is directly determined during the movement using a laser. It is assumed that the oscillator is near a stable fixed-point z so that the effect of the Casimir force can be approximated linearly (weak force regime). This assumption

is valid at distances above several hundred nanometers (typically > 100 nm). However, the weak force assumption will be broken at distances below 100 nm, where the Casimir force extends the amplitude of the oscillator to the nonlinear regime [25]. In a dynamic setup the derivative of the Casimir force is measured, rather than the force itself, from the shift of the resonance frequency ω_r .

Most of the modern experiments are performed with the use of microelectromechanical systems (MEMS), which are characterized by small separation between bodies and sufficiently large surfaces of the interacting bodies. Under these conditions (small distance and large area) dispersion forces (DFs) manifest themselves most distinctly. Irreversible adhesion (stiction) of moving MEMS elements induced by the DF results in malfunction and failure of MEMS devices [26–28]. During operation of the device, stiction can occur due to the pull-in instability, and even during the fabrication stage can be induced by capillary forces in the presence of humidity. The stiction problem seriously restricts applications of MEMS. It is worth to note that strong adhesion by chemical interactions or by capillary forces can be excluded by special preparation of the surfaces, but a relatively weak adhesion due to the DFs cannot be excluded in principle since they are omnipresent. The DF adhesion is responsible for many natural phenomena (for example, the firm grip of geckos on walls and ceilings [29]), and for many artificial processes (for example, stiction of a polymeric film to surfaces [30]). It is also fair to say that the dispersion forces could control the performance of NEMS, and actuation of these devices with the vdW/Casimir forces taken into account is a dynamically developing field [31–35].

Nonetheless, there have been reported difficulties when measuring Casimir force at short distances between surfaces. In reality, when two solids get into contact, they are still separated in practice by an average distance $d \sim 5\text{--}10$ nm due to finite roughness of the contacting bodies. This distance range corresponds to the transition between vdW and Casimir forces that corresponds to the transition between retarded and un-retarded interactions. This range was studied much less than the range $d > 100$ nm, though the DF at shorter distances plays more important role. In addition, the poor knowledge of the forces in the transition region is also related to the same pull-in instability that leads to stiction of the MEMS elements and in general interacting surfaces. During the force measurements, the force is balanced by a spring of a known stiffness (k) as it is shown schematically in Figure 1-4a. In some experiments, the spring is a cantilever that is widely used in atomic force microscopy (AFM) [24,37–39]. In other experiments, this role belongs to a torsional

rod [40,41] or to a string [42]. Such set-ups have as main disadvantage the loss of stability at short separations.

In practice, the shortest jump-to-contact distance ever reported was 12 nm for AFM Casimir force measurements with a stiff cantilever [43], which was significantly smaller than the minimal distance from previous high precision experiments (40 nm [44] to 150 nm [45]). In more stable experiments, the surface force apparatus [46] with much stiffer springs can be used, but even in this case the shortest separation distance (in vacuum) was about 8.5 nm [47]. On the other hand, using a stiffer cantilever in Casimir force measurements would reduce the force resolution. Although, there are measurements at short separations between good conductors (Au-Au) [43,47], similar short range measurements were also reported between a smooth nitrogen doped SiC and Au surfaces [48]. The reason of low accuracy in the short-distance experiments is the absolute uncertainty in the separation $\delta d \sim 1$ nm which gives a large relative error in the force measurement $\sim \alpha(\Delta d/d)$, where $2 < \alpha < 3$ for the sphere-plate configuration. Due to the pull-in instability, the data in the short-distance range $d \sim 10$ nm are limited and have poor accuracy ~ 20 – 30 %, even though in this range the role of the dispersion forces is crucial for many physical phenomena.

The theoretical prediction and modeling of the force at short distances is as difficult as is its measurement. The problem occurs due to non-additivity of the dispersion forces and the description of the contribution of surface roughness. In condensed bodies (in contrast with gases) the atoms in the neighborhood cause an essential change in the properties of the electronic shells, and the presence of a medium between the interacting atoms affects the EM fluctuations through which the interaction is established [49]. If the root-mean-square (rms) roughness is small in comparison with the distance between the bodies, the roughness contribution can be evaluated as a perturbative effect [50,51]. However, if the roughness amplitude is comparable with the separation gap, there has been a reliable manner to predict the force [52].

The Casimir force is typically small, even in the case of perfectly conducting plates. Therefore, not surprisingly, it took many years before it was unambiguously measured. For coherent overview of the topic, we briefly review some of the important earlier experiments. The very first effort to measure the van der Waals force between macroscopic bodies in sphere-plate geometry was done by Derjaguin et al. in the 1950s [53]. The group found difficult to align the setup for parallel plates, and they instead examined a spherical lens and a plate using the PFA approximation. Later, it was found that for a certain range of experiments, measuring in the

sphere-plate geometry is fairly accurate for separations significantly less than the sphere radius. The experiments of Derjaguin et al. for separations of 0.1–0.4 μm between quartz plates provoked the development of Lifshitz's theory [53]. The comparison with Lifshitz theory was complicated due to the incomplete information about the imaginary part of the dielectric function for the quartz over all of its different absorption regions. Forces measured much later were found to fit well with the Lifshitz theory in the retarded regime of large separations, but detailed comparison of theory and experiments were again hindered by insufficient data for the permittivities required in the Lifshitz theory.

The experiments reported by Lamoreaux [16,42] and Mohideen et al. [11,24,54] in 1997 and 1998 marked the beginning of a new era of much more precise measurements of Casimir forces. Lamoreaux performed Cavendish-type experiments employing torsion balances, while the Mohideen's group pioneered the use of the AFM where the Casimir force was determined from the deflection of a cantilever. The agreement of these experiments with theory appears to be on the order of several percent. This great improvement in accuracy compared to earlier experiments stems in part from the availability of much better mechanical, surface characterization, and data acquisition tools.

Another very significant step in the direction of improved accuracy of Casimir force measurements was reported by Chan et al. [25,41], who measured the Casimir force based on the shift in the frequency of a periodically driven MEMS torsional oscillator (MTO) due to the torque exerted on a plate by a metallic sphere. Two years later Decca et al. [40] reported the first highly accurate measurement of the Casimir force between two dissimilar metals on an MTO system. Later on, De Man et al. [38,55] have described AFM Casimir force measurements that allow for continuous calibration together with compensation of residual electrostatic effects. Experiments measuring the force in air between a gold-coated sphere and a glass coated with either gold or a conductive oxide demonstrated firstly that residual electrostatic effects could be effectively eliminated, and secondly as expected the Casimir force was substantially reduced when the gold surface was replaced by the conductive oxide film.

So far, most of the works on the Casimir effect between surfaces have dealt with good conductors or dielectric materials separated by vacuum or air (permittivity $\epsilon=1$), and the force was therefore attractive. However, if the intervening dielectric media is different, the force can be repulsive under certain conditions. Such a repulsion was predicted by Dzyaloshinskii et

al. [49,56] and can be applied, for instance, to explain the tendency of liquid helium to climb the walls of its container: the helium vapor is repelled by the walls and helium liquid moves in to fill the gap left by the vapor. The repulsive force resulting from regions of different permittivity appears to be well known among colloid scientist [57] as it has been directly observed by Munday et al. [58], who measured a repulsive force between a gold sphere and a glass surface immersed in bromobenzene. Consequently, there has been a growing interest in the control of Casimir forces, and in particular in the possibility of realizing repulsive Casimir forces between vacuum-separated objects. This in turn has led to the consideration of metamaterials and topological insulators that might be engineered to have permittivities and permeabilities that would allow a degree of control of the Casimir force that would not be possible with naturally occurring materials. In the case of metamaterials, the studies indicate that the repulsive force between two objects might only be possible at short distances, in which case a metamaterial surface cannot be assumed to have continuous translational symmetry. Even in this case, however, a repulsion cannot result in a stable equilibrium [59].

1.4. The effect of contact potential and distance upon contact

The Casimir force is notoriously difficult to compute for arbitrary geometries. The geometry plays an important role in the context of theory/experiment comparison for the Casimir force. Precise experiments are indeed performed between a plate and a sphere, whereas most exact calculations are devoted to the geometry of two parallel plates. The estimation of the force in the sphere-plate geometry, as we explained earlier, involves the PFA. Moreover, the effect of patch potentials and the roughness of the involved surfaces are the factors that one has to take into account when analyzing the experimental data, and since Lifshitz theory does not account any of these, it is a necessity to employ theoretical modifications. The effect of electrostatic forces and roughness of the surfaces are discussed briefly as in the following.

It is often assumed that the surface of a conductor is an equipotential. While this could be true for a perfectly clean surface of a homogenous conductor cut along one of its crystalline planes, it is not the case for any real surface which can be polycrystalline, stressed, or chemically contaminated, putting aside different materials. Experiments show that even with precautions for extreme cleanliness, typical surface potential variations are on the order of at least a few millivolts [60]. This is most likely due to local variations in

the surface crystalline structure, giving rise to varying work functions and hence varying-potential patches. It is well known that the work function of a metal surface depends on the crystallographic plane along which it lies. As an example, for gold the work functions [61] are 5.47, 5.37, and 5.31 eV for surfaces in the $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$ directions respectively. This variation can be related to different electron masses, hence Fermi energies, for different axes. Therefore, we define a contact potential (V_0) which exists between samples of two dissimilar electrically conductive materials (with different electron work functions) that have been brought into thermal equilibrium with each other, usually through a physical contact. The contact potential may not be small if poor conducting materials are used, where contributions due to trapped charges can exist. If V_0 is large enough to produce significant electrostatic force with respect to the Casimir force, then the effect of V_0 cannot be neglected from the Casimir force curves, besides an interfering signal of laser-surface scattered light which also has to be filtered out at larger separations. Therefore, the effect of V_0 should be removed or minimized while performing the force measurement by applying a compensating voltage.

Although, the Lifshitz theory can be applied for two flat parallel plates separated by a gap d , in reality each plate is likely to be rough and the theory cannot be applied directly. When the separation of the plates is much larger than their root-mean-square (rms) roughness (w), one can calculate a correction to the force due to the roughness using perturbation theory. But even in this case the problem is far from trivial [51,62,63]. The roughness correction was calculated in [52] in comparison to force measurements.

Even though we can measure forces in the scale of pico-Newtons, if the distance between surfaces is not known, we will not reach high accuracy [64]. The roughness of the interacting bodies restricts the minimal separation upon contact d_0 between the bodies. The absolute distance between two bodies is a parameter of high importance for the determination of dispersive forces. It becomes difficult to determine the forces when the separation gap approaches nanometer dimensions. The complication stems from the presence of surface roughness, which manifest itself on the same scale. The so-called distance upon contact has also special significance for adhesion, which under dry conditions is mainly due to Casimir/van der Waals forces. The plate-sphere separation, which is measured with respect to the point of contact of the surfaces, is given by [52,65]

$$d = d_{\text{piezo}} + d_0 - d_{\text{defl}}$$

d_{piezo} is the piezo movement, d_0 is the distance upon contact due to substrate and sphere roughness, and $d_{\text{defl}} = mF_{\text{pd}}$ is the cantilever deflection correction. F_{pd} is the photodiode difference signal and m the deflection coefficient. Nowadays, the smoothest surfaces have in many cases only a few nm top-to-bottom roughness. But even such a small roughness may lead to large errors in the force measurement. In fact, in modern measurements of the dispersion forces d_0 is the main source of errors [52,65,66]. The accurate amount of d_0 can be measured by fitting the electrostatic curves during electrostatic calibration of the system and combining with topography roughness measurements, e.g., by the AFM [52,65].

One way to theoretically describe the effect of surface roughness on the Casimir force is an expansion for “weakly rough” surfaces [50,51]: this assumes that deviations from an ideal geometry are small on the scale of the separation distance; it is an expansion of w/d . This is a very useful approximation since not only it is easy to estimate, but also it relies only on the assumption of the ratio w/d being small. It takes the non-additive nature of the Casimir force into account, and it does not have to rely on a pairwise additive summation by taking the statistical average. However, by itself it has turned out to be insufficient to reproduce experimental data [67]. At separations below 30–40 nm the experimental data significantly deviate from the results of this approximation. In 2012, Broer et al. [68] presented a reliable model to solve this discrepancy, which is based on a statistical analysis of AFM topography data [52,65].

1.5. Micro/Nano Electro Mechanical systems and Casimir effect

Applications of the Casimir effect varies in a wide range of domains of physics and related disciplines: from the theory of molecular forces to cosmology. One of the focuses is on fundamental physics as, for example, what the Casimir force can tell us about the quantum vacuum, and any possible relationship between zero-point energy and cosmological observations as the dark energy. In general, the evaluation of fluctuation induced phenomena on different scales (cosmological, mesoscopic, nanometric) and under different conditions (vacuum or medium assisted) is of great interest for the benefit of science in the future. New developments have been devoted to observations of the Casimir force in complex geometries and novel materials (phase-change materials, nanoparticles, carbon nanotubes, liquids, metamaterials etc.) with

Figure 1-5. An example of a MEMS in comparison with the same system in a very large scale. The same mechanism has completely different behavior only because of different size.

a view to applications, especially in nano-devices [69]. Understanding the Casimir force in a whole variety of flexibly shaped boundaries will open novel techniques in the engineering of nano-mechanical systems. Moreover, implementation of Casimir phenomena resulting in sticking and adhesion phenomena may revolutionize the engineering possibilities of micro and nanomachines [70].

Mechanical devices with dimensions in the order of tens of microns, known as MEMS, already have a pervasive presence in new technologies [69]. They are massively used as sensors, actuators, accelerometers and microswitches thanks to their fast response time and high sensitivity to external perturbations. By extending the size scale to submicron dimensions, we enter the world of the so-called NEMS. MEMS devices are not only able to measure the Casimir force very precisely, but they are also affected by the same phenomena when functioning. Experimental techniques based on recent technological developments in nanotechnology including atomic force microscopy, and MEMS devices are widely employed for the measurement of Casimir forces [70].

The vulnerability of these devices to the Casimir force and to what extent this force has to be considered in the design of MEMS/NEMS explains the importance of understanding this effect both fundamentally and from the engineering point of view. In the absence of electrostatic actuation, the Casimir force is the dominant interaction in particular when the surface to

distance ratio of the involved miniaturized bodies is very large. In other words, by the advancement in micro/nanofabrication and consequent reduction of the scale of microdevices into the submicron range, the role of Casimir and electrostatic forces could become comparable [71,72]. Although the electrostatic forces can be utilized to actuate MEMS/NEMS, the Casimir force is omnipresent and, in principle, it can always influence the actuation dynamics of operating devices leading to chaotic behavior towards stiction [73,74].

1.6. Thesis outline

The chapters of this thesis are arranged as follows. After an overview of the fundamental and practical aspects of the Casimir force in this chapter (chapter 1), chapter 2 presents the Lifshitz theory in more details for calculating the Casimir force using the optical data of the interacting surfaces as input.

Chapter 3 covers our work on Casimir forces and associated electrostatics from conductive SiC and Ru surfaces. In this chapter, we compare the results from different faces of SiC (Si- and C-terminated faces) using Ruthenium as a metallic reference. This work shows how surface characterization is of substantial importance in this field. We also provide the details of the measurement set-up with comprehensive discussions about the different behavior of two faces of SiC.

Chapter 4 deals with the thorough description of electrostatics and Casimir forces from Bi_2Se_3 films with different thicknesses, which were measured experimentally for the first time with Atomic Force Microscopy (AFM) in the sphere-plate geometry. In fact, we explored the decisive role of thickness on the Casimir force from the well-known Topological Insulator Bi_2Se_3 . Our work, as the first effort to measure the Casimir force in this complicated system, brings the attention to the effect of electrostatic forces when dealing with thin Bi_2Se_3 films.

Chapter 5 contains our theoretical studies on the non-equilibrium Casimir effect in the operation of MEMS/NEMS with input the measured optical data of the interacting material. First, we focus on how the non-equilibrium Casimir forces, which is the most general case when the interacting components have different temperatures, can influence the actuation of devices towards unstable and chaotic behavior in strong correlation with their optical properties, and associated conduction state, as well as applied electrostatic potentials. In the second part of this chapter, we present a more in-depth discussion about the uncertainty in calculating the Casimir force in the

non-equilibrium situation, which is known for more than 15 years now, due to the extrapolation of the measured optical properties at low frequencies via the Drude or the Plasma models.

1

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Chapter 2

Lifshitz theory

2.1. Abstract

Following the work of Hendrik Casimir in 1948 showing the existence of electromagnetic zero-point energy would lead to an attractive force between two uncharged perfectly conducting parallel plates, Evgeny Lifshitz in 1955 generalized this theory to the case of dielectric media and finite temperature. The Lifshitz theory is valid for arbitrary macroscopic bodies regardless of their chemical composition. It includes retardation effects and its restriction is the distances between the bodies which must be much larger than the interatomic distances. The remarkable feature of Lifshitz theory is that it expresses the force in terms of the macroscopic dielectric functions of the bodies. The theory correctly describes the interaction between two bodies due to quantum fluctuations for all separations, covering both the Casimir (long-range) and van der Waals (short-range) regimes which are the same force dominant in different separation range.

2.2. Understanding of the fundamental idea

The most general theory of the van der Waals interaction between any objects was developed by Lifshitz in 1954-1955 [1]. The theory automatically takes into account relativistic retardation effects. We can calculate the force by having the knowledge of the dielectric and magnetic permeabilities of the condensed

bodies and solving the Maxwell equations for their given configurations. One of the assumptions in Lifshitz theory is that the gap between two bodies is filled with vacuum, therefore this approach is based on the averaging of the Maxwell stress tensor in vacuum with respect to electromagnetic fluctuations in thermodynamic equilibrium.

