Dynamic nuclear spin polarization in resonant laser spectroscopy of a quantum dot

A. Högele 1, M. Kroner 2, C. Latta 2, M. Claassen 3, I. Carusotto 4, C. Bulutay 2,5, and A. Imamoglu 2

1 Fakultät für Physik and CeNS, Ludwig-Maximilians-Universität München, D-80539 München, Germany
2 Institute of Quantum Electronics, ETH Zürich, CH-8093, Zürich, Switzerland
3 Department of Applied Physics, Stanford University, Stanford, California 94305, USA
4 INO-CNR BEC Center and Dipartimento di Fisica, Università di Trento, I-38123 Povo, Italy and
5 Department of Physics, Bilkent University, Ankara, 06800, Turkey
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Resonant optical excitation of lowest-energy excitonic transitions in self-assembled quantum dots lead to nuclear spin polarization that is qualitatively different from the well known optical orientation phenomena. By carrying out a comprehensive set of experiments, we demonstrate that nuclear spin polarization manifests itself in quantum dots subjected to finite external magnetic field as locking of the higher energy Zeeman transition to the driving laser field, as well as the avoidance of the resonance condition for the lower energy Zeeman branch. We interpret our findings on the basis of dynamic nuclear spin polarization originating from non-collinear hyperfine interaction and find an excellent agreement between the experimental results and the theoretical model.

The basic principles of optical nuclear spin orientation in solids have been studied extensively in bulk semiconductors [1] and attracted a revived attention by recent optical studies of semiconductor quantum dots (QDs). Dynamic nuclear spin polarization (DNSP) in self-assembled InGaAs QDs has been reported for quasi-resonant [2] and non-resonant excitation [3, 4]. On the basis of these experiments and related theoretical studies, a comprehensive picture of unidirectional optical orientation of QD nuclear spins effected by light-polarization selective pumping was developed. Early experiments carried out on positive and negative trions [2, 4] as well as neutral excitons [5] had been used to demonstrate bistability of DNSP as a function of magnetic field or incident laser power and polarization [3, 4, 6, 7], and to study nuclear spin buildup and decay dynamics [3, 4].

In stark contrast to non-resonant excitation however, bidirectional nuclear spin orientation independent of photon polarization was observed in resonant laser scattering of elementary excitations in neutral [10] and negatively charged QDs [10, 12]. A particularly striking feature of resonant DNSP using the higher energy Zeeman transition at external magnetic fields exceeding 1 T is the flat-top absorption spectra, stemming from active locking of the QD resonance to the laser frequency [10, 11]. Remarkably, neutral and negatively charged QDs showed similar spectral signatures in resonant spectroscopy despite substantially different energy level diagrams: for both charge states, the locking of the coupled electron-nuclear spin system to the incident laser (dragging) was observed over tens of μeV detunings to either side of the resonance [10].

In this Letter, we carry out a comprehensive experimental and theoretical analysis of dragging in resonantly driven QD transitions. We develop a microscopic model based on the effective non-collinear hyperfine coupling that was first proposed by Ref. [13] to explain nuclear spin relaxation in self-assembled QDs. Our experiments demonstrate that the nature of resonant DNSP depends drastically on whether the blue (higher energy) or the red (lower energy) Zeeman transition is resonantly excited; while the blue transition exhibits locking of the QD resonance to the incident laser, nuclear spin polarization ensures that the resonance condition is avoided for the red transition [14]. We also find that while the frequency range over which blue Zeeman transition locking takes place varies from QD to QD, the dependence of the corresponding dragging width on laser power, scan speed and the magnetic field is similar for all QDs.

A key requirement for dragging is the presence of an unpaired electron spin with a long spin-flip-time, either in the initial or the final state of the optical transition; this condition is satisfied by fundamental neutral (X⁰), single-electron (X⁻) and single-hole (X⁺) charged QD transitions. The Overhauser field [1] experienced by this unpaired electron facilitates the feedback that modifies the QD transition energy. However, whether or not this feedback leads to resonance seeking (as in the blue Zeeman branch) or resonance avoiding (as in the red Zeeman branch) excitations depends on the spin orientation of the electron that couples to the incident laser field.

