Mode locking of hole spin coherences in CsPb(Cl, Br)₃ perovskite nanocrystals

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The spin physics of perovskite nanocrystals with confined electrons or holes is attracting increasing attention, both for fundamental studies and spintronic applications. Here, stable CsPb(ClₓBr₁₋ₓ)₃ lead halide perovskite nanocrystals embedded in a fluorophosphate glass matrix are studied by time-resolved optical spectroscopy to unravel the coherent spin dynamics of holes and their interaction with nuclear spins of the ²⁰⁷Pb isotope. We demonstrate the spin mode locking effect provided by the synchronization of the Larmor precession of single hole spins in each nanocrystal in the ensemble that are excited periodically by a laser in an external magnetic field. The mode locking is enhanced by nuclei-induced frequency focusing. An ensemble spin dephasing time T₂ of a nanosecond and a single hole spin coherence time of T₂ = 13 ns are measured. The developed theoretical model accounting for the mode locking and nuclear focusing for randomly oriented nanocrystals with perovskite band structure describes the experimental data very well.

Received: 12 July 2022  
Accepted: 19 January 2023  
Published online: 08 February 2023

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Optical orientation, optical alignment and anisotropic exciton Zeeman splitting were observed. The coherent spin dynamics of electrons and holes in CsPbBr₃ NCs were observed, and the picosecond spin dynamics of carriers in CsPbI₃ NCs were reported. Despite the progress in crystal growth, perovskite NCs still suffer from insufficient long-term stability of optical properties. A promising approach here is to synthesize NCs embedded in glass, providing protection by encapsulation.

As a result, perovskite NCs in a glass matrix may be suitable for quantum technologies. For reference, one may compare NCs with the established system of self-assembled (In,Ga)As/GaAs quantum dots (QDs) singly charged with an electron or a hole. Using all-optical approaches one can orient and manipulate the spins of the charge carriers on the picosecond timescale at high operation frequencies.

There are two complementary concepts: the first one uses a single spin in a QD for establishing a quantum bit, the second one exploits a QD ensemble which may be suited for a quantum memory with sufficiently strong light-matter interaction. Here, we focus on a NC ensemble, that...
provides information about the average spin properties and their dispersion due to inhomogeneity. The inhomogeneity obstacle, however, can be overcome by applying a periodic laser excitation that synchronizes the electron spin Larmor precession in different NCs subject to a transverse magnetic field. This spin mode locking (SML) effect, initially discovered in singly charged (In,Ga)As QDs, allows one to uncover the spin coherence of individual carriers, which is typically prevented due to the faster spin dephasing in the ensemble. The interaction of resident carrier spins with the surrounding nuclear bath and hyperstrated so far only for (In,Ga)As QDs. This collective phenomenon of spin dynamics homogenization has been demonstrated so far only for (In,Ga)As QDs.

In this paper we demonstrate the SML effect for holes in a totally different class of QDs, namely perovskite CsPb(Cl, Br)3 NCs synthesized in a glass matrix. We provide detailed information on the spin dynamics of confined holes correlated with the appearance of spin mode locking. We measure the g-factor, spread of g-factors, longitudinal spin relaxation time $T_1$, transversal spin coherence time $T_2$, and inhomogeneous spin dephasing time $T_2^*$. Exploiting the hyperfine interaction with nuclear spins, we implement dynamic nuclear polarization and optically-detected nuclear magnetic resonance (ODNMR) to identify the involved nuclear isotopes. A theoretical model of the SML in perovskite structures is developed to account for the inverted band structure and for the dominating role of the hole-nuclear interaction, compared to self-assembled QDs. As an additional challenge, the random orientation of NCs in the ensemble is considered.

