Nuclear induced frequency focusing in the mode-locking of hole spin coherences in $CsPb(Cl,Br)_3$ perovskite nanocrystals

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The spin physics of lead halide perovskite crystals is attracting increasing attention both for fundamental studies and spintronic applications. Here, stable $CsPb(Cl_{0.56}Br_{0.44})_3$ lead halide perovskite nanocrystals (NC) embedded in a fluorophosphate glass matrix, similar to colloidal quantum dots (QD), are studied by time-resolved optical spectroscopy to unravel the coherent spin dynamics of holes and their interaction with nuclear spins of the ^{207}Pb isotope [1]. We demonstrate the spin mode-locking (SML) effect [2] and nuclear induced frequency focusing leading to the synchronization of the hole spin Larmor precession frequencies of the NC ensemble.

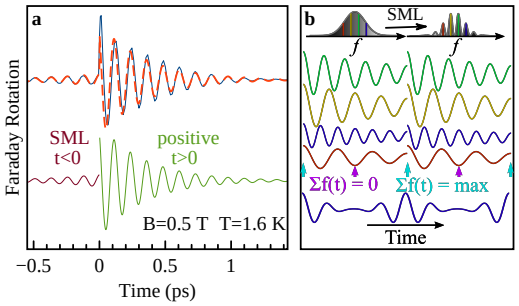

Fig. 1. **a** Spin precession signal of a nanocrystal ensemble (blue) with fit (dashed red) and the fit components ($t > 0$) green and ($t < 0$) red below. Red curve originates from SML effect. **b** Illustration of the SML effect. Due the SML the spin precession frequency Gaussian distribution changes to a comp-like one. The sum of frequencies results in the SML effect.

In a magnetic field the carrier spins in the NCs precess proportional to the magnetic field strength. However, due to confinement effects and shape anisotropy the coupling of each NC to the magnetic field differs, thus the Larmor precession frequency distribution is broad. The superpositions of the broad frequency distribution leads to a short ensemble coherence time (T_2^*), below a nanosecond. We overcome this limitation with use of the SML effect. By that the Larmor precession is synchronized by the period laser excitation, enhancing NCs with in phase precession and suppression of out of phase ones, see Fig.1**b**.
It is known that the hyperfine interaction is a major limitation of the spin coherence. However, additional to the SML the nuclear induced frequency focusing takes place which polarizes the nuclear spin in such a way that, due to the emerging Overhauser field, the Larmor precession of the NCs gets tuned that they become commensurable with the SML. Thus prolonging the ensemble spin coherence. Despite the short T_2^* below a nanosecond we observe the spin coherence ten times longer and can derive a spin coherence T_2 of 13 ns (Fig.1**a**).

References

[1] Kirstein, E., Kopteva, N. E., Yakovlev, D. R., Zhukov, E. A., Kolobkova, E. V., Kuznetsova, M. S., Belykh, V. V., Yugova, I. A., Glazov, M. M., Bayer, M., Greilich, A. Mode locking of hole spin coherences in $CsPb(Cl,Br)_3$ perovskite nanocrystals. Nat. Commun. 14, 699 (2023).

[2] Greilich, A., Yakovlev, D. R., Shabaev, A., Efros, A. L., Yugova, I. A., Oulton, R., Stavarache, V., Reuter, D., Wieck, A., Bayer, M. Mode Locking of Electron Spin Coherences in Singly Charged Quantum Dots. Science 313, 341-345 (2006).