The fundamental idea of the theory is based on the fluctuation-dissipation theorem (FDT) [2] which is an implement in statistical physics to predict the behavior of systems, and it is applicable to both classical and quantum mechanical systems. The FDT is an extensive outcome of statistical thermodynamics that quantifies the relation between the fluctuations in a system and the response of it to the applied perturbations. The FDT makes the linear relaxation response of a system in its non-equilibrium state connected to its statistical fluctuation properties in equilibrium [3]. The electromagnetic (EM) fluctuations are omnipresent inside the bodies and can be extended beyond the material boundaries. A notable example of this fundamental idea is thermal radiation. However, one must know that electromagnetic fluctuations exist even at zero temperature termed as zero-point quantum fluctuations. The sources of these fluctuations are the electric polarization $\mathbf{P}(\omega, \mathbf{r})$ or equivalently the electric current density $\mathbf{J}(\omega, \mathbf{r}) = -i\omega\mathbf{P}(\omega, \mathbf{r})$ [4]. In the case of metals, the point $\mathbf{r}$ where the density of electrons is smaller than that of its surrounding attract electrons and it changes the current in order to increase the density at $\mathbf{r}$. The correlations of the fluctuating currents are related to the dissipation in the medium according to FDT as following:

$$\langle \mathbf{J}_\alpha(\omega, \mathbf{r}) \mathbf{J}_\beta^*(\omega', \mathbf{r}') \rangle = \omega \epsilon''(\omega) \left(\frac{\hbar\omega}{2} + \frac{\hbar\omega}{e^{\hbar\omega/kT} - 1} \right) \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}') \delta_{\alpha\beta} \quad (2-1)$$

With $\alpha, \beta = x, y, z$ being the vector components. The imaginary part $\epsilon''(\omega)$ of the frequency dependent permittivity $\epsilon(\omega)$ is associated with the dissipation of the EM fields in the interacting bodies. In fact, FDT proves that the existence of dispersion forces is principally related to the dissipation in the materials of interacting bodies. The term $(\frac{\hbar\omega}{2})$ represents the zero-point energy part of the equation and $(\frac{\hbar\omega}{e^{\hbar\omega/kT} - 1})$ shows how the thermal fluctuations evolve within this theorem [5,6]. The electromagnetic field is described with Maxwell equations and by using Green function, one can express the solutions of the Maxwell equations as following [2]:

$$\mathbf{E}_\alpha(\omega, \mathbf{r}) = \frac{i}{\omega} \int d\mathbf{r}' \mathbf{G}_{\alpha\beta}(\omega, \mathbf{r}, \mathbf{r}') \mathbf{J}_\beta(\omega, \mathbf{r}'), \quad (2-2)$$

Here, $E_\alpha(\omega, \mathbf{r})$ represents the electric field and it is expressed through the components of the Green Function Tensor (GFT). Combining the equations (2.1) and (2.2) with some established properties of the GFT [7], yields the following relation for the correlation function of the electric field:

$$\langle E_\alpha(\omega, \mathbf{r}) E_\beta^*(\omega', \mathbf{r}') \rangle = 2\pi\omega\hbar \coth\left(\frac{\hbar\omega}{2kT}\right) \text{Im} G_{\alpha\beta}(\omega, \mathbf{r}, \mathbf{r}') \delta(\omega - \omega') \quad (2.3)$$

The correlation functions of the magnetic field can also be derived by applying Maxwell's equations. The Green tensor, which is the solution of the Maxwell equations, is expressed via the equation for a point charge:

$$\left[\partial_\alpha \partial_\beta - \delta_{\alpha\beta} \left(\nabla^2 + \frac{\omega^2}{c^2} \varepsilon(\omega, \mathbf{r}) \right) \right] G_{\alpha\gamma}(\omega, \mathbf{r}, \mathbf{r}') = 4\pi \frac{\omega^2}{c^2} \delta_{\alpha\beta} \delta(\mathbf{r} - \mathbf{r}') \quad (2.4)$$

In Equation (2.4), $\varepsilon(\omega, \mathbf{r})$ expresses the dielectric function of the involved materials. In other words, the force between the surfaces of the objects can be described by the normal component of the averaged Maxwell stress tensor via the correlation functions (Equation 2.3), or via the Green function of the system (Equation 2.4). Due to the symmetry around the x and y axes, only the component of the Maxwell stress tensors perpendicular to the surfaces needs to be considered. The real and imaginary frequency presentations are formulated as the following:

2.3. Real-frequency description

The explicit form of the Green functions can be effortlessly derived for two parallel plates, which are interacting via long wavelength fluctuations. The simplest configuration contained two semi spaces of different materials identified by the dielectric functions $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$, separated by a small gap, which is filled with the material characterized by the dielectric function $\varepsilon_0(\omega)$. For parallel plates, the force acting on each plate is calculated via the Green functions. The final result for the force (per unit area) can be formulated as

$$F(T, z) = \frac{1}{2} \frac{\hbar}{\pi^2} \int_0^\infty d\omega \coth\left(\frac{\hbar\omega}{2kT}\right) \text{Re} \int_0^\infty dq q |k_0| g(\mathbf{q}, \omega) \quad (2.5)$$

where the wave vector in the gap is $K = (\mathbf{q}, k_0)$ with $\mathbf{q}$ being the in-plane vector, and the $k_0 = (\varepsilon_0\omega^2/c^2 - \mathbf{q}^2)^{1/2}$ component. The function $g(\mathbf{q}, \omega)$ is given by

$$g(\mathbf{q}, \omega) = \sum_{v=s,p} \frac{r_1^v r_2^v e^{2ik_0 z}}{1 - r_1^v r_2^v e^{2ik_0 z}} \quad (2.6)$$

The coefficients $r_{1,2}^v$ are the Fresnel reflection coefficients (see Equation 2.7) for the inner surfaces of the plates 1 and 2. In this format, $v=s$ and $v=p$ denote the transverse electric (TE) and transverse magnetic (TM) polarization, respectively. The factor $g(\mathbf{q}, \omega)$ describes multiple reflections from the inner surfaces of the bodies 1 and 2. The frequency dependent factor $\coth(\hbar\omega/2kT)$ emerges from the FDT. Finally, the Fresnel reflection coefficients are given by

$$r_i^s = \frac{k_0 - k_i}{k_0 + k_i}, \quad r_i^p = \frac{\varepsilon_i k_0 - \varepsilon_0 k_i}{\varepsilon_i k_0 + \varepsilon_0 k_i} \quad (2.7)$$

With:

$$k_0 = \sqrt{\varepsilon_0(\omega) \frac{\omega^2}{c^2} - \mathbf{q}^2}, \quad k_i = \sqrt{\varepsilon_i(\omega) \frac{\omega^2}{c^2} - \mathbf{q}^2}. \quad (2.8)$$

The real frequency representation of Equation 2.5 is inoperable for force calculations because the integrand is a fast-oscillating function due to the factor $e^{ik_0 z}$. However, there are cases where the real frequency representation is required to be considered as for instance, when the involved bodies have different temperatures leading to non-equilibrium situations [6,8,9].

2.4. Imaginary-frequency description

The fast oscillations problem in Equation 2.5 is normally avoided by the contour rotation in the frequency complex plane. As the result of this mechanism, only the poles of the function $\coth(\hbar\omega/kT)$ contribute to the integral. These poles are positioned at following frequencies:

$$\omega_n = i\zeta_n = i \frac{2\pi n k T}{\hbar}, \quad n = 0, 1, 2, \dots \quad (2.9)$$

where ζ represents the imaginary part of frequency, which is known as the *Matsubara* frequency. After the transformation, the Lifshitz formula is given the form of:

$$F(T, z) = \frac{kT}{\pi} \sum_{n=0}^{\infty} {}' \int_0^{\infty} d\mathbf{q} \, \mathbf{q} \, |k_0| \sum_{v=s,p} \frac{r_1^v r_2^v e^{2ik_0 z}}{1 - r_1^v r_2^v e^{2ik_0 z}} \quad (2.10)$$

where prime signifies that the term corresponding term $n=0$ must be multiplied with a factor of 1/2. Contrary to the real frequency representation, here the quantities $k_0 = \sqrt{\varepsilon_0(i\zeta)(\zeta^2/c^2 + \mathbf{q}^2)}$ and $g(\mathbf{q}, i\zeta_n) = \sum_{v=s,p} (r_1^v r_2^v e^{2ik_0 z} / 1 - r_1^v r_2^v e^{2ik_0 z})$ no longer oscillate as a function of frequency. According to the

relation $\coth(\hbar\omega/kT) = 1 + 2/[\exp(\hbar\omega/kT) - 1]$, only the first term on the right side stays with the limit $T \rightarrow 0$, associated with the zero-point energy. At separation distance much shorter than λ_T (i.e., in this study $d = 200$ nm), the contribution of thermal fluctuations becomes negligible. In this fashion, the Matsubara frequency can be considered as a continuous variable, and the sum in Equation 2.10 transforms to an integral if we substitute $\frac{kT}{\pi} \sum_{n=0}^{\infty} \rightarrow \frac{\hbar}{2\pi^2} \int_0^{\infty} d\zeta$. Consequently, Equation 2.10 takes the form:

$$F(z) = \frac{\hbar}{2\pi^2} \int_0^{\infty} d\zeta \int_0^{\infty} d\mathbf{q} \, \mathbf{q} \, k_0 \sum_{v=s,p} \frac{r_1^v r_2^v e^{-2k_0 z}}{1 - r_1^v r_2^v e^{-2k_0 z}} \quad (2.11)$$

The reflection coefficients in this case depend only on the dielectric functions at imaginary frequencies $\varepsilon(i\zeta)$. It is not feasible to directly measure these functions, however, they can be modulated via the measurable function $\varepsilon''(\omega)$ through the Kramers–Kronig relation [8,10,11],

$$\varepsilon(i\zeta) = 1 + \frac{2}{\pi} \int_0^{\infty} \frac{\omega \varepsilon''(\omega)}{\omega^2 + \zeta^2} \quad (2.12)$$

The function $\varepsilon(i\zeta)$ is real and positive as its general characteristics, and it decreases with increasing ζ . It also indicates that the dispersion forces are utterly regulated by $\varepsilon''(\omega)$, which is responsible for the dissipation in the material. As it is aforementioned, in order to calculate the force with the Lifshitz formula, one has to know the dielectric function of the material at imaginary frequencies $\varepsilon(i\zeta)$, which is calculated via $\varepsilon''(\omega)$ according to Equation 2.12. Next section is devoted to this problem.

2.5. Different approaches for extrapolation of dielectric function

The experimental data available for $\varepsilon''(\omega)$ are always bounded by low and high frequencies. The low-frequency cut-off ω_{cut} is specifically crucial in the case of metals, such as Au, which shows substantial absorption owing to conduction charge carriers in the infrared range. As a result, $\varepsilon''(\omega)$ is large in metals at low frequencies which contribute remarkably to $\varepsilon(i\zeta)$. Hence, a critical step in the evaluation of $\varepsilon(i\zeta)$ is extrapolation of the dielectric function $\varepsilon''(\omega)$ to low frequencies $\omega < \omega_{cut}$, where the experimental data are not attainable. One of the most established method which can solve this problem is the Drude model. This model defines the dielectric function in low optical frequencies as:

$$\epsilon(\omega)_D = \epsilon_0 - \frac{\omega_p^2}{\omega(\omega + i\omega_\tau)}. \quad (2.13)$$

The second term in Equation 2.13 is determined by the plasma frequency ω_p , and the relaxation frequency ω_τ . The ratio ω_p^2/ω_τ is a manifestation of static conductivity (for $\omega \rightarrow 0$) in the material [8,10,11]. The Drude model is normally used for extrapolating in the low optical frequency regime $0 < \omega < \omega_1$ ($= 0.03$ eV). In the high optical frequency range $\omega > \omega_2$, which is vital only at separations smaller than 10 nm, the imaginary part of the permittivity is extrapolated as an inverse power law $\epsilon''(\omega) \sim 1/\omega^3$. Consequently, for frequencies $\omega < \omega_1$ and $\omega > \omega_2$, $\epsilon''(\omega)$ is extrapolated as below:

$$\begin{aligned} \omega < \omega_1 (= 0.03 \text{ eV}): \epsilon''(\omega) &= \frac{\omega_p^2 \omega_\tau}{\omega(\omega^2 + \omega_\tau^2)}, \\ \omega > \omega_2 (= 9.34 \text{ eV}): \epsilon''(\omega) &= \frac{A}{\omega^3} \end{aligned} \quad (2.14)$$

where A is found to match the value of $\epsilon''(\omega)$ at $\omega = \omega_2$ between experimental data and the extrapolation. Using the Drude model, the dielectric function $\epsilon(i\zeta)$ in all frequencies can be formulated as:

$$\epsilon(i\zeta) = 1 + \frac{2}{\pi} \int_{\omega_1}^{\omega_2} \frac{\omega \epsilon''_{\text{exp}}(\omega)}{\omega^2 + \zeta^2} d\omega + \Delta_L \epsilon(i\zeta) + \Delta_H \epsilon(i\zeta) \quad (2.15)$$

With,

$$\Delta_L \epsilon(i\zeta) = \frac{2}{\pi} \int_0^{\omega_1} \frac{\omega \epsilon''_L(\omega)}{\omega^2 + \zeta^2} d\omega, \quad \Delta_H \epsilon(i\zeta) = \frac{2}{\pi} \int_{\omega_2}^{\infty} \frac{\omega \epsilon''_H(\omega)}{\omega^2 + \zeta^2} d\omega \quad (2.16)$$

The integrals of the extrapolations can also be developed analytically:

$$\Delta_H \epsilon(i\zeta) = \frac{2}{\pi} \int_{\omega_2}^{\infty} \frac{\omega \epsilon''(\omega)}{\omega^2 + \zeta^2} d\omega = \frac{2\omega_2^3 \epsilon''(\omega_2)}{\pi \zeta^2} \left[\frac{1}{\omega_2} - \frac{\frac{\pi}{2} \arctan(\omega_2/\zeta)}{\zeta} \right] \quad (2.17)$$

$$\Delta_L \epsilon(i\zeta) = \frac{2}{\pi} \int_0^{\omega_1} \frac{\omega \epsilon''(\omega)}{\omega^2 + \zeta^2} d\omega = \frac{2\omega_p^2 \omega_\tau}{\pi(\zeta^2 - \omega_\tau^2)} \left[\frac{\arctan(\omega_1/\omega_\tau)}{\omega_\tau} - \frac{\arctan(\omega_1/\zeta)}{\zeta} \right] \quad (2.18)$$

However, the Drude model, which is used to calculate the Casimir force through the Lifshitz theory, accompanies to deviations from experimental force results. This discussion remains an open problem up to today in Casimir physics [12–14]. This issue for metals is very well addressed by the dissipation-less plasma model (having infinite absorption at the frequency $\omega = 0$ and zero anywhere else) at separations above 160 nm [8,10,11]. The plasma model formula is straightforward to apply. At low optical frequencies

$\omega < \omega_1$, the term $\Delta_L \varepsilon(i\zeta)$ from the Drude model (Equation (2.15)) is replaced by ω_p^2/ζ^2 , yielding:

$$\varepsilon(i\zeta)_P = 1 + \frac{2}{\pi} \int_{\omega_1}^{\omega_2} \frac{\omega \varepsilon''_{\text{exp}}(\omega)}{\omega^2 + \zeta^2} d\omega + \frac{\omega_p^2}{\zeta^2} + \Delta_H \varepsilon(i\zeta) \quad (2.19)$$

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Chapter 3

Casimir forces and electrostatics from conductive SiC-Si/C and Ru surfaces

3

3.1. Abstract

Comprehensive knowledge of Casimir forces and associated electrostatics from conductive SiC and Ru surfaces can be essential in diverse areas ranging from micro/nanodevice operation in harsh environments to multilayer coatings in advanced lithography technologies. Hence, the Casimir force was measured between an Au-coated microsphere and N-doped SiC samples with Si- and C-terminated faces, and the results were compared with the measurements using the same microsphere and a metallic Ruthenium surface. Electrostatic calibration showed that the Si- and C-faces behave differently with a nearly $\sim 0.6\text{--}0.7\text{ V}$ difference in the contact potentials $V_0^{\text{Si/C}}$. We attribute this to a higher incorporation of N on the C-terminated face in the near surface region resulting in the formation of NO_x and an increased work function compared to the Si-terminated surface which is in agreement with x-ray photoelectron spectroscopy data. Notably, the contact potential of the SiC-C face ($V_0^{\text{C}} \sim 0.1\text{ V}$) was closer to the metallic Ru-Au system ($V_0^{\text{Ru}} \sim 0.05\text{ V}$). However, the measured optical properties of the SiC-Si/C terminated surfaces with ellipsometry did not show any substantial differences indicating that the effective depth of the Si/C terminating surface layers are significantly smaller than the photon penetration depth not leading to any differences in the calculated forces via Lifshitz theory. Nonetheless, the

measured Casimir forces, after compensation of the electrostatics contributions, showed differences between the Si/C faces, whereas the comparison with the Lifshitz theory prediction shows better agreement for the SiC-Si face. Finally, comparison of the Casimir forces below 40 nm separations between the SiC-Si/C and Ru surfaces indicated that the short-range roughness effects on the Casimir force increase in magnitude with increasing metallic behavior of the plate surface. Therefore, not only the material optical properties but also the conductive state and roughness of the surface layers must be carefully taken into account in short range Casimir interactions between more complex dielectric materials.

3.2. Introduction

Nowadays, the Casimir force that originates from the perturbation of electromagnetic vacuum fluctuations is still a topic of relentless research [1–10], though its proposition in 1948 by the Dutch physicist Hendrik Casimir almost dates back 70 years [2]. The interest stems from a multitude of research fields ranging from fundamental physics in search of new forces beyond the standard model to micro/nanodevices for technology applications [1,3–6]. Lifshitz and co-workers in the 1950s [7,8] considered the general case of flat dielectric plates by exploiting the fluctuation-dissipation theorem, which relates the dissipative properties of the plates (due to optical absorption by many microscopic dipoles) and the resulting electromagnetic (EM) fluctuations. The theory describes the attractive interaction due to quantum fluctuations for all separations covering both the Casimir (long-range) and van der Waals (short-range) regimes [1,3–9]. As devices enter the submicron range within the realm of nanoelectromechanical systems, a deep understanding of Casimir forces at nanoscale separations between real materials is inevitable.

The dependence of the Casimir force on the type of material is an important topic since in principle one can tailor the force by a suitable choice of materials [6,8,10–21]. In this sense, silicon-based semiconductors appear as a promising choice since the materials properties can be modified and controlled allowing also the tuning of the Casimir interactions [6,21]. Different doping and charge carrier densities in semiconductors can change the Casimir force [10]. However, in microelectromechanical systems applications for industry, e.g. automotive and space technologies [22–24], the micro-sensors are required to operate in harsh environments. Because the latter can be a challenge for Si-based sensing devices, silicon carbide (SiC) is considered an

excellent substitute for Si due to its outstanding properties if attributes such as high durability combined with high stiffness and low thermal expansion are necessary. SiC is currently also utilized for precise instrumentation frames and mirrors, and additionally, there is the possibility for usage in macro/nano assembly technologies via direct (optical) bonding concepts [25–27]. In addition, because SiC exhibits high hardness, chemical inertness, and ability to survive operation at high temperatures and harsh environments, it is well suited to be used as protective coating of micro-machined parts.

SiC is a material that exhibits strong polytypism. All polytypes have identical planar arrangement, while their difference lies in the stacking of the planes so that the difference in the stacking periodicity of similar planes results in different types of polytypes (≥ 250). The relatively low residual stress level in the layers, the high stiffness, and the excellent etch-stop properties allow even the fabrication of free standing SiC microstructures via standard Si bulk micro-machining techniques [25–27]. The terminating layers of SiC can be either Si or C and that might alter the associated Casimir interaction for this material in potential applications, taking also the associated electrostatic characteristics (e.g. contact potentials) into consideration. Although Casimir force measurements on Si-terminated nitrogen doped SiC have been performed in the sphere-plate geometry, which gave reasonable agreement with predictions of the Lifshitz theory [28], the effect of the surface termination layer has remained so far unexplored.

Therefore, measurements and analysis of the Casimir forces at short ranges (< 100 nm) from both Si/C faces are necessary to gauge the effect of the surface termination on both the Casimir force and the associated surface electrostatics. In order to minimize charging and electrostatic effects, since SiC is insulating, our measurements were performed on highly doped conductive SiC samples. Moreover, any roughness on the termination layers of the SiC and/or on the colloid probe can have significant influence on the Casimir forces, as studies for metal coatings [29–34] have shown, whereas its magnitude in relation to the conductivity of a dielectric system still remains unexplored. In fact, increasing the minimum separation between interacting surfaces from zero, for an ideal smooth system, to d_0 (distance upon contact due to roughness) for a rough system between the mean average planes of the interacting surfaces could prevent the Casimir force to induce stiction in actuating systems [30,35–37]. On the other hand, in direct bonding applications, the strong Casimir force between relatively smooth surfaces ($d_0 < 2\text{--}3$ nm) would be highly beneficial [16]. The aforementioned brought us to investigate also the effect of surface roughness from the different faces of SiC on

the Casimir force, and compare the forces to those of thin film ruthenium (Ru) surfaces with comparable smoothness to that of SiC. The chosen Ru surfaces, which are relatively resistant to oxidation and are heavily used in multilayer coatings for mirrors in advanced lithography technologies [21], allow one to evaluate the relative effect of material conductivity changes at short interaction ranges, where the roughness effects are manifested as deviations from the Lifshitz theory predictions for flat surfaces.

3.3. Optical characterization of SiC and Ru samples

Conductive nitrogen (N)-doped SiC with distinguished Si and C terminated faces were provided from Norstel AB fabricated using the hot-wall technique. The Ru samples were obtained from the Industrial Focus Group XUV Optics, University of Twente, The Netherlands. Because the Casimir force is strongly influenced by the optical properties of the interacting bodies [38,39] we measured the optical properties of all the samples under study. Therefore, we performed Ellipsometry measurements in a wide range of frequencies using the J.A Woollam Co., Inc. Ellipsometers VUV-VASE (0.5–9.34 eV) and IR-VASE (0.03–0.5 eV) at three different incident angles (55°, 65°, 75°) for SiC, and (70°, 75°, 80°) for Ru with respect to the sample surface. The optical data were analyzed as in [40], and subsequently the frequency dependent dielectric function $\epsilon(\omega)$ was obtained. The latter allows calculation of the dielectric function $\epsilon(i\zeta)$ at imaginary frequencies ζ (see Figure 3-1 and Equation (2-15), which is the necessary input for the Casimir force predictions via the Lifshitz theory.

