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Magnetic-field-induced resonance in four-wave mixing in GaAs

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The frequency-domain four-wave-mixing response of excitons in GaAs in a magnetic field is theoretically and experimentally studied. For linearly cross-polarized excitation beams, the application of a magnetic field induces a resonance that leads to the observation of a narrow dip in the primary resonance. The primary resonance has a width given by the exciton spin-flip rate while the narrow dip has a width determined by the recombination rate. The results reflect the dynamics of two observables defined in terms of the appropriate Fourier components of the net population and the net population transfer between the two oppositely spin-oriented excitonic states, similar to the normal decay modes of the system. The relative amplitude of these observables is determined by the relative polarization of the incident fields and the strength of the applied magnetic field.

The resonant coherent nonlinear optical response observed, for example, to the lowest order in cw four-wave mixing (FWM), depends sensitively on the nature of the relaxation of the system. The relaxation processes determine the characteristic parameters such as the widths of the various resonances that contribute to the nonlinear response, and can also lead to more profound effects such as the well-known pressure-induced resonance,[1] predicted by Bloembergen, Lotem, and Lynch.[2] Using an amplitude rather than density-matrix description,[3] it was seen that these extra resonances result essentially from the collision-induced removal of destructive interference between independent quantum-mechanical amplitudes. Resonant features were also observed in simple closed two-level systems coupling to a reservoir through spontaneous emission[4] or state-specific collisions[5] which lead to an opening of this system, where "open" and "closed" are defined in terms of whether or not the population is effectively conserved.

In this paper, we report a resonant feature observed on the excitonic nonlinear response in GaAs induced by the presence of a magnetic field. In this system, excitons of opposite spin orientations, created independently by circularly polarized (σ^- or σ^+) light, are coupled by processes leading to excitonic spin flips. Using a density-matrix formalism, we demonstrate that the relaxation of spin-polarized exciton populations (either σ^+ or σ^-) can be described as a superposition of two independent observables (similar to the normal decay modes of the system) characterized by distinct relaxation rates: the total population decays with the recombination rate, and the net population transfer between the two oppositely spin-oriented states decays with a rate dominated by spin flip. In the absence of a magnetic field, these two normal decay modes