Results

The optical properties of CsPb(Cl0.56Br0.44)3 nanocrystals embedded in a fluorophosphate glass are shown in Fig. 1a. At a cryogenic temperature of $T = 5\,\text{K}$, the transmission spectrum of the NCs, with about 8 nm size, shows a pronounced exciton resonance at 2.743 eV, which is broadened due to NC size dispersion. The exciton resonance can be also traced by the spectral dependence of the time-resolved Faraday ellipticity (TRFE) amplitude. The exciton population dynamics is measured by time-resolved differential transmission ($\Delta T/T$), showing a decay time of 160 ps, which corresponds to the exciton lifetime (inset in Fig. 1a).

To study the coherent spin dynamics of resident carriers we use the pump-probe technique with spin-sensitive detection of the Faraday ellipticity, whose potential has been approved for lead halide perovskite crystals, polycrystalline films, and CsPbBr3 NCs. The pump laser pulses are circularly polarized and, according to the selection rules, generate spin polarization of electrons and holes along the light $\mathbf{k}$ vector. The oriented carrier spins precess at the Larmor frequency around the magnetic field applied in the Voigt geometry $B_V(B_\perp k)$. The coherent spin dynamics is then detected through the Faraday ellipticity of the linearly-polarized probe pulses.

A typical TRFE signal, measured at $T = 1.6\,\text{K}$ in $B_V = 1\,\text{T}$, is shown in the upper part of Fig. 1b. It shows oscillations with the Larmor frequency $\omega_L$, which is a linear function of $B_V$, $\omega_L = \mu_B B_V /\hbar$. This allows us to evaluate the g-factor of $|g| = 1.20$. $\omega_L$ has no offset at zero magnetic field (Fig. S4a), which is a strong argument in favor of the presence of resident carriers in the NCs rather than of carriers bound in an exciton. In the latter case, an offset equal to the electron-hole

![Fig. 1 | Spin mode locking in CsPb(Cl, Br)3 NCs.](image-url)
halide perovskite semiconductors due to their net magnetic field increases up to 0.5 ns at low magnetic fields. The hole interaction with the nuclear spin interaction compared to the electron-nuclear one is specified (Supplementary Notes 1 and 2). Note, that the stronger hole-nuclear interaction of the Larmor spin precession in the ensemble of NC carriers studied NCs (Supplementary Note 4).

Obtained for spin coherence times both negative and positive charging29,31,40. It is caused by synchronization of the Larmor spin precession in the ensemble of NC carriers with the periodic laser pumping, termed the spin mode locking effect41, an effect similar to the resonant spin amplification42.

The SML effect results from the efficient accumulation of spin polarization from carrier spins which Lamor frequencies \( \omega \) are commensurate with the laser repetition period \( T_R \). These spin precession modes fulfill the phase synchronization condition (PSC): \( \omega = N \omega_0 - 2\pi g N / T_R \), with \( N \) being an integer. If the single spin coherence time exceeds \( T_R \), the spin polarization of these modes accumulates. Modes fulfilling the PSC are shown schematically in Fig. 1c, assuming a Gaussian distribution of the Lamor frequencies centered around \( N \). The same effect was observed for \( n = 1 \) in Fig. 1d by the green line with the spread of Lamor frequencies \( \Delta \omega \). The spin polarization of these modes accumulates. Modes fulfilling the PSC are shown schematically in Fig. 1d, assuming a Gaussian distribution of the Lamor frequencies centered around \( N \). Within this distribution, the five essential PSC modes (red needles) are located.

The signal resulting from the sum of the five PSC modes is shown at the bottom of Fig. 1c. The maxima of the PSC mode signal coincide with the pump pulse arrival times, in between the signal is reduced to zero.

Spin mode locking in nanocrystal ensembles

The most striking feature of the TRFE signal (blue) in Fig. 1b is the presence of an oscillating signal at negative time delays, before the pump arrival. Its amplitude has a maximum at small negative delays and decays with increasing negative delay with the same dephasing time as at positive delays. For such a signal to occur, optically-induced spin coherence should be present in the NCs for times exceeding the repetition period of the laser pulses \( T_R = 13.2 \) ns, and can therefore be obtained for spin coherence times \( T_2 > T_R \). The same effect was observed in ensembles of singly charged \((n, Ga)As/GaAs QDs with both negative and positive charging\(^{39,40} \)). It is caused by synchronization of the Lamor spin precession in the ensemble of NC carriers with the periodic laser pumping, termed the spin mode locking effect41, an effect similar to the resonant spin amplification42.