Moreover, for the Casimir force calculations using the Lifshitz theory in the frequency range that is not covered by the experimental optical data, the Drude model was used to extrapolate at low frequencies (see Equation (2-11)) [41]. After fitting the optical data, the Drude parameters, namely, the plasma and relaxation frequencies (ω_p , ω_τ) were (0.138 ± 0.008 eV, 0.074 ± 0.001 eV) for the SiC-Si face, (0.156 ± 0.008 eV, 0.078 ± 0.001 eV) for the SiC-C face, and (5.98 ± 0.07 eV, 0.09 ± 0.002 eV) for the Ru, respectively. From the plasma frequency ω_p we can estimate the concentration of conduction carriers $N_e = m^* \omega_p^2 / 4\pi e^2$ in SiC, if the effective carrier mass m^* is close to the mass of the free electrons. The estimated N_e for SiC-Si, SiC-C and Ru were ~ 1.4 (which is similar 32ort h value for the SiC samples in our previous studies [28]), $\sim 1.8 \times 10^{19} \text{ cm}^{-3}$ and $\sim 2.6 \times 10^{22} \text{ cm}^{-3}$, respectively. The phonon polariton absorption peak for SiC, which is due to the absorption of

Figure 3-1. Imaginary Dielectric function of SiC-Si, SiC-C and Ru at imaginary frequencies, which were calculated using the Drude model to extrapolate at low frequencies. The zoomed-in area shows that there is a small difference in the calculated dielectric function for the SiC-Si/C terminated layers. The inset shows the imaginary part of the frequency dependent dielectric function.

infrared light by transverse optical phonon modes, was slightly higher for the SiC-Si face as compared to that of the SiC-C face. However, the difference between the dielectric function of the SiC-Si/C surfaces is negligible (see Figure 3.1) implying that the calculated Casimir force via Lifshitz theory will not show any significant variation. This is because the penetration depth (skin effect) of the photons during ellipsometry measurements, which give the averaged value for $\epsilon(\omega)$ within this depth, is not sensitive enough to capture the influence of the Si/C surface termination layers.

3.4. Direct and inverse atomic force microscopy morphology measurement

The morphology of the surfaces of all samples was measured using a Bruker Multimode 8 atomic force microscope (AFM) operated in tapping mode to minimize the surface damage by the AFM tip. Figure 3-2 shows the AFM topography of the SiC-Si/C and Ru surfaces. The root-mean-square (rms) roughness was measured over a scan area of $1 \times 1 \mu\text{m}^2$, which is comparable to the effective Casimir force interaction area $\sim \pi dR$ in the sphere-plate geometry (for sphere radius $R \sim 10 \mu\text{m}$ and separations $d < 100 \text{ nm}$). The topography analysis yielded for the SiC-Si, SiC-C, and Ru surfaces $w_{\text{SiC-Si}} = 0.22 \text{ nm}$, $w_{\text{SiC-C}} = 1.02 \text{ nm}$, and $w_{\text{Ru}} = 0.45 \text{ nm}$, respectively. Therefore, the surfaces of

Figure 3-2. The topography (3D height images) of (a) Ruthenium, (b) SiC-C face, (c) SiC-Si face and (d) the Au coated sphere. The inverse AFM image of the sphere shows a repeated pattern since the same contact area of the sphere is scanned by multiple sharp tips. © Height distribution of the three samples which yields from its positive width the maximum contribution $d_{0,pl}$ to the separation upon contact. The C face is only chemically polished and contains several scratches. (f) Height distribution of the sphere as obtained by inverse AFM.

all samples are smooth enough ($w \leq 1$ nm) to give only limited contribution to the separation upon contact between the sample and sphere surfaces, which is mainly limited by the roughness of the sphere.

Furthermore, in order to measure the Casimir force in the sphere-plane geometry, a borosilicate sphere with radius $R = 10.1 \pm 0.6 \mu\text{m}$ was glued to the end of a tipless cantilever. The spheres were coated with Au using a Cressington 208 HR Sputter Coater. The sample stage in the coater can be rotated allowing coating of the spheres with homogenous thickness Au films which ensure electrical contact with the rest of the cantilever. The latter is necessary for the electrostatic calibration of the force measuring system (see

Appendix 3). The topography of the sphere is imaged using the so-called reverse AFM (see Figure 3-2), where the sphere is scanned on top of a grid with inverted sharp tips (TGT1 grating from NT-MDT Spectrum Instruments, <https://www.ntmdt-si.com/>). This was performed in order to obtain topography information of the sphere surface as close as possible to the real interaction area between sphere-plate during the force measurements.

Indeed, as it is shown in Figure 3.2, the sphere is considerably rougher than the planar SiC and Ru samples. Hence, the roughness of the sphere yields the dominant contribution to limit the force measurement at separations $d_0 \approx d_{0\text{sph}} + d_{0\text{plt}}$ [16] with $d_{0\text{sph}}$ and $d_{0\text{plt}}$ the maximum positive full widths of the highest peaks in the height histograms (negative widths in the histograms correspond to valleys) (e.g. see Figure 3.2 for the sphere) for both the sphere surfaces and planar samples, respectively. These estimations yielded a maximum distance upon contact d_0 for the Au/SiC-Si, Au/SiC-C, and Au/Ru interacting surfaces with values 30.8 ± 2 nm, 31.2 ± 2.1 nm and 35 ± 2.3 nm, respectively. However, because $d_{0\text{sph}} \gg d_{0\text{plt}}$, the actual separation distance is more likely $d_0 \approx d_{0\text{sph}} + \omega_{\text{plt}}$ with ω_{plt} the rms surface roughness of the sample surfaces. The latter yields for the actual distance upon contact d_0 between surfaces the values 30.2 nm, 31.0 nm and 30.5 nm for the Au/SiC-Si, Au/SiC-C, and Au/Ru systems, respectively.

3.5. Electrostatic and XPS-analysis of material surfaces

The measurement of the electrostatic force between the sphere and the plate is crucial for the precise measurement of the actual Casimir force. This is because one obtains from these measurements the contact potential V_0 between the two interacting bodies, and more accurate values for the cantilever spring constant than those obtained from noise measurements (see Appendix 3) [30]. The knowledge of V_0 is necessary in order to apply potential compensation during the force measurements and minimize the electrostatic contributions on the measured force besides that of the genuine Casimir force. Hence, in order to obtain the contact potential, we applied various voltages V between the sphere and plate in the range $[-3 \text{ V}, 3 \text{ V}]$. The potential was applied to the sample with the tip grounded, while the tests with inverse polarity gave similar results. From the measurements of the electrostatic forces at larger separations (≥ 100 nm), where the Casimir force plays a negligible role (by comparing to Casimir force predictions using Lifshitz theory for the size of the spheres used for the force measurements),

we assigned the contact potential V_o to the minimum of the cantilever deflection versus applied voltage V .

The contact potential V_o (typically <1 V) always exists between two dissimilar conductive materials (even between the same material prepared under different conditions), which are in thermal equilibrium, due to differences in work functions [31]. From the electrostatic force measurements in Figure 3.3, we obtained a considerable difference between the measured contact potential V_o for the Au/SiC-Si ($V_o^{\text{Si}} \sim -0.7$ to -0.8 V) and Au/SiC-C ($V_o^{\text{C}} \sim -0.1$ V) interacting surfaces. For V_o^{Si} similar values were also found in our previous studies of Si-terminated (with comparable doping) SiC samples [28]. The measured contact potential for Au/Ru is $V_o^{\text{Ru}} = 0.05$ V in our setup. The contact potential difference $\Delta V_o = V_o^{\text{Si}} - V_o^{\text{C}} = (\sim 0.6-0.7$ V) between the Si/C-faces is significant even when taking into account that the C-face is relatively rougher than the Si face, and only chemically polished. However, besides trapped charges and any patch potentials (areas with different surface potential) caused by roughness, the electrostatic difference ΔV_o between the two SiC faces has to be attributed to other reasons.

Since the contact potential is linked to the work function difference between the two materials involved for the contact potential measurement and the work function consists of a bulk and surface part [42], the obtained contact potential difference between the Si- and C-face of SiC can be mainly explained by the difference of the status of the respective surface. In other words, because the bulk part of the work function is the same for the Si- and C-face the surface part of the work function must differ. According to the measured contact potential values, the surface part of the work function of the Si-terminated surface is by $0.6-0.7$ V lower than the one with the C-terminated surface. This has to be related to differences in the chemical environment of the atoms present at the surface (and perhaps subsurface region) as well as the concentration of the types of atoms at the surface and subsurface region. Importantly, one has to consider that the above measurements were conducted under ambient conditions. This implies that oxidation of the surface needs to be taken into account when looking for an adequate explanation of the obtained data.

To obtain information on the types of different chemical species and their relative amounts present at the (sub)surface regions at the Si and C faces of SiC and with this to present an explanation of the observed contact potential differences, we conducted x-ray photoelectron spectroscopy (XPS) measurements (Figure 3-4, see also in Appendix 3 Fig. A.3.1 and Tables A.3.1 and A.3.2). The XPS data show that the chemical environment for C, N, O and

Figure 3-3. Cantilever deflection versus applied voltage in the range $[-3\text{V}, 3\text{V}]$. The contact potential V_0 is assigned at the minimum of the cantilever deflection: (a) Au/SiC-C, and (b) Au/SiC-Si systems. The different curves correspond to different sphere-plate separations in the range $\geq 100\text{ nm}$ where the Casimir force has negligible contribution. The lines are only a guide for the eye.

Si is different on the Si- and C-faces. Besides the expected higher amount of C and Si on the C and Si faces, respectively, the XPS results indicate that the amount of N (the dopant of the SiC samples) as well as O (due to oxidation at ambient conditions) is noticeably higher on the C face (see Appendix 3 Fig. A.3.1 and Tables A.3.1 and A.3.2).

It should be noted that only a relative comparison of the amount of N and O on the two different sample surfaces is possible and that we did not perform an absolute quantification. The higher amount of nitrogen is based on the model that N prefers to occupy a C site because of the similarities in

Figure 3-4. Comparison of XPS spectra for the Si- and C-face of SiC. (a-c) Si 2p, (d-f) C 1s, (g-i) N 1s and (j-l) O 1s core level spectra. The peak assignment is detailed in Table SM2 in the Appendix 3.

the covalent radii between the C and N atoms (covalent radius: Si: 0.110 nm, C: 0.077 nm, and N: 0.075 nm) [43]. Moreover, Si-N bonds are energetically more favorable than C-N ones so that N should rather incorporate in the C planes within SiC making more Si-N bonds compared to Si-C ones. The higher amount of oxygen means that the oxidation of the C face of 4H SiC proceeds much easier and faster compared to the Si face [33]. This can be on the one side explained by the difference in electronegativity of Si and C atoms when bound with O atoms (e.g. difference in Pauling electronegativity is $\Delta X_{\text{C-Si}} = X_{\text{C}} - X_{\text{Si}} \approx 0.65$) [44]. On the other side – and that is most likely the more relevant factor for the stronger oxidation of the C face – the increased presence of N on the C face will result in a higher amount of NO_x compared to the Si face and thus, an overall higher amount of oxidation of the C face.

From previous investigations for adsorption of NO on a metal surface [44], it turned out that the work function increases compared to the bare metal surface. This is assigned to the dipole moment of NO (N being positively charged and O negatively) and the fact that NO binds with its N atom adsorbed on the metal surface. Transferring this knowledge to our system leads to the conclusion that a larger amount of NO_x at the surface should also result in a higher work function, which is indeed what we obtained from the contact potential measurements.

Figure 3-5. (a) Height and (b) Potential profiles of the SiC-Si face from KPFM measurements. The EFM images of the SiC-Si face are also shown with an applied potential of 2 volts (c) and -0.5 volts (d) between the sample and the tip.

The electrostatic findings for the contact potential of SiC generated further interest to perform a microscopic analysis of the SiC surfaces via Electric and Kelvin Probe Force Microscopy (EFM / KPFM) as shown in Figure 3-5 [34]. Using EFM, one can measure electric field gradient distributions above the sample surface. It is also used to identify trapped charges and more importantly provide an electric polarization map of the sample surface.

For EFM measurements the phase shift signal ($\sim$ electric field gradient) is usually measured [35]. On the other hand, KPFM shows the local variation of the work function of materials, and it has been also previously used for this purpose in Casimir research [46,47]. However, KPFM images can change with adsorption layers like water, oxide layers, electrostatic charges, dopant concentration, and/or surface dipole moments [35]. Hence, we measured with KPFM the surface potential distribution over a scan area of $1 \times 1 \mu\text{m}^2$ for the three different samples (Figure 3-5a). For the SiC-C and Ru samples, the potential scale of the KPFM measurements did not match with the measured contact potential V_0 from the electrostatic calibration, while the

SiC-Si face shows a relatively comparable scale variation with the measured . Moreover, after performing EFM measurements, the SiC-Si face responded to the applied bias potential (Figure 3-5b). In fact, by changing the applied DC bias potential from 2 V to -0.5 V bright regions became darker and vice versa indicating the presence of surface charges. The EFM and KPFM results show that the surface of the SiC-Si face is electrically different from those of the SiC-C and Ru samples (see in Appendix 3 Fig. A.3.2) deviating from a metallic behavior, as it was also concluded from the obtained contact potentials via electrostatic calibration.

3.6. Casimir force analysis

In order to calculate the force from experimentally obtained cantilever deflection data, the electrostatic calibration [30] of the cantilever-sphere spring constant was performed, which yielded $k = 1.83 \pm 0.02$ N/m (see Appendix 3) [36]. Moreover, the contribution of the separation dependent repulsive hydrodynamic drag force in the thin gap between sphere-plate surfaces is negligible. Indeed, this force is given by the expression [37] $F_h(z) = -(6\pi\mu R^2/z)(dz/dt)f^*$, where μ is the dynamic viscosity of the surrounding fluid (e.g., $\mu \approx 1.983$ kg/ms for air at $T = 300$ K), dz/dt is the velocity of the sphere, and f^* is the correction for the deviation from standard Reynolds flow due to fluid slip of the interacting surfaces. Assuming that no slip occurs ($f^* = 1$), then the piezo approach/retraction speed of $dz/dt = 300$ nm/s yields a repulsive hydrodynamic force $F_h \approx 3.2 \times 10^{-7}$ nN at short separations (e.g., $z \approx d_0 \approx 35$ nm). The latter is negligible in comparison to Casimir forces of the order of pN to nN at the separations of interest ≤ 100 nm.

Furthermore, the Casimir force curves from both the theoretical calculation and the experimental data for the different samples are presented in Figure 3-6. The relative thermal correction at $T = 300$ K for a separation below 100 nm can be neglected, so that we can use the convenient integral representation of the Lifshitz formula (see Equation (2-11) to calculate the Casimir force [48]. Moreover, for completeness, we performed also force calculations via the Lifshitz theory using a variation of the dielectric function for the Au coating of the sphere within its maximum limits by considering the handbook data with the maximum plasma frequency of 9.0 eV. As it is shown (see in Appendix 3 Fig. A.3.3) the force difference for the Au-SiC system is negligible to play any role. One can also consider the plasma model to extrapolate at low frequencies (see Equation (2-19)) since this an unresolved

issue for more than 15 years in the Casimir field, e.g., a signature of either an inconsistency in the Lifshitz theory or a contribution of electrostatic surface potentials [46–48]. However, the results, at the short separations we probe (< 100 nm), will not change more than the variation we considered above for the Au coating of the spherical probe since at short separations (< 100 nm) the difference between Drude and Plasma models is not significant [49]. Furthermore, for a clear comparison between the experimental results and the Lifshitz theory calculation, we also calculated the relative force error $|F_{\text{exp}} - F_{\text{Lif}}|/F_{\text{exp}}$. The cantilever deflection was translated to a Casimir force using the electrostatically determined spring constant $k = 1.83$ N/m, and for the Lifshitz theory calculations we used the measured optical data of SiC-Si/C and Ru from Fig. 1, as well as those for Au from previous measurements [38].

The maximum absolute force difference is up to $\sim 60\%$ between the force measured at the closest distance $d_0 \sim 30$ – 35 nm, and the Lifshitz theory for Ru. The measured deviation is due to the sphere roughness contribution to the Casimir force at separations below 40 nm, where the high surface peaks lead to a rapid increase of the force, as it was observed in the past for the rough Au-Au systems [15,20,29]. Strong deviation was also observed for the SiC-C face, for which at the shortest separation the deviation due to roughness is $\sim 40\%$, while that for the SiC-Si face is about ~ 10 – 20% . The roughness contribution appears to be of less importance as the conductivity of the probing sample reduces (since the sphere was the same for all surfaces), taking also into account that the SiC-Si/C and Ru surfaces have low rms surface roughness ($w \leq 1$ nm) over the effective Casimir force interaction area of $\sim 1 \times 1 \mu\text{m}^2$. Beyond 40 nm separations, the force data and Lifshitz theory data deviate only within a range of 10% .

Beyond the separation regime, where surface roughness enhances the Casimir force (< 40 nm), one can describe the dependence of the Casimir force with an average power-law behavior $F_c \sim z^{-m}$ having an exponent $m < 3$ for the sphere-plate geometry. In fact, the obtained values for the exponents from the force data (and also from theory) were for the Ru, SiC-Si and SiC-C system 2.74 (theory: 2.53), 2.77 (theory: 2.70), and 2.65 (theory: 2.69), respectively. These values are in good agreement with the results of previous studies on Si-terminated SiC [28,40] and other surfaces [39] that yielded $m < 3$ for the sphere-plate geometry over a diverse variety of interacting systems, and separation ranges $< 1 \mu\text{m}$.

From the power-law behavior one can estimate the relative force error which occurs because of both the uncertainty in the actual surface separation or the roughness contribution in d_0 , and the error for the measured spring

Figure 3-6. The experimental Casimir force data together with the Lifshitz theory calculation in the separation 30-100 nm. (a) Ru, (b) SiC-Si/C faces. (c) Relative force error comparing the experimental Casimir force data with Lifshitz theory calculations using the measured optical data for the three samples shown in Figure 3.1.

constant k . The estimated relative error for the Casimir force was calculated as $\Delta F_c/F_c \approx [(\Delta k/k)^2 + (m \Delta z/z)^2]^{1/2}$. The latter is roughly $\sim 5\%$ for all three samples at the separation $z = 100$ nm and decreases with increasing separation for each sample. For the smoothest sample (SiC-Si face) at the shortest separation of $z = 30.2 \pm 1.8$ nm, the relative error is $\Delta F_c/F_c \sim 15\%$ (see also e.g.,

error bars in Figure 3-6b). For the other surfaces, the relative estimated error at the shortest separations was also comparable. In any case, the estimated errors are significantly less than the force variation due to surface roughness in Figure 3-6c, which can reach a level close to ~60 % for the most conductive surface at shortest separations of ~30 nm.

The comparison of the force data obtained with Lifshitz theory is only indicative due to the following limitations. First of all, the difference in charge carrier density for the two different faces of SiC, which was distinctly shown by electrostatic measurements was not translated into differences of the force calculated with the Lifshitz theory. This is because the ellipsometry measurement of the optical properties is not sensitive enough to variations of the dielectric permittivity of the different terminated surface layers. Similar interesting observations have been made in the past for other systems, where the surface layers were different from the bulk of the samples [50].

