Generation of Strong Isolated Transient Magnetic Fields for Magneto-Optical Spectroscopy

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Traditional optical spectroscopy relies on the interaction of the electric field (EF) component of light with the atomic or molecular (transition) electric dipole (ED) moment. Magnetic dipole (MD) interactions contribute to the optical spectrum by ~2 orders of magnitude less than the ED interactions, therefore usually remain concealed. MD transitions follow different selection rules from ED transitions giving complementary information about the matter. In this work, we propose a novel type of magneto-optical spectroscopy based on the selective probe of the magnetic-field (MF)–MD interactions.

In order to investigate predominantly MD transitions, it is necessary to enhance the MF-component and thus to improve the MF/EF-contrast. This can be achieved through (1) spectral selectivity, e.g. by choosing a system exhibiting sufficient spectral separation between the ED and MD transitions, and (2) through spatial separation of the EF and MF components of the optical pulses. The optical spectrum of Eu^{3+} ions incorporated in different hosts exhibits a MD-allowed transition at 527.5 nm and an ED-allowed one at 532 nm [1], making this system a model spectroscopic target. To achieve spatial separation of the MF- and EF-components of the optical fields we employ two different approaches.

First, the spatial isolation the MF- from EF-component relies on the generation of azimuthally polarized beams (APB). Under external focusing, the APB develops a doughnut-shaped intensity profile with a longitudinal MF-component propagating along the optical axis as the polarization of the EF oscillates along the circular beam perimeter. Additionally, MF enhancement can be achieved through the interaction of the oscillating EF with thin-film ring-shaped metal apertures, resulting in oscillating ring-currents in the metal. The second method of achieving the spatial separation relies on two counter-propagating beams forming a standing wave, where the MF- and EF-components of the optical field are separated in space. By varying a phase delay between the beams, the components of the EF or MF of the light can be shifted with respect to thin-film samples containing Eu^{3+} ions.

We experimentally demonstrate the generation of narrowband wavelength-tunable APBs for the sole excitation of MD and ED transitions in Eu^{3+} ions. Wavelength tunability is achieved by stimulated Raman scattering governed spectral shift of fs μJ pulses in an air-filled photonic crystal fiber [2] and subsequent spectral focusing when generating second harmonics (SHG) in a 2.5-cm long β-barium borate (BBO) crystal [3]. The APBs are generated by employing a birefringent spiral varying retarder in combination with a set of half and quarter waveplates [4]. As a probe of Eu^{3+} ions embedded on surfaces of solid samples and selectively excited via MD or ED transitions we will employ time resolved luminescence spectroscopy.

In the case of the counter-propagating beams, the interaction of the EF and MF with thin films containing Eu^{3+} ions will be investigated by means of pump-probe transient XUV-absorption spectroscopy.

References

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