We studied individual InGaAs QDs embedded in a field effect device [15]. Two samples distinct by the thickness of the tunnel barrier between the heavily n-doped back contact and the QD layer (25 nm and 35 nm in samples A and B, respectively) were employed to probe the fundamental exciton transitions in resonant laser scattering experiments at 4.2 K [16]. Representative spectra measured on the neutral exciton X⁰ in sample A subjected to a moderate magnetic field of B₂ = 4.5 T are shown in the upper panels of Fig. (a) and (d). The spectra instantly reveal drastic departures from a two-level Lorentzian with resonance frequency ω_X: the blue Zeeman optical transition of X⁰ shows flat-top absorption (Fig. (c)), also reported earlier for the negative trion in Faraday [10] and Voigt [11] configurations. In contrast, the spectral shape of the red Zeeman resonance is tri- angular (Fig. (d)) with maximum contrast that is a factor of ~ 10 lower than its blue counterpart. From direct comparison it becomes apparent that the blue transition is locked to the laser at frequency ω_L and can be dragged to positive and negative laser detunings Δ = ω_X − ω_L by tens of μeV, dependent on the scan direction (grey and blue spectra in Fig. (c)). In contrast, the red transition avoids the resonance with the laser...
FIG. 1: Quantum dot nuclear spin polarization in resonant laser scattering. Level diagrams of the blue (a) and red (b) Zeeman transitions of a neutral exciton $X^0$ in a finite magnetic field applied along the growth direction $z$: resonant laser field couples dipole allowed and dipole forbidden transitions (straight and diagonal arrows, respectively) of the exciton-nuclear spin manifold. For both Zeeman branches the lower states of the manifold are electron/hole spin singlets ($|\uparrow\downarrow\rangle$) and sense the nuclear field $I_n$ of $N$ nuclei via the Overhauser shift $\pm (A/N) I_n$ with the hyperfine coupling constant $A/2$. Change in the nuclear spin polarization occurs through spin-flip assisted diagonal transitions followed by spin preserving radiative decay (wavy arrows): finite laser detunings lead to an imbalanced competition between the bidirectional nuclear spin diffusion processes within the manifold (horizontal arrows). The coupled exciton-nuclear spin system reaches steady state by locking the blue Zeeman transition to the laser (dragging) or, alternatively, pushing the red Zeeman transition away from the laser resonance (anti-dragging). The corresponding spectra for opposite scan directions are color-coded in (c) and (d) as grey and blue/red for initial red and blue laser detunings (upper panel: experiments on sample A, lower panel: simulations; note the factor of $\sim 10$ difference in ordinate scales).

using DNSP (Fig. 1(d)), resulting in a triangular line shape.

We systematically measure the same qualitative response for the blue (red) Zeeman transitions of $X^0$ as well as of both trions, $X^+$ and $X^-$, in Faraday and Voigt magnetic field geometries, as demonstrated in Fig. 2. There are, however, quantitative variations in the efficiency of DNSP from sample to sample and even from dot to dot within one sample [17]. The thickness of the tunnel barriers in samples A and B plays a crucial role for electron spin exchange with the Fermi reservoir via co-tunneling [18] and thus for electron-spin pumping at magnetic fields exceeding $\sim 0.3$ T [19] as well as the efficiency of DNSP [16].

Our findings demonstrate that effects of bidirectional DNSP are omnipresent in resonant laser spectroscopy of QD excitons and call for a unified explanation that goes beyond directional DNSP mediated by flip-flop terms of the Fermi-contact hyperfine interaction. Obviously, the model should be insensitive to the details of the initial and final QD states, such as charge configuration or the presence of dark exciton states [20], yet capture marked signatures and differences in the response of the blue and red Zeeman transitions to a near-resonant laser. Recently, Yang and Sham [14] proposed that non-collinear hyperfine interaction between heavy-holes and the nuclei, induced by heavy-light-hole coupling, provides an excellent qualitative description of the signatures related to DNSP in resonant laser scattering [21]. On the other hand, recent experiments [22] demonstrate that non-collinear hyperfine interaction between the electron and the nuclei plays a significant role in determining QD nuclear spin dynamics even in the absence of optically generated holes; this interaction is induced by large quadrupolar fields in strained self-assembled QDs which ensure that nuclear spin projection along $B_z$ is not a good quantum number. The resulting effective non-collinear interaction between the QD electron and the nuclei is [13]:

