Stimulated Raman spin-coherence and spin-flip induced hole burning in charged GaAs quantum dots

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High-resolution spectral hole burning (SHB) in coherent non-degenerate differential transmission spectroscopy discloses spin-trion dynamics in an ensemble of negatively charged quantum dots. In the Voigt geometry, stimulated Raman spin coherence gives rise to Stokes and anti-Stokes sidebands on top of the trion spectral hole. The prominent feature of an extremely narrow spike at zero detuning arises from spin population pulsation dynamics. These SHB features confirm coherent electron spin dynamics in charged dots and the linewidths reveal spin spectral diffusion processes.

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Single electron spin localized in semiconductor quantum dots (QDs) has attracted a great deal of interest due to its potential use in quantum applications [1]. Experimental and theoretical efforts have been focused on controllable coherent spin dynamics and possible decoherence mechanisms [2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14]. In this paper, we report spectral hole burning (SHB) in coherent differential transmission (DT) spectroscopy induced by spin-trion dynamics in an ensemble of negatively charged QDs. Spin coherence induced SHB by stimulated Raman excitation and spin relaxation induced SHB due to the population pulsation dynamics are observed in the QD system. Features of SHB not only disclose important spin dynamics observed from transient spectroscopy [2] and phase modulation techniques [11], but also provide information on spin spectral diffusion (SD) processes [14] which are not easily revealed by previous methods.

The interface fluctuation GaAs/Al0.3Ga0.7As QDs are molecular beam epitaxy grown with growth interrupts, and modulation Si doping in the barrier incorporates excess electrons [15]. The pump \( E_1(\omega_1) \) and probe \( E_2(\omega_2) \) optical fields are derived from two frequency-stabilized and independently tunable CW lasers with a mutual coherence bandwidth of 20 neV, which is crucial for this experiment as discussed below. The sample is kept inside a superconducting magnetic liquid helium flow cryostat and the temperature is maintained at 4.5 K. The DT signal is homodyne detected with the probe field by a photodiode and extracted by a lock-in amplifier.

A nonlinear degenerate DT spectrum with \( \omega_1 = \omega_2 \) [Fig. 1(a)] shows the trion and exciton ensemble resonances. Their assignments are confirmed by both photoluminescence and transient quantum beats studies (data not shown). The trion binding energy (i.e., the separation between exciton and trion resonances) is measured to be 2.9 meV, in agreement with earlier reports of photoluminescence [15] and transient spectroscopy [2]. The ensemble trion inhomogeneous broadening width (\( \sim 2.5 \) meV) can be estimated from the broad Gaussian profile of the trion resonance.

To study SHB with non-degenerate DT spectroscopy, the pump beam is fixed near the trion ensemble peak and the probe beam is detuned (\( \Delta = \omega_2 - \omega_1 \)). A narrow spectral structure appears, exhibiting a double-Lorentzian-like shape, i.e., a narrower Lorentzian peak on top of a broader one [Fig. 1(c)]. As established below, the narrower Lorentzian peak is due to the trion population relaxation dynamics and its linewidth (\( \sim 12 \) meV) gives the trion population relaxation rate. The broader Lorentzian peak (\( \sim 50 \) meV) underneath this resonance is due to trion population relaxation and coherence decay broadened by the trion SD process. This complex lineshape and its unfolded SHB features with a magnetic field in the Voigt geometry are the focus of our study.

The dynamics of the QD ensemble in the presence of
characterized by its resonance energy $\epsilon$. $H$ is the total Hamiltonian including the interaction with the coherent optical fields. The second term on the RHS is a generalized relaxation term that describes population decay and pure dephasing. The last term is due to various SD processes [17]. Eq. (1) without the SD term reduces to the standard OBE.

