Nonlinear optical response of hole–trion systems in quantum dots in tilted magnetic fields

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Received XXXX, revised XXXX, accepted XXXX
Published online XXXX

Key words: nonlinear optical spectroscopy, spin, decoherence, magnetooptical Kerr effect

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We discuss, from a theoretical point of view, the four wave mixing spectroscopy on an ensemble of p-doped quantum dots in a magnetic field slightly tilted from the in-plane configuration. We describe the system evolution in the density matrix formalism. In the limit of coherent ultrafast optical driving, we obtain analytical formulas for the single system dynamics and for the response of an inhomogeneously broadened ensemble.

The results are compared to the previously studied time-resolved Kerr rotation spectroscopy on the same system. We show that the Kerr rotation and four wave mixing spectra yield complementary information on the spin dynamics (precession and damping).

1 Introduction. The properties of confined spins in semiconductor nanostructures are interesting because of their expected important role in quantum computing and spintronic devices. In particular, the hole spin attracts much attention because of its enhanced coherence time. In systems that efficiently couple to optical fields, like quantum wells and self-assembled quantum dots, an interesting possibility is to study the spin dynamics using optical spectroscopy tools. A method which is particularly suited for this purpose is the time-resolved Kerr (or Faraday) rotation (TRKR, TRFR) [1,2,3], where the evolution of the occupations of the hole and trion Zeeman states is traced by investigating the rotation of the polarization plane of the reflected or transmitted probe pulse.

In a recent work [4], we proposed a general description for the dynamics of a confined hole-trion system in a tilted magnetic field. We showed that a TRKR experiment provides rich information about the rates of spin precession and decoherence (both longitudinal and transverse). In particular, the optical response at slightly tilted magnetic fields contains contributions related both to the hole spin relaxation (longitudinal decoherence) with respect to the hole spin quantization axis, as well as the dephasing of spin coherences (transverse decoherence) with respect to this axis. Under favorable experimental conditions (a sufficient separation of time scales), the corresponding two decoherence rates can be deduced from a single run of an experiment, although in a realistic system the latter may be convoluted with (or even dominated by) the inhomogeneous dephasing.

Although the TRKR or TRFR method may be the most obvious choice for the investigation of the spin dynamics the spin precession of the trion and hole in a tilted field will lead to transitions between optically active and inactive states which should be manifested in any form of the optical response. In particular, signatures of the spin-related dynamics should be visible in the four wave mixing (FWM) nonlinear spectroscopy. As the FWM spectroscopy is one of the most widely used methods in the investigation of semiconductors and their nanostructures [5,6] it may be interesting to study how the spin precession and decoherence affect the FWM response and what information on the spin-related kinetics can be extracted from this kind of experiments.
In this contribution, we study the FWM response of the same system of confined holes as discussed in our previous work [4]. We show that the FWM signal is also affected by the spin dynamics in a way that allows one to extract the Larmor frequencies of electrons and holes. The available information on the decoherence rates is less specific than that contained in the TRKR response and is available only as long as the hole spin dephasing times are not much longer than the lifetime of the optical coherence. On the other hand, the spins undergo partial refocusing in the two-pulse optical echo experiment which leads to the appearance of components in the optical response that depend on the inherent spin dephasing rates but are insensitive to the inhomogeneous distribution of the Larmor frequencies. In this way, the FWM method may be a valuable experimental tool to study the spin dynamics, complementary to the TRKR or TRFR techniques.

The paper is organized as follows. In Sec. 2 we define the model under study. In Sec. 3 we derive the FWM response of the system. Next, in Sec. 4 we discuss the result and compare the spin-related information contained in the FWM signal to that available from the TRKR response.

2 Model. We study a system similar to that discussed in our previous work [4], consisting of an ensemble of quantum dots or trapping centers in quantum wells with one confined hole in each of the dots. The system is excited with a pump pulse at the time \( t = -\tau \) and a probe pulse at \( t = 0 \). Unlike in our previous work, we are now interested in the FWM response from the system. Experimentally, the relevant third-order response is isolated by choosing the appropriate excitation and detection directions (see Ref. [4] for a discussion). In the modeling, we assume the pump and probe pulses to have phases \( \phi_1 \) and \( \phi_2 \), respectively, and calculate the terms in the response of an inhomogeneously broadened system that carry the phase \( 2\phi_2 - \phi_1 \). Out of many possible configurations of the polarizations of the excitation and detection, we choose the circularly co-polarized one, with both pulses having the \( \sigma_+ \) polarizations and the detection being performed at the same polarization. The system is placed in a magnetic field tilted by the angle \( \vartheta \) to the normal, which defines the trion spin quantization. The hole spin is quantized along the axis at an angle \( \varphi \) from the normal to the sample, defined by the components of the hole g-tensor equal to 0.26. We will assume that the electron spin coherence is dominated by the latter. The trion Larmor frequency is 0.16\( \text{ps}^{-1} \) and the hole spin dephasing is slow compared to the trion lifetime so that the electron spin coherence is dominated by the latter.