$$\delta_z = \omega_0^2 + A I_z,$$

where $\omega_0$ is the energy of the QD exciton, $A$ is the hyperfine coupling constant, and $I_z$ is the nuclear spin component along the laser field direction.
FIG. 2: Optical transitions in quantum dots of sample B at $B = 4.0$ T as a function of quantum dot charge and magnetic field orientation: positive and negative trions, $X^+$ and $X^-$, exhibit characteristic flat-top resonances on the blue Zeeman transition and triangular lineshapes on the red Zeeman transition (left and right panels in a and b, respectively) in magnetic field oriented (a) parallel (Faraday) and (b) perpendicular (Voigt) to the sample growth axis.

The fact that $\hat{H}_{\text{nc}}$ could explain dragging is at first sight surprising since its dominant effect appears to be nuclear spin diffusion. However, a careful inspection shows that

\begin{equation}
\hat{H}_{\text{nc}} = \sum_i A_i^{\text{nc}} \hat{I}_i \hat{S}_z.
\end{equation}

with $A_i^{\text{nc}} = A_i B_i^Q \sin(2\theta_i)/(2\omega^2_0)$, and $\hat{S}, \hat{I}_i$ spin operators of the electron spin and the $i$-th nucleus, respectively. Here, $\omega^2_0$ denotes the nuclear Zeeman energy, $B_i^Q$ the strength of the quadrupolar interaction and $\theta_i$ is the angle between the major quadrupolar axis of the $i$-th nucleus and the $z$-axis. For the coupling strength of the electron to the $i$-th nucleus we assumed $A_i = A/N$, where $A$ is the maximal Overhauser field splitting and $N$ the number of nuclei. To determine $B_i^Q$ and $\theta_i$, we first employed molecular statics with Tersoff type force fields [23] to obtain the realistic structure for more than one million atoms hosting $N \approx 32,000$ QD nuclei. The atomistic strain and nuclear quadrupolar distributions are extracted over this relaxed structure [24]. Fig. 3(a) shows the distribution of the biaxial strain $\varepsilon_B \equiv \varepsilon_{xx} - (\varepsilon_{xx} + \varepsilon_{yy})/2$ which is primarily responsible for the nuclear quadrupolar shifts. Based on this distribution, we determine $A_i^{\text{nc}}$ for a line-cut along the QD taken through the center and the [010] axis, cf. Fig. 3(b). Averaging over this distribution for nuclei that lie within the Gaussian QD electron wavefunction, we obtain $A_i^{\text{nc}} \approx 1.3 \times 10^{-4}$ $\mu$eV, consistent with [23].

The same Hamiltonian also leads to a small polarization term whose direction is determined by the sign of the optical detuning [14, 17]. To explain this, we consider the energy level diagrams of $X^0$ in Fig. 1(a) and (b), each showing a ladder of two-level quantum systems coupled by nuclear spin-flip processes. Here we adopt a mean-field description of the nuclear spins by neglecting the quantum fluctuations in the Overhauser field ($I_z = \langle I_z \rangle$) and limit ourselves to effective spin $1/2$ nuclei for simplicity. For a given nuclear spin polarization $I_z$, e.g. $| \uparrow \downarrow \rangle$, we can label the two level system by the states $| 0, I_z \rangle$ and $| \uparrow \downarrow \rangle$. The ground states $| 0, I_z \rangle$ differ by an energy $\delta_1 = \omega^2_2$. The corresponding energy differences for the excited states are $\delta_2 = \omega^2_0 + A_1 I_z$ and $\omega^2_2 - A_1 I_z$ for the blue and red Zeeman transition, respectively. The transition rate associated with hyperfine-assisted laser coupling is given for the blue Zeeman branch (Fig. 1(a)) by