Second, comparing the experimental data with the theoretical prediction shows that there is a considerable deviation from the Lifshitz theory for flat surfaces at short separations (< 40 nm). The reason for this discrepancy is due to the considerable effect of roughness on the Casimir force due to the high surface peaks [20,29], which becomes more pronounced for material systems with increasing surface conductivity (Figure 3-6c). The surface roughness also has a strong effect in determining the actual separation distance between the interacting bodies [16,29]. The experimental data for Ru appear to be noisier at larger separations as compared to both SiC samples. The latter can be attributed to possible degradation of the probe since all force measurements for the three samples were performed by one individual probe.

3.6. Conclusion

In summary, the comprehensive knowledge of the Casimir force as well as the associated electrostatic characteristics from differently terminated conductive SiC and Ru surfaces can be essential in diverse micro/nanotechnologies for operation in harsh environments and the design of advanced coatings in optics. Therefore, we performed Casimir force measurements between the same Au-coated microsphere and N doped SiC with both Si-terminated and C-terminated faces and compared the results to relatively flat and metallic Ru surfaces. Electrostatic calibration measurements showed a ~0.6–0.7 V difference in the contact potential V_0 between SiC-Au for the two faces of SiC. We attributed this to the higher incorporation of N and subsequently

the formation of NO_x on the C face which was confirmed by XPS measurements. On the other hand, the measured optical properties of the SiC-Si/C terminated surfaces with ellipsometry did not show any substantial differences indicating that the effective depth of the different Si/C surface layers is significantly smaller than the skin depth prohibiting as a result any substantial differences to appear in force calculations via Lifshitz theory.

However, the Casimir force measurement after minimization of electrostatics contributions showed differences between the Si/C faces, while the comparison with the Lifshitz theory calculations show better agreement for the SiC-Si face. In addition, short-range force measurements at separations less than 40 nm were mainly limited by the surface roughness of the sphere. In fact, the comparison of the Casimir forces below 40 nm between the SiC-Si/C and Ru surfaces indicates that the short-range roughness effects increase in magnitude with increasing metallic behavior of the sample surface. Although comparisons with Lifshitz theory are limited for the samples under considerations, our results indicate that surface layers and surface roughness must be taken into consideration in Casimir and electrostatic force measurements as dimensions and surface separations decrease from the micro to the nano regimes.

Appendix 3

XPS data analysis:

The x-ray photoelectron spectroscopy (XPS) measurements were performed in an ultrahigh vacuum system equipped with a preparation and analysis chamber containing an Al_{Kα}/Mg_{Kα} twin-anode x-ray gun and a hemispherical energy analyzer (Thermo Fisher). Al_{Kα} x-rays with a photon energy of 1461.6 eV were used for the XPS measurements.

Figure A.3.1 XPS survey spectra for the Si- and C-face of SiC. In addition to the expected elements from the SiC samples (Si, C, N), additional peaks from the stainless-steel holder (e.g., the peaks at ~576 eV, which belong to Cr 2p), from oxidation of Si (O 1s at around 530 eV) and/or surface plasmons (e.g., the peaks at ~122 eV, ~174 eV for Si) are present.

Table A.3.1 Chemical composition of the SiC samples obtained from the spectra in Figure SM1.

Sample/Element (at%)	Si	C	O	N
SiC Si-face	67.53	23.61	7.66	0.58
SiC C-face	37.51	40.85	17.21	2.65

Table A.3.2 Peak position, full width at half maximum (FWHM) and area are summarized for the spectra shown in Figure 4 in the main part.

	Peak label	C-face			Si-face		
		Pos (eV)	FWHM (eV)	Area (%)	Pos (eV)	FWHM (eV)	Area (%)
C 1s	I. (C-Si)	283.40	1.67	18.86	283.40	1.54	47.12
	II. (C-C/C-H)	285.78	1.84	69.88	285.41	1.81	45.34
	III. (C-Si _x O _y)	287.53	2	7.17	286.76	1.68	4.34
	IV. (C-N _x)	289.53	1.88	4.09	288.61	2.43	3.20
N 1s	I. (N-Si/N-H/N-C)	398.62	3.75	44.48	398.69	3.51	67.58
	II. (NO _x)	403.51	5.81	55.52	403.24	5.69	32.42
O 1s	I. (O-Si _x)	531.13	2.64	24.49	531.18	2.29	31.04
	II. (O _x -N/OH ⁺ /H ₂ O)	533.00	2.64	75.51	533.44	2.33	68.96
Si 2p	I. (Si-Si/Si-H/Si-N)	101.18	2.27	83.10	100.96	2.16	96.98
	II. (Si-O _x)	103.30	2.26	16.90	103.16	1.85	3.02

From the XPS results we draw the following tentative conclusions:

1. The amount of Si on the Si-face is significantly higher as compared to the C-face as expected. The same holds for the amount of carbon which is much higher on the C-face as compared to the Si-face.
2. The amount of nitrogen on the C-face is higher as compared to the Si-face. This is in line with literature reports [43,51] which reason that the incorporation of N in the C-face is preferred due to the similar covalent radii of carbon and nitrogen as well as the fact that the formation of Si-N bonds is energetically preferred over C-N ones, i. e. when N incorporates into the C-face it will form more Si-N bonds.
3. The amount of oxygen on the C-face is – from a relative point of view – higher as compared to the Si-face, i. e. oxidation proceeds faster/easier on the C-face and the oxidation of the more abundant nitrogen atoms on the C-face (see Figure 4 (g-i)) results in an increased amount of NO_x. Furthermore, nitrogen bonded to carbon could contribute to the higher binding energy peak at around 403 eV in the N 1s spectrum. However, from the C 1s data for the C-N peak at around 289.5 eV we can conclude that this is only to a very limited extent the case.

4. Due to the limited resolution, we have in performing XPS measurements with our lab-based setup, it should be noted that the peaks having a FWHM > 2 eV will most likely consist of more than peak. That means chemically different species will contribute to the N 1s and O 1s peaks but we are not able to resolve them and thus, we list different possibilities to explain one peak position.

AFM and KPFM data for SiC and Ru surface:

Figure A.3.2 AFM topography (left) and potential from KPFM images (right) for (a) the SiC-C surface, and (b) the Ru surface.

Electrostatic calibration of the cantilever spring constant:

In order to calculate the force from experimentally obtained cantilever deflection data, the electrostatic calibration of the cantilever-sphere spring constant was performed. Initially, the Au coated sphere-cantilever yielded a spring constant $k = 1.93 \pm 0.21$ N/nm from noise measurements (thermal tuning) [36]. This is only an indication of the expected spring constant within ~ 10 % error. The spring constant can also be determined more accurately (with

error < 5 %) after performing electrostatic force measurements from an Au test sample and fitting the data with the parabolic equation

$$\Delta_t = \left(\frac{X(z)}{k} \right) (V - V_0)^2 + A(z) \text{ with } X(z) = -2\pi\epsilon_0 \sum_{m=0}^7 A_m t^{m-1}. \quad (1)$$

In Eq. (SM3.1) the parameter Δt is the total deflection of the cantilever, $A(z)$ represents a deflection offset, $t = z/R$ with z the surface separation and R the sphere radius, and the coefficients from A_0 to A_7 have the values 0.5, -1.1826, 22.2375, -571.366, 9592.45, -90200.5, 383084 and -300357, respectively [43]. The fitting parameters yielded for the spring constant of the probe $k = 1.83 \pm 0.02$ N/m, and an offset $A(z)$ of 1.120 ± 0.001 nm.

Different extrapolation results:

As it is shown below in Figure A.3.3, the force difference for the Au-SiC system with different extrapolation is negligible to be considered. Using the plasma model to extrapolate at low frequencies (see Equation (2-19)), which is an unresolved issue for many years in the Casimir physics, is another signature of either an inconsistency in the Lifshitz theory or a contribution of electrostatic surface potentials. Making a solid conclusion requires further studies on both issues [46–48]. However, the results, at the short separations we investigate here in this study (<100 nm), does not change more than the variation we considered above for the Au coating of the spherical probe. The reason is the difference between Drude and Plasma models is not significant at short separations (<100 nm) [49].

Figure A.3.3 shows the Casimir force calculation for the Au-SiC system via the Lifshitz theory using a variation of the dielectric function for the Au coating of the sphere within its maximum limits by considering the handbook data (solid line) with the maximum plasma frequency of 9.0 eV. These are compared with the measured dielectric function of Au, sample 3 in ref [38] (dash line).

Figure A.3.3 Casimir force calculation for the Au-SiC system via the Lifshitz theory using a variation of the dielectric function for the Au coating of the sphere within its maximum limits by considering the handbook data (solid line) with the maximum plasma frequency of 9.0 eV. These are compared with the measured dielectric function of Au, sample 3 in ref [38] (dash line). The inset shows the absolute difference of the calculated force between the two cases, which is negligible for our SiC – Au system.

The work described in this chapter has been published as:

Z. Babamahdi, V. B. Svetovoy, M. Enache, M. Stöhr, and G. Palasantzas, Comparison of Casimir Forces and Electrostatics from Conductive SiC-Si/C and Ru Surfaces, Phys. Rev. B **100**, (2019).

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Chapter 4

Casimir effect from Topological Insulator (Bi_2Se_3)

4

4.1. Abstract

We explored here the decisive role of film thickness on Casimir and electrostatic forces from topological insulating Bi_2Se_3 films. The forces were measured for the Au- Bi_2Se_3 system under ambient conditions to simulate realistic applications. It is shown that the electrostatic forces associated with a non-zero contact potential of $V_0 \sim 200$ mV played a dominant role, and could not be compensated for thinner films (≤ 20 nm thick). However, for thicker films the measured force, after voltage compensation, started to approach the genuine Casimir force in very good agreement with Lifshitz theory. Our results point out that any gradual deviation from theory for thinner films underlies the presence of strong electrostatic effects.

4.2. Topological Insulators

The first discovered, topologically distinct state is the quantum Hall state in which the conductivity perpendicular to the bias direction was found to change in quantized steps [1]. Thouless and Kosterlitz in the early 1970s explained this observation theoretically by connecting it to the state's topology, being different from that of its environment (vacuum or air) [2]. Their work

led to the understanding of such systems in which an ordering is omnipresent that cannot be disturbed by thermal fluctuations. Therefore, a crossover state at the edge of the quantum Hall state can be expected to match the difference in topology. Later on, the explanation of electrons traveling via skipping orbits along this edge fitted to the bigger picture of topological classification that predicts such an edge state to be present. Interest in this novel class of materials has grown dramatically since the discovery of two-dimensional TI (Hg-Te) [3]. In this system, the presented edge states do not carry a charge current, but do carry a net spin current. These edge states originate from required crossover states at the interface of the topologically distinct phases. In these states, the spin and momentum of the carriers are directly related and interestingly the generated currents flow dissipationless.

Afterwards, various theoretical groups found out that topological band structures can be present in ordinary three-dimensional systems, this paved the way towards topological insulators. It was theoretically proposed that binary compounds with heavy elements Sb, Se, Te, and Bi could give rise to three-dimensional topological insulators where the one-dimensional edge states are converted into two-dimensional surface states. Such surface states have been experimentally observed in the binary compounds Bi₂Se₃, Bi₂Te₃, and Sb₂Te₃ [3–9] by angular-resolved photoemission spectroscopy (ARPS). In the binary compounds, the topological surface states can be addressed independently from the bulk since these states are located inside the bulk band gap. Furthermore, the charge carriers have their momentum connected to their spin similar to the quantum spin Hall state, which is known as ‘spin–momentum locking’. This property is protected by topology and thus cannot be disturbed by any perturbation that still preserves time-reversal symmetry.

4.3. Three-dimensional topological Insulators

The group of topological insulators of interest in this thesis belongs to the class of (V)–(VI)-semiconductors in which (V)-elements Sb and Bi and (VI)-elements Se and Te are bonded with a stoichiometry (V)₂(VI)₃. The arrangement of this family of compounds is in a rhombohedral crystal structure. The super unit cell of the crystal structure of these compounds consists of 15 atomic layers (~3 nm) with inverted-repeating quintuple layer basic units (~1 nm) that are separated by a Van der Waals gap (VdW gap). The basic unit of five atomic layers within a supercell, termed as a quintuple layer (QL) has a height

Figure 4.1. (a) Rhombohedral unit cell of Bi_2Se_3 . The red box indicates one QL. (b) The top view of the unit cell along the z direction. A, B and C sites indicate the possible locations each atom can occupy. (c) The sideview of the unit cell and the stacking order of Bi and Se atomic layers [10].

of roughly 1 nm. The basic units are stacked as $(\text{VI})^{\text{B}}-(\text{V})^{\text{C}}-(\text{VI})^{\text{A}}-(\text{V})^{\text{B}}-(\text{VI})^{\text{C}}$ where A, B, and C correspond to the lattice sites that the atoms can occupy, see Figure 4.1.

The first experimentally discovered 3D topological insulator was $\text{Bi}_{1-x}\text{Sb}_x$. The surface states of this 3D topological insulator have been shown using surface-sensitive techniques such as angular-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy (STM). The protection against backscattering of charge carriers in the topological surface states has been observed using STM measurements [11]. However, the small band gap and the complicated surface state structure found for this compound limit room-temperature applications since the bulk will contribute to the charge transport in this material easily and masking of the surface states will occur [12]. Soon after, the other 3D topological insulators Sb_2Te_3 , Bi_2Te_3 and Bi_2Se_3 were experimentally explored. In this thesis, we studied the Casimir effect on Bi_2Se_3 . It has been reported that the compound Bi_2Se_3 usually suffers from Se vacancies that act as electron donors to the system [13–15]. Moreover, antisite defects, i.e. atoms exchanging position in the crystal lattice contribute to the n-type character [14,15]. Growing in Bi-rich or Se-rich conditions can

lead to lower charge-carrier densities [16,17], but will always lead to n-type Bi_2Se_3 where the Fermi level is usually located in the bulk conduction band, yielding an intermixing of bulk and surface states in non-surface-sensitive (transport) investigations.

Growth of Bi_2Se_3 on $\text{Al}_2\text{O}_3(0001)$ having a mismatch of $\sim 14\%$ has a few advantages: the chemical stability of the substrate is noticeable, the substrate can withstand high temperatures (without having any reactions with the grown material), and it is insulating. Nonetheless, it is not recommended to grow directly on this substrate at higher temperatures, because of the poor sticking coefficient of Bi and Se to the substrate at such high temperatures. Hence, a two-temperature process has to be adopted in which the substrate is normally passivated by a monolayer of Se which was grown at low temperature. Afterwards, 3 QL of Bi_2Se_3 is grown as the seed layer at the same temperature. In due course, the temperature is increased to escalate the surface mobility of the atoms and therefore improve the Bi_2Se_3 growth. It has been observed that triangular features taking shape in this sort of growth are noticeably larger in feature size [18]. This indicates the high quality of these films and can be connected to the lower density of nucleation sites owing to the inertness of Al_2O_3 . For this type of growth, it has been observed that the grain boundaries in the first QL can affect the charge carrier density although such defects are not proceeded in the further subsequent layer due to the Van der Waals-nature growth of these class of materials. These interfacial defects have been found to introduce a significant amount of charge carriers and pin the mobility due to the induced disorder [19]. The disorder is especially detectable in charge transport measurements on films with thicknesses below 100 nm. Besides, it has been reported that Se diffusion through the material is involved in the thickness-dependent charge-transport properties [20], and more importantly, casts doubts on the origin of the thickness-independent transport channels, as reported earlier [18,19].

4.4. Casimir force and three-dimensional topological Insulators

Understanding the Casimir force between conductive materials provides accessibility to micrometer and sub-micrometer range physics, which has been of strong theoretical and experimental interest for the last two decades [21–24]. On the other hand, in recent years three dimensional topological insulators (TI), possessing a conductive surface and an insulator

bulk with surface states located within the bulk band gap, have been studied massively [25–27]. Although these challenging systems are not yet thoroughly understood, they provide specific properties that can be highly promising for future micro/nanoelectromechanical systems [28,29]. Therefore, a natural question to ask is how the Casimir force, which is omnipresent between condensed media, would manifest itself in these novel materials as they interact with other materials. So far, there has been a number of theoretical works predicting the Casimir force exerted by TIs [30,31]. In fact, it was predicted only theoretically that the Casimir force can be tuned and, in some cases becomes repulsive, by controlling the sign of the topological magnetoelectric polarizability (TMEP) and the distance between the surfaces [32]. Moreover, how a TI would influence the Casimir force, taking into account possible electrostatic contributions in the nanoscale vicinity of a metal surface, still remains unexplored. Hence, the absence of Casimir experiments with TIs does not allow to draw meaningful conclusions about how this intriguing class of materials interacts with other materials. Indeed, it has been observed both theoretically and experimentally that metals in the proximity of TIs give rise to potential variations, which are of great importance for the preservation of the topological surface state [33–35]. Consequently, and in view of future applications, experimental studies are necessary to shed light not only on the electrostatics of the TI surface when approaching a metal, but also more interestingly on the associated Casimir force [9, 10].

Therefore, we investigated Casimir forces from thin films of Bi_2Se_3 , a well-known 3D topological insulator, when they are exposed to ambient conditions leading to non-ideal behavior, something which is very likely to occur in applications. Our measurements show that when the Bi_2Se_3 surface is approaching an Au surface, it acts as a poor conductor most likely due to its surface oxidation upon exposure to air as surface analysis using X-ray photoelectron spectroscopy (see Figure 4.4 and Table 4.1 and 4.2) indicated. This is an important insight on how to balance electrostatic effects in sub-micron devices made of such materials, although under ideal circumstances 3D TIs have surface conductivity [28]. Accordingly, due to surface oxidation upon exposure to air the studied system cannot be considered as a perfect TI with a conductive surface caused by the surface states. More interestingly, our study revealed that the measured Casimir force is dependent on the thickness of the Bi_2Se_3 film resulting in a dramatic deviation from Lifshitz theory predictions when the thickness of Bi_2Se_3 is as low as 10 nm. Consequently, we measured the forces for Bi_2Se_3 samples with different thicknesses of 10, 20, 30 and 100 quintuple layers (QLs where the thickness of each QL ≈ 1 nm).

The contact potential for the Au/Bi₂Se₃ system was $V_o \approx -200$ mV for all samples with different Bi₂Se₃ thicknesses. Moreover, repetitive measurements showed that V_o remained constant and stable over a period of six months. Figure 4.2 shows the three sets of measurements plotted together to stress the consistency of the contact potential independent of the age of the samples. In addition, the electrostatic measurements yielded similar results at different locations on the surface of the Bi₂Se₃ samples (for all thicknesses studied). However, translating the measured contact potential V_o between the two surfaces into the work function difference between them is not trivial, since the work function difference between materials always consists of a bulk contribution, a surface contribution, and an image charge contribution. In equilibrium, very small surface charges can build up a voltage of the order of the work function difference [37].

The Bi₂Se₃ films were fabricated by Molecular Beam Epitaxy (MBE) on Al₂O₃(0001) substrates in a custom-designed SVTA MOS-V-2 MBE system at Rutgers University (USA) following the methods as described in [36]. Furthermore, in order to measure the contact potential V_o , which is the electrostatic potential existing between electrically conductive materials due to work function differences and trapped surface charges, we performed electrostatic calibration using Atomic Force Microscopy (AFM) in contact mode. The knowledge of V_o is of crucial importance, since it should be applied as the compensation voltage to suppress electrostatic contributions which in turn enables a more precise measurement of the genuine Casimir force. In the force setup, the plate is the Bi₂Se₃ film, and the spherical probe was made by attaching a borosilicate sphere with a 10 μ m radius at the end of a tipless cantilever. The process was followed by coating the probe with 60 nm Au using a Cressington 208 HR sputter coater. The Au layer is sufficiently thick to be optically opaque (infrared skin depth for Au is ~ 22 nm) and simultaneously has sufficiently low roughness to allow short range force measurements. By applying various voltages in the range $[-1.5$ to 1.5 V] to the plate making direct electrical contact with Ag paste to the surface and the sphere being grounded, we can estimate the minimum deflection of the cantilever corresponding to the contact potential V_o between Au and Bi₂Se₃ (see Fig. 4.2).