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The signal resulting from the sum of the five PSC modes is shown at the bottom of Fig. 1c. The maxima of the PSC mode signal coincide with the pump pulse arrival times, in between the signal is reduced to zero.
PSC modes have to satisfy two commensurability conditions: $\omega_h = 2mN/T_D$ and $\omega_g = 2mN/T_p$, with $M$ being an integer. In the experiment this leads to the emergence of signal bursts at multiple times of $T_D$. This behaviour is indeed also found in the studied perovskite NCs, as one can see from the red colored signal in Fig. 1b and its zoom in Fig. 1c, measured with $T_D = 0.1T_p = 1.32$ ns. Two bursts at $1.32$ ns and $+2.64$ ns are seen very prominently. A scheme of the PSC mode spectrum modulation for the two-pump case is given in Fig. 2a.

### Nuclei-induced frequency focusing

In semiconductors, the spin dynamics of electrons and holes are strongly influenced by their interaction with the nuclear spin system in which they are embedded. Polarized nuclei provide the Overhauser field $B_0$, which shifts the carrier Larmor precession frequency to $\omega = 2g_\mu_B(B_0 + B_N)/\hbar$. Thereby, spin precession modes in Fig. 1d, that do not match the PSC, can be shifted in their frequency such that they satisfy the PSC. This effect was found experimentally in (In,Ga)As QDs and identified as nuclei-induced frequency focusing (NIFF) of the carrier spin coherence. The NIFF mechanism represents a positive feedback loop, that provides the required nuclear polarization in each QD, so that a large dot fraction in the ensemble is pushed into PSC modes. Due to the very slow nuclear spin relaxation at cryogenic temperatures, the NIFF configuration can be kept for hours. In an ideal, very efficient, system it is the case for (In,Ga)As QDs with strong carrier-nuclei interaction, NIFF results in identical amplitudes of the TRFE signal at negative and positive time delays, meaning that all precession frequencies are focused on the PSC modes. This is obviously not the case for the studied perovskite NCs, see Fig. 1b. Still, NIFF is present as can be evidenced by the relaxation dynamics of the bunch amplitude at $-1.32$ ns delay when the second pump is blocked. In Fig. 2b we demonstrate such a measurement. We find the decay time of $r = 0.5$ s, which is related to the repolarization of the nuclear spin system from the two-pump to the one-pump protocol, sketched in Fig. 2a. Without nuclear involvement the amplitude of the signal would decay within the carrier spin coherence time $T_2$, after switching off the second pump, as was the case for hole spins in (In,Ga)As QDs.

### Dynamic nuclear polarization and ODNMR

To further justify the involvement of nuclear spins, we apply the method of dynamic nuclear polarization (DNP) in a tilted magnetic field. The spin polarization of optically-oriented carriers is transferred via the hyperfine interaction to the nuclear spin system. The Overhauser field of the polarized nuclei $B_0$ acts back on the carriers and shifts their Larmor precession frequency, the detailed scheme can be found in Supplementary Note 2 and ref. In the shift of the Larmor frequency allows one to measure the nuclear spin polarization.

One can see in Fig. 2c, that for $\sigma^-$ pump the Larmor precession becomes faster compared to the $\sigma^+$ case. The relative frequency shift is considerable, which is typical for the strong hole-nuclear spin interaction in perovskites. It allows us to confirm that the TRFE signal is related to holes and to evaluate the Overhauser field of $B_0 = 5.8$ mT. The sign of the observed Larmor frequency change corresponds to a positive hole $g$-factor.

