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Fine structure of excitons in mixed halide perovskite single crystal revealed by photon echo quantum beats

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Photon echo spectroscopy overcomes the significant inhomogeneous broadening of approximately 16 meV in bulk mixed-halide perovskite $\text{FA}_{0.9}\text{Cs}_{0.1}\text{PbI}_{2.8}\text{Br}_{0.2}$ [1], revealing a much narrower homogeneous linewidth of about 16 μeV . This enables direct measurement of the fine structure splitting of excitonic states, arising from the Zeeman effect in an external magnetic field and electron-hole exchange interactions. These splittings manifest as oscillations in the time-resolved photon echo signal, caused by quantum beats between coherently excited exciton states. In Voigt geometry, the applied magnetic field induces mixing between bright and dark exciton eigenstates, allowing us to address and measure the splittings between all four excitonic states. By systematically varying the magnetic field strength and orientation, and combining this with polarization-sensitive excitation and detection, we extract electron and hole g-factors of $g_e = 3.4$ and $g_h = -1.0$, respectively, and determine a zero-field splitting of 0.5 meV between the bright ($J = 1$) and dark ($J = 0$) exciton states.

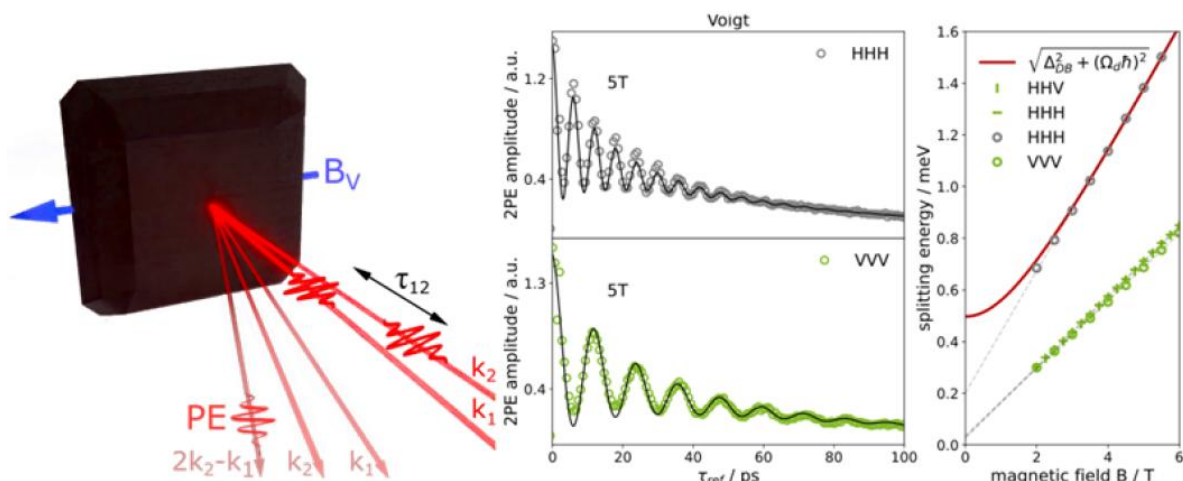


Fig. 1. Photon echo oscillations in magnetic field $\mathbf{B}$ for different polarization configurations. Left: Recorded photon echo decay at $B = 5$ T and temperature $T = 2$ K. The legend indicates the horizontal (H||B) and vertical (V⊥B) polarization of the first and second excitation pulse and detection. Right: Evaluated quantum-beats oscillation frequencies as a function of the applied magnetic field in Voigt (circles) and Faraday (bars) geometry.