Booklet for the Ph.D. thesis

Exotic magneto-optical effects in solids

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Introduction

The entanglement between the spin, lattice and charge degrees of freedom results in intriguing cross-effects in solids. Beside the fundamental interest, the manipulation of different physical properties by external stimuli, such as temperature, pressure, electric or magnetic field, is also attractive for technological applications. The existence of considerably large cross-effects is a prerequisite for such applications. Cross-effects with sufficiently large magnitude for application emerge in those phases of matter where the different degrees of freedom are strongly coupled and order cooperatively. In this context the so-called multiferroic materials are investigated extensively, since the coexistence of magnetism and ferroelectricity allows the effective control of the ferroelectric polarization by magnetic field and the magnetization via electric field.

Using the magneto-optical effects in solids, either the intensity or the polarization state of light can be manipulated by external magnetic fields. The polarization rotation of a light beam upon transmission through a magnetic material or reflection from its surface are termed as Faraday effect and magneto-optical Kerr effect, respectively. These phenomena are caused by the inequivalence between the propagation of the two circularly polarized states of the light as a consequence of the broken time reversal symmetry. More precisely, the spin imbalance corresponding to the finite magnetization of the material results in an asymmetry in the charge excitation spectra for the left- and right circularly polarized photons owing to the spin-orbit coupling. Therefore, the magneto-optical spectroscopy is an efficient tool to deduce the spin polarized band structure of solids and can provide information about fundamental physical quantities such as the strength of the Coulomb interaction, the crystal field splitting, the spin-orbit coupling and the magnetic exchange interactions. This method when combined with microscopy is widely used for the high-sensitivity detection of the local magnetization and recently for the investigation of spin dynamics on the picosecond time scale.

In the last decades, novel magneto-optical effects have been explored in materials with low spatial symmetry. The so-called directional anisotropy, when the speed and the absorption of the light are different for counter propagating beams irrespective of their polarization, emerges if both the time reversal and spatial inversion symmetry are lost. Though this phenomenon would be interesting from technological point of view, its small magnitude in usual materials do not make them attractive for real applications.

The goal of the research

My PhD work had dual-purpose. My first aim was to reveal the nature of corss-coupling effects between the different degrees of freedom in solids. In this study, I have used magneto-optical and optical spectroscopy to probe the charge, lattice and spin excitations in compounds with complex magnetic order over a broad energy range. Following the structure of the dissertation, I have investigated the origin of the magneto-optical activity in metals both with collinear and non-collinear spin structure in the region of the inter- and intraband charge excitations. Then, I have studied magneto-elasticity, associated with the crystal symmetry lowering induced by magnetic ordering in chromium spinel oxides, via the observation of magnetically induced splittings for the infrared active lattice vibrations. Finally, I have investigated the magnetic field dependence of the absorption spectra for spin excitations in a recently recently synthetized multiferroic oxide $Ba_2CoGe_2O_7$ to reveal the nature of the spin dynamics when strongly coupled to electric polarization.

The second target of my PhD work was to find materials which exhibit exotic magneto-optical effects and the tailoring of novel compounds with unique optical functionalities. In $Ba_2CoGe_2O_7$, I found giant directional anisotropy in its multiferroic phase. Moreover, I demonstrated that due to its special non-centrosymmetric crystal structure a chiral state can be established by the application of external magnetic fields. In this phase, I have studied the chirality induced polarization rotation and directional anisotropy.

Experimental techniques

In order to study the excitations of charge, lattice and spin degrees of freedom I performed optical and magneto-optical spectroscopy over a wide photon energy range.

For conventional optical spectroscopy, I have used commercial spectrometers. On the other hand, I have developed a unique setup for magneto-optical spectroscopy which combines a grating and an FT-IR spectrometer to cover the energy range of E=0.1-4 eV. I have implemented a polarization modulation technique to measure the polarization rotation with a precision of $\theta = \pm 0.005\,^\circ$.

I have investigated the spin excitations by terahertz time-domain spectroscopy (THz-TDS), which is a recently developed methodology based on femtosecond LASERs. In contrast to conventional electron spin resonance experiments performed in a cavity resonator, this allows the continuous variation of the excitation energy and offers straightforward way to determine the change in the polarization state of light via the direct measurement of the complex transmission coefficients.

Thesis points

The major achievements of my Ph.D. work are summarized in the following thesis points:

1. I have studied the spin-wave excitations of the multiferroic oxide $Ba_2CoGe_2O_7$ over a broad region of the temperature-magnetic field phase diagram by means of terahertz time-domain spectroscopy. On the basis of the selection rules determined from the polarized absorption spectra I have concluded, that one of the two magnon modes, observed sub-terahertz frequencies, can be excited both by the electric and the magnetic component of the light, while the other mode is only sensitive to the magnetic component of the radiation. I have found large directional dichroism, $\Delta\alpha/\alpha \sim 50\,\%$, in the former case. I have shown that the large directional dichroism is the consequence of the entanglement between the electric and the magnetic dipole excitations i.e. the so-called electromagnon nature. [1]