In any case, to better interpret the measured contact potential, the chemical composition of the surface of Bi₂Se₃, and the work function of the sample were studied by performing X-ray Photoelectron Spectroscopy (XPS) and Ultraviolet Photoelectron Spectroscopy (UPS), respectively (see Fig. 4.3). As reference sample, Au (111) was used ($E_F = 26.26$ eV). The measured work

Figure 4.2. Cantilever deflection versus applied voltage in the range $[-1.5 \text{ V}, 1.5 \text{ V}]$. Different colors correspond to different sets of measurement, and within the same color different data points at a specific voltage correspond to different separations (more than 100 nm) between the interacting surfaces, where the electrostatic force is the dominant force and the Casimir effect can be neglected. The minimum of all the data points meet at -200 mV which is considered as the contact potential between Bi_2Se_3 and Au.

Figure 4.3. (1) Oxygen 1s core level spectra for the (a) Bi sample, (b) Se sample and (c) Bi_2Se_3 sample (film thickness 10 nm). Bi and Se samples are used as a reference. The O 1s envelopes look different for all the samples. The peak positions for the Se sample are shifted towards higher binding energies compared to the other two samples. (2) (a) UPS spectra (He I, photon energy 21.2 eV) for the Bi_2Se_3 sample (film thickness 10 nm) to determine the work function (sample was biased with -5 V). The black arrow shows the secondary electron-cut off (SECO). (b) UPS spectra for the Au (III) single crystal which was used as a reference. The measurement conditions were the same as for the Bi_2Se_3 sample.

function of Bi_2Se_3 using UPS (see Fig. 4.3(2)) yielded a value of 3.42 eV, which is smaller than those reported in previous studies with values of 5.40 eV [38]

Figure 4.4. Survey spectra of Bi sample (black), Se sample (red) and Bi₂Se₃ sample (blue). The spectra are plotted with an offset for clarity. The black arrows point to the main core levels.

Table 4.1. Chemical composition of each sample as obtained from the survey spectra (Figure 4.4)

Sample/Element (at%)	O	C	Bi	Se
Bi sample	18.43	41.76	39.80	--
Se sample	8.98	44.76	--	46.26
Bi ₂ Se ₃ sample	14.19	31.27	22.01	32.52

Table 4.2. Peak positions, FWHM and area for O 1s core levels for the Bi sample, Se sample, and Bi₂Se₃ sample.

O 1s	Bi sample				Se sample			Bi ₂ Se ₃ sample		
	Peak label	Pos (eV)	FWHM (eV)	Area (%)	Pos (eV)	FWHM (eV)	Area (%)	Pos (eV)	FWHM (eV)	Area (%)
	I.	530.54	2.19	38.09	532.42	2.08	62.26	530.94	1.94	36.43
	II.	531.83	1.93	26.45	533.79	2.23	37.74	532.68	2.09	50.63
	III.	533.03	2.11	35.53	--	--	--	534.31	2.12	12.95

and 5.60 eV [39]. Nevertheless, a decrease of the work function of Bi_2Se_3 with 1.4 eV due to air exposure was previously reported [38], which is in good agreement with our measurement. Figure 4.3(1) shows the O 1s core level spectrum of the 10 nm thick Bi_2Se_3 sample, which was exposed to air.

The XPS measurements were performed in an ultrahigh vacuum system. For the XPS measurements a Mg K_{α} source was used with an excitation energy of 1254 eV. A pass energy of 50 eV, and an energy step of 0.05 eV was used for all the acquired spectra. All the spectra were calibrated with respect to the C 1s peak position which was set to 284.8 eV. A Thermo Scientific setup was used for the XPS/UPS measurements which is equipped with an Alpha 110 analyzer, a Vacuum Generator (VG) twin Mg/Al anode x-ray source and a VG HA030005 ultraviolet source. The survey spectra of the samples are plotted in Figure 4.4 and Table 4.1 and 4.2 consist of the details of the XPS measurements on the 10 nm Bi_2Se_3 sample.

Table 4.1 and 4.2 show the chemical compositions of the samples and the Peak positions, FWHM and area for O 1s core levels for the studied samples.

Moving on to the surface status of the samples, Figure 4.5 shows the surface topography of the 100 nm thick Bi_2Se_3 sample (measured by AFM in TM mode, Bruker Multimode 8), where the triangular features indicate the high-quality growth of the Bi_2Se_3 films. The topography of the thinner Bi_2Se_3 samples is also shown in Figure 4.5. Using also reverse AFM (with a dedicated TGT1 grating from NT-MDT spectrum instruments), we can measure the morphology of the sphere surface that interacts with the Bi_2Se_3 surfaces during force measurements. From the topography of the involved surfaces the root-mean-square (rms) roughness and the height of peaks can be derived, which yield the actual distance between surfaces. The measured distance upon contact d_0 , which is strongly affected by the roughness of the sphere, can be estimated as $d_0 \approx d_{0sph} + \omega_{plt}$, where d_{0sph} is the maximum positive full width of the height distribution and ω_{plt} is the rms surface roughness of the Bi_2Se_3 surface (see Figure 4.5) [40]. We obtained for $d_0 = 28 \pm 1.7$ nm, 30 ± 2.1 nm, 27.5 ± 1.8 nm, and 28 ± 2.3 nm for the 10, 20, 30, and 100 nm $\text{Bi}_2\text{Se}_3/\text{Au}$ systems, respectively.

Furthermore, the optical properties of the interacting materials are necessary as an input for the Casimir force calculations via the Lifshitz theory. Therefore, the dielectric functions of the interacting bodies were measured by ellipsometry in a wide frequency range (corresponding to wavelengths from 140 nm to 33 μm) at J.A Woollam Co. Inc (USA) (see Figure 4.6). The UV-VASE and IR-VASE measurements were performed at incidence angles of 50° and 70° with respect to the surface of the samples.

Figure 4.5. Topography of the surface of the Bi_2Se_3 samples with thicknesses (a) 10 nm, (b) 20 nm, (c) 30 nm and (d) 100 nm. The rms roughness of the films are 0.61, 0.53, 0.77 and 2.85 nm for 10 nm, 20 nm, 30 nm and 100 nm Bi_2Se_3 samples, respectively. The histogram of one of the spheres (e) used in the measurements shows that the height of the peaks is significantly larger than that of the samples supporting our considerations for the distance upon contact .

Since the thickness of the Bi_2Se_3 films are in the range 10, 20, 30 and 100 nm, the optical response contribution from the substrate is not negligible. For this reason, we performed the same optical measurements for an Al_2O_3 substrate similar to that used for the film growth. The approach to analyse the optical data and calculate the Casimir force, is to consider the Bi_2Se_3 samples as a two layer system where the contribution of the substrate is taken into

Figure 4.6. Imaginary part of the frequency dependent dielectric function for Bi_2Se_3 and the substrate Al_2O_3 . The peak at (1.96 eV) for Bi_2Se_3 is associated with transitions from the valence band (v) to the conduction band (c) and the strong peaks in Al_2O_3 are the phonon polaritons absorption peaks. The inset shows the calculated dielectric function of Bi_2Se_3 and Al_2O_3 at imaginary frequencies, which is the necessary input for the calculation of the Casimir force with Lifshitz theory.

4

account [41]. Accordingly, we extracted the data corresponding to Bi_2Se_3 from the raw ellipsometry data using the WVASE® software by performing multiple fitting steps (point by point and then multiple Gaussian dispersion relations, where the final fits for the whole spectrum are shown in Figure 4.7. The calculated dielectric functions at imaginary frequencies $\epsilon(i\zeta)$ for both the Bi_2Se_3 and Al_2O_3 , which are the necessary inputs in Lifshitz theory, are illustrated in the inset of Figure 4.6. Since the measured optical data do not cover the whole frequency range $[0, \infty]$ that is used in the Lifshitz theory, we have extrapolated the data at low frequencies using the Drude model for Au. Notably, since we do not see Drude tail neither for Bi_2Se_3 nor Al_2O_3 , we can consider the $\Delta_L \epsilon(i\zeta)$ in Equation (2-15) to be zero. We have been used the approach of a double layer system explained in [41] to calculate the force between Au and thin Bi_2Se_3 film on a Al_2O_3 substrate.

Ellipsometry measurement parameters of Ψ and Δ were collected over a wide spectral region (130 nm–33 μm) for a single Sapphire substrate and for Bi_2Se_3 on Sapphire sample. Variable angle measurements were collected to reduce parameter correlation during fitting. An optical model consisting of two-layers representing the optical dispersion relations of our samples have been constructed for the data fitting, one for the Sapphire substrate

Figure 4.7. Data fitting in order to extract the optical data corresponding to Bi_2Se_3 which were grown on the Al_2O_3 substrate. (a) Psi and (b) Delta vs the photon energy for the measured and fitted data. (c) and (d) are representing the zoomed data in the low-frequency region to show the good fit, and (e) indicates the index of refraction and extinction coefficient values vs photon energy for Bi_2Se_3 .

and another for the Bi_2Se_3 thin film on top. For the substrate, the dielectric constants of sapphire from the measured sapphire sample (from Woollam) are used. A very good fit is shown here with $\text{MSE} = 4.16$.

In order to compare the theoretical force calculation with the experimental force measurement, the knowledge of the spring constant of the cantilever-sphere system is necessary which can be also obtained from the electrostatic calibration of the system [42]. The values of the spring constant k for the probes used for the force measurements of the Bi_2Se_3 samples with thicknesses 10, 20, 30 and 100 nm were $k = 1.62 \pm 0.03$, 1.48 ± 0.007 , 1.58 ± 0.04 and 1.89 ± 0.02 N/m, respectively. Moreover, the separation dependent hydrodynamic drag force has a negligible effect as compared to the Casimir force for the separations of interest between the interacting surfaces (< 100 nm) [42,43]. In addition, the thermal correction at $T = 300$ K can also be neglected for separations < 100 nm. The calculated and measured Casimir forces for the different thicknesses of Bi_2Se_3 are shown in Figure 4.8, where for consistency both the 10 and 100 nm thick Bi_2Se_3 samples were characterized optically and the data were used for the force calculations. From Figure 4.8, it is evident that there is a significant discrepancy between theory and experiment, especially for the case of the 10 nm thick Bi_2Se_3 sample. As the film thickness increases, the measured force data approach the theoretical prediction as it is the case for the 100 nm thick Bi_2Se_3 . The strong deviations at the shorter separations (< 30 nm) can be explained by the roughness effect on the Casimir force due to the high peaks [44].

Figure 4.8. Experimental Casimir force data together with the Lifshitz theory calculation for separations below 100 nm for different thicknesses of Bi₂Se₃. The experimental data for the shortest separation are restricted by surface roughness leading to a deviation from the power law behavior. The lower inset shows the variation of the power law exponent vs. film thickness.

The deviation of the force measurements from theory for thinner films is related to electrostatic contributions, which cannot be nullified by applying the compensating voltage. Therefore, as the film thickness decreases, we observe a clear trend going from the genuine Casimir force to a state where the force is dramatically influenced by electrostatics. Beyond the shortest separations where the roughness plays a significant role (> 30 nm), the force curves show a power law behaviour $F_c \sim z^{-m}$ with exponent $m < 3$, which is anticipated for the sphere-plate geometry in the separation range $< 1 \mu\text{m}$. The obtained power law values were $m \approx 1.87, 2.03, 2.6$ and 2.68 for the 10, 20, 30, and 100 nm Bi₂Se₃/Au interacting systems, respectively (inset also Figure 4.4). The value of the exponent from the Casimir force calculations is $m \approx 2.7$ for all the Au / Bi₂Se₃ systems, which is close to the experimental values for Bi₂Se₃ samples with film thicknesses ≥ 30 nm. For thinner films, the exponents start to approach values closer to that for electrostatic dominating forces (with $\sim z^{-1}$ in the sphere-plane geometry) [24,42,45].

One would expect for thinner Bi₂Se₃ films that the conductive properties of the surface will be more prominent, but the force measurements rather show the opposite trend.

This behavior can be explained by an increase of the electron density and a decrease of the electron mobility as the thickness decreases [46]. This implies that the electrical properties of Bi_2Se_3 are thickness-dependent and consist of bulk and surface-interface contributions. The latter contribution diminishes as the film thickness increases, becoming negligible for thick films like 100 nm [46]. The charge transport in the surface-interface layer can lead to accumulation of electrons and the formation of a kind of patch potential effect on the surface [47]. Our analysis indicates that the effect of electrostatics from TIs could vary with film thickness and having a more significant signature for thinner films, though more detailed analysis is necessary. This is because surface-interface states become more dominant for thinner Bi_2Se_3 films. Instead of being conductive, they are insulating which is probably due to surface oxidation.

In conclusion, we have explored the role of film thickness on Casimir and electrostatic forces for the topological insulator Bi_2Se_3 . The measurements were done under ambient conditions to simulate realistic conditions in applications. In fact, the electrostatic forces associated with a non-zero contact potential V_o played a dominant role and could not be compensated for the thinner films (< 20 nm), while above 30 nm thickness the obtained force after voltage compensation was closer to the genuine Casimir force. Hence, the measurements for the thickest film (100 nm) showed very good agreement with Lifshitz theory predictions, while there is a trend of deviation for thinner films underlying the significance of strong electrostatic effects.

The work described in this chapter has been published as:

Z. Babamahdi, V. B. Svetovoy, D. T. Yimam, B. J. Kooi, T. Banerjee, J. Moon, S. Oh, M. Enache, M. Stöhr, and G. Palasantzas, *Casimir and Electrostatic Forces from Bi₂Se₃ Thin Films of Varying Thickness*, Phys. Rev. B **103**, L161102 (2021).

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Chapter 5

Non-equilibrium Casimir effect in MEMs and NEMs

5.1. Abstract

Micro and nanoelectromechanical systems have attracted widespread attention from the scientific and technology point of view for sensor technologies, accelerometers, microswitches etc. Because Casimir forces are omnipresent, they could always influence the actuation of devices and under certain conditions can draw moving elements together into permanent adhesion, which is a phenomenon termed as stiction. As a result, the design of devices can be challenging due to the occurrence of chaotic behavior driven by the Casimir forces, which it can cause abrupt changes in their dynamical behavior leading to possible stiction. Therefore, detailed knowledge about the phenomena of stiction due to Casimir interactions, and its relation to chaotic motion during actuation is necessary in order to improve the long-term performance of devices. It is shown here that the non-equilibrium Casimir forces, which are the most general case when the interacting components have different temperatures, can influence the actuation of devices towards unstable and chaotic behavior in strong correlation with their optical properties, and associated conduction state, as well as applied electrostatic potentials. The situation becomes more complicated by the uncertainty in calculating the Casimir, which is known for more than 15 years now, due to the extrapolation of the measured optical properties at low frequencies via the Drude

or the Plasma models. The latter models can predict either enhanced or suppressed chaotic behavior depending strongly also on the conductivity of the interacting materials.

1) Dependence of non-equilibrium Casimir forces on material optical properties towards chaotic motion during device actuation

We have considered the effect of Casimir forces in microswitches, which are essential MEMS/NEMS components. These systems are typically constructed from two electrodes of which one is fixed, and the other is suspended by a mechanical spring governed by the Hooke's law [1]. The application of a bias voltage between the electrodes actuates them towards each other, but it is also possible that the moving component to become unstable and collapse (pulls-in) onto the other [1,2]. The design of MEMS could always be quite challenging due to the occurrence of chaotic behavior, which causes abrupt changes in their dynamical behavior, and as a result device malfunctions related to possible stiction of moving components. Obviously, more detailed knowledge about the phenomena of stiction, and its relation to chaos is necessary to improve the long-term performance, and design of MEMS devices operating under thermal equilibrium and/or non-equilibrium conditions [3–8]. Despite of several studies, which have been performed on effect of optical properties on the actuation dynamic of microswitches [4,6], it still remains unexplored how significant is the influence of thermal non-equilibrium conditions on the performance microdevices in view also of recent advances to measure thermal effects with MEMS [8]. Indeed, besides the optical properties of material coatings on device components, when MEMS operate under ambient conditions additional factors like thermal non-equilibrium conditions, due to temperature differences between the interacting bodies, can influence the Casimir forces [7]. Consequently, the dynamical behavior of actuating components can also be affected with unknown long-term effects. Therefore, here we aim to explore to what extent thermal non-equilibrium conditions influence the actuation dynamics of microswitches towards chaos, which prohibits the long-term prediction of stable device operation. This will be accomplished by taking also into account the optical properties of different materials ranging from poor to good conductors in order to compare how significant is the choice of materials, and any applied electrostatic potentials that lead to additional attractive interactions.

5.2. Optical properties of materials and device actuation

In order to make realistic the results of this research in understanding the thermal effects on the actuation of a wide range of microdevices, we considered the effect of optical properties for gold (Au) as a good conductor, and highly nitrogen-doped silicon carbide (SiC) as a poor conductor. In fact, Au as a good conductor is widely used for Casimir force measurements, while highly doped with nitrogen (N) conductive SiC is a promising material for device operating under harsh environments allowing also integration with Si based technologies. Both materials were optically characterized in the past with the same equipment at J. A. Woollam Co., Inc. using the ellipsometers VUV-VASE (0.5–9.34 eV and IR-VASE (0.03–0.5 eV) [9,10]. Figure 5-1 shows the real and the imaginary parts of the measured dielectric functions of these materials. For comparison, after analysis of the optical data, the static conductivity ratio ω_p^2/ω_τ , in terms of the Drude model with ω_p the plasma frequency and ω_τ the relaxation frequency (see Appendix A), obtained for these materials the values $\omega_p^2/\omega_\tau|_{\text{SiC}} = 0.4 \text{ eV}$ [31], and $\omega_p^2/\omega_\tau|_{\text{Au}} \approx 1600 \text{ eV}$ [9,10]. The latter indicates a conductivity contrast of almost four orders of magnitude to justify these materials as suitable candidates for our analysis.

The equation of motion for the electromechanical system (inset Fig. 5-1(a)), where both the fixed and movable plates are considered to be coated with same material (Au or SiC), is given by

$$M \frac{d^2 z}{dt^2} + \left(\frac{M\omega_0}{Q} \right) \frac{dz}{dt} = -F_{\text{res}} + F_{\text{elec}} + F_{\text{Cas}} + \varepsilon F^\circ \cos(\omega t), \quad (1)$$

where M is the mass of the movable plate, and $(M\omega_0/Q) (dz/dt)$ is intrinsic energy dissipation in the actuating system associated with a quality factor Q . The frequency ω_0 is assumed to be that of dynamic mode atomic force microscope (AFM) cantilevers or MEMS (typically $\omega = 300 \text{ kHz}$) [11–13]. The parameter ε was introduced to distinguish between the conservative (frictionless) autonomous operation of the actuating system ($\varepsilon = 0$), and the non-conservative driven system by an external force ($\varepsilon = 1$) in presence of dissipation (friction) having a finite quality factor Q . In fact, for a conservative system we consider MEMS with a high-quality factor $Q > 10^4$ [11–13], so that we can neglect any dissipation effects. Moreover, for the study of non-equilibrium effects we considered initial separations d (where any elastic restoring force is zero) between the actuating plates in the range 850 nm to $2.5 \mu\text{m}$. We also assumed plates with flat surfaces having lateral dimensions $L_x = L_y = 10 \mu\text{m}$,

Figure 5-1. Imaginary (a) and real (b) parts of the frequency-dependent dielectric functions for both the Au and SiC systems measured with ellipsometry. The inset in (a) shows the schematic of the MEMS under consideration.

though any nanoscale roughness only gives significant contributions at short separations below 100 nm [14].