\begin{equation}
W_{\pm}(I_z) = \left( \frac{\Omega A_i^{\text{nc}}}{4\omega_0^2} \right)^2 \frac{\Gamma_0}{4\delta_2^2 + \Gamma_0^2 + \Omega^2/2}.
\end{equation}

where $\Omega$ is the laser Rabi frequency and $\Gamma_0$ the radiative decay rate [25]. A remarkable feature of $W_{\pm}(I_z)$ is its dependence on the sign of the laser detuning entering through the effective optical detuning $\delta_\pm = \Delta - A_1 (I_z \pm 1) \mp \omega^2_2$: when the incident laser field is red (blue) detuned, the transition rate $W_{+}(I_z)$ ($W_{-}(I_z)$) dominates over $W_{-}(I_z)$ ($W_{+}(I_z)$) and ensures that the Overhauser field increases (decreases). This directional DNSP will in turn result in a decrease of the effective detuning $\delta$ from $\Delta - A_1 I_z$ to $\Delta - A_1 (I_z + 1)$ for a red detuned laser and to $\Delta - A_1 (I_z - 1)$ for a blue detuned laser. If initially $I_z \ll N/2$, then DNSP will continue until $\delta \approx 0$.
parametric plot in (c) depicts the dragging width obtained for incre-
sions leading to pure nuclear spin diffusion at rate which includes nuclear-spin-flip assisted spontaneous emis-
us, respectively). The steady state nuclear spin polarization

dependent on the steady-state solution. The absorption

differentiation away from the laser resonance. The experiments validate

determination of scan speed. Both the detuning step \( \Delta_n \) and the waiting

time constant \( t_c \) used for signal integration after each step contribute to the

effective scan speed of the laser detuning: for a given \( \Delta_n \) the maximum

While a laser scan across the blue transition leads to a posi-
tive feedback of the nuclear spins to ensure locking condition,
a scan across the red Zeeman line causes an anti-dragging ef-
fect. To understand this, we note that the effective optical detun-
ing in this case is \( \delta_{\pm} = \Delta + A_{\pm}(I_L \pm 1) \mp \omega^2_2 \). The simple

device change in the effective optical detuning renders the exact

To obtain a quantitative prediction, we consider the rate

equation

\[
\frac{dI_z}{dt} = W_+(I_z)(\frac{N}{2} - I_z) - W_-(I_z)(\frac{N}{2} + I_z) - \Gamma_d I_z
\]

which includes nuclear-spin-flip assisted spontaneous emission

processes leading to pure nuclear spin diffusion at rate \( \Gamma_d \) [17]. The steady-state solution exhibits bistability due to the

nonlinear dependence of the rates \( W_\pm(I_z) \). For a given initial laser detuning \( \Delta \) the solution of the rate equation yields the

steady state nuclear spin polarization \( I_z \) and thus the effective optical detuning \( \delta \) as established within the integration

time \( t_c \) of the experiment. The absorption spectrum is calcu-

lating by varying the laser detuning in discrete steps \( \Delta_n \). The

rate equation (3) is symmetric with respect to the laser detun-
ing. In order to account for the asymmetry observed experi-
mentally for the two scan directions, we refined the model by

including terms for spin-flip Raman scattering processes that

arise from the Fermi-contact hyperfine interaction as well as an

unbalanced telegraph noise in the resonance condition [17].

We find excellent agreement between theory and experiment, as demonstrated in Fig. 4.

