Theory of Spin Beatings in the Faraday Rotation of Semiconductors

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We show from first principles that the coherent excitation of the Zeeman-split exciton spin states, leading to quantum beatings, is entirely due to correlation between two excitons of opposite spin, and is, thus, beyond the phase-space filling and the mean-field effects between excitons. This explains the ultrafast terahertz oscillations in the delay time between the pump and probe pulses in the presence of a magnetic field observed by the recent time-integrated measurements of the optically induced Faraday rotation in diluted magnetic semiconductor quantum wells.

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Ultrafast nonlinear spectroscopy experiments have led to exciting new studies of optical coherence in semiconductors. From the observed coherent polarization mixing, it becomes possible to study the exciton-exciton interaction [1–3]. In this paper, we study a different problem of coherence of the spin-split states in a dilute magnetic semiconductor [4] and the role of exciton-exciton correlation in polarization mixing with the twin purposes of gaining a deeper understanding of coherence in semiconductors and studying the carrier spin dynamics in a magnetic semiconductor. We confine our attention to the low magnetic field case of Zeeman splitting as opposed to the high-field Landau levels [5,6].

Recent measurements of spin dynamics using femtosecond resolved magneto-optical techniques reveal coherent ultrafast oscillations in the Faraday rotation of the transmitted polarization [4]. These oscillations decay on a time scale of the phase-coherence lifetime long before spin scattering of the electrons becomes important. The measurements were carried out with magnetic barrier coupled [$Zn_{1-x}Cd(Mn)_xSe$] double quantum wells (MCDQW), which leads to a large Zeeman splitting of the degenerate spin states due to an enhancement of the effective g factor in the coupling with magnetic Mn^{++} ions in the barrier [7]. The oscillation frequency of the signal in the pump and probe (PP) geometry with respect to the pulse delay corresponds to the splitting of the symmetrical heavy-hole exciton transitions in the double well and can be *tuned* with the strength of the magnetic field.

We will show that the oscillatory Faraday rotation is not mere quantum beatings of the two opposite-spin excitons but results from a coherent excitation of the two excitons through their correlation. Thus this phenomenon does not occur in the mean-field approximation. In the larger context of biexciton effects on coherence transfer through mixed polarizations, this theoretical explanation of a coherent oscillation demonstrates the importance of the exciton-exciton correlation without necessarily the binding of the two excitons. There is a sharp distinction between the quantum beats in Faraday rotation under study here and the luminescence interference of the two exciton spin states [8]. The former involves the final state of a single exciton carrying the spin coherence, whereas the latter involves radiations from two spin states.

We consider a semiconductor quantum-well model with only heavy-hole excitons. We neglect the light-hole excitons. The *sp-d* exchange interaction of the electron spin with the magnetic ions in the barrier is treated in a mean-field approximation [7]. The Zeeman splitting is then tunable due to the magnetic field controlled magnetization. In the Faraday geometry, the propagation of the pump and probe pulses is along the growth axis which coincides with the applied external magnetic field. The rotation of the polarization plane of the linearly polarized probe pulse across the sample thickness is calculated in terms of the change of the optical constant to second order in the pump pulse with circular or plane polarization. The third-order polarizability needed (to first order in the probe field and second order in the pump field) is calculated using a recently developed theory [9]. The equation of motion for the contribution of the exciton at level n and spin σ to the third-order polarizability has the natural frequency of the exciton, $\omega_{n,\sigma}$, and a dephasing rate Γ. It is driven by (1) the phase-space filling term [10,11], arising from the Pauli exclusion of the constituents of two excitons, (2) the mean-field (Lorentz field and self-energy) term [10] due to other excitons, and (3) the exciton-exciton correlation via a time-dependent (force-force) correlation function [9] $F^{n'\sigma';n'',\sigma''}_{\tilde{n},\tilde{\sigma};n,\sigma}(t)$.

The contributions of the three source terms are calculated for a two-dimensional Hubbard model with a conduction band and a heavy-hole band and long-range Coulomb interaction between the electrons in both bands. In the low field, Zeeman limit, the magnetic field comes in only through the large g factor. In the exciton-exciton correlation function, the interactions among the four particles (two electrons and two holes) are treated on an equal basis and the ensuing many-body effects are computed with controlled numerical precision for a finite system size of the square lattice with 36–100 sites by enumerating all possible many-body excited states. The numbers of sites used exhibit already the convergent *short-time* behavior of the infinite system. We have also neglected the contribution of higher energy excitonic bound and unbound states

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as a source term for the pump-induced polarizability. No further approximation, such as a decoupling scheme for the correlation function, is necessary.

