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# Scientific summary

## Optical preparation and detection of spin coherence in molecules and crystal defects

This thesis presents the results of research aimed at exploring several possibilities of optically-induced electron spin coherence in molecules and crystal defects. In our approach we have performed a theoretical investigation of underlying fundamentals and forthcoming requirements (Chapter 2-4), as well as an experimental investigation of a system demonstrating optically induced electron spin coherence (Chapter 5). We believe that the scientific progress of our work contributes to a better characterization of various materials with opto-electronic functionality and provides better probing tools. Also, we believe our work is relevant for new quantum-information functionalities based on the selective coupling of photons to electronic spin states.

For atoms and crystals with a high symmetry, the optical selection rules for electronic transitions are well covered in physics textbooks. However, how optical selection rules change when the high symmetry is gradually distorted is less understood. In Chapter 2, we present a detailed analysis of how a gradual symmetry distortion leads to a complete alteration of optical selection rules. As a model system, we consider the transitions between  $1s$  and  $2p$  sublevels of the hydrogen atom, which get distorted by placing charged particles in its environment. Upon increasing the distortion, part of the optical selection rules evolve from circular via elliptical to linear character, with an associated evolution between allowed and forbidden transitions. Our presentation combines an analytical approach with quantitative results from numerical simulations, thus providing insight in how the evolution occurs as a function of the strength of the distortion. This study provides a useful theoretical framework for more complex systems. Moreover, it paves the road for manipulation of optical selection rules. The ability to manipulate selection rules allows to better control the interaction of photons and electrons, potentially allowing for new ways to control the flow of information

within the field of quantum information.

Organic molecules are increasingly used for opto-electronic devices, because of their chemical tunability, low-cost and ease of processing. In such devices, the ratio of singlet to triplet excitons can be an important performance parameter. Moreover, because of the many interesting spin-related phenomena discovered in organic semiconductors and molecules, their application into organic spintronics is very promising. Both for organic opto-electronics and spintronics, being able to control and probe triplet spin states will be of great value. Optical polarization proves to be a promising candidate for this. One way to study correlations between spin and optical polarization is the Time-Resolved Faraday Rotation (TRFR) technique. Within this all-optical non-invasive pump-probe technique, a pump pulse induces spin polarization, after which the spin dynamics are probed by the polarization rotation of a probe pulse. In Chapter 3, we investigate how this technique allows for optical control and probing of triplet-exciton spin dynamics in metal-organic molecules. In our theoretical analysis, an ultrashort polarized pump pulse brings the molecular system in a superposition of triplet excited state sublevels. We derive how the polarization of a comparable but detuned probe pulse is affected upon transmission, from which the requirements for polarization rotation follow. Using the results of *ab initio* calculations, we calculate the time dependence of the polarization rotation angle and of the expectation value of the total electronic angular momentum. Both show an oscillation with a frequency corresponding to the sublevel splitting, implying that the oscillation of polarization rotation is a suitable measure for the coherent spin dynamics.

Color centers in wide-bandgap semiconductors are attractive systems for quantum technologies since they can combine long-coherent electronic spin and bright optical properties. Several suitable centers have been identified, most famously the nitrogen-vacancy defect in diamond. In Chapter 4, we propose to use the TRFR technique (usually being applied to materials with strong spin-orbit coupling) to characterize spin-active color centers in materials with negligible spin-orbit coupling, like silicon carbide and diamond. The fundamentals and scenario for a TRFR experiment are worked out in detail for a homogeneous ensemble of *c*-axis divacancies (pairs of missing neighboring silicon and carbon atoms along the growth axis) in silicon carbide. Under certain circumstances, the material is birefringent (two different refractive indices). Moreover, one of the indices of refraction oscillates as a function of time in the presence of coherences. Due to this time-dependent birefringence, a probe pulse will undergo a polarization rotation as a function of the pump-probe delay time. This polarization rotation is

a measure for the spin coherence of the triplet excited state.

Integration of semiconductor color centers in communication technology is hindered by the fact that their optical transitions lie outside telecom wavelength bands. Several transition-metal impurities in silicon carbide do emit at and near telecom wavelengths, but knowledge about their spin and optical properties is incomplete. In Chapter 5, we present all-optical identification and coherent control of molybdenum-impurity spins in silicon carbide with transitions at near-infrared wavelengths. Our results identify spin  $S = 1/2$  for both the electronic ground and excited state, with highly anisotropic spin properties that we apply for implementing optical control of ground-state spin coherence. Our results show optical lifetimes of  $\sim 60$  ns and inhomogeneous spin dephasing times of  $\sim 0.3$   $\mu$ s, establishing relevance for quantum spin-photon interfacing.