Remarkably, the model also reproduces the dependence of DNSP in resonant laser scattering on key experimental parameters. Fig. 4(c) shows how the dragging width evolves as a function of scan speed. Both the detuning step \( \Delta_n \) and the waiting time constant \( t_c \) used for signal integration after each step contribute to the effective scan speed of the laser detuning: for a given \( \Delta_n \) the maximum width increases non-linearly with \( t_c \), as exemplified in Fig. 4(a) for \( \Delta_n = 1.84 \mu eV \). The non-linearity in the functional dependence makes our simulations highly sensitive to the set of parameters that determine the DNSP dynamics; in particular, it allows us to extract a value for \( A_{nc}^\theta \) for a given set of \( N \) and \( A \). Fig. 4(c) demonstrates that the full dynamic range of the experiment is correctly captured with the following set of parameters: \( \hbar \Gamma_0 = 0.7 \mu eV \), \( \Omega = 0.5 \mu eV \), \( B = 4.5 \mu T \). The non-collinear hyperfine coupling constant found from simulations is in good agreement with that obtained independently from atomistic calculations and nuclear spin decay measurements [22]. The same set of parameters was also used to reproduce the external magnetic field and the laser power dependence [17] and to calculate the absorption spectra in Fig. 1.

Our results establish quadrupolar interaction induced non-
collinear hyperfine coupling as the mechanism responsible for resonant bidirectional DNSP that is ubiquitous for self-assembled QDs. An obvious extension of our work will be to carry out similar experiments in interface or droplet QDs where quadrupolar interactions are vanishingly small.

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[21] Experimentally determined values of hole-hyperfine interaction would suggest that the resulting nuclear spin-flip rates would be an order of magnitude too small to explain the relative insignificance of the directional reverse Overhauser effect. In addition, the large apparent variation in heavy-light-hole mixing in QDs as indicated by the measured in-plane hole $g$-factors would suggest that dragging features would change appreciably from QD to QD, if the mechanism was due to hole-hyperfine interaction.
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[25] By carrying out a complete numerical analysis, we have confirmed that the saturation of $W_\pm(I_z)$ is correctly taken into account in Eq. (2).
SUPPLEMENTARY ONLINE MATERIAL

Description of the experiment
A voltage applied to the Schottky top gate was used to tune the QD into stable charge configurations with the ground state being singly positively charged, neutral or singly negatively charged \[1, 2\]. Single electrons were injected into the QD from the Fermi reservoir, whereas in absence of a hole reservoir in the device, single hole injection was ensured by the presence of a weak non-resonant laser. The sample was cooled in a bath cryostat to liquid helium temperature (4.2 K) and finite magnetic fields were applied in Faraday or Voigt configuration, parallel or perpendicular to the sample growth axis \(z\), respectively.

Resonant absorption of the neutral exciton \(X^0\) as well as positive and negative trions, \(X^+\) and \(X^-\), was probed with a tunable narrow-band laser in differential transmission spectroscopy \[3\]. Absorption spectra were recorded by setting the gate voltage or the laser energy to a specific detuning \(\Delta = \omega_X - \omega_L\), waiting for a time \(t_c\) and monitoring the transmission signal with a lock-in amplifier. After each measurement, the voltage/laser energy was changed by a discrete detuning step \(\Delta_n\).

In finite magnetic fields, QDs in both samples A and B showed pronounced dragging. QDs in sample A, for example, exhibited dragging of both \(X^0\) and \(X^-\) in the range of tens of \(\mu\)eV (Fig. 1) in contrast to QDs of sample B with sub \(10 \mu\)eV scale for both trions (Fig. 2). The thickness of the tunnel barrier strongly affects electron spin exchange with the Fermi reservoir via co-tunneling \[4\] and thus the efficiency of DNSP \[5\]. In particular, in sample B the dragging widths of \(X^-\) are reduced due to strong spin pumping at magnetic fields exceeding \(0.3 \ T\) \[6\]. In Voigt geometry the spin pumping leads to a significant reduction in the transmission contrast and consequently in the dragging efficiency even at the edge of the charging plateau. However, the \(X^0\) of the same QD from sample B exhibited a \(\approx 20 \ \mu\)eV dragging width, consistent with the findings from sample A. For \(X^+\) the reduced DNSP range is consistent with the presence of a non-resonant laser which not only injects holes into the ground state of \(X^+\) but also non-geminate electron spins opening up an additional nuclear spin decay channel. The nuclear spin polarization is therefore reduced for (photo-generated) single-hole-charged initial states.

