Magnetoelectric multiferroics: From static via dynamic magnetoelectric effect to nonlinear light-matter interaction

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The main goal of my PhD work was to study the dynamic magnetoelectric effect in various multiferroic and magnetoelectric compounds. My aim was not only to observe this effect but also to control it by static fields as well as by the oscillating electromagnetic fields of the THz radiation. Multiferroics with simultaneous ferroelectric and magnetic order attracted interest in the past two decades as they allow magnetic-field control of electric polarization, and electric-field control of magnetic states. The latter effect, if realized in an insulating magnet, provides a huge technological advantage for information storage. In a multiferroic memory device, the energy dissipated by Joule heating is expected to be 2–3 orders of magnitude smaller as compared to for example spin-transfer-torque magnetic random access memories.

The magnetoelectric coupling gives rise to intriguing phenomena at finite frequencies as well. Notably, magnetoelectric crystals can be transparent in one, but absorbing in the opposite light propagation direction, thus, these materials may gain applications in optical diodes.

Despite the advantages of the electric field control of the magnetism, it has some drawbacks as it requires contacts and its speed can be limited. The optical manipulation of magnetism can help to overcome these issues. Contactless and fast all-optical access of writing and reading magnetic states of matter, attracted enormous interest in last two decades. Magnetoelectric materials may allow a new combination of these two fields as the coupled electric and magnetic dipole excitations provide a new handle to manipulate magnetic states.

I demonstrated isothermal control of the directional dichroism, i.e. absorption difference for counter-propagating light beams, in a multiferroic $Ba_2CoGe_2O_7$ single crystal by an electric field and tiny rotation of the crystal with respect to a magnetic field. By studying the electric-field ($\boldsymbol{E} \parallel [100]$) and temperature dependence of the absorption contrast using Fourier-Transform Infrared and time-domain THz spectroscopy, I revealed that the domains of the easy-plane antiferromagnetic structure play an essential role in the electric-field effect. By studying spectra upon magnetic field rotation around [100] and [010] axes, I proposed specific scenarios of domain transformations.

In multiferroic Y-hexaferrite $BaSrCoZnFe_{11}AlO_{22}$ and Z-hexaferrite $Ba_{0.5}Sr_{2.5}Co_2Fe_{24}O_{41}$, I found purely electric-dipole-active magnons, so-called electromagnons, by measuring and comparing their THz and Raman spectra. I studied their temperature and magnetic field dependence, and correlated their features with the changes in the static magnetic structures. I developed microscopic selection rules based on the exchange-striction mechanism of electromagnon activations, which allowed me to explain the temperature and magnetic-field dependences of their spectral strength. Further, I investigated the influence of intense THz radiation on electromagnons hexaferrites, while I observed only unclear spectral changes.

In a magnetoelectric $LiCoPO_4$ single crystal, I measured directional dichroism as it is defined, i.e. by the reversal of the light propagation direction. Applying this method, I identified antiferromagentic domains following magnetoelectric annealing in electric ($\boldsymbol{E} \parallel [010]$) and magnetic ($\boldsymbol{H} \parallel [100]$) fields selecting either of the domains. Furthermore, I selected a magnetoelectric domain in $LiCoPO_4$ single crystal by intense THz radiation tuned to frequencies of magnetoelectric excitations. I found that counter-propagating light pulses selected the same domain, which implies that the asymmetry introduced by the sample holder is important. Therefore, I ascribed the observed effect to temperature gradient and subsequent propagation of magnetoelectric quasiparticles, which must be nonreciprocal in the magnetoelectric domains.