FIG. 2: The calculated SHB lineshape of 2-level system with different spectral diffusion rates, where $\Lambda_t = 1000\Gamma_t$ and $\gamma_t = \Gamma_t/2$. (a) $\Gamma_t^{SD} = 0$. (b) $\Gamma_t^{SD} = \Gamma_t$. (c) $\Gamma_t^{SD} = 10\Gamma_t$.

the radiation field and various interactions with the environment is described by the modified optical Bloch equation (OBE) [16]:

$$ih\frac{\partial \rho(t)}{\partial t} = [H, \rho(t)] + \frac{\partial \rho(t)}{\partial t}\bigg|_{\text{relax}} + \frac{\partial \rho(t)}{\partial t}\bigg|_{\text{SD}}$$ (1)

where $\rho(t)$ is the density matrix of each ensemble member

$$P^{(3)}_{NL} \approx -\frac{iN|\mu|^4|E_1|^2E_2^*}{2\hbar^2\Lambda_t} \left\{ \frac{1}{\Gamma_t + \Gamma_t^{SD} + i\Delta} \left[ \frac{1}{\Lambda_t + 2(\gamma_t + \Gamma_t^{SD}) + i\Delta} \right] + \frac{\sqrt{\pi}}{\Gamma_t + \Gamma_t^{SD}} \left[ \frac{1}{\Lambda_t (\Gamma_t + i\Delta)} \right] \right\}$$ (3)

where $\Gamma_t (\gamma_t)$ is trion population (coherence) decay rate, $N$ is the total number of excited charged QDs, $\Lambda_t$ is the ensemble inhomogeneous broadening width of the trion resonance and $\mu$ the optical dipole of a single dot. We have assumed a redistribution kernel $W_t(\epsilon, \epsilon') = \Gamma_t^{SD}\exp(-|\epsilon-\epsilon|^2/(\Lambda_t)^2)/(\sqrt{\pi}\Lambda_t)$ where $\epsilon$ is the ensemble averaged resonance energy [21]. Eq. (3) holds when the approximation of plasma dispersion function is taken [22] because $\Gamma_t, \gamma_t, \Gamma_t^{SD}, \Delta \ll \Lambda_t$ and the pump frequency is fixed near the center of the ensemble trion spectrum.

The SD process significantly changes the trion SHB lineshape and linewidth, which can be discussed in three regimes, as schematically shown in Fig. 2. $\Gamma_t = 2\gamma_t$ is assumed only for the theoretical calculation in Fig. 2 to simplify discussions, since pure dephasing has been found negligible for trion states at 4.5 K [23]. When $\Gamma_t^{SD} \ll \Gamma_t$, the trion SHB in the DT spectrum reduces to the standard Lorentzian squared with width $\sim \Gamma_t$. Secondly, when $\Gamma_t^{SD} \approx \Gamma_t$, the trion lineshape is $(2\Gamma_t^2 + \Delta^2)(\Gamma_t + \Gamma_t^{SD})^2 + \Delta^2)^{-1}[(\Gamma_t + 2\Gamma_t^{SD})^2 + \Delta^2]^{-1}$ where the linewidth is considerably broadened by the SD process. Finally, when $\Gamma_t^{SD} \gg \Gamma_t$, the lineshape changes to a double Lorentzian-like profile with larger FWHM of $2(\Gamma_t + \Gamma_t^{SD})$ and smaller FWHM of $2\Gamma_t$. The physical origin of the narrower Lorentzian is due to the population pulsed effect [24]. This explains the observed lineshape.
order spin population $\rho$ of the trion, which oscillates at the detuning frequency $\Delta$ (known as population pulsations [24]). In the presence of the spin population relaxation process, the standard OBE predicts a hole burning at $\Delta = 0$ with a linewidth given by the spin relaxation rate $\sim \Gamma_s [23]$. However, as shown in Figs. 3 and 4 the measured linewidth is orders of magnitude larger than $\Gamma_s \sim 0.1$ neV measured by the phase modulation technique for these QDs [11]. We will establish below that the extra broadening is likely to be due to SD processes involving the spin states.