Non-collinear hyperfine interaction
The effective non-collinear hyperfine interaction Hamiltonian stems from the fact that the quadrupolar interaction Hamiltonian for a nuclear spin with strain axis tilted by an angle \(\theta\) from the \(z\)-axis (in the \(x - z\) plane)

\[
\hat{H}_{\text{quad}} = B_Q (I_x^2 \cos^2 \theta + (I_z I_x + I_x I_z) \sin \theta \cos \theta + I_x^2 \sin^2 \theta)
\]

(1)
does not commute with the dominant \(\hat{H}_{\text{Fem}} = \sum_i A_i I_z^2 S_z\) term of the Fermi-contact hyperfine interaction \(\hat{H}_{\text{Fem}}\). To obtain an analytic expression for \(A_i^{\text{nc}}\), we assume \(\theta \ll 1\) and use a Schrieffer-Wolff (SW) transformation to obtain \(\hat{H}_{\text{hyp}} = \hat{H}_{\text{Fem}} + \hat{H}_{\text{hyp-quad}}\) where

\[
\hat{H}_{\text{hyp-quad}} = A_i^{\text{nc}} \hat{S}_z [\hat{I}_x^2 \hat{I}_z + \hat{I}_x^2 \hat{I}_z] ,
\]

(2)
with \(A_i^{\text{nc}} = A_i B_Q \sin(2\theta_i) / (2 \omega_{0i}^n)\). In \(\hat{H}_{\text{hyp-quad}}\) we have only kept the terms that describe processes which leave the electron spin-state unchanged, since contributions that flip the electron spin will be negligible at high external fields.

Finally, we note that even for large \(B_z\), the dominant role of flip-flop terms of Fermi-contact hyperfine interaction is to induce indirect interaction between the QD nuclei \[7\]: the primary effect of this interaction, in the presence of fast optical dephasing of the electronic spin resonance, is to ensure that the nuclear spin population assumes a thermal distribution on timescales fast compared to the polarization timescale determined by \(A_i^{\text{nc}}\). In this limit, the dynamics due to \(\hat{H}_{\text{hyp-quad}}\) will be indistinguishable from that described by \(\hat{H}_{\text{Fem}}\).

Averaging over the distribution for nuclei that lie within the Gaussian QD electron wavefunction, we obtain for cations (In and Ga with 9/2 and 3/2 nuclear spins, respectively, and \(A_i^{\text{nc}} = 0.3 \text{ eV} \approx 0.0062 \cdot A_{i^{\text{nc}}}^{\text{In}},Ga\) and for anions (As with 3/2 nuclear spin and \(A_i^{\text{nc}} = 0.3 \mu\text{eV} \approx 0.0424 \cdot A_{i^{\text{nc}}}^{\text{As}}\). For a fully polarized \(\text{In}_0\gamma\text{Ga}_{0.3}\text{As}\) system we determine the value for the maximum Overhauser splitting due to the non-collinear hyperfine coupling as \(A_i^{\text{nc}} = 0.0002 \cdot 0.7 \cdot \frac{3}{2} A_{i^{\text{nc}}}^{\text{In}} + 0.3 \cdot \frac{3}{2} A_{i^{\text{nc}}}^{\text{Ga}}\) + 0.0424 \cdot 0.5 \cdot \frac{3}{2} A_{i^{\text{nc}}}^{\text{As}} \approx 4.14 \mu\text{eV} and obtain an average value of \(A_i^{\text{nc}} \approx 1.3 \cdot 10^{-4} \mu\text{eV} with \(N = 3.2 \cdot 10^4\).

