Circularly polarized emission from self-trapped excitons in $Cs_2AgInCl_6$ crystals

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Halide double perovskite or elpasolite semiconductors, with the common chemical formula of $A_2BB'X_6$, have seen a renewed interest in the past couple of years because of their potential applications in photovoltaics, white light generation and UV light detection. Here, A is a monovalent cation (like Cs^+), B is a monovalent metal ion (such as Na^+, Ag^+), B′ is a trivalent metal ion (like Bi^{3+}, In^{3+}, Sb^{3+}), and X is some halide ion (like Br^-, Cl^-, I^-). One such elpasolite system is $Cs_2AgInCl_6$, which on one hand shows bright white light emission from self-trapped excitons at room temperature upon antimony (Sb) doping [1] and on the other hand shows antiferromagnetic order at low temperatures (< 15 K) when doped with iron (Fe) [2]. A soft lattice ($\hbar\omega_{\text{phonon}} \sim 20$ meV) and a large electron-phonon coupling (Huang-Rhys factor of ~ 40) in undoped $Cs_2AgInCl_6$ favors a strong Jahn-Teller distortion of the $AgCl_6$ octahedron in the excited state to form self-trapped excitons [1]. At room temperature excitonic emission at the bandgap is predicted to be weak, because the transition is parity-forbidden. Nevertheless, we observe bright photoluminescence (PL) around 500 nm at low temperatures (< 150 K) [Figure 1 (a)]. The PL emission is rather broad, which indicates that indeed the emission is due to self-trapped excitons, which are strongly coupled to the lattice. Remarkably, below 15 K, we see a significant reduction of the integrated PL intensity [Figure 1 (b)], which is possibly related to a local distortion.

Magneto-optics has been an essential tool to investigate the exciton parameters in many semiconductors, including self-trapped excitons in alkali halides [3]. We measured the PL emission of $Cs_2AgInCl_6$ single crystals as a function of magnetic fields up to 27 T in a 1.4 - 100 K temperature range. Due to the broad PL spectrum, we do not observe the diamagnetic shift of the emission in our field range. Yet, the emission exhibits a pronounced degree of circular polarization [DCP = $(I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$], where σ^+ and σ^- correspond to right- and left-handed circularly polarized light, respectively.

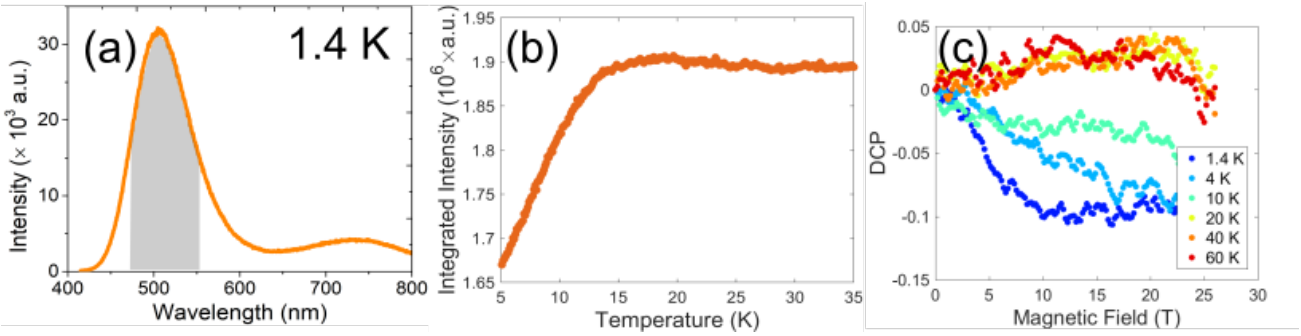

Figure 1: (*a*) *PL spectrum of $Cs_2AgInCl_6$ single crystal at* 1.4 *K.* (*b*) *Integrated intensities of the PL spectra with temperature.* (*c*) *Degree of circular polarization of the shaded PL peak in figure* (*a*) *at different temperatures.*

The main low-temperature emission peak around 500 nm shows a negative DCP below 10 K, which becomes nearly zero around 20 K and above [Figure 1 (c)]. The temperature-dependent behaviour of the PL intensity and the DCP at low temperatures (1.4 - 15 K) supports the local distortion hypothesis. Polarized Raman scattering studies confirm that no global structural phase transition is involved. We are in the process of further investigating this phenomenon through X-ray diffraction experiments.

References

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