2. I have investigated the character of the spin excitations in $Ba_2CoGe_2O_7$ by measuring the natural optical rotation and ellipticity of the transmitted light in the terahertz frequency region. I have found that magnetic fields applied along the [100] or the [010] directions can induce chirality in this material. I have demonstrated that the rotation of the field direction from [100] to [010] axis can reverse the chirality of the crystal as expected from a symmetry argument. As a further consequence of the field-induced chiral state in this multiferroic material, I found exceptionally strong magnetochiral effect, as large as $\Delta\alpha/\alpha \sim 20\,\%$. [5]

3. I have studied the lowering of the symmetry upon the magnetic phase transitions in various ACr_2O_4 chromium spinel oxides by investigating the far-infrared reflectivity of the optical phonon modes. $MnCr_2O_4$ and $CoCr_2O_4$ remain cubic down to T=10 K within the accuracy of the experiments, while the symmetry of $FeCr_2O_4$, $NiCr_2O_4$ and $CuCr_2O_4$ reduced at least to orthorhombic in the magnetically ordered phase. I have pointed out that the lattice symmetry is lowered in the magnetically ordered phase of those compounds in which the A-site cations have orbital degeneracy at high temperatures. [2]

4. I have participated in the development of a broad band magneto-optical spectrometer covering the photon energy range E=0.1-4 eV which allows the determination of the magneto-optical Kerr parameters with the accuracy of $\theta = \pm 0.005°$. Using this spectrometer I measured the magneto-optical Kerr effect in $CuCr_2Se_4$ at various temperatures. In $CuCr_2Se_4$, from the Kerr and the reflectivity spectra I have evaluated the diagonal and the off-diagonal elements of the optical conductivity tensor, which were compared to theoretical band structure calculations. Based on the agreement between my experimental results and the band structure calculations, I have proposed that the low-energy excitation, clearly discerned in the off-diagonal conductivity at E≈0.5 eV, corresponds to a hybridization induced gap, which is responsible for the nearly half-metallic band structure and the large off-diagonal conductivity of $CuCr_2Se_4$. [3]

5. In the case of the electron and hole doped $Nd_2Mo_2O_7$ compounds, I have investigated the origin of the anomalous Hall effect by measuring the off-diagonal conductivity, σ_{xy}, at optical frequencies. To reveal the low-energy behavior of the magneto-optical spectra in doped $Nd_2Mo_2O_7$, I used the same home-designed broad band spectrometer introduced in the previous thesis point. I have found that the magnitude and the peak position in the low-energy part of the off-diagonal conductivity spectra (deduced from the magneto-optical spectra) correlate with the anomalous Hall effect measured in the dc limit. I proposed that the two effects have a common origin, namely a band anticrossing point determines the low-energy behavior of $\sigma_{xy}(\omega)$. Based on this assumption, I have reproduced the experimental spectra based on a two band model of a band anticrossing point. [4]

Publications related to the thesis points

[1] I. Kézsmárki, N. Kida, H. Murakawa, S. Bordács, Y. Onose, and Y. Tokura
Enhanced Directional Dichroism of Terahertz Light in Resonance with Magnetic Excitations of the Multiferroic $Ba_2CoGe_2O_7$ Oxide Compound
Physical Review Letters 106, 057403 (2011)

[2] S. Bordács, D. Varjas, I. Kézsmárki, G. Mihály, L. Baldassarre, C. A. Kuntscher, K. Ohgushi, Y. Tokura
Magnetic-order-induced crystal symmetry lowering in ACr_2O_4 ferrimagnetic spinels

Physical Review Letters 103, 077205 (2009)

[3] S. Bordács, I. Kézsmárki, K. Ohgushi, and Y. Tokura
Experimental band structure of the nearly half-metal $CuCr_2Se_4$: an optical and magneto-optical study
New Journal of Physics 12, 053039 (2010)

[4] S. Iguchi, S. Kumakura, Y. Onose, S. Bordács, I. Kézsmárki, N. Nagaosa, and Y. Tokura
Optical Probe for Anomalous Hall Resonance in Ferromagnets with Spin Chirality
Physical Review Letters 103, 267206 (2009)

[5] S. Bordács, I. Kézsmárki, N. Kida, H. Murakawa, L. Demkó, Y. Onose, R. Shimano, S. Miyahara, N. Furukawa, and Y. Tokura
Chirality of Matter Shows up via Spin Excitations
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Further publications

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Magneto-optical characterization of the ferromagnetic-paramagnetic phase boundary in the composition-spread epitaxial film of $Sr_{1-x}Ca_xRuO_3$
Applied Physics Express 1, 113001 (2008)

[10] L. Demkó, I. Kézsmárki, M. Csontos, S. Bordács, and G. Mihály
Improved thermal relaxation method for the simultaneous measurement of the specific heat and thermal conductivity
The European Physical Journal B 74, 27 (2010)

[11] L. Demkó, B. Dóra, T. Vojta, S. Bordács, H. Yamada, M. Kawasaki, Y. Tokura, and I. Kézsmárki
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submitted to Nature Physics