Before going into details, we summarize here the key results of our calculation. The Faraday rotation of the linearly polarized probe pulse decays smoothly (with no oscillatory behavior) as a function of the delay time from the pump pulse which is circularly polarized. The optical transitions from the total angular momentum $\pm 3/2$ states in the heavy-hole band to the $\pm 1/2$ spin states in the conduction band are excited, respectively, by the $\sigma = \mp 1$ circularly polarized light. Thus a circularly polarized pump pulse excites only excitons of only one spin direction, whose exponential decay in pump-probe delay time creates a smooth decrease of the change in optical constant responsible for the Faraday rotation. A linearly polarized pump pulse can be resolved into a left-handed and a right-handed circularly polarized pulse. In the two source terms of the third-order polarizability due to phase-space filling and to the mean-field effect, the net contribution to the Faraday rotation of the probe pulse is essentially zero, because each circularly polarized component of the pump pulse excites only excitons of the corresponding spin, producing Faraday rotation of opposite sign to that due to the other component. The third source term, the exciton-exciton correlation term, yields the only nonzero Faraday rotation, which, in addition, oscillates with the frequency of the Zeeman splitting of the exciton level as a function of the delay time. This beating arises from the correlation of the two excitons of opposite spins coherently excited by the two components of the pump pulse.

Let the Faraday rotation of a *linearly* polarized probe pulse plane wave propagating through the sample thickness L be denoted by the angle of rotation Θ_F of the polarization plane. In the experimental setup [4,12], the transmitted light from the sample is split into two linearly orthogonal polarizations in the probe direction and rotated by an angle $\Theta_0 \sim \pi/4$, and the difference *intensity* is measured in an optical bridge. Without the pump beam, the bridge is balanced by adjusting Θ_0, so that the two linear contributions cancel. Normalizing the output gives the following result for the *Faraday rotation* $\Theta_F(\tau_d)$;

$$\Theta_F(\tau_d) = \frac{L}{2I_0}\,\mathrm{Re}\Big\{\sum_{n,\sigma}\sigma\kappa_{n,\sigma} \times \int \langle B_{n,\sigma,\mathbf{k}_1}\rangle_t\,[\Omega_{\sigma,\mathbf{k}_1}(t,0)]^*\,dt\Big\}, \quad (1)$$

in terms of only the *pump-induced* corrections for the exciton polarizability $\langle B_{n,\sigma,\mathbf{k}_1}\rangle_t$ at time t, where B is the exciton annihilation operator at level n and spin σ. The propagation of the light pulse in the $\mathbf{k}_1$ direction for the σ-circularly polarized plane wave is represented by the Rabi frequency $\Omega_{\sigma,\mathbf{k}_1} = 2(\mu_\sigma E_{\sigma,\mathbf{k}_1})$, where μ_σ is the interband electric dipole matrix element. Equation (1) includes an average of the linear phase over the wave packet with intensity $I_0 \equiv \int |\Omega_{\sigma,\mathbf{k}_1}(t,0)|^2\,dt$ and depends on the delay $\tau_d \equiv t_{\mathrm{probe}} - t_{\mathrm{pump}}$ between the pump and the probe pulse maxima:

$$\kappa_{n,\sigma} = \frac{4\pi}{V}\frac{\omega_p}{n_{L,\sigma}c}|\mu_\sigma|^2\,\alpha^*_{n,\sigma}, \quad (2)$$

with $n_{L,\sigma}$ being the *linear* index of refraction of the semiconductor, the central frequency of the light pulse at ω_p, $\alpha_{n,\sigma}$ the exciton wave function at zero relative distance, and V the system volume. In the quasiresonant approximation [13], i.e., for a central frequency of the external laser field ω_p near the fundamental $1s$-exciton resonances with energies $\omega_{1s,\sigma=\pm 1}$ and spectrally limited pulses, to a good approximation, only first-order contributions arising from these resonances need to be taken into account as source terms for the third-order polarizability. In the rotating frame, the magnetic field induced Zeeman splitting is given by $\omega_\sigma = -\sigma\omega_B/2$ ($\omega_B > 0$). From here on, we drop the exciton indices.