Similar to the trion SD given in Eq. (2), the effects of spin SD processes on the spin population and coherence are given by,

$$
\frac{\partial \rho_{x,x}(\epsilon_t, \epsilon_s)}{\partial t}_{\text{SD}} = W_s(\epsilon_t, \epsilon_s; \epsilon'_t, \epsilon'_s) \rho_{x,x}(\epsilon'_t, \epsilon'_s) d\epsilon'_t d\epsilon'_s - \Gamma_s(\epsilon_t, \epsilon_s) \rho_{x,x}(\epsilon_t, \epsilon_s)
$$

(5)

$$
\frac{\partial \rho_{x,x}(\epsilon_t, \epsilon_s)}{\partial t}_{\text{SD}} = -\Gamma_s(\epsilon_t, \epsilon_s) \rho_{x,x}(\epsilon_t, \epsilon_s)
$$

(6)

where the density matrix for each QD in the ensemble is now characterized by two variables, i.e., the zero field trion resonance energy $\epsilon_t$ and the spin Zeeman splitting $\epsilon_s$ in the external magnetic field plus the local (e.g., nuclear Overhauser) field. Similar to the description of trion SD, $W_s$ is the redistribution kernel and $\Gamma_s(\epsilon_t, \epsilon_s) = \int W_s(\epsilon'_t, \epsilon'_s; \epsilon_t, \epsilon_s) d\epsilon'_t d\epsilon'_s$ is the spin SD rate. The qualitative feature of the redistribution kernel function depends critically on the SD mechanism. Local nuclear field fluctuation induced SD only affects the spin Zeeman splitting: $W_s = f(\epsilon_t, \epsilon_s) \delta(\epsilon_t - \epsilon'_t)$. We note that the inhomogeneous broadening of $\epsilon_s$ induced by the nuclear field is given by $\Lambda_s \sim 0.1$ meV $< \gamma_t [28]$.

We also note that two quantum dots are equally excited if the dependence in their resonance frequency for spin to trion transition is much smaller than the trion broadening $\gamma_t$. Thus, $\rho_{x,x}(\epsilon_t, \epsilon_s) = \rho_{x,x}(\epsilon'_t, \epsilon'_s)$ if $|\epsilon_t - \epsilon'_t| \pm |\epsilon_s - \epsilon'_s|/2 < \gamma_t$. It can then be shown that the two terms on RHS of Eq. (5) cancel each other and hence this mechanism has a negligible effect on the linewidth of the ultra-narrow central spike associated with the spin population pulsation dynamics. On the other hand, if the SD process is due to the interdot transfer of non-equilibrium spin population, it is more reasonable to assume a redistribution kernel $W_s = \Gamma_s^{SD} \exp[-(\epsilon_s - \bar{\epsilon}_s)^2/\Lambda_s^2 - (\epsilon_t - \bar{\epsilon}_t)^2/\Lambda_t^2]/(\pi \Lambda_s \Lambda_t)$ where $\bar{\epsilon}_t$ ($\bar{\epsilon}_s$) is the ensemble averaged trion (spin) resonance energy [21]. In the vicinity of zero detuning $\Delta \ll \gamma_t$, the DT signal is determined by

$$
E_{NL} \simeq \frac{N \sqrt{\pi} |\mu|^4 |E_1|^2 E_2^*}{8h^3 \Lambda_s (2\gamma_t + \Gamma_s^{SD})} 2\Gamma_s + \Gamma_s^{SD} \Delta^2 + (2\Gamma_s + \Gamma_s^{SD})^2
$$

(7)

shown as the sharp central spike with linewidth of $2\Gamma_s + \Gamma_s^{SD}$ in Fig. 4 (Theory). The experimental data at various magnetic fields are shown in Fig. 4 (Experiment), and the measured linewidth of the sharp central spike is plotted in Fig. 4(b) from which we extract $h\Gamma_s^{SD} \sim 0.2$ meV.
In addition to the sharp central spike, the newly emerged SHB features in Voigt geometry also include two symmetric sidebands as highlighted by the ellipse regions in Fig. 3(b). These sidebands can be understood from the perturbation pathway:

\[
\rho^{(0)}_{\tilde{x}, \tilde{x}} \rightarrow \rho^{(1)}_{\tilde{x}, \tilde{x}} \rightarrow \rho^{(2)}_{\tilde{x}, \tilde{x}} \rightarrow \rho^{(3)}_{\tilde{x}, \tilde{x}}
\]  