The variation of the TRFE signals by the DNP effect allows us to use it for optical detection of nuclear magnetic resonance (ODNMR). For that, the time delay is fixed and the TRFE amplitude is measured as a function of the radio frequency of additional radiation with $f_{RF} = 0.1$–10 MHz. The nuclear spin system is depolarized when the energy $\hbar f_{RF}$ matches to the Zeeman splitting of a nuclear isotope $\hbar g_B nI$, i.e., for a NMR. Here, $\mu_B$ is the nuclear magneton and $g_B$ is the nuclear $g$-factor (Supplementary Table 1). In the experiment we detected as a resonant decrease of the TRFE amplitude, as shown in the inset of Fig. 2d at $B = 0.2$ T. From a linear fit of the resonance frequency dependence on the magnetic field $f_{RF}(B)$, we evaluate the gyromagnetic ratio $g = \mu g_B / \hbar = 8.80$ MHz/T, see Fig. 2d. One can conclude from the Supplementary Table 1 that among the nuclear isotopes present in the CsPb(Cl, Br)$_3$ NCs only the $^{207}$Pb isotope matches this value. The dominant role of $^{207}$Pb on the carrier spin dynamics was identified earlier in FA$_0$.Cs$_0$.Pb$_{230}$Br$_{0.2}$ crystals and the theoretical analysis establishes it as common feature for lead halide perovskites.

### Theory of spin mode locking in perovskite NCs

The theory of spin mode locking was previously developed for self-assembled quantum dots and successfully applied to explain the experimental results. However, the discovery of SML in perovskite nanocrystals provides new theoretical challenges. First, the generation of spin coherence in perovskite structures occurs on the basis of other selection rules. Circularly polarized light generates an exciton with an electron and a hole, both having spin $1/2$, instead of an exciton with an electron (1/2) and a heavy hole (3/2). Second, perovskite colloidal nanocrystals in an ensemble are randomly oriented. For a complete theoretical description, the anisotropy of the $g$-factor for a single NC and the possibility of generating spin coherence through positively and negatively charged trions have to be considered. Taking all these facts into account requires a significant adjustment of the theoretical model.

The model, which details are given in Supplementary Notes 6–10, has two key parts: the first one considers the spin coherence generation and its dynamics in magnetic field for a single nanocrystal with arbitrary oriented crystallographic axes. The second part accounts for the inhomogeneity of the NC ensemble, namely the NCs random orientation and the $g$-factor spread.

Without loss of generality we consider resident holes, that can be created by photo-charging, as in the experiment the SML of holes is observed. The resident carrier spin polarization, providing the pump-probe signal, is generated by a mechanism involving trions. The oriented spin polarization components are tilted relative to the direction of the magnetic field, precess about it in time, and decay with the spin coherence time $T_2$. For $T_2 > T_1$ the spin polarization accumulates by pumping with a train of optical pulses. The SML is an ensemble effect resulting from a large number of oscillating signals with frequencies commensurable with $T_1$, therefore, the model takes into account the spread of the $g$-factors (Supplementary Note 8).

The experimentally observed spin dynamics, measured at $B = 0.35$ T (black dots in Fig. 3a), are modeled numerically using Supplementary Eqs. 28–61. The signal decay, at negative and positive time delays, is described by the $T_2^*$, that is given by the $g$-factor spread, $\Delta g$, and nuclear spin fluctuations, $\delta B_0$. The ratio $S_2/S_0$ of the amplitudes before pulse arrival $S_0$ and after pulse arrival $S_2$ is determined by $T_2$, the optical pulse area $\Theta$ and the NIFF effect. Technically, we calculate the spin polarization distribution $S_2$ as a function of the Larmor frequency parameterized by the hole $g$-factor $\omega = g_\mu_B B_0 / \hbar$ (Fig. 3b).