For a simple, analytic discussion, we assume short symmetric, nonoverlapping pulses with width $\tau_p \ll 1/\Gamma$ and a delay $\tau_d > \tau_p$. The nonlinear polarizability in the probe direction can then be written with $\tilde{\Omega}_{\sigma,\mathbf{k}_2} \equiv \int_{-\infty}^{\infty}\Omega_{\sigma,\mathbf{k}_2}\,dt$ as follows:

$$\tilde{B}_{\sigma,\mathbf{k}_1}(t,\tau_d) \equiv \sum_n \alpha^*_{n,\sigma}\langle B_{n,\sigma,\mathbf{k}_1}(\tau_d)\rangle_t = \sum_{\sigma',\sigma_1,\sigma_2}\Theta(\tau_d)e^{i(\omega_{\sigma_2}-\omega_{\sigma_1})\tau_d}e^{-2\Gamma\tau_d}\tilde{\Omega}_{\sigma_1,\mathbf{k}_2}(\tilde{\Omega}_{\sigma_2,\mathbf{k}_2})^*\int_{-\infty}^{t}\chi^{\sigma',\sigma_1}_{\sigma,\sigma_2}(t,t')\Omega_{\sigma',\mathbf{k}_1}(t')\,dt'. \quad (3)$$

The polarization-dependent third-order susceptibility $\chi^{\sigma',\sigma_1}_{\sigma,\sigma_2}(t,t')$ includes the three source terms discussed above.

We have calculated the specific rotatory power $\Theta_F(\tau_d)/L$ for the various pump-induced source terms separately. The pump field is chosen to be either $\pm$ circularly polarized or linearly polarized in the x (or y) direction. The probe field is fixed with linear x polarization. The central frequency of the light pulses is centered at the zero-field degenerate exciton resonance. The contributions from the three source terms are as follows.

(i) Phase-space filling (PSF).—The PSF contribution depends only on the density of the induced exciton population. For a sufficiently short test pulse with $t_p\Gamma \ll 1$ we find

$$\chi^{\sigma',\sigma_1}_{\sigma,\sigma_2}(t,t') = -\frac{|\alpha_{1s,\sigma}|^2}{4}\sum_{n,n'}\alpha^*_{n,\sigma}\alpha_{n',\sigma}C^{n',\sigma;1s,\sigma}_{n,\sigma;1s,\sigma} \times \chi_{L;n,\sigma}(t-t')\delta_{\sigma,\sigma'}\delta_{\sigma,\sigma_1}\delta_{\sigma,\sigma_2}, \quad (4)$$

with the coefficient [13] $C^{n',\sigma',n'',\sigma''}_{\tilde{n},\tilde{\sigma};n,\sigma}$ arising from the Pauli exclusion of the constituents of two excitons scattering from states $(\tilde{n},\tilde{\sigma};n,\sigma)$ into states $(n',\sigma',n'',\sigma'')$. Here the linear susceptibility is given by $\chi_{L;n,\sigma}(\tau) = (i/2)\Theta(\tau)e^{-i(\omega_{n,\sigma}-i\Gamma)\tau}$. For a spectrally limited test pulse at the $1s$-exciton resonances the summation over the exciton contributions can be limited to the relevant terms $n = 1s$. The result is the well-known bleaching of the exciton resonance due to Pauli blocking. As $\mathrm{Re}\chi_{L;1s,\sigma}(\tau) \equiv 0$ for $\omega_B = 0$ and symmetrical pulses, no zero-field Faraday rotation is observed. Because of the symmetry of the linear susceptibility $\mathrm{Re}\chi_{L;1s,\sigma}(\tau) \sim \sigma$, the total Faraday signal is $\sim \sum_\sigma \Omega_{\sigma,\mathbf{k}_2}(\Omega_{\sigma,\mathbf{k}_2})^*$, which is *independent* of the polarization configuration of the pump field.

(ii) Mean field exciton-exciton interaction (MF).—The mean-field term [9] is given by

$$\chi^{\sigma',\sigma_1}_{\sigma,\sigma_2}(t,t') = -\frac{|\alpha_{1s,\sigma}|^2}{8}\sum_{n,n'}\alpha^*_{n,\sigma}\alpha_{n',\sigma}\beta^{n',\sigma;1s,\sigma}_{n,\sigma;1s,\sigma} \times \delta_{\sigma,\sigma'}\delta_{\sigma,\sigma_1}\delta_{\sigma,\sigma_2}e^{-i(\omega_{n,\sigma}-i\Gamma)t}e^{i(\omega_{n',\sigma}-i\Gamma)t'} \times \int_{t'}^{t} e^{-2\Gamma t''}\,dt'', \quad (5)$$

with the coefficient [13] $\beta^{n',\sigma';n'',\sigma''}_{\tilde{n},\tilde{\sigma};n,\sigma}$ being the mean-field term due to other excitons. Again, focusing on the $1s$ response only, for a circularly polarized pump field the mean-field contribution results in a symmetrical signal, which is finite even at zero applied magnetic field (cf. Fig. 1). For linearly polarized excitation, the net signal is zero due to cancellation in the total Faraday signal $\sim \sum_\sigma \sigma\tilde{\Omega}_{\sigma,\mathbf{k}_2}(\tilde{\Omega}_{\sigma,\mathbf{k}_2})^*$. Therefore a mean-field description fails to describe the experimental results [4] in the coherent regime of the Faraday rotation. In the low spin-split limit, we expect a ratio