In Eq. (1) the Casimir force F_{Cas} is opposed by the elastic restoring force $F_{\text{res}} = -K(d-z)$ where K is the spring constant, and F_{elec} is the electrostatic force acting between two plates

$$F_{\text{elec}}(z) = \frac{1}{2} \frac{\epsilon_0 A V^2}{z^2}, \quad (2)$$

where V is applied voltage between the two plates with respect to the conduct potential difference due to differences in the work function of the material coatings of the plates, and ϵ_0 is the permittivity of vacuum. The Casimir force F_{Cas} is given by the expression

$$F_{\text{Cas}}(T_1, T_2, z) = F_0(z) + F_{\text{th}}^{\text{neq/eq}}(T_1, T_2, z), \quad (3)$$

where the contribution of the zero-point fluctuations ($T=0$), $F_0(z)$ is separated from the contribution $F_{\text{thermal}} = F_{\text{th}}^{\text{neq/eq}}(T_1, T_2, z)$ due to thermal fluctuations. $F_0(z)$ has been calculated via Lifshitz theory using the dielectric response function at imaginary frequencies $\epsilon(i\zeta)$, where we extrapolated at low optical frequencies using the Drude model (see Equations (2-11_18)). The chosen materials show significant optical contrast for the dielectric function at imaginary frequencies $\epsilon(i\zeta)$, which is the necessary input for the calculation of the Casimir force via the Lifshitz theory (see Equation (2-15)).

According to Ref. 11, the thermal force between two bodies in both configurations for thermal equilibrium (eq) and non-equilibrium (n-eq) conditions is given by (see Appendix 5)

$$F_{\text{th}}^{\text{neq/eq}}(T_1, T_2, z) = F_{\text{th}}^{\text{neq/eq}}(T_1, 0, z) + F_{\text{th}}^{\text{neq/eq}}(0, T_2, z). \quad (4)$$

The first and second terms at the right side of Eq. (4) describe the thermal equilibrium/non-equilibrium of the two bodies at different temperatures T_1 and T_2 respectively. The thermal part of the force $F_{\text{th}}^{\text{neq/eq}}(T, z)$ for each single body can be split into two contributions having the form

$$F_{\text{th}}^{\text{neq/eq}}(T, z) = F_{\text{th}}^{\text{neq/eq, PW}}(T, z) + F_{\text{th}}^{\text{neq/eq, EW}}(T, z), \quad (5)$$

where the indices PW and EW denote propagating and evanescent waves respectively. The thermal parts in Equations (3)–(5) can be calculated using the Lifshitz theory in the real frequency representation considering the PW and EW contributions. The PWs satisfy the condition $ck_{\perp} < \omega$ (with $k_{\perp}$ the in-plane wave vector), which is valid for real photons that propagate both in the vacuum gap and inside the bodies. EWs satisfy the condition $\omega \leq ck_{\perp}$, and they exist only along the boundary planes, as well as their electromagnetic field decreases exponentially within the gap separating the bodies. Figure 5-2 shows calculations of the Casimir force, where comparisons are made between the contribution of zero-point fluctuations F_0 and the total force $F_{\text{Cas}} = (F_0 + F_{\text{thermal}})$ for both the Au-Au and SiC-SiC systems. Clearly the thermal effects appear to influence more the poor conducting system leading to an increasing force with surface separation by increasing temperature differences. As a result, one can anticipate that for the poor conducting SiC-SiC system, the non-equilibrium effects vs. separation will likely play more important role on actuation properties than those of the good conductors.

5.3. Conservative actuating system ($\epsilon = 0$)

In order to investigate the thermal effect on the actuation of the microsystem, we introduced the bifurcation parameter $\delta_{\text{Cas}} = F_{\text{Cas}}^m / kd$ that represents the ratio of the minimum Casimir force $F_{\text{Cas}}^m = F_{\text{Cas}}(z = d)$ to the maximum restoring force. The parameter δ_{Cas} is useful to determine when there is a stable periodic solution for the system, which corresponds to sufficient restoring force to prevent stiction of the plates [6]. Using δ_{Cas} , Eq. (1) assumes the more convenient form for $\epsilon = 0$.

$$\frac{d^2\lambda}{dT^2} + \left(\frac{1}{Q}\right) \frac{d\lambda}{dT} = -(1 - \lambda) + \delta_v \frac{F_{\text{elec}}}{F_{\text{elec}}^m} + \delta_{\text{Cas}} \frac{F_{\text{Cas}}}{F_{\text{Cas}}^m} \quad (6)$$

with $\lambda = z/d$, $T = \omega_0 t$, and $\delta_v = F_{\text{elec}}^m / F_{\text{res}}^m = \epsilon_0 AV^2 / 2kd^3$ the corresponding electrostatic bifurcation parameter [15–17].

Figures 5-3 and 5-4 compare the strength of thermal effect between equilibrium and non-equilibrium conditions for both poor (SiC) and good (Au) conducting materials. Indeed, for the good conductive system Au-Au, as Figures 5-2_5-4 shows the thermal correction F_{thermal} has significant contribution at large separations ($> 4\mu\text{m}$), while at shorter separations it decreases the Casimir force [18–21]. As Figure 5-4 shows, for the Au-Au system the thermal correction vs. T decreases up to 300 K, and then it starts to increase at larger separations with the non-equilibrium effects contributing more to the force. By contrast, for the poor conductive system SiC-SiC the non-equilibrium state has stronger effect before 300 K, and after this temperature the influence of thermal equilibrium increase. Nevertheless, for the SiC-SiC system the thermal effects show significantly different contribution for both equilibrium and non-equilibrium situations.

The bifurcation diagrams in Figure 5-5 illustrate how the thermal effect can influence the stable and unstable regions. If the restoring force is strong enough ($\delta_{\text{Cas}} < \delta_{\text{Cas}}^{\text{Max}}$), then there are two equilibria points for the system. The stationary points closest to the initial separation d are stable centers with periodic solutions around them. While the ones closer to the fixed plate are unstable saddle points, and the motion around them will lead to stiction on the fixed plate due to the stronger Casimir force. These plots illustrate that the thermal contribution is likely to make significant change on the actuation, and performance of MEMS. The width of the stable area changes not only by considering the thermal contributions but also by changing the temperature between the interacting bodies. Clearly the thermal effects are more prominent for the case of the poor conductive system (SiC-SiC). The

Figure 5-2. Comparison between F_0 (zero-point fluctuations contribution) and the $F_{Cas} = (F_0 + F_{thermal})$ for both the Au-Au and SiC-SiC systems. The calculations for the thermal contribution $F_{thermal}$ were performed under non-equilibrium conditions for $T_1 = 300$ K and $T_2 = 400$ K.

Figure 5-3. $\alpha_{thermal} = F_{Cas}/F_0$ vs z for non-equilibrium conditions for both Au-Au and SiC-SiC systems, where for the calculations we considered one plate at $T_1 = 300$ K.

Figure 5-4. $\alpha_{thermal} (= F_{Cas}/F_0)$ vs $T_2(K)$ for both equilibrium ($T_1 = T_2$) and non-equilibrium ($T_1 = 300$ K) situations. The separation distance between plates is $z = 1 \mu m$, while that in the inset is $z = 2 \mu m$.

Figure 5-5. Bifurcation diagrams δ_{Cas} vs $\lambda(=z/d)$ for non-equilibrium conditions ($T_1=300$ K, $T_2=1$ K and $T=400$ K), $\delta_d=0$, and different initial actuation distances d : (a) and (b) $d=1\mu\text{m}$; (c) $d=850$ nm, (d) $d=2.5\mu\text{m}$. The solid and dashed lines represent the stable and unstable equilibrium points, respectively.

thermal effect is also clearly demonstrated if one considers different initial separations d and compares the bifurcation diagrams in Figures 5-5(c) and 5-5(d).

Besides the bifurcation analysis, the sensitive dependence of the actuation dynamics on the thermal effect is reflected by the Poincare maps in Figure 5-6. The homoclinic orbit separates unstable motion (leading to stiction within one period for the autonomous system) from the periodic closed orbits around the stable center point.

When the thermal effects decrease the Casimir force, as Figure 5-6 shows, they produce a wider stable region (red elliptic in shape central area). In fact, for the Au-Au system the stable area increases by increasing the temperature for both small and large separations indicating a decreasing Casimir force due to thermal effects. The situation is the reverse for the SiC-SiC system where the stable area decreases by increasing temperature for both small and large separations indicating an increasing Casimir force due to thermal effects. Nevertheless, the positive benefit of good conductors towards increased actuation stability due to thermal non-equilibrium effects on Casimir forces can be compromised even by a weak voltage application (or due to imperfect

Figure 5-6. Contour plots $d\lambda/dt$ vs. λ for the conservative system. The elliptical in shape homoclinic orbit contains the initial conditions that lead to stable oscillations. Au-Au system: (a) $\delta_{Cas} = 0.07$, $\delta_v = 0$, and $d = 850$ nm; (b) $\delta_{Cas} = 0.06$, $\delta_v = 0$, and $d = 2.5$ μ m. SiC-SiC system: (c) $\delta_{Cas} = 0.068$, $\delta_v = 0$, and $d = 850$ nm. (d) $\delta_{Cas} = 0.05$, $\delta_v = 0$, and $d = 2.5$ μ m. For the calculations we used 150×150 initial conditions (λ , $d\lambda/dt$), and considered $T_1 = 300$ K and T_2 as indicated.

compensation of contact potentials) that lead to additional attractive forces. This behavior is depicted Figure 5-7, which shows that any application of voltage strongly reduces the size of the region enclosed by the homoclinic orbit, and consequently the range of initial conditions that favor stable motion as the material conductivity increases.

Figure 5-7. Threshold curve $\alpha(=\gamma\omega_0 d/F_0)$ vs. driving frequency ω/ω_0 (with ω_0 the natural frequency of the system) comparing the influence of zero-point fluctuation and thermal effects under non-equilibrium conditions with $T_1=300$ K, $T_2=1$ K or 400 K. (a) Au-Au system with $\delta_{Cas}=0.075$ and (b) SiC-SiC system with $\delta_{Cas}=0.058$. The initial distance was in both cases $d=1\mu\text{m}$.

5.4. Non-conservative actuating systems ($\epsilon=1$)

Here we performed calculations to investigate the existence of chaotic behavior in dissipative microsystems undergoing forced oscillation via an applied external force $F_0 \cos(\omega, t)$ [22]. The chaotic behavior occurs if the separatrix (homoclinic orbit) of the conservative system splits. The latter can

be addressed by the so-called Melnikov function and Poincare map analysis. In the driven system, the unstable equilibrium turns into an unstable periodic orbit. If we define the homoclinic solution of the conservative system as $\varphi_{\text{hom}}^c(T)$, then the Melnikov function for the torsional system ($\varepsilon = 1$) is given by:

$$M(T_0) = \frac{1}{Q} \int_{-\infty}^{+\infty} \left(\frac{d\varphi_{\text{hom}}^c(T)}{dT} \right)^2 dT + \frac{\tau_0}{\tau_{\text{res}}^{\text{MAX}}} \int_{-\infty}^{+\infty} \frac{d\varphi_{\text{hom}}^c(T)}{dT} \cos\left[\frac{\omega}{\omega_0}(T + T_0)\right] dT \quad (7)$$

The separatrix splits if the Melnikov function has simple zeros so that $M(T_0) = 0$ and $M'(T_0) \neq 0$. If $M(T_0)$ has no zeros, the motion will not be chaotic. The conditions of nonsimple zeros, $M(T_0) = 0$ and $M'(T_0) = 0$ gives the threshold condition for chaotic motion. If we define

$$\mu_{\text{hom}}^c = \int_{-\infty}^{+\infty} \left(\frac{d\varphi_{\text{hom}}^c(T)}{dT} \right)^2 dT \text{ and } \beta(\omega) = \left| H \left[\text{Re} \left(F \left\{ \frac{d\varphi_{\text{hom}}^c(T)}{dT} \right\} \right) \right] \right|, \quad (8)$$

then the threshold condition for chaotic motion $\alpha = \beta(\omega)/\mu_{\text{hom}}^c$ with $\alpha = (1/Q)(F_0/F_{\text{res}}^{\text{MAX}})^{-1} = \gamma\omega_0 d/F_0$ obtains the form

$$\alpha = \frac{\gamma\omega_0 d}{F_0} = \left| H \left[\text{Re} \left(F \left\{ \frac{d\varphi_{\text{hom}}^c(T)}{dT} \right\} \right) \right] \right| / \int_{-\infty}^{+\infty} \left(\frac{d\varphi_{\text{hom}}^c(T)}{dT} \right)^2 dT \quad (9)$$

with $\gamma = I\omega_0/Q$, and $H[\dots]$ denoting the Hilbert transform [22,23].

Figures 5-8 shows the threshold curves $\alpha = \gamma\omega_0 d/F_0$ vs. the driving frequency ratio ω/ω_0 . For large values of α (above the curve) the dissipation dominates the driving force ($\alpha \sim \gamma/F_0$) leading to a regular motion, which approaches asymptotically the stable periodic orbit of the conservative system. However, for parameter values below the curve, the splitting of the separatrix leads possibly to chaotic motion. Clearly for the good conductive system Au-Au, the non-equilibrium thermal effects lead to reduction of the area below the threshold curve suggesting a reduction of the possibility for chaotic motion with increasing temperature. However, the inverse behavior takes place for the poor conducting SiC-SiC, where the area below the threshold curve increases due the increment of the Casimir force. The latter indicates enhanced possibility for chaotic motion that increases with increasing temperature and/or strengthening of the non-equilibrium state. By comparing Figures 5-8(a) and 5-8(b), it becomes obvious that by the weakening the dielectric function the thermal part become stronger to change stable actuation to chaotic motion, and consequently limit the long-term device predictability.

Since we study the occurrence of chaotic motion in terms of the sensitive dependence of the motion on its initial conditions, we present in Figure 5-9

Figure 5-8. Contour plot of the transient times to stiction using Poincare phase maps $d\lambda/dt$ vs. λ for the non-conservative system. (a) Left column is for the Au-Au system with $\delta_{Cas} = 0.075$; (b) right column is for the SiC-SiC system with $\delta_{Cas} = 0.058$. For the other parameters we used $\delta_v = 0$, $d = 1 \mu m$, $\alpha = 0.35$ and $\omega/\omega_0 = 0.5$. For both systems we considered also the effect of zero-point fluctuations (F_0), and for the non-equilibrium effects $T_1 = 300 K$ and T_2 as indicated. For the calculations we used 150×150 initial conditions (λ , $d\lambda/dt$). The central elliptical in shape (red) area shows the initial condition for which the device continues to follow stable motion after 100 oscillations.

the Poincare maps to show how the thermal correction can change the dynamical prediction towards chaos for both the good and poor conductive systems. When chaotic motion occurs (with decreasing value of $\alpha \sim \gamma/F_0$), there is a region of initial conditions where the distinction between qualitatively different solutions is unclear. For chaotic motion there is no longer a simple smooth boundary between the elliptical like stable area (red), and the outer area with unstable motion (blue region) towards stiction on the

fixed plate. As a result, if the motion is chaotic then stiction could take place after several periods prohibiting the long-term prediction of the actuation state for the device. As a result, chaotic the behavior introduces significant risk for stiction, and this is more prominent to occur when the Casimir force tends to increase due to the thermal effects.

As Figure 5-9(a) shows for the Au-Au system, with increasing temperature the thermal contribution clearly suppresses chaotic motion leading to stiction of the moving plate on the fixed one, since the central elliptical like (red) area increases in size. On the other hand, the inverse takes place for the SiC, where the increasing Casimir force with the thermal corrections leads to strong reduction of the stable actuation area and therefore to enhancement of chaotic behavior and possibility to stiction. Therefore, any consideration of thermal effects on the actuation of Casimir oscillators must be performed in close synergy with the material optical properties, Nevertheless, the influence of additional the attractive electrostatic forces by voltage application has to be carefully considered also for the driven dissipative systems. In fact,

Figure 5-9. Contour plot of the transient times to stiction using Poincare phase maps $d\lambda/dt$ vs. λ for the non-conservative system. (a) Left column is for the Au-Au system with $\delta_{Cas} = 0.075$; (b) right column is for the SiC-SiC system with $\delta_{Cas} = 0.058$. For the other parameters we used $\delta_v = 0$, $d = 1\mu m$, $\alpha = 0.35$ and $\omega/\omega_0 = 0.5$. For both systems we considered also the effect of zero-point fluctuations (F_0), and for the non-equilibrium effects $T_1 = 300K$ and T_2 as indicated. For the calculations we used 150×150 initial conditions (λ , $d\lambda/dt$). The central elliptical in shape (red) area shows the initial condition for which the device continues to follow stable motion after 100 oscillations.

Figure 5-10. Contour plot of the transient times to stiction using Poincare phase maps $d\lambda/dt$ vs. λ for the non-conservative system. (a) Left column is for the Au-Au system with $\delta_{Cas} = 0.07$; (b) right column is for the SiC-SiC system with $\delta_{Cas} = 0.05$. For the other parameters we used $\delta_v = 0.02$, $d = 1 \mu m$, $\alpha = 0.75$ and $\omega/\omega_0 = 0.5$. For both systems we considered also the effect of zero-point fluctuations (F_0), and for the non-equilibrium effects $T_1 = 300 K$ and T_2 as indicated. For the calculations we used 150×150 initial conditions (λ , $d\lambda/dt$). The central elliptical in shape (red) area shows the initial condition for which the device continues to follow stable motion after 100 oscillations.

Figure 5-10 demonstrates the sensitive dependence of chaotic motion on the applied electrostatic potential for both the Au and SiC systems. Again, it is confirmed that any voltage application will strongly influence the chaotic behavior of the system having a dramatic effect for the higher conductivity materials, where any positive effects due to the thermal contributions on the Casimir forces are highly compromised.

5.5. Conclusion part (I)

In conclusion, we investigated thermal non-equilibrium effects on the actuation of an autonomous and a driven microswitch under the influence of Casimir forces between interacting materials. For the latter we considered Casimir interactions between good and poor conductors, in both cases, the

Drude model was used to extrapolate at low optical far-infrared frequencies, where measured optical data were not available. Furthermore, for the frictionless autonomous systems, the bifurcation and phase portrait analysis show that the non-equilibrium conditions can have significant impact on the stable and unstable operating regimes depending on the material optical properties. Indeed, at separation below $4\text{ }\mu\text{m}$, for systems with the high conductivity materials, an increasing temperature difference between the actuating components can enhance the stable operating region. This is due to the reduction of the thermal effect on the Casimir force, while for the poor conductive materials the opposite takes place.

For a dissipative but periodically driven system, the Melnikov function and Poincare portrait analysis have shown that for the poor conductive systems (e.g., SiC) the thermal non-equilibrium effects, which will increase the Casimir forces, lead to increased possibility for chaotic motion and possible stiction with increasing temperature differences between the actuating components. Therefore, in this case the thermal non-equilibrium effects could make impossible the long-term prediction of the actuation state of a device with increasing temperature differences. However, for good conducting systems (e.g., Au-Au), the thermal fluctuations reduce the possibility of chaotic behavior with increasing temperature in comparison to a system without thermal fluctuations. Nevertheless, the positive benefit of good conductors towards increased actuation stability and reduced chaotic behavior due to non-equilibrium Casimir forces can be easily compromised by any voltage application. Therefore, thermal non-equilibrium Casimir forces can influence the actuation of devices towards unstable and chaotic behavior in strong correlation with their optical properties, and associated conduction state, as well as applied electrostatic potentials.

II) Sensitivity of non-equilibrium Casimir forces on low frequency optical properties towards chaotic motion of microsystems: Drude vs. Plasma model

It has been shown that the low optical frequency range, which is not accessible by experimental measurements [9,10,24–31], is playing significant role for an effective stable operation of devices [32–38]. In fact, Casimir force measurements have revealed deviations from force predictions of dissipative models (e.g., the Drude model) [32–36], which lead to finite absorption at frequencies $\omega > 0$ and singular absorption $\sim 1/\omega$ for $\omega \rightarrow 0$ (static limit). On

the other hand, the Plasma (P) model [32–36], which can also be thought as having infinite absorption at the frequency $\omega \sim 0$, and zero anywhere else, allowed calculations of the Casimir force that described the measured force data more precisely at separations above 160 nm [32–36,38]. Recently, we have shown that for systems at thermal equilibrium the choice of the Drude or Plasma models to describe the optical properties at low optical frequencies (far-infrared and below) in the range where any measured optical data are not available, lead to remarkable different results regarding the stability and emerging chaotic motion of MEMS [37,38].