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**Fig. 3** Calculation of spin polarization and TRFE signal. a Time resolved Faraday ellipticity (dots) measured at $B_0 = 0.35$ T and its modeling (red line). b Spectral distribution of precessing modes for $T_2 = 28$ ns. Inset: illustration of precessing mode with NIFF for $T_2 = 13$ ns (green) and without NIFF for $T_2 = 28$ ns (red).
distribution width is defined by \(2\Delta \xi\) with \(\Delta \xi = 0.1\). The multiple peaks correspond to synchronized precession modes, with the width of each peak determined by \(1/T_2 + 1/T_2',\) where \(T_2 = h/(g_\mu \beta \sqrt{\Delta B_\xi^2})\) is related to the nuclear spin fluctuations. The modeled SML signal, corresponding to this distribution, is shown by the red line in Fig. 3a. It reproduces well all features of the experimental data. Finally we note, that the presented theoretical approach for the description of the SML effect is universal and applicable for different crystal phases of the NCs.

In order to highlight the role of the hole-nuclear interaction for the measured SML, we perform modeling without and with account for the NIFF. Without NIFF, we get a hole spin coherence time \(T_2 = 28\) ns for \(\theta = \pi\) as best fit parameter. The nuclear polarization changes the spin polarization distribution by increasing \(T_2'\) and narrowing the precession modes. This results in an increase of the ratio \(S_p/S_0,\) and to match its experimental value of 0.1 a shorter spin coherence time should be taken, to keep the mode width the same (insert of Fig. 3b). With NIFF we get \(T_2 = 13\) ns, which is at the lower limit of the times required for the SML to occur. It is remarkable that due to the involvement of NIFF we are able to observe the SML effect for such a short-lived hole spin coherence.

**Discussion**

In summary, we have demonstrated the outstanding potential of perovskite NCs embedded in a glass matrix, using their spin properties as exemplary feature. We find a very long hole spin lifetime up to the microsecond range. We present an additional class of materials, in which the effects of spin mode locking and nuclei-induced frequency focusing are observed. This allows one to measure the hole spin coherence time \(T_2\) disregarding the random NC orientation and variation of NC sizes and shapes. It opens the pathway for implementation of multi-pulse protocols for manipulating the spin coherence, also with involvement of strong hole-nuclear interaction provided by the inverted band structure of the lead halide perovskites.

Despite our main goal here to report on the SML effect in perovskite NCs, we would like to compare our discovery with other systems. In the studies of colloidal CsPbBr$_3$ NCs (grown in solution) the SML effect was not observed.

Methods

**Samples**

The studied CsPb(Cl, Br)$_3$ nanocrystals embedded in a fluorophosphate glass matrix were synthesized by rapid cooling of a glass melt enriched with the components needed for the perovskite crystallization. Samples of fluorophosphates (FP) glasses of 60Ba(IO$_3$)$_2$-15NaPO$_3$-12AlF$_3$-1Ga$_2$O$_3$-4Cs$_2$O-8PbF$_2$ (mol. %) composition doped with 16 mol.% NaCl, 3.4 mol.% BaBr$_2$ were prepared using the melt-quench technique. The glass synthesis was performed in a closed glassy carbon crucible at temperatures of 1000–1050 °C. About 30 g of the mixed powder was melted in the crucible during 20 min. Then the glass melt was cast on a glassy carbon plate and pressed to form a plate with a thickness of about 2 mm. The formation of the crystalline phase in glass can be carried out directly in the process of cooling the melt. During the pouring out of the transparent melt spontaneous precipitation of NCs with sizes smaller than the Bohr exciton radius occurs. By a subsequent heat treatment at a temperature just above the glass transition temperature (400–429 °C), it is possible to sequentially grow NCs to large sizes. The details of the method are given in ref. 26.

The NC size of 8 ± 1 nm is evaluated from a scanning transmission electron microscopy technique, see Supplementary Note 11. The concentration of NCs, of about 6 × 10$^{13}$ cm$^{-3}$, is evaluated from optical absorption measurements. The composition of the CsPb(Cl$_{0.6}$Br$_{0.4}$)$_3$ NCs is estimated from the XRD data, see Supplementary Note 11. At \(T = 5\) K the exciton resonance, mainly broadened due to the NC size dispersion, is at 2.743 eV. The band gap lies between that of bulk CsPbBr$_3$ and bulk CsPbCl$_3$. Additional for NCs, compared to the bulk case, the optical transition is shifted due to the quantum confinement$^{12,48}$. For optical experiments, the sample with a thickness of 500 mm was polished from both sides. The remarkable advantage of NCs in a glass matrix versus wet-chemistry synthesized NCs is the encapsulation resulting in long-term stability. Also the possibility to have optically flat sample surfaces with diminished light scattering is important for the polarization sensitive optical techniques used in this study. The effect of phase separation due to a laser excitation is excluded by low temperatures and relatively low laser powers applied resonantly to the NCs transitions. An additional long-term stability test is provided, in the Supplementary Note 11, to prove our claims.