(8)

associated with stimulated Raman spin excitations [2]. The Stokes and anti-Stokes sidebands appear respectively at \( \Delta = \pm \mu_B g^S_e B / \hbar \) and their separation as a function of the magnetic field gives the ensemble averaged electron g-factor \( \bar{g}^S_e \approx 0.13 \) (see Fig. 3(a)), in agreement with earlier reports [2]. As the sideband feature is associated with the Raman spin coherence, it can be inferred from Eq. (6) that spin SD process will broaden the homogeneous linewidth of the sidebands from \( \gamma_s \) to \( \gamma_s + \Gamma^{SD}_s \), where \( \gamma_s \) is the spin decoherence rate. The ensemble averaged sideband lineshapes are a convolution of the homogeneous lineshape with the inhomogeneous broadening of the spin Zeeman energy. In the vicinity of \( \Delta = \pm \mu_B g^S_e B / \hbar \), the nonlinear DT signal is

\[
E_{NL} \approx \frac{N|\mu|^4|E_1|^2E_2^2(\gamma_s + \Gamma^{SD}_s)}{8\hbar^3x\lambda_1\lambda_2\lambda_3(\gamma_s + \Gamma^{SD}_s)} \times \int \exp\left(-\left(\epsilon - \mu_B g^S_e B\right)^2/\Lambda_s^2\right) \left(\gamma_s + \Gamma^{SD}_s\right)^2 \left((\Lambda - \epsilon)^2\right) d\epsilon.
\]  

(9)

At finite magnetic field, the spin inhomogeneous broadening \( \Lambda_s(B) = \Lambda_s(0) + \mu_B \Delta g^S_e B \) has the contribution from the inhomogeneity of the electron g-factor \( \Delta g^S_e \) in addition to the nuclear induced zero field inhomogeneous broadening \( \Lambda_s(0) \). The sideband linewidth measured at various magnetic fields is shown in Fig. 5(b), which is consistent with that calculated using Eq. (9) with parameters as \( h(\gamma_s + \Gamma^{SD}_s) \approx 0.18 \mu eV \) and \( \Lambda_s \approx 0.64 B \mu eV / T + 0.1 \mu eV \). As \( \gamma_s \ll \Gamma^{SD}_s \) from the theoretical investigation of both the nuclear and phonon induced spin decoherence [12, 13], we can get \( \hbar \Gamma^{SD}_s = 0.18 \mu eV \) which agrees with the value of \( \Gamma^{SD}_s \) extracted from the central narrow spike. In addition, the spin g-factor variation \( \Delta g^S_e \) is determined to be 0.01 from \( \Lambda_s \), so \( \Delta g^S_e / g^S_e \approx 0.1 \), in agreement with transient spin quantum beat measurements [2].

In summary, we have shown in this paper that SD from the trion state complicates the trion SHB profile, and makes an important contribution to the double-Lorentzian-like lineshape. In the Voigt geometry, we have found a complex lineshape arising from spin dynamics in the spectral hole burning: a narrow central spike and two symmetric Stokes and anti-Stokes sidebands. They have been theoretically identified to result as consequence of spin population pulsation dynamics and stimulated Raman spin coherence, respectively. Moreover, a spin SD process is observed in contributions to the SHB lineshape, and has been theoretically identified as interdot transfer of the non-equilibrium spin population. Possible mechanisms include the interdot spin flip-flop interactions of the electrons or spin conserved electron tunneling to adjacent neutral dots. Their quantitative estimates can not be determined without a detailed calculation which is beyond the scope of the paper. Nevertheless, the existence of these decoherence mechanisms revealed by our experiments will have important impacts on the efforts towards spin based quantum applications in these kinds of QDs.

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