However, it still remains unexplored how the manner one could describe or equivalently model the optical properties in the low frequency range can affect actuation of devices towards chaotic motion under the influence of thermal non-equilibrium Casimir forces taking into account the conductivity of interacting materials, and possible residual electrostatic interactions due to uncompensated contact potentials. This topic will be explored here for both good and poor conductive materials that are used in microdevices and can lead to irreversible adhesion of moving parts due to stiction on a long-term during operation. In fact, the design of these devices can always be quite challenging due to the occurrence of chaotic behavior, which causes abrupt changes in their dynamical behavior resulting in device malfunctions. As a result, the present study will provide essential knowledge for the design of actuating devices operating under non-equilibrium conditions, and add new functionalities to MEMS/NEMS architectures taking into account detailed modeling of material optical properties.

5.6. Material systems and device actuation

For our purpose we have chosen gold (Au) and highly doped silicon carbide (SiC) in order to cover wide range of materials with different optical properties and associated conductivities. Au is used due to its high conduction ratio $\omega_p^2/\omega_{\tau|\text{Au}} \approx 1600 \text{ eV}$ [10,39] and its frequent use in devices, while as a poor conductor we used nitrogen doped SiC with a conductivity ratio $\omega_p^2/\omega_{\tau|\text{SiC}} = 0.4 \text{ eV}$ [31]. Notably, SiC is also suitable for operation in harsh environments and an important system that is compatible with Si-based technologies. Both materials were optically characterized with the same ellipsometry equipment (J. A. Woollam Co., Inc. ellipsometers VUV-VASE (0.5–9.34 eV) and IR-VASE (0.03–0.5 eV)) [10,31,39]. Details for the Drude and Plasma models and the frequency dependent dielectric functions of

the materials in our study are shown in Chapter 2, Equations (2-11_19). The corresponding dielectric functions at imaginary frequencies $\epsilon(i\zeta)$, which are the necessary inputs for calculating the contribution of the zero-point fluctuations ($T=0$) on the Casimir force via the Lifshitz theory, are calculated as it is explained in Equation (2-15) [10,31,39].

In order to understand the influence of the non-equilibrium Casimir force on MEMS actuation, we have considered in Figure 5-1a (inset) a typical microswitch, which it is a well-known essential device. It is constructed from two electrodes of which one is fixed, and the other is suspended by a mechanical spring governed by the Hooke's law [1]. The elastic restoring force $F_{\text{res}} = -k(d-z)$ of the spring with stiffness k counterbalances both the attractive Casimir and electrostatic forces. After applying a bias voltage V between the planar electrodes and/or due to uncompensated contact potentials of the material coatings, it is produced an electrostatic force F_{elec} in Eq. (2). Furthermore, it is possible to express the non-equilibrium Casimir force F_{Cas} between the plates as in Eq. (3). $F_0(z)$ is separated from the thermal part $F_{\text{th}}^{\text{neq}}(T_1, T_2, z)$ due to thermal fluctuations. $F_0(z)$ has been calculated via the Lifshitz theory using the dielectric functions at imaginary frequencies for both the Drude and Plasma models (see Chapter 2 for the extrapolations of the measured optical data via the Drude and Plasma models). The chosen materials (Au and SiC) show significant optical contrast for the dielectric function at imaginary frequencies $\epsilon(i\zeta)$ at frequencies $\zeta < 1$ eV, which it will manifest in Casimir force variations for nanoscale separations $c/2\zeta > 10$ nm.

According to [7], the thermal force between the two bodies in both configurations in and out of thermal equilibrium can be presented as in Eq. (4). The first and the second term at the right side of Eq. (4) describe the bodies at temperature T_1 and T_2 , respectively as it is written in Eq. (4).

Equations 2-4 can be presented using the Lifshitz formula in real frequencies as a sum of contributions from propagating and evanescent waves. The propagating waves satisfy the condition $ck_{\perp} < \omega$, ($k_{\perp}$ is in-plane wave vector), which is valid for real photons. They propagate both in the vacuum gap and inside the bodies. The corresponding out of plane wave vector k_0 is real. Evanescent waves satisfy the condition $\omega \leq ck_{\perp}$, and they may propagate only along the boundary planes. The electromagnetic field of an evanescent wave decreases exponentially with distance from the interface between the vacuum gap and the interacting bodies, and the corresponding out of plane wave vector k_0 is imaginary.

In this study, our main goal is to investigate the influence of optical properties at low frequencies on the dynamical actuation of systems with strong

and weak conductivity. Therefore, we have focused on identical interacting bodies and ignored mix states between good and poor conductors. The latter is highly interesting, but it which will be given elsewhere for several systems including Au, SiC, Ru, and Phase change materials. Moreover, the consideration of identical bodies simplifies the process of calculations to gain better insight into these complex situations. Indeed, according to [7], $F_{th}^{neq}(T, z)$ can be separated in symmetric and anti-symmetric parts, which in system with identical bodies the anti-symmetric terms disappear (see Appendix 5).

Finally, the equation of motion for the micro electromechanical system (Figure 5-1a), where the fixed and moving plates are considered to be coated by Au or SiC, is given by Eq. (1) where the parameter ϵ was introduced to distinguish between the conservative frictionless autonomous operation of the actuating system ($\epsilon=0$), and the non-conservative driven system by an external force ($\epsilon=1$) in presence of friction having a finite quality factor Q . Finally, in each case we assumed flat surfaces, because any nanoscale roughness will give significant contribution at separations below 100 nm [14], while we have considered different initial distances between the plates in the range $d=850$ nm to $2.5\mu\text{m}$. In all cases the lateral dimensions of the plates were $L_x=L_y=10\mu\text{m}$.

5.7. Conservative systems ($\epsilon=0$)

For our stability analysis we introduced the bifurcation parameter $\delta_{Cas} = F_{Cas}^m / kd$ [6,40,41] which is the ratio of the minimum of Casimir force $F_{Cas}^m = F_{Cas}(z=d)$ to the maximum restoring force. In this manner, we are able to compare the force influence for different non-equilibrium thermal conditions. The locus of the equilibrium points is obtained from Eq. (1) if we set $F_{total} = -F_{res} + F_{elec} + F_{Cas} = 0$. The solution yields for the bifurcation parameter δ_{Cas} [6,41,42]

$$\delta_{Cas} = \left(-F_{res}(z) + \delta_v \frac{F_{elec}(z)}{F_{elec}^m} \right) \left(\frac{F_{Cas}^m}{F_{Cas}(z,T)} \right) \quad (10)$$

where $\delta_v = F_{elec}^m / F_{res}^m = \epsilon_0 AV^2 / 2kd^3$ is the corresponding electrostatic bifurcation parameter [15–17]. The critical points, where stiction occurs, are also characterized by the condition $dF_{total}/dz=0$ [6,41,42]. Therefore, the use of δ_{Cas} allows to determine when there is a stable periodic solution for the device that corresponds to sufficient restoring force to prevent stiction of the plates [6]. Using δ_{Cas} , Eq. (1) assumes the more convenient form

$$\frac{d^2\lambda}{dT^2} + \left(\frac{1}{Q}\right)\frac{d\lambda}{dT} = -(1-\lambda) + \delta_v \frac{F_{elec}}{F_{elec}^m} + \delta_{Cas} \frac{F_{Cas}}{F_{Cas}^m} + \varepsilon \frac{F_0}{F_{res}^{Max}} \cos\left(\frac{\omega}{\omega_0} T\right) \quad (11)$$

With $\lambda = z/d$, and $T = \omega_0 t$.

5.8. Casimir force and actuation for good conductivity microsystems (Au-Au)

According to Figures 5-12a and 5-13a, the extrapolation with the Drude model at low frequencies results in a large thermal correction for the Au-Au system, which it does not occur for the Plasma model. The predicted thermal correction of the Drude model has significant value even at shorter separations ($<1\mu\text{m}$). Although several experimental studies are in disagreement with

Figure 5-12. Comparison between F_0 (zero-point fluctuations) and the F_{Cas} ($=F_0 + F_{thermal}$) for the (a) Au-Au, and (b) SiC-SiC systems using both the Drude and Plasma models. The calculations for the thermal contribution $F_{thermal}$ were performed under non-equilibrium conditions for $T_1 = 300$ K and $T_2 = 400$ K.

Figure 5-13. $\alpha_{\text{thermal}} = F_{\text{Cas}}/F_0$ vs. z for non-equilibrium conditions for the (a) Au-Au, and (b) SiC-SiC systems using the Drude and Plasma models.

the Drude model predictions [19–21,33], there are investigations that show agreement with the Drude model prediction [43]. Moreover, the thermal correction is opposite in sign to the main contribution in the Casimir force from zero-point fluctuations (F_0) within a wide range of separations, and consequently leads to a decrease in magnitude of the total Casimir force. As it emerges from Figure 5-13a, for the Drude model, and ignoring the sign of the thermal correction, the F_{thermal} becomes weak with decreasing temperature. The same takes place also for the Plasma model but the sign of the F_{thermal} remains positive for most of the separations. For the good conductive Au-Au

Figure 5-14. $\alpha_{\text{thermal}} = F_{\text{Cas}}/F_0$ vs T_2 (K) for both equilibrium ($T_1 = T_2$) and non-equilibrium ($T_1 = 300$ K) situations in (a) Au-Au, and (b) SiC-SiC systems using the Drude and Plasma models. The separation distance between plates is $z = 1 \mu\text{m}$, while that in the inset is $z = 2 \mu\text{m}$.

system, at large separations ($> 4 \mu\text{m}$), the key factor that affects the strength of the thermal contribution is the magnitude of the temperature, while for smaller separations ($< 2.5 \mu\text{m}$), the influence of the optical properties at low frequencies overcomes the effect of the magnitude of the temperature.

Finally, Figure 5-14a provides a comparison between thermal equilibrium and non-equilibrium situations for both the Drude and Plasma models vs temperature.

For the Drude model the magnitude of total Casimir force is stronger for systems at thermal equilibrium for temperatures below 300 K. While for the Plasma model below 300 K the effect of the thermal part is too weak without any difference between thermal equilibrium and non-equilibrium situations. However, by increasing T_2 and consequently strengthening the contribution of the thermal component the magnitude of total Casimir force becomes stronger at equilibrium conditions, which is the opposite for the Drude model.

Furthermore, Figures 5-14a–c illustrate the stability of the microsystem operating with different initial separations d . If the restoring force is strong enough ($\delta_{\text{Cas}} < \delta_{\text{Cas}}^{\text{Max}}$), then there are two equilibria for the system. The stationary points closest to d are the stable centers around which periodic solutions exist, while the points closer to the fixed plate are unstable saddle points so that motion around them will lead to stiction on the fixed plate due to stronger Casimir forces. Considering the negative sign of F_{thermal} in the Drude model, and increasing its magnitude by increasing the temperature, the stability of the system will increase at higher temperatures (e.g., a system at 1 K loses its stability sooner than at 400 K). However, for the Plasma model the thermal effect is negligible at short separations and will not change the device operation. As the initial separation increases, because of the positive contribution of the thermal effect to the Casimir force the stability of the system will decrease with increasing temperature (e.g., a system at 400 K loses its stability sooner than at 1 K).

Besides the bifurcation diagrams, the sensitive dependence of the actuation dynamics on the thermal effect is reflected by the Poincare maps in Figure 5-16. For the conservative system the homoclinic orbit separates unstable motion, leading to stiction within one period, from the periodic closed orbits around the stable center point. Due to the negative sign of the thermal component in the Au-Au system if we use the Drude model, which leads to decrement of the total force, the stable area (elliptical in shape red area) increases by increasing temperature in absence of any applied voltage. However, by applying a voltage the effect of the positive effect of the thermal component can be counterbalanced leading to reduced stable region. On the other hand, for the Plasma model (at separations $> 2 \mu\text{m}$) the positive contribution to the Casimir force of the thermal component leads to reduction of the stable operating area, and this effect is amplified by an additional non-zero electrostatic voltage contributing an extra attractive force between the interacting plates.

Figure 5-15. Bifurcation diagrams δ_{Cas} vs $\lambda (=z/d)$ for non-equilibrium conditions ($T_1=300\text{ K}$, $T_2=1\text{ K}$ and 400 K), $\delta_v=0$, and different initial actuation distances d : (a) $d=850\text{ nm}$ and (b) $d=1\text{ }\mu\text{m}$ and (c) $d=2.5\text{ }\mu\text{m}$ for the Au-Au system; (d) $d=850\text{ nm}$ and (e) $d=1\text{ }\mu\text{m}$ and (f) $d=2.5\text{ }\mu\text{m}$ for the SiC-SiC system. The solid and dashed lines represent the unstable and stable equilibrium points, respectively.

5.9. Casimir force and actuation for poor conductivity microsystems (SiC-SiC)

For this system the thermal effects can generate considerably more significant changes in the Casimir force and therefore into device motion in comparison to the high conductivity materials. As it is shown in Figure 5-13b, unlike to the Au-Au system, here for both the Drude and Plasma models the thermal component for the most range of separations has positive sign and it increases the magnitude of the Casimir force. The key factor which determines the strength of the thermal effect is the magnitude of the temperature, and its

Figure 5-16. Contour phase space plots $d\lambda/dt$ vs. λ for the Au-Au system. For the calculations, we used 200×200 initial conditions $(\lambda, d\lambda/dt)$. The elliptical in shape homoclinic orbit encloses the initial conditions that lead to stable oscillations. $\delta_{Cas} = 0.06$, and $d = 2 \mu\text{m}$: $\delta_v = 0$ (left column), and $\delta_v = 0.012$ (right column) using both the Drude and Plasma models. Here we considered $T_1 = 300 \text{ K}$, and $T_2 = 1 \text{ K}$ or 400 K as indicated.

effect is stronger than the influence of the low optical frequencies (using either the Drude or the Plasma model for the extrapolation). Figure 5-14b indicates the opposite results to those for the Au-Au microsystem if we compare the thermal equilibrium and non-equilibrium situations. The total Casimir force is stronger for the Drude model and also, at low temperature, the magnitude of total Casimir force is in any case stronger for non-equilibrium conditions for both the Drude and Plasma modes.

Figure 5-17. Contour phase space plots $d\lambda/dt$ vs. λ for the SiC-SiC system. For the calculations, we used 200×200 initial conditions $(\lambda, d\lambda/dt)$. The elliptical in shape homoclinic orbit encloses the initial conditions that lead to stable oscillations. $\delta_{Cas} = 0.06$, and $d = 2 \mu\text{m}$: $\delta_v = 0$ (left column), and $\delta_v = 0.012$ (right column) using both the Drude and Plasma models. Here we considered $T_1 = 300 \text{ K}$, and $T_2 = 1 \text{ K}$ or 400 K as indicated.

Figures 5-15d–f illustrate the changes of stability for the SiC-SiC system due to the thermal contributions. Unlike to the Au-Au system, for the SiC-SiC system both the Drude and Plasma models generate significant influence in the bifurcation diagrams, where the temperature difference plays the dominant role. As a result, decrement of the temperature leads to increased stable operation region as comparison for 1 K and 400 K operation indicates. Furthermore, we illustrate the dynamical behavior using Poincare maps. Indeed, Figure 5-17 illustrates that, by increasing the temperature the stable

region (elliptical in shape red area) decreases, and the sensitivity to temperatures changes are higher for the Drude model. This is due to the extrapolation in low frequencies of the dielectric function for real frequencies, which is the vital input for the calculation of the thermal component. On the other hand, there is no extrapolation for the Plasma model at real frequencies, and consequently the influence of the low frequency range on the thermal component is absent.

As a result, in each column of Figure 5-17 (for systems at the same temperature) the stable area is smaller for the Drude model, and the change in stable area is sharper at $T = 400$ K by switching from the Drude to Plasma models. Despite the same scenario with the Au-Au system regarding the extrapolations at low frequencies, the reverse behavior takes place for the SiC-SiC system because of the opposite sign of the thermal component in comparison to F_0 in the Drude model for the Au-Au system (leading to larger phase space area for stable Au-Au actuation).

5.10. Non-conservative systems ($\epsilon = 1$)

Here we performed calculations to investigate the existence of chaotic behavior in microsystems undergoing forced oscillation via an applied external force $F_0 \cos(\omega t)$ [44], and compare the influence of thermal effects for both the Drude and Plasma models. Chaotic behavior could occur if the separatrix (homoclinic orbit) of the conservative system splits. This behavior can be addressed by the so-called Melnikov function and Poincare map analysis [23,44]. In a driven system, the unstable equilibrium turns into an unstable periodic orbit.

Figure 5-18 shows the threshold curves $\alpha = \gamma\omega_0 d/F_0$ vs. driving frequency ratio ω/ω_0 . For large values of α (above the threshold curve) the dissipation dominates the driving force ($\alpha \sim \gamma/F_0$) leading to motion, which asymptotically approaches the stable periodic orbit of the conservative system ($\epsilon = 0$). However, for parameter values α below the threshold curve, the splitting of the separatrix could lead to chaotic motion. Clearly for the Au-Au system (good conductors), the thermal contribution calculated via the Plasma model (Figure 5-18a) is not able to change the stability of device since the curves with and without thermal fluctuation are similar to each other. However, for the Drude model (Figure 5-18b), the thermal contribution decreases the possibility for chaotic motion, which is further augmented by increasing in magnitude of the temperature difference between the actuating components.

Figure 5-18. Threshold curve $\alpha (= \gamma \omega_0 d / F_0)$ vs. driving frequency ω / ω_0 (with ω_0 the natural frequency of the system) to compare the influence of zero-point fluctuations and thermal effects under non-equilibrium conditions with $T_1 = 300$ K, and $T_2 = 1$ K or 400 K for the Au-Au system with $\delta_{Cas} = 0.07$, $\delta_v = 0$ and $d = 1 \mu\text{m}$: (a) Plasma model, and (b) Drude model. Similar plots are shown for the SiC-SiC system with $\delta_{Cas} = 0.06$, $\delta_v = 0$ and $d = 1 \mu\text{m}$: (c) Plasma model, and (d) Drude model.

Furthermore, according to Figures 5-18c and 5-18d, for the SiC-SiC system (poor conductors) the contribution of the thermal component for both the Drude and Plasma models can change the occurrence of chaotic behavior. The extrapolation at low frequencies with the Drude model has stronger effect on the threshold condition for chaotic behavior, which is enhanced by increasing temperature difference between the actuating components. The occurrence of chaotic motion can be confirmed by investigating the sensitive dependence of the motion on its initial conditions via the Poincare maps, as it shown in Figures 19-22. When the possibility for chaotic motion to occur increases with decreasing value of α from the Melnikov analysis, there is a region of initial conditions where the distinction between qualitatively different solutions is unclear.

For chaotic motion there is no a simple smooth boundary between the red (lighter gray) and the blue (Dark gray) regions (as it is the case for conservative motion in Figures 5-16 and 5-17). As a result, if the motion is chaotic then stiction can still take place after several periods affecting the

Figure 5-19. Contour plot of the transient times to stiction using Poincare phase maps $d\lambda/dt$ vs. λ for the non-conservative Au-Au system using the Drude model with $d=1\mu\text{m}$, $\alpha=0.5$, $\omega/\omega_0=0.6$, $\delta_{\text{Cas}}=0.07$, and $\delta_v=0$ (left column), and $\delta_v=0.012$ (right column). In both cases we considered also the effect of zero-point fluctuations (F_0), and the non-equilibrium effects for $T_1=300\text{K}$ and $T_2=1\text{K}$ or 400K (shown in the plots). For the calculations we used 200×200 initial conditions (λ , $d\lambda/dt$).

long-term stability of the device. Therefore, chaotic behavior introduces significant risk for stiction, and this more prominent to occur when the magnitude of the Casimir force increases limiting our ability to predict the long-term behavior of the actuating systems.