**Magneto-optical measurements**

For low-temperature optical measurements we use a liquid helium cryostat with a variable temperature from 1.6 K up to 300 K. At \(T = 1.6\) K the sample is immersed in superfluid helium, while at 4.2–300 K it is held in helium gas. A superconducting vector magnet equipped with three orthogonal pairs of split coils to orient the magnetic field up to 3 T along any arbitrary direction is used. The magnetic field parallel to \(k\) is denoted as \(B_{L}\) (Faraday geometry), perpendicular to \(k\) as \(B_{V}\) (Voigt geometry). If not stated otherwise, the \(B_{L}\) is oriented horizontally. The angle \(\alpha\) is defined as the angle between \(B_{L}\) and the light wave vector \(k\), where \(\alpha = 0°\) corresponds to \(B_{V}\).

**Transmission spectra**

A white-light halogen lamp is used to measure the optical transmission. The sample was thinned to 43 mm in order to avoid saturation of the light absorption in the spectral range of the exciton resonances. The spectra are detected by a 0.5 m monochromator equipped with a silicon charge-coupled-device camera.

**Time-resolved Faraday ellipticity (TRFE)**

The coherent spin dynamics are measured using a degenerate pump-probe setup$^{39}$. A titanium-sapphire (Ti:Sa) laser generates 1.5 ps long pulses in the spectral range of 700–980 nm (1.265–1.771 eV), which are frequency doubled with a beta barium borate crystal to the range of 350–490 nm (2.530–3.542 eV). The laser spectral width is about 1 nm (about 2 meV), and the pulse repetition rate is 76 MHz (repetition period \(T_{rep} = 13.2\) ns). The laser output is split into two beams, pump and probe, which pulses can be delayed with respect to one another by a mechanical delay line. The laser photon energy is tuned to be in resonance with the exciton transition, e.g. at 2.737 eV at \(T = 1.6\) and 5 K. The pump and probe beams are modulated using photo-electric modulators (PEM). The probe beam is linearly polarized and its amplitude is modulated at 94 kHz, while the pump beam is either helicity modulated at 50 kHz between \(\sigma^+\)/\(\sigma^-\) circular polarizations or amplitude modulated at 100 kHz with the circular polarization fixed at either \(\sigma^+\) or \(\sigma^-\). The signal was measured by a lock-in amplifier locked to the difference frequency of the pump and probe modulation. For measuring by spin inertia technique, the modulation frequency of the pump helicity \(f_m\) was varied from 1 kHz to 5 MHz by an electro-optical modulator (EOM). For measuring the spin dynamics induced by the pump pulses, the polarization of the transmitted probe beam was analyzed with respect to its ellipticity (Faraday ellipticity) using balanced photodiodes and a lock-in amplifier. In \(B_{L} \neq 0\), the Faraday...
ellipticity amplitude oscillates in time reflecting the Larmor precession of the charge carrier spins. The dynamics of the Faraday ellipticity signal can be described by a decaying oscillatory function:

$$A(t) = S \cos(\omega t) \exp(-t/T^2).$$  \hspace{1cm} (1)

Here $S$ is the amplitude at zero delay, $\omega_S$ is the Larmor precession frequency, and $T^2$ the spin dephasing time. For application of a two-pump pulse scheme, after the delay line the so far not modulated linearly polarized pump beam was passed through a 50/50 nonpolarizing beam splitter. The deflected beam was sent via two mirrors over a certain distance and merged with the transmitted pump beam via a second nonpolarizing 50/50 beam splitter. By careful adjustment both pump beams were brought aligned parallel to each other, to excite the same sample spot with 300 μm diameter.