The simulations presented in this contribution are performed for \( T = 4 \text{ K} \) and for the magnetic field of 7 T tilted at an angle \( \pi/2 - \vartheta = 4^\circ \) from the system plane. For the in-plane and perpendicular components of the hole g-tensor equal to 0.04 and 0.6, respectively, this yields the hole Larmor frequency \( \omega_h = 0.030 \text{ps}^{-1} \) and the hole spin quantization axis oriented at \( \varphi = 44^\circ \) from the normal direction. The trion Larmor frequency is 0.16\( \text{ps}^{-1} \) (corresponding to the electron g-factor of 0.26). We will assume the decoherence times \( T_{\gamma_1} = 1.2 \text{ ns} \) and \( T_1 = 2 \text{ ns} \) and no additional pure dephasing effects (\( \gamma_0 = \kappa_0 = 0 \)).

3 The FWM response. Our theoretical analysis is based on the method developed in Ref. [4]; the system evolution is studied in the density matrix formalism, with the optical and spin-related dephasing included via a Lindblad dissipator in the evolution equation. In the limit of coherent ultrafast optical driving, this approach yields analytical formulas for the single system dynamics, which allows one to perform averaging over an inhomogeneous distribution of various parameters in the ensemble.

For a \( \sigma_+ \) probe, the only element of the density matrix just after the probe pulse that carries the \( \exp(2i\phi_2) \) phase dependence is

\[
\rho_{31}(t) = \rho_{13}(0^-)e^{2i\phi_2} \sin^2\frac{\alpha_2}{2},
\]

where \( \phi_2 \) is the phase of the probe pulse, \( \alpha_2 \) is its area, and \( 0^- \) denotes the time instant just before the arrival of the pulse. The evolution of the system state is then calculated using the Master equation in the Lindblad form, like in Ref. [4]. The \( \sigma_+ \) interband coherence at a time \( t > 0 \) is

\[
\rho_{31}(t) = \sum_{\pm} d_{\pm} e^{i\lambda_{\pm} t} \rho_{13}(0^+) e^{-iEt/\hbar},
\]

where the amplitudes and the exponents are given by

\[
\begin{align*}
d_1 &= \frac{\cos \varphi + 1}{4}, & \lambda_1 &= \frac{2i\omega_h - (2i\omega_h - \beta)}{4}, \\
d_2 &= \frac{\cos \varphi - 1}{4}, & \lambda_2 &= \frac{2i\omega_h + (2i\omega_h - \beta)}{4}, \\
d_3 &= \frac{\cos \varphi + 1}{4}, & \lambda_3 &= \frac{-2i\omega_h - (2i\omega_h - \beta)}{4}, \\
d_4 &= \frac{\cos \varphi - 1}{4}, & \lambda_4 &= \frac{-2i\omega_1 + (2i\omega_h - \beta)}{4}.
\end{align*}
\]

Here \( E \) is the interband transition energy (at zero field) and the dephasing constants are

\[
\Gamma = 4\gamma_0 + 2\gamma_1 + \kappa_0 + \kappa_+ + \kappa_-
\]
and
\[ \beta = (\kappa_- - \kappa_+) \left[ (\cos \varphi - 1) \sin^2 \varphi + 1 \right]. \]

Now, we need to find \( \rho_{13}(0^-) \). For a \( \sigma_+ \)-polarized pump, just after the pump pulse (at \( t = -\tau^+ \)), the only non-zero element linear in the pump amplitude is \( \rho_{31} \). By solving the Master equation one finds at \( t = 0^- \)
\[ \rho_{13}(0^-) = \sum_i c_i e^{i \lambda_i^1 \tau} e^{i E \tau / h}, \]
(1)
where
\[ c_i = \rho_{13}(-\tau^+) d_i. \]

Thus, at the time \( t \) one finds
\[ \rho_{31}(t) = \frac{1}{2} \sin \alpha_1 \sin^2 \frac{\alpha_2}{2} e^{i (2 \phi_2 - \phi_1)} e^{-i E (t - \tau) / h} \sum_{ij} d_i d_j e^{i \lambda_i^1 t + i \lambda_j^1 \tau}. \]

When averaging over the distribution of interband energies \( E \) in a typical QD ensemble, the exponent produces a narrow echo peak around \( t = \tau \). Since the spin-related evolution (frequencies \( \lambda_i \)) is slow on this time scale, we can put \( t = \tau \) under the summation. Then the magnitude of the time-integrated response is proportional to
\[ \text{FWM} \sim \left| \sum_i d_i e^{i \lambda_i^1 \tau} \right|^2 = \frac{1}{8} \left( 1 + \cos \omega \tau \right) e^{-\Gamma \tau / 2} \left[ (\cos \varphi - 1)^2 e^{-\beta \tau / 2} + (\cos \varphi + 1)^2 e^{\beta \tau / 2} + 2(1 - \cos^2 \varphi) \cos \omega_h \tau \right]. \]
(2)