For the Au-Au system, if we compare the Poincare maps of Figures 5-19 (Drude model) and 5-20 (Plasma model), it is evident that for the Drude model the thermal corrections are more effective to suppress the occurrence of chaotic motion, and enhance stable actuation with further increment of the temperature difference. At the same time the presence of an electrostatic voltage can reduce or even fully compensate the positive effect of the thermal

Figure 5-20. Contour plot of the transient times to stiction using Poincare phase maps $d\lambda/dt$ vs. λ for the non-conservative Au-Au system using the Plasma model with $d = 1 \mu\text{m}$, $\alpha = 0.5$, $\omega/\omega_0 = 0.6$, $\delta_{\text{Cas}} = 0.07$, and $\delta_v = 0$ (left column), and $\delta_v = 0.012$ (right column). In both cases we considered also the effect of zero-point fluctuations (F_0), and the non-equilibrium effects for $T_1 = 300 \text{ K}$ and $T_2 = 1 \text{ K}$ or 400 K (shown in the plots). For the calculations we used 200×200 initial conditions (λ , $d\lambda/dt$).

component, and subsequently drive the system to chaotic motion and stiction. On the other hand, for the Plasma model the effect of the thermal contribution is rather weak to alter the chaotic behavior in presence or absence of any electrostatic forces.

According to Figures 5-21 and 5-22, for the SiC-SiC system the thermal contribution for both the Drude and Plasma models are able to make significant changes in the dynamics of the actuating system towards chaotic motion. By comparing the Poincare maps in Figures 5-21 and 5-22 it is obvious that the strength of the thermal contribution is higher for the Drude model in absence and/or presence of electrostatic voltage leading to increased chaotic

Figure 5-21. Contour plot of the transient times to stiction using Poincare phase maps $d\lambda/dt$ vs. λ for the non-conservative SiC-SiC system using the Drude model with $d = 1 \mu\text{m}$, $\alpha = 0.5$, $\omega/\omega_0 = 0.6$, $\delta_{\text{Cas}} = 0.055$, and $\delta_v = 0$ (left column) and $\delta_v = 0.012$ (right column). In both cases we considered also the effect of zero-point fluctuations (F_0), and the non-equilibrium effects for $T_1 = 300 \text{ K}$ and $T_2 = 1 \text{ K}$ or 400 K (shown in the plots). For the calculations we used 200×200 initial conditions (λ , $d\lambda/dt$).

behavior. This is because of the differences in extrapolating at real frequencies between the Drude and Plasma models.

Figure 5-22. Contour plot of the transient times to stiction using Poincare phase maps $d\lambda/dt$ vs. λ for the non-conservative SiC-SiC system using the Plasma model with $d=1\text{ }\mu\text{m}$, $\alpha=0.5$, $\omega/\omega_0=0.6$, $\delta_{\text{Cas}}=0.055$, and $\delta_v=0$ (left column), and $\delta_v=0.012$ (right column). In both cases we considered also the effect of zero-point fluctuations (F_0), and the non-equilibrium effects for $T_1=300\text{ K}$ and $T_2=1\text{ K}$ or 400 K (shown in the plots). For the calculations we used 200×200 initial conditions (λ , $d\lambda/dt$).

5.11. Conclusion part (II)

In conclusion, we investigated the sensitivity of non-equilibrium Casimir forces to optical properties at low frequencies via the Drude and Plasma models during actuation of microsystems. For the latter we explored how the thermal effects influence the stable and chaotic motion for both autonomous conservative and non-conservative driven systems assuming both good and poor conductors, whose static conductivity is different by more than three orders of magnitude. For both material systems we used the Drude and

Plasma models to extrapolate the optical properties at low frequencies, where optical measurements are not feasible. In fact, for the conservative actuating system bifurcation and phase space analysis show that the system motion is strongly influenced by the thermal non-equilibrium effects depending on the modeling of the optical properties at low frequencies, where also the presence of residual electrostatic forces can drastically alter the actuating state of the system, depending strongly on the material conductivity. For non-conservative systems the Melnikov function approach is used to explore the possible presence of chaotic motion rendering predictions of stable actuation or malfunction due to stiction over long times rather impossible. Moreover, it is shown that the thermal contributions produce the opposite effect for the emerging chaotic behavior of the Au-Au and SiC-SiC systems, if the Drude model is used to model the low optical frequencies. On the other hand, using the Plasma model then only for the poor conducting SiC-SiC system the possibility of chaotic motion is enhanced, while for the good conducting Au-Au system at small separations (less than $2\text{ }\mu\text{m}$) the chaotic behavior will remain unaffected. Therefore, the modeling of the low frequency regime for the materials under consideration in combination with thermal effects, and applied (or uncompensated) electrostatic voltages must be taken very carefully into account for the design of actuating devices.

Appendix 5: Brief theory of the non-equilibrium Casimir force

As it has been shown in [7], when both components of the actuating device are made from the same material, the non-equilibrium thermal force between them is given by

$$F_{th}^{neq}(T_1, T_2, z) = \frac{1}{2} [F_{th}^{eq, PW}(T_1, 0, z) + F_{th}^{eq, EW}(T_1, 0, z)] + \frac{1}{2} [F_{th}^{eq, PW}(0, T_2, z) + F_{th}^{eq, EW}(0, T_2, z)]. \quad (B.1)$$

The contribution of the propagating waves in thermal equilibrium can be written as

$$F^{eq, PW}(T, z) = \frac{-\hbar}{\pi^2} \int_0^\infty d\omega \frac{1}{\exp(\frac{\hbar\omega}{k_B T}) - 1} \int_0^k dk_\perp k_\perp k_0 \sum_{v=s,p} \frac{\text{Re}(r_1^v r_2^v \exp(2id k_0)) - |r_1^v r_2^v|^2}{|D_v|^2}, \quad (B.2)$$

while for the evanescent component we have

$$F^{eq, EW}(T, z) = \frac{\hbar}{\pi^2} \int_0^\infty d\omega \frac{1}{\exp(\frac{\hbar\omega}{k_B T}) - 1} \int_k^\infty dk_\perp k_\perp \text{Im}(k_0) \exp(-2d \text{Im}(k_0)) \sum_{v=TE,T} \frac{\text{Im}(r_1^v r_2^v)}{|D_v|^2}. \quad (B.3)$$

Equations B.1 to B.3 describe the force per unit area, with ω the real frequency, and $D_v = 1 - r_1^v r_2^v \exp(2i k_0 z)$. Both coefficients r_1^v , r_2^v are defined in Appendix A with $k_i (i = 0, 1, 2) = \sqrt{\epsilon_i(\omega)(\omega^2/c^2) + k_\perp^2}$, and $\epsilon(\omega) = \text{Re}[\epsilon(\omega)] + i\text{Im}[\epsilon(\omega)]$.

The works discussed in this chapter have been published as

“F. Tajik, Z. Babamahdi, M. Sedighi, A. A. Masoudi, and G. Palasantzas, *Dependence of Non-Equilibrium Casimir Forces on Material Optical Properties toward Chaotic Motion during Device Actuation*, *Chaos* 29, 93126 (2019)”

“F. Tajik, A. A. Masoudi, Z. Babamahdi, M. Sedighi, and G. Palasantzas, *Sensitivity of Nonequilibrium Casimir Forces on Low Frequency Optical Properties toward Chaotic Motion of Microsystems: Drude vs Plasma Model*, *Chaos* 30, 023108 (2020).”

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Samenvatting

Thans geldt de Casimirkracht, een kracht die zijn origine vindt in de verstoringen van elektromagnetische vacuüm fluctuaties, nog steeds als een onderwerp van onophoudelijk onderzoek, ofschoon dat de stelling door de Nederlandse fysicus Hendrik Casimir in 1948 reeds 70 jaren terug gaat. De belangstelling komt voort uit de veelvoud aan onderzoeksvelden variërend van fundamentele natuurkunde in de zoektocht naar nieuwe krachten verder reikend dan het standaard model, tot micro/nanoschakelingen in technologische toepassingen. Alhoewel de Casimirkracht in de laatste twee decennia uitgebreid bestudeerd is, is het echter niet geëxploreerd in veel gebieden die complexe materialen betrekken. Deze zijn van groot belang voor toepassing. Feitelijk kan uitgebreide kennis van Casimirkrachten en de daarbij geassocieerde elektrostatica van complexe geleidende oppervlakten essentieel zijn in diverse velden reikend van de werking van micro/nanoapparaten in barre omstandigheden tot multilaag coatings in geavanceerde lithografie technieken. In dit proefschrift worden verslag gelegd van onze inspanningen om de effecten van oppervlakte compositie, ruigheid en elektrostatische aandeel op de gemeten Casimirkracht aan te tonen benevens het analyseren van de invloed der optische eigenschappen en verschillende manieren van data extrapolatie op de theoretische berekeningen.

Wij hebben Casimirkrachtmetingen uitgevoerd en elektrostatica van geleidende SiC en Ru oppervlakten daarbij betrokken. De elektrostatische kalibratie toonde aan dat de Si- en C- fasen zich verschillende gedragen met een bijna $\sim 0,6\text{--}0,7\text{ V}$ verschil in contactpotentiaal $V_0^{\text{Si/C}}$ wat kan worden toegeschreven aan een hogere incorporatie van N aan de C-terminerende fase. Echter, de met ellipsometrie gemeten optische eigenschappen van de SiC-Si/C terminerende oppervlakten vertoonden geen wezenlijke verschillen, beduidend dat de effectieve diepte der Si/C terminerende oppervlakte laag is

significanter kleiner dan de penetratie diepte van een foton, zonder te leiden tot enig verschil in de via de Lifshitz theorie berekende krachten. Desalniettemin, de gemeten Casimirkrachten, na compensatie voor de elektrostatische componenten, toonde de verschillen tussen de Si/C fasen, terwijl de vergelijking met de prognose door de Lifshitz theorie een betere overeenkomst voor de SiC-Si toont. Tenslotte indiceerde de vergelijking van de Casimirkrachten voor sub 40 nm scheiding tussen de SiC-Si/C and Ru oppervlakten dat de korte-afstandseffecten op de Casimirkracht toeneemt in grootte met toenemend metallisch gedrag van de oppervlakte plaat. Vanwege die reden is voorzichtigheid geboden met het in overweging nemen van de korte-afstand Casimirinteractie tussen complexere diëlektrische materialen, niet louter voor de optische eigenschappen van het materiaal, doch ook voor de geleidende staat en ruigheid der oppervlakte lagen.

Gegeven de potentie van de nieuw ontdekte topologische isolatoren in de opkomende technologische apparaten, hebben wij de beslissende rol der laagdikte van de topologisch niet-geleidende Bi₂Se₃ lagen op de Casimir- en elektrostatische krachten onderzocht. De krachten werden gemeten voor Au-Bi₂Se₃ systemen onder standaard omgevingscondities om realistisch toepassing te simuleren. Aangetoond werd dat de elektrostatische krachten behorend aan het niet-nul contact potentiaal van $V \sim 200$ mV een dominante rol speelden terwijl dit niet gecompenseerd kon worden voor dunnere lagen (dunner dan 20 nm in dikte). Echter, voor dikkere lagen begon de gemeten kracht, na potentiaal compensatie, de echte Casimirkracht te benaderen in zeer goede overeenkomst met de Lifshitz theorie. Onze resultaten beduiden dat enig geleidende afwijking van de theorie voor dunnere lagen ten grondslag ligt aan de aanwezigheid van sterker elektrostatische effecten.

De gedetailleerde kennis over de fenomenen van stictie door de Casimirinteractie en haar verbondenheid aan chaotische beweging tijdens aandrijving, is noodzakelijk om de lange termijn prestatie van apparaten te verbeteren. Hier is aangetoond dat de niet-evenwichts Casimirkrachten, die het algemeenste geval zijn als de interacterende componenten verschillende temperaturen hebben, de aandrijving van apparaten naar instabiel en chaotisch gedrag in sterke correlatie met hun optische eigenschappen en bijbehorende geleidingsstaat kan beïnvloeden, zowel als uitgeoefende elektrostatische potentialen. Het geheel compliceert, door de extrapolatie van de gemeten optische eigenschappen bij lagere frequenties via het Drude model of Plasma modellen, wanneer de onzekerheid in de Casimirkracht, die reeds meer dan 15 jaren bekend is, berekend wordt. De laatste genoemde modellen kunnen beide versterkt of onderdrukt chaotisch gedrag

voorspellen, ook sterk afhankelijk van de geleiding van de interacterende materialen.

Het onderzoek gepresenteerd in dit proefschrift belicht het belang van de eigenschappen der oppervlakten in een micro-nano systeem waar Casimir-krachten zich manifesteren. De gecombineerde experimentele en theoretische onderzoeken in dit werk kunnen het pad vrij maken voor toekomstige wetenschappers om klein geschaalde technologieën te ontwerpen en te engineeren, evenals het nieuwe perspectieven mag brengen voor onderzoekers de fundamentele studies op krachten tussen complexe materialen najagend.

Summary

Today, the Casimir force that originates from the perturbation of electromagnetic vacuum fluctuations is still a topic of relentless research, though its proposition in 1948 by the Dutch physicist Hendrik Casimir dates back almost 70 years. The interest stems from a multitude of research fields ranging from fundamental physics in search of new forces beyond the standard model to micro/nanodevices for technology applications. Although the Casimir force has been extensively studied over the last two decades, yet it has not been explored in many areas involving complex materials, which are of high importance for applications. In fact, comprehensive knowledge of Casimir forces and associated electrostatics from complex conductive surfaces can be essential in diverse areas ranging from micro/nanodevice operation in harsh environments to multilayer coatings in advanced lithography technologies. In this thesis, our efforts to show the effect of surface composition, roughness, and electrostatic contribution on the measured Casimir force are reported together with analyzing the influence of the optical properties and different ways of data extrapolation on the theoretical calculations.

We have performed Casimir force measurements and associated electrostatics from conductive SiC and Ru surfaces. The electrostatic calibration showed that the Si- and C-faces behave differently with a nearly $\sim 0.6\text{--}0.7\text{ V}$ difference in the contact potentials $V_0^{\text{Si/C}}$ which can be attributed to a higher incorporation of N on the C-terminated face. However, the measured optical properties of the SiC-Si/C terminated surfaces with ellipsometry did not show any substantial differences, indicating that the effective depth of the Si/C terminating surface layers are significantly smaller than the photon penetration depth, without leading to any differences in the calculated forces via Lifshitz theory. Nonetheless, the measured Casimir forces, after compensation of the electrostatics contributions, showed differences between the Si/C faces,

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whereas the comparison with the Lifshitz theory prediction shows better agreement for the SiC-Si face. Finally, comparison of the Casimir forces below 40 nm separations between the SiC-Si/C and Ru surfaces indicated that the short-range roughness effects on the Casimir force increase in magnitude with increasing metallic behavior of the plate surface. Therefore, not only the material optical properties but also the conductive state and roughness of the surface layers must be carefully taken into account in short range Casimir interactions between more complex dielectric materials.

Given the potential of the newly discovered topological insulators in the emerging technological devices, we explored the decisive role of film thickness on Casimir and electrostatic forces from topological insulating Bi_2Se_3 films. The forces were measured for the Au- Bi_2Se_3 system under ambient conditions to simulate realistic applications. It is shown that the electrostatic forces associated with a non-zero contact potential of $V_0 \sim 200$ mV played a dominant role, and could not be compensated for thinner films (less than 20 nm in thickness). However, for thicker films the measured force, after voltage compensation, started to approach the genuine Casimir force in very good agreement with Lifshitz theory. Our results point out that any gradual deviation from theory for thinner films underlies the presence of strong electrostatic effects.

The detailed knowledge about the phenomena of stiction due to Casimir interactions, and its relation to chaotic motion during actuation is necessary in order to improve the long-term performance of devices. It is shown here that the non-equilibrium Casimir forces, which are the most general case when the interacting components have different temperatures, can influence the actuation of devices towards unstable and chaotic behavior in strong correlation with their optical properties, and associated conduction state, as well as applied electrostatic potentials. The situation becomes more complicated by the uncertainty in calculating the Casimir, which is known for more than 15 years now, due to the extrapolation of the measured optical properties at low frequencies via the Drude or the Plasma models. The latter models can predict either enhanced or suppressed chaotic behavior depending strongly also on the conductivity of the interacting materials.

The research presented in this thesis highlights the importance of the characteristics of the surfaces involved in a micro-nano system where Casimir force manifests itself. The combined experimental and theoretical studies in this work can pave the path for the future scientist to design and engineer small-size technologies, as it may bring new perspective for the researchers pursuing fundamental studies for forces between complex materials.

Acknowledgements

Here, I am going to express my gratitude and regards to my family, friends and colleagues without whom the completion of this thesis would not be possible.

First and foremost, thank you *George* for offering me this position and then for more than four years of support, encouragement, supervision and of course educating me to be a better researcher/person. *Bart*, I really enjoyed being part of the group you are leading. Moreover, your animal-loving attitude inspired me to become vegetarian. *Vitaly*, without your scientific input and collaboration, this thesis would not be like this, thank you for patiently guiding me through the theoretical aspects of our work. And *Gert*, I already missed all our culture-oriented discussions and of course, thanks for all the technical supports.

Furthermore, I would like to thank my assessment committee, prof. L. M. Woods, prof. dr. Steven Hoekstra and prof. dr. Katja Loos for taking the time and effort to evaluate my thesis.

Being a member of the NMI group, I was delighted spending time with colleagues who became friends: *Xiaotian*, you were the best officemate ever, *Mehdi*, *Jamo*, *Paul*, *Bin*, *Lijuan*, *Xukia*, *Fatemeh*, *Farnaz*, *Daniel*, *Dennis*, *Sytze*, *Weiteng*, *Heng*, *Laura*, *Razihe*, *Julien* and *Majid*.

But the fun and the joy in the last four years in Groningen was not limited to NMI, outside the group I had shared a lot of memorable and unforgettable moments with friends, that even thinking about them makes me cheerful. *Talieh*, the highlight of our friendship was the moment we found out we are twins being born the same day (how is it possible?) and then all the

laughs and fun we had together, my PhD life would not be as joyful without you. *Hamid*, thank you for being such a caring friend for me and of course thanks for all the soups. *Dennis*, you are such a life-saver, I am still thinking of a nice wine to pay back your most recent favor: moving to Utrecht. I am furthermore delighted and grateful for the time I shared with *Martijn*, *Julian*, *Pep*, *Azadeh*, *Soheil*, *Masoud*, *Pouya*, *Carmem*, *Obed*, *Alexey*, *Antonio*, *Jorge*, *Christian*, *Hamoon*, *Fatemeh* and *Farnaz*.

My dear friends *Koorosh* and *Xu*, you know that you are granted an honor to be my paranymphs but thank you for accepting such a challenge. Honestly, it was after moving to Utrecht that I realized you both are such great friends/cooks. Thank you for always being there for me.

Tim, I am always the better version of myself when I am with you. The best moment of my PhD life is undoubtedly when I met you at the Seminar in Germany. Thank you for all the support and love.

Finally, my dear *family*, thank you for all the care, trust, support and love you gave me all my life. And to the one amazing woman: my beautiful, loving and selfless *Mami*, you are the sunshine of my life, I cannot think of any glamorous word to be even close enough to describe your unconditional love. You are simply extraordinary. I love you immensely from the bottom of my heart.

Zahra Babamahdi
Utrecht, the Netherlands
August 4, 2021

List of publications

1. Z. Babamahdi, V. B. Svetovoy, M. Enache, M. Stöhr, and G. Palasantzas, *Comparison of Casimir Forces and Electrostatics from Conductive SiC-Si/C and Ru Surfaces*, Phys. Rev. B **100**, (2019).
2. Z. Babamahdi, V. B. Svetovoy, D. T. Yimam, B. J. Kooi, T. Banerjee, J. Moon, S. Oh, M. Enache, M. Stöhr, and G. Palasantzas, *Casimir and Electrostatic Forces from Bi₂Se₃ Thin Films of Varying Thickness*, Phys. Rev. B **103**, L161102 (2021).
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