**Pump-probe time-resolved differential transmission**

Here, for excitation by the pump beam, the same scheme as in TRFE is used, but with linear polarization. The pump is amplitude modulated by a photo-elastic modulator at 100 kHz. The linearly polarized, unmodulated probe beam is separated into a reference beam, which is routed around the cryostat and the test beam, which is transmitted through the sample. The signal is recorded with respect to the intensity difference between the transmitted and the reference beams, using a balanced photodetector.

**Optically-detected nuclear magnetic resonance (ODNMR) with TRFE detection**

In this technique nuclear magnetic resonances are measured via resonant decrease of dynamic nuclear polarization by radio frequency (RF) radiation. For that optical detection of the TRFE amplitude at fixed magnetic field is used. A small RF coil of about 5 mm diameter having 5 turns is placed close to the sample surface, similar to ref. 49. The coil is mounted flat at the sample surface and the laser beam is transmitted through the bore of the coil. The RF induced magnetic field direction is perpendicular to its surface and parallel to $\mathbf{k}$. The RF in the frequency range $f_{\text{RF}}$ from 100 Hz up to 10 MHz is driven by a frequency generator, with an applied voltage of 10 V leading to an oscillating field amplitude of about 0.1 mT. The RF is terminated by internal 50 Ω resistors, but not frequency matched to the circuit. In the low frequency range up to 5 MHz the current is nearly frequency independent, as the inductive resistance of the coil is small compared to the internal termination. For observation of DNP it is essential to tilt the external magnetic field direction away from the Voigt geometry, to have a nonzero scalar product $\mathbf{B} \cdot \mathbf{S}$ and apply a constant pump helicity.

**Data availability**

The data on which the plots in this paper are based and other findings of this study are available from the corresponding authors upon justified request.

**Code availability**

The code on which the calculations within this paper are based and other findings of this study are available from the corresponding author upon justified request.

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Acknowledgements

The authors are thankful to A.L. Efros, M. A. Semina, and M. O. Nestoklon for fruitful discussions. We thank M. L. Skorikov for help with experiment. The authors would like to sincerely thank V. Brandt for performing the TEM characterization. We acknowledge financial support by the Deutsche Forschungsgemeinschaft via the SPP 2196 Priority Program (Project A 65/26-1) and the International Collaborative Research Centre TRR 160 (Projects A1 and B2). M.M.G. acknowledges support of the Russian Foundation for Basic Research (Grant No. 19-52-12038). The low-temperature sample characterization was partially supported by the Ministry of Science and Higher Education of the Russian Federation (Contract No. 075-15-2021-598 at the P. N. Lebedev Physical Institute). M.S.K. and I.A.Y. acknowledge the Saint-Petersburg State University (Grant No. 9182694) and the Russian Foundation for Basic Research (RFBR Project No. 19-52-12059). Synthesis of the samples have been supported by Priority 2030 Federal Academic Leadership Program.

Author contributions

D.R.Y., A.G. and E.K. conceived the experiment. E.K., E.A.Z. and A.G. built the experimental apparatus and performed the measurements. E.K., N.E.K., V.V.B., A.G. and D.R.Y. analyzed the data. N.E.K., I.A.Y. and M.M.G. provided the theoretical description. E.V.K. and M.S.K. fabricated and characterized the sample. All authors contributed to interpretation of the data. N.E.K., E.K., V.V.B., A.G. and D.R.Y. wrote the manuscript in close consultation with M.B.

Funding

Open Access funding enabled and organized by Projekt DEAL.

Competing interests

The authors declare no competing interests.

Additional information

**Supplementary information** The online version contains supplementary material available at https://doi.org/10.1038/s41467-023-36165-0.

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**Peer review information** Nature Communications thanks the anonymous reviewer(s) for their contribution to the peer review of this work.

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