Next, we discuss the derivation of the rate equation for the neutral exciton blue Zeeman transition. We consider the limit of a large external magnetic field where \(\omega_{2i} < \omega_X < \Omega\), with \(\Omega \approx \Gamma_0\) (\(\omega_{2i}\) and \(\omega_X\) are the electron and nuclear Zeeman energies, \(\Omega\) the laser Rabi frequency and \(\Gamma_0\) the radiative decay rate). In this limit, all nuclear spin-flip processes, including those described by \(\hat{H}_{\text{Fem}}\) are energetically forbidden to first order in perturbation theory. Eliminating \(\hat{H}_{\text{Fem}}\) by a SW transformation we arrive at the following correction terms to the laser-exciton coupling

\[
\hat{H}_{\text{Fem-}\text{laser}} = \frac{1}{2} \sum_i \Omega_i A_i^{\text{nc}} / 2 \omega_{0i}^n \left( (\hat{\sigma}_{0X} - \hat{\sigma}_{X}\hat{I}_y) \hat{I}_y \right)
\]

(3)
with \(\hat{\sigma}_{0X} = \hat{X}_0\). Here, \(\hat{X}_0\) and \(\hat{X}_0\) denote the exciton and vacuum state, respectively. Application of the same SW transformation to the Liouvillian term leads to nuclear-spin-flip assisted spontaneous emission terms with maximum rate \(\approx \Gamma_0 (A_i^{\text{nc}} / 4 \omega_{0i}^n)^2\). In the limit \(\omega_{2i} \approx \Gamma_0\) of interest, the denominator in Eq. (3) should be modified to take into account broadening of the excitonic spin states due to spontaneous emission.

The rates for spin-flip Raman scattering processes arising from the Fermi-contact hyperfine interaction take place at a rate \(\approx \Gamma_0 / (A_i^{\text{nc}} / 4 \omega_{0i}^n)^2\); given that \(\omega_{2i} \approx 1000 \\omega_{2i}\) and \(A_i^{\text{nc}} \approx 0.02 A_i\), we conclude that the latter processes will take place at a rate that is \(\approx 300\) times slower.
FIG. 1: (a) Simulations of dynamic dragging as in Fig. 1 of the manuscript yet without spectral jitter: the solid and dashed lines represent steady state solutions for scans (t_e = 0.2 s and Δ_n = 0.23 μeV) with and without spin-flip Raman scattering processes according to Eqn.s 6 and 4, respectively. The slight asymmetry to positive laser detunings is a result of directional dynamic nuclear spin polarization stemming from spin-flip Raman processes. (b) Comparison between experimental spectra (dark cyan) and results of the simulation (blue) for sequential data acquisition (t_e = 60 s and Δ_n = 0.46 μeV): each data point of the spectra was obtained by (i) erasing the nuclear spin polarization in a voltage region with strong co-tunneling, (ii) subsequently establishing a finite laser detuning, and (iii) integrating for the time t_e at this specific detuning. The Lorentzian spectra in (a) and (b) are calculated with Γ_0 = 0.73 μeV and shown in black for reference.

Modelling of the experimental data

To obtain a quantitative prediction, we consider the rate equation

\[ \frac{dI_x}{dt} = W_+(I_x)(\frac{N}{2} + I_x) - W_-(I_x)(\frac{N}{2} - I_x) - \Gamma_d I_x, \tag{4} \]

where

\[ \Gamma_d = \Gamma_0 \left( \frac{A_{nc}^2}{4\omega_Z^2} \right)^2 \frac{\Omega^2/4}{\delta^2 + \Gamma_0^2/4 + \Omega^2/2} \tag{5} \]

is the rate at which nuclear-spin-flip assisted spontaneous emission, leading to pure nuclear spin diffusion, takes place. Here, as well as in the following equation, \( \delta = \Delta - A_1 I_x \). The rate equation yields symmetric dragging (dashed spectra in Fig. 1(a)) and anti-dragging to either side of the resonance for the blue and red Zeeman branches, respectively, qualitatively similar to the results of Yang and Sham [5].

