Coherent nonlinear optical spectroscopy of spin effects in semiconductors and magnetic materials

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Among a broad variety of nonlinear optical phenomena, quite a distinguished role can be attributed to those processes which lead to the frequency conversion. The simplest examples are coherent processes of 2nd, 3rd and higher order optical harmonics generation. They are defined by higher order susceptibilities and therefore are more symmetry sensitive in comparison to linear optical phenomena. For example, second harmonic generation (SHG) is strongly anisotropic even in cubic crystals. In this talk we are going to discuss how magnetic field, spins and spin ordering can contribute to harmonics generation in several groups of semiconductors and magnetic materials.

Until recently, there were no detailed studies of harmonics generation at low temperature in broad spectroscopic range in applied magnetic fields. We performed such studies in fields up to 10 T at 2 K and higher temperatures. In GaAs, we found that application of a magnetic field induces new features in the optical SHG spectrum above the band gap [1]. A series of sharp SHG lines was observed in the spectral range from 1.52 to 1.77 eV that we attributed to Landau-level quantization of the band energy spectrum. Calculations revealed that magneto-optical spatial dispersion that comes together with the electric-dipole term is the dominant mechanism of this nonlinearity.

Basically different mechanisms of optical SHG induced by an external magnetic field were identified experimentally by studying the diluted magnetic semiconductors (Cd,Mn)Te [2]. For paramagnetic compounds the SHG response is governed by spin quantization of electronic states. The mechanisms can be identified by the distinct magnetic field dependence of the SHG intensity which scales with the spin splitting in the paramagnetic case as compared to the B^2 dependence in the diamagnetic case.

The third group of materials we discuss is europium chalcogenides EuSe and EuTe [3]. They are magnetic semiconductors with crystallographic and magnetic structures distinctly different from those of GaAs and CdTe. $4f^7$ states of Eu^{2+} ions lie above the $4p^6$ and $5p^6$ states of Te^{2-} ions in EuSe and EuTe, respectively. Analysis of the electronic structure and selection rules showed that SHG is forbidden in the electric dipole and electric quadrupole approximations. Nevertheless in EuSe and EuTe we observed SHG signals in the presence of an applied magnetic field in the vicinity of the band gap of 2.2-2.4 eV. Detailed study allowed us to conclude that the SHG signals arise due to the *ferromagnetic* component of the magnetic structure. ZnO is known for its complicated exciton structure which we studied in zero and applied magnetic field. We found that the applied magnetic field can induce new contributions to the SHG by itself and due to the so-called magneto-Stark effect.

We shall discuss some examples of harmonics generation by spontaneous spin ordering (no magnetic field applied) in antiferromagnetic and ferromagnetic materials.

This work is supported by the RFBR, the DFG-RFBR joint project, and the Programs of the Russian Academy of Sciences.

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