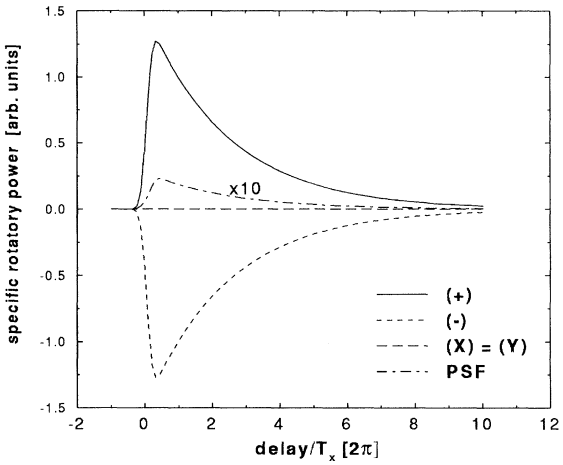

FIG. 1. Specific rotatory power vs delay between pump and probe pulse for different pump-pulse polarizations and $\omega_B = 0.2\omega_x$. This figure contains the MF contribution and the PSF contribution only.

of the above two mean-field contributions to be approximately $|\Theta^{(\mathrm{PSF})}_F/\Theta^{(\mathrm{MF})}_F| \approx C_{\omega_B}/4\beta \ll 1$, with the microscopic parameters $\beta = (|\alpha_{1s,\sigma}|^2/2)\beta^{1s,\sigma;1s,\sigma}_{1s,\sigma;1s,\sigma}$ and $C := \sum_{n'}\alpha^*_{1s,\sigma}\alpha_{n',\sigma}C^{n',\sigma;1s,\sigma}_{1s,\sigma;1s,\sigma}$. The relative weight can be seen in Fig. 1, where we have plotted the rotatory power with respect to the delay between the pulses.

(iii) Exciton-exciton correlation (XX): parallel spins.—The exact parallel-spin exciton-exciton contribution requires the knowledge of the corresponding two-exciton correlation function [9]. However, for parallel-spin correlation, no *polarization mixing* can be observed. We can derive an analytical expression for the nonlinear susceptibility for arbitrary polarization couplings

$$\chi^{\sigma',\sigma_1}_{\sigma,\sigma_2}(t,t') = -\frac{i}{8}\alpha_{1s,\sigma_1}\alpha^*_{1s,\sigma_2}\sum_{n,n'}\alpha^*_{n,\sigma}\alpha_{n',\sigma}e^{-\Gamma(t-t')} \times e^{-i(\omega_{n,\sigma}t-\omega_{n',\sigma'}t')}\int_{t'}^{t}\int_{t'}^{t''} e^{-2\Gamma t''}F^{n',\sigma';1s,\sigma_1}_{n,\sigma;1s,\sigma_2}(t''-\tilde{t})_{B=0}\,d\tilde{t}\,dt''. \quad (6)$$

For the $1s$ contribution, the two-exciton correlation function $F^{\sigma',\sigma_1}_{\sigma,\sigma_2}(\tau)_{B=0}$ is, in general, complex [9], and the fast time dependence of the correlation function requires a direct numerical calculation of $F(\tau)$ on the time scale of the pulse duration t_p. In our model calculation, the parallel spin contributions are displayed in Fig. 2. The effect of parallel-spin correlation is basically a correction to the mean-field contribution with symmetry relations easily derived from Eq. (6) and will be discussed elsewhere.

(iv) Exciton-exciton correlation (XX): opposite spins.—The most interesting correlation contribution arises if the interaction of electrons with opposite spins is taken into account. From Eqs. (3) and (6) we have two different *spin-dependent polarization-mixing* contributions to the Faraday rotation. A contribution to the susceptibility Eq. (6) results from the polarization-mixing interaction depending on the *density* of the opposite spin type exciton population ($\sigma = \sigma'$) and gives a result similar to the parallel-spin XX terms; i.e., a partial cancellation occurs for linearly polarized excitation. However, the contribution with ($\sigma = -\sigma'$) leads to a coupling of excitons with opposite total spins, and the Faraday rotation signal of Eq. (1) shown in Fig. 3 is uniquely modulated by the splitting frequency ω_B. The modulations are absent for circularly polarized pump fields. A π phase shift for orthogonal linearly polarized fields is observed as $\Omega_{\sigma,\mathbf{k}_2}\Omega^*_{-\sigma,\mathbf{k}_2}$ changes sign. The striking symmetry of the Faraday rotation signal $\sim \sin(\omega_B\tau_d)$ is due a partial cancellation of $\sim \cos(\omega_B\tau_d)$ contributions in Eq. (6). This can be seen by neglecting the residual field dependence of the susceptibility in Eq. (6) in the limit $\omega_B t_p \ll 1$, i.e., for beating periods much larger than the pulse duration. The cor-

