Spintronics and thermoelectrics in exfoliated and epitaxial graphene

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Summary

Spintronics in epitaxial graphene

One of the long term goals of the field of spintronics is the use of the electron spin as a carrier for information. To create a network of communicating spin devices, a spin accumulation must be maintained between all the connections of the network. It would then be possible to perform logic operations by manipulating the orientation of the spins. To achieve these goals, it is necessary to first expand existing knowledge of the behavior of spin carriers in the solid state environment.

One important parameter that characterizes the performance of a spintronic material is its spin relaxation length, which is the typical distance electrons can travel before losing their initial spin orientation due to interactions with the environment. Applying a magnetic field induces spin precession, an effect used in Hanle precession experiments to determine spin transport properties. The spin relaxation time, the spin diffusion coefficient and the spin relaxation length can be determined from the experimentally measured Hanle curve. This is done by a curve fit with the solution of the Bloch equation, which describes the spin dynamics in the material. Investigating the Hanle curve gives valuable insights about the suitability of the material for spintronic applications. Moreover, it can also be used as a characterization tool to obtain information about various processes in the material.

Graphene is an interesting material for spintronics because of its a spin relaxation length of several micrometers at room temperature, longer than in any other material. Most graphene in research is produced by repeatedly cleaving graphite until an atomic monolayer is reached, a method known as mechanical exfolation. It is also possible to grow wafer scale quantities of good quality monolayer graphene by thermal decomposition of a silicon carbide (SiC) substrate. This material, called epitaxial graphene due to its correlation with the underlying SiC, is typically grown on either the silicon terminated (0001) face or the carbon terminated $(000\bar{1})$ face of the SiC crystal. Epitaxial graphene on SiC(0001) has a nonconducting, carbon monolayer in between the SiC and the graphene, called the buffer layer. Due to charge doping, it has a great influence on the transport properties of the graphene. The buffer layer

is absent in epitaxial graphene on SiC(000$\bar{1}$).

To investigate the spin transport properties of epitaxial graphene on SiC, we developed a fabrication method to prepare nonlocal spin valve devices on a standard epitaxial graphene chip. We measured these devices in Hanle precession experiments and observed a very narrow Hanle curve, which is associated with a high spin relaxation time if one applies conventional Hanle analysis by fitting with the standard Bloch equation. We also observed a very low spin diffusion coefficient, which was much lower than the charge diffusion coefficient that we independently obtained using standard Hall measurements.

We explained these observations with a spin transport model for a diffusive channel with coupled localized states. We described the coupled system by an effective Bloch equation with an increased precession frequency and a reduced spin relaxation time, resulting in a narrowing of the Hanle curve. We then showed that this effect was absent in epitaxial graphene on SiC(0001) where the buffer layer was removed by hydrogen intercalation and could thus conclude that the effect of the localized states is strongly associated with the buffer layer.

In a different experiment, we observed not only a narrow Hanle curve, but also a change of the general shape of this curve, thereby making the fitting procedure with the solutions of the Bloch equation unusable. We showed that a shape change occurs when the coupling rate to the localized states is in a regime where it is comparable to the relaxation rate. We compared epitaxial graphene that was grown by sublimation with epitaxial graphene grown by chemical vapor deposition, and observed a strong difference between the two. Thus, we extracted information about the localized states associated with the structural differences related to different growth methods.

Next, we offered a more detailed explanation by extending the model with the assumption that the localized states have a distribution of trapping times or, similarly, coupling rates. Based on a model for materials with dispersive charge traps, we incorporated a power-law distribution of trapping times to explain some typical features of the changing shape of the Hanle curve. We simulated the temperature evolution of the Hanle curve and estimated the energy dependence of these trapping times from the Hanle precession experiments.

With an experimental study on epitaxial graphene on SiC(000$\bar{1}$), we observed an increasing deviation between the charge diffusion coefficient and the spin diffusion coefficient, though the saturated effect was less strong than on SiC(0001). Thus, we concluded that localized states can also be present in a system without a buffer layer, probably due to charge traps at the graphene-substrate interface.

Graphene thermoelectrics

Thermoelectric phenomena are the combined effect of heat and charge flows. A well-known example is the generation of an electrical current due to a heat gradient, an effect called the Seebeck effect. The strength of this effect in a given material is given by its Seebeck coefficient. Efficient thermoelectric materials are exploited in thermocouples and can be used to generate electrical power from waste heat. The reciprocal of the Seebeck effect is the Peltier effect, which can be described as the transport of heat by an electric current. Its effectiveness is described by the Peltier coefficient, which is closely linked to the Seebeck coefficient. The Peltier can be used to heat or cool an interface between two materials with different Peltier coefficients.

The thermoelectric coefficients in graphene are tunable, because they are determined by the energy dependence of the conductivity. In graphene, the energy of the electons can be controlled with an applied gate voltage, making it possible to change the charge carrier density and even their polarity. At the Dirac point, the energy level at which the charge carriers switch from negatively charged electrons to positively charged holes, the Seebeck and Peltier coefficient change sign. When inducing the Peltier effect at a graphene-metal junction, this manifests itself as a transition from cooling to heating of the interface.

We designed and fabricated a device to measure the electronic cooling and heating via the Peltier effect of a graphene-gold interface. The measured temperature modulation was up to 15 mK, which we probed with very sensitive thermocouples directly at the junction. We quantified the measured temperature differences by estimating all the heat flows in the device. We showed that the temperature changes were related to the Peltier effect, because of its sign change for electron and hole transport and because of the linear scaling with the applied current, which is different from the common Joule heating that scales quadratically.

The measurement of the Peltier effect in graphene can be used to investigate fundamental thermodynamic relations in graphene, such as the validity of Onsager reciprocity, which describes the equivalence of the Seebeck and the Peltier coefficient. The combination of this electrical measurement scheme and our simple and intuitive model could also be of use for investigating refrigeration effects in more complex thermoelectric nanomaterials.