

The Faraday Effect and Phase Transition in the $\text{CH}_3\text{NH}_3\text{PbI}_3$ Halide Perovskite Single Crystal

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The spin degree of freedom of charge carriers in halide-perovskite semiconductors can be highly useful for information photonics applications. The Faraday effect is known to be the best indicator of paramagnetism of the material and of the spin-light interaction. In this work, the Faraday effect is demonstrated, for the first time, in a hybrid organic–inorganic halide perovskite MAPbI_3 ($\text{MA}^+ = \text{CH}_3\text{NH}_2^+$). The Faraday rotation and birefringence are measured across the tetragonal–cubic phase transition at 327 K. The Faraday rotation is strongly suppressed below the phase transition temperature due to anisotropy (linear birefringence) of the tetragonal crystal phase. The situation changes drastically above the phase transition temperature, when the crystal becomes optically isotropic. The emerging Faraday rotation obeys the Curie law, demonstrating its population-related paramagnetic nature. This observation opens new prospects for application of these systems and for their investigations using methods of the polarization noise spectroscopy applicable to optically anisotropic materials.

However, Faraday effect has not been found for the most studied and practically important halide perovskite material – MAPbI_3 ($\text{MA}^+ = \text{CH}_3\text{NH}_2^+$).

At room temperature, MAPbI_3 belongs to the tetragonal crystal family with $I_{4/mcm}$ space group.^[15,16] In this connection, the absence of Faraday effect in this material at room temperature is not surprising because it is suppressed by linear birefringence of the crystal. Above 327 K, the crystal undergoes a phase transition to the cubic phase with $Fm\bar{3}m$ space group.^[17]

In this work, we show that Faraday effect in these samples can be correctly measured at elevated temperatures in the cubic phase of the crystal. We have also carried out complementary polarization measurements around the phase transition and investigated temperature dependence of the Faraday rotation (FR). This quantity obeys

the Curie law, which is a proof of the paramagnetic nature of involved spin centers.

1. Introduction

Halide perovskites are novel semiconductors with unique combination of properties. Cheap solution synthesis, bandgap tunability across the full visible wavelength range and defect tolerance^[1–4] makes them a promising materials for different optoelectronic applications ranging from photovoltaics^[5] to white light emission^[6,7] and lasing.^[8–9] Recent studies have shown spin-optical properties of halide perovskites that make them suitable for applications in spintronics, such as spin-polarized exciton quantum beatings,^[10] spin control of the lasing threshold,^[11] coherent spin manipulation^[12] and other related magneto-optical phenomena,^[13–14] thus opening a way to spinoptronic applications.^[15] In particular, a large Verdet constant and a strong Faraday effect was found in 3D halide perovskites.^[16,17]

2. Results

Figure 1a shows the optical density (OD) spectra of the sample in tetragonal phase at $T = 320$ K. This spectrum demonstrates a typical interband absorption of the MAPbI_3 semiconductor. Recent study has shown that the binding energy of the exciton in tetragonal phase is of the order of several meV,^[18] so that no excitonic effects are to be expected at these temperatures. The FR was found to be close to zero in the whole studied spectral range in contrast to the MAPbBr_3 crystal demonstrating strong FR even at room temperature.^[14]

When the MAPbI_3 crystal is heated above $T = 327$ K, it transits into the cubic phase, and its magneto-optical properties change dramatically. Figure 1b shows the FR and OD spectra for $T = 333$ K. As is seen, the OD spectrum remains practically the same (showing that no noticeable changes in the bandgap structure occurs across the phase transition^[21]), while a pronounced FR arises. The FR reaches its maximum at 1.48 eV and is still detectable at 1.33 eV, that was tested in additional experiments. The FR angle, as expected for linear magneto-optics, was proportional to the magnetic field B and sample thickness L : $\theta = VBL$, where V is the Verdet constant. For $E = 1.48$ eV, we obtained $V = 28 \text{ rad}\cdot\text{T}^{-1}\cdot\text{m}^{-1}$, which is half as much as for MAPbBr_3 , and

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The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adom.202302095>

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