Charged and Neutral Excitons in Perovskite $CsPbBr_3$ Nanocrystals and Bulk Material in High Magnetic Fields: Spin Structure, Spin Dynamics and Exciton Parameters

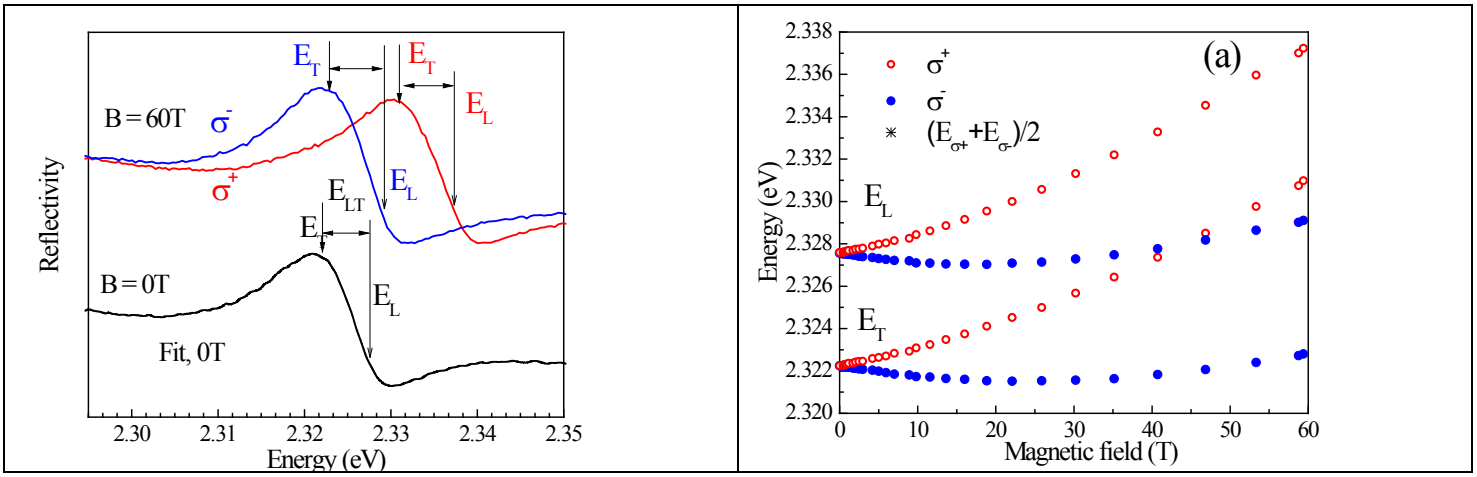
Yakovlev, D.R. (TU Dortmund University, Germany), Semina, M.A. (Ioffe Institute, St. Petersburg, Russia), Crooker, S.A. (NHMFL-Los Alamos)

Introduction

There is a strong recent interest in perovskite materials and their nanostructures in view of photovoltaic applications. Despite the fact that some of these materials, namely inorganic perovskites ($CsPbBr_3$, $CsPbCl_3$, etc.) are known from the 1970s, information on their optical and magneto-optical properties is rather limited, especially in the field of exciton properties and spin dynamics.

Experimental

Reflectivity and polarized photoluminescence has been measured on $CsPbBr_3$ high-quality bulk samples and colloidal nanocrystals (NCs). Experiments in magnetic fields up to 10 T have been performed in Dortmund and up to 65 T in Los Alamos. Oscillator strength, diamagnetic shift, Zeeman splitting of the ground state (1s) exciton were measured. Also 2p exciton state has been addressed and identified by two-photon excitation and its diamagnetic shift in fields to 10 T has been measured. Coherent spin dynamics of carriers has been measured by pump-probe Kerr rotation in Dortmund and the measured g-factors for electrons and holes have been compared with the exciton g-factor measured from measurements to 65 T in Los Alamos.



Results and Discussion

Reflectivity spectra of bulk $CsPbBr_3$ in zero and 60 T magnetic field measured at 1.6 K are shown in the left panel. Strong resonance of the 1s exciton-polariton is clearly seen, the evaluated parameters are energies of the transverse and longitudinal polaritons: E_T = 2.322 eV, and E_L = 2.327 eV, and longitudinal-transverse splitting of 5-4 meV. In magnetic fields both energies show a diamagnetic shift of 0.4 μeV/T^2 and Zeeman splitting (evaluated exciton g-factor is 2.45). As the exciton binding energy exceeds the LO phonon energy, the question of the effective dielectric constant describing the exciton properties is left open. For full evaluation of the exciton parameters we have measured the energy of the 2p exciton at 2.347 eV and its diamagnetic shift of 10 μeV/T^2. This allows us to perform modeling without fitting parameters and estimate effective dielectric constant of 7.8 for the 1s state and 9.8 for the 2p state, and also determine the exciton binding energy of 29.5 meV with high precision.

For $CsPbBr_3$ colloidal nanocrystals, polarized photoluminescence has been measured to 65 T and magnetic field dependence of the circular polarization degree has been analyzed. In turns out that the induced circular polarization is considerably smaller than the expected equilibrium polarization. Clarification of this difference requires further studies, which will be continued in TU Dortmund.

References

[1] D. R. Yakovlev, S. A. Crooker, M. A. Semina, J. Rautert, D. Dirin, M. V. Kovalenko, and M. Bayer, *Exciton-polaritons in $CsPbBr_3$ revealed by high magnetic fields and two-photon spectroscopy*, under preparation