Taking into account uni-directional spin-flip Raman scattering processes that arise from the Fermi-contact hyperfine interaction, we arrive at a refined rate equation model:

\[ \frac{dI_x}{dt} = W_+(I_x)(\frac{N}{2} + I_x) - W_-(I_x)(\frac{N}{2} - I_x) - \Gamma_d I_x - \Gamma_{sf} \left( \frac{N}{2} + I_x \right) \tag{6} \]

with

\[ \Gamma_{sf} = \Gamma_0 \left( \frac{A_i}{4\omega_Z^2} \right)^2 \frac{\Omega^2/4}{\delta^2 + \Gamma_0^2/4 + \Omega^2/2}. \tag{7} \]

Spin-flip Raman scattering processes at rate \( \Gamma_{sf} \) give rise to the asymmetry in the spectra for forward and reverse scan directions (compare solid and dashed spectra in Fig. 1(a)), in agreement with experimental findings. The argument for the asymmetry holds when the nuclear spin polarization is erased before a sudden jump to a finite detuning and subsequent build-up of DNSP [5]: locking of the resonance extends further for positive laser detunings (Fig. 1(b)). Moreover, spin-flip Raman scattering processes ensure small but finite nuclear spin polarization at high laser powers, as shown in the inset of Fig. 2(a).

The normalized absorption spectra depicted in Fig. 1(a) are calculated from steady state solutions of Eqn.s 4 and 6 with the following parameters: \( h\Gamma_0 = 0.7 \) μeV from the radiative decay rate \( 1/\Gamma_0 = 1.2 \) ns determined from saturation [10], \( \Omega = 0.5 \Gamma_0, B = 4.5 \) T, step size \( \Delta_n = 0.23 \) μeV.
and dwell time $t_c = 0.2$ s, as used in the experiments, and $\omega_0^2/\omega_2^2 = 1000$, $N = 3.2 \times 10^4$, $A_i = 240 \mu eV/N$, $A_{hc}^i = 0.45 \times 10^{-4} \mu eV$. Intrinsic decay of the nuclear spin polarization is negligible for the ground state of $X^0$ \[7, 11\]. Here, we omitted the unbalanced telegraph noise in the resonance condition used to calculate the spectra in Fig. 1 of the manuscript. This jitter in the resonance condition with an amplitude of $0.5 \mu eV$ (smaller than the linewidth) and timescales longer than $t_c$ was included in simulations in order to account for the asymmetry observed experimentally for the two scan directions. It is consistent with the spectral fluctuations in resonant QD spectroscopy \[10\]. Based on experimental observations for the QDs in sample A, it is reasonable to assume these fluctuations to be unbalanced with a small weight on the higher energy side of the resonance. However, comparing the observed traces for different QDs in different samples reveals that the spectral jitter can actually appear on either side of the resonance.

The dependence of the dragging width on the magnitude of the laser power and the external magnetic field provides further confirmation of the model. Fig. 2 shows how the dragging width evolves as a function of laser power and magnetic field. The effect of dragging is inhibited at low incident powers, increases until reaching a maximum below the saturation at $\Omega \simeq \Gamma_0$, and vanishes in the limit of high excitation powers (Fig. 2a). This is consistent with the prediction of Eqs. 4 and 6: the maximum dragging width is expected at $\Omega < \Gamma_0$ for non-vanishing $\Gamma_d$ and $\Gamma_{sf}$. Both $X^0$ and $X^-$ reveal the same dependence when the dragging width on the ordinate is normalized to the total linewidth $\Gamma_{tot} = \sqrt{\Gamma_0^2 + \Omega^2/2}$ and the abscissa is expressed in units of $\Omega/\Gamma_0$. The results of the model reproduce our experimental findings (solid line in Fig. 2a) predicting $\sim 10\%$ of nuclear spin polarization at maximum (inset of Fig. 2a). The model overestimates the dragging width at powers below saturation but gives perfect agreement above saturation for $\Omega = \Omega/2$. We speculate that the scaling factor of 2 stems from the line broadening $\Gamma \simeq 2 \Gamma_0$ that we typically find in our samples as a result of spectral fluctuations \[10\]. The monotonic sublinear increase of the dragging width with magnetic field, as measured on $X^0$ close to saturation, is clearly reproduced by our model (solid line in Fig. 2b) and provides further confirmation for the quantitative nature of our analysis.

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