In order to take into account also the inhomogeneity of the Landé tensors at different trapping centers this result should be averaged over a certain distribution of the Larmor frequencies \( \omega_h \) and \( \omega_\lambda \), which we will assume to be Gaussian and characterized by the variances \( \sigma_{\omega_h}^2, \sigma_{\omega_\lambda}^2 \). We assume here that the distribution of Landé tensors is uncorrelated to the spectral positions of the trion transitions.

In Fig. 1(a), we show the calculated time-integrated FWM signal in the absence of inhomogeneous distribution of the Larmor frequencies. As follows from Eq. (2), oscillations at various frequencies are present in the optical signal, corresponding to combinations of the hole and trion frequencies. A more transparent picture is obtained after performing a Fourier-transform of this signal, as shown in Fig. 1(b). Here one can see the zero-frequency line, as well as lines at the frequencies \( \pm \omega_h, \pm \omega_\lambda \), and \( \pm \omega_\lambda \pm \omega_h \).

In a real system, the optical response is affected by the inhomogeneous distribution of the relevant parameters. In the present case, the Larmor frequencies are usually not identical for each hole-trion system due to variations of the \( g \)-factors in the nanostructure. In Fig. 2 we show the results of a simulation performed for the same parameters as in the previous case but with the additional effect of inhomogeneous broadening of the Larmor frequency, which is assumed to be equal to 20% of their average values. Due to this inhomogeneity effect, the oscillations in the time integrated response are damped and the signal is dominated by the monotonic decay (except for short delay times). Correspondingly, all the non-zero frequency peaks in the Fourier spectrum are strongly broadened. However, the central peak remains unaffected.

4 Discussion and conclusions. An interesting point in the discussion presented above is the presence of a zero-frequency component which produces a central peak in the Fourier transform, composed of two Lorentzian contributions: one with the width \( (\Gamma + \beta) / 2 \) and another one with \( (\Gamma - \beta) / 2 \). Interestingly, as follows from Eq. (1), there are no zero-frequency components in the polarization evolution between the pulses. Therefore, the non-oscillatory part of the FWM response can be interpreted as a result of partial refocusing of the Larmor precession by the probe pulse.

The presence of this zero-frequency component, which is insensitive to the inhomogeneous distribution of the \( g \)-factor, opens a possibility of extracting useful information on the spin-related system parameters. If the trion life-
time is sufficiently long compared to the hole spin decoherence times ($\beta$ not too small compared to $\Gamma$) then the two Lorentzian components of the central line, with the widths $\Gamma \pm \beta$ can be separated by fitting, from which the values of $\Gamma$ and $\beta$ can be deduced. The value of $\beta$, along with the detailed balance relation, allows one to extract the rates $\kappa_{\pm}$, hence the longitudinal decoherence time. On the other hand, $\Gamma$ involves both the spin-related rates $\kappa_{\pm,0}$ and the optical lifetime and dephasing rates $\gamma_{0,1}$. The latter, however, can often be deduced independently for a given system, which can allow one to find also the value of $\kappa_0$ and to calculate the intrinsic transverse spin decoherence time $T_2$.

This should be compared with the information available from a TRKR experiment [3]. There, the hole-related response originates from the occupations of the hole Zeeman sublevels after the trion recombination. Therefore, the TRKR experiment allows one to extract the hole spin-related information even if the exciton lifetime and coherence time are very short, as was indeed the case in the experiments [1]. However, as the information on the transverse spin dephasing in that experiment is deduced from the spin precession signal, it is convoluted with the inhomogeneous effect which may even completely dominate the decay of coherent precession signal. Therefore, in a TRKR experiment essentially only the inhomogeneous transverse decoherence time $T^*_2$ is available.

Thus, we have shown that a FWM experiment performed on an ensemble of quantum dots or other trapping centers doped with excess holes can provide useful information on the properties of hole spin precession and decoherence. Although this information is only available under favorable conditions related to the various decoherence rates in the system (long exciton coherence) it is insensitive to inhomogeneous effects, in particular to a variation of $g$-factors. Therefore, we conclude that the FWM experiments may be a useful tool to extract spin-related information which is complementary to that available from the TRKR study.

Acknowledgments. This work was supported in part by the Alexander von Humboldt Foundation within a Research Group Linkage Grant. P.M. acknowledges support from the TEAM programme of the Foundation for Polish Science, co-financed from the European Regional Development Fund.

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