Spin-Flip Raman Scattering on Electrons and Holes in Two-Dimensional $(PEA)_2PbI_4$ Perovskites

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The class of Ruddlesden-Popper type $(PEA)_2PbI_4$ perovskites comprises two-dimensional (2D) structures which are promising materials for photovoltaic and optoelectronic applications as their optical properties are determined by excitons with a large binding energy of about 260 meV. In 2D perovskites, a similar degree of optical spin control can be achieved as in conventional III-V and II-VI semiconductors, whose band structure is inverted compared to lead halide perovskites. We present our recent studies using spin-flip Raman scattering to measure the Zeeman splitting of electrons and holes in a magnetic field up to 10 T (see Fig. 1a)[1]. From the recorded data, the electron and hole Landé factors (g-factors) are evaluated (see Fig. 1b), their anisotropies are measured, and the hole sign is determined. The electron g-factor value changes from +2.11 out-of-plane to +2.50 in-plane, while the hole g-factor ranges between −0.13 and −0.51. Spin-flips of resident electrons and holes have been observed through their interaction with photogenerated excitons, as well as double spin-flip processes in which a resident electron and hole interact with the same exciton. Furthermore, we demonstrate the hyperfine hole-nuclei interaction in 2D perovskites by means of the dynamic nuclear polarization detected in corresponding changes of the hole Zeeman splitting (see Fig. 1c). Due to the small g-factor of the hole, we are able to achieve an Overhauser field value of $B_{\mathrm{N,h}} = 0.6\,\mathrm{T}$.

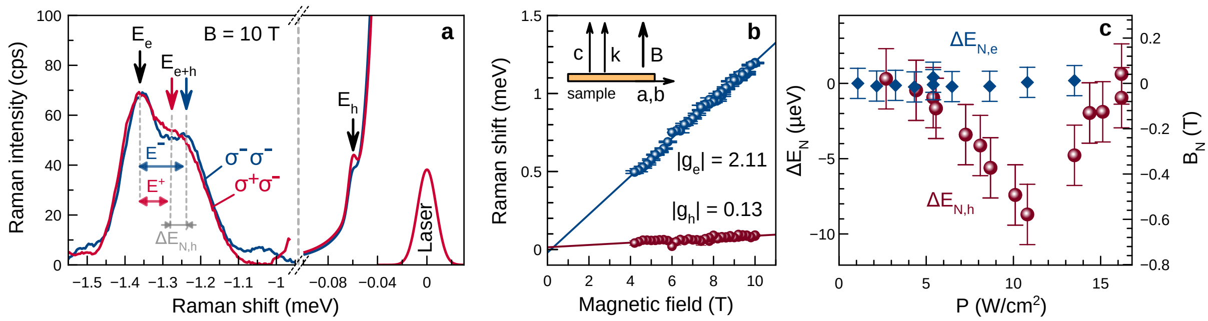

Figure 1: (a) Spin-flip Raman spectrum in the anti-Stokes spectral range in cross polarizations for $B = 10$ T. The hole E_{h}, electron E_{e}, and their double spin-flip $E_{\mathrm{e+h}}$ lines are highlighted by arrows. (b) Electron and hole Raman shift as function of the magnetic field in out-of-plane geometry ($\mathbf{B} \parallel \mathbf{k}$). (c) Power density dependences of the energy splitting $\Delta\ E_{\mathrm{N}} = E^+ - E^-$ (the superscript indicates the excitation polarization) for the electron and hole shifts. Right axis gives the corresponding Overhauser field B_{N}.

[1] C. Harkort, D. Kudlacik, N. E. Kopteva, D. R. Yakovlev, M. Karzel, E. Kirstein, O. Hordiichuk, M. Kovalenko, M. Bayer, *arXiv*:2302.02349 (2023).