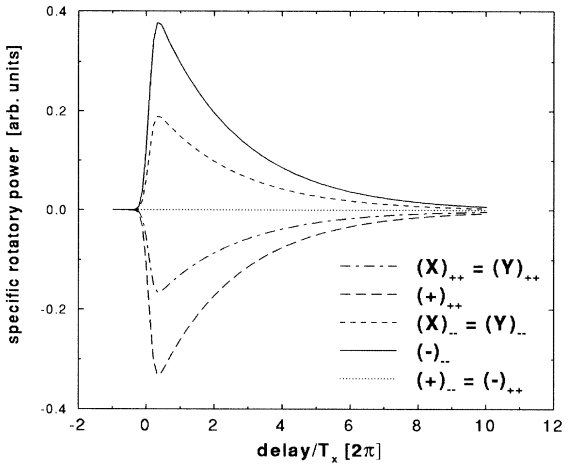

FIG. 2. Specific rotatory power for the pulse parameters in Fig. 1 for parallel-spin exciton-exciton correlation (XX) only. The lack of mirror symmetry is due to the spin dependence of the correlation effects.

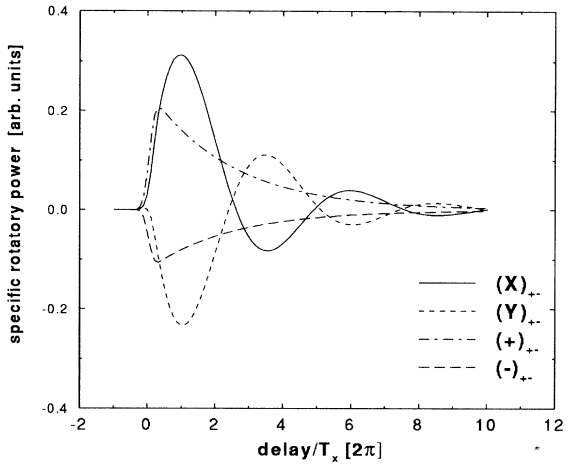

FIG. 3. Specific rotatory power for the pulse parameters in Fig. 1 due to polarization-mixing exciton-exciton correlation only. Spin beatings are evident for linearly polarized pump fields with the frequency of the Zeeman splitting of the exciton transitions.

relation, which, in the short-time regime, arises mainly from unbound two-exciton states, is important for the relative strength of the signals. Possible *bound biexciton* states present in a quantum well have *no significant effect* on the short-time domain of the convolution in Eq. (1). This shows that some important aspects of a microscopic description of exciton-exciton correlation [9–11,14,15] cannot be reproduced by a phenomenological few-level model [3].

Here is a physical picture of the processes which lead to the observed oscillations in Faraday rotation. The effective magnetization in the barrier gives rise to a large Zeeman splitting of the two lowest energy excitonic states with opposite spins. Only the linearly polarized (as opposed to the purely circularly polarized) pump field can create simultaneously both these two excitons as well as inducing the spatially homogeneous *spin coherence*, $\sim \Omega_{\sigma,\mathbf{k}_2}\Omega^*_{-\sigma,\mathbf{k}_2}$, which oscillates with the Zeeman frequency ω_B. Conservation of *total angular momentum* demands that the probe pulse of one circular polarization will produce in this spin coherence an exciton of the opposite spin. Therefore the Coulomb correlation between the two opposite spin excitons is essential to the production of the photon of polarization opposite to that of the probe. This photon carries the temporal oscillations of the spin coherence as a phase information which is detected by the Faraday rotation.

In conclusion, we have presented a first-principles analysis of the pump-induced Faraday rotation in semiconductor quantum wells using a previously developed calculational scheme [9]. We have demonstrated that a recently observed *spin-beating* phenomena for linearly polarized pump fields can be explained by coherent polarization mixing in semiconductors, in which the Coulomb interaction between excitons of opposite spins is essential. For circularly polarized pump-field excitation, the Faraday rotation signal is dominated by the mean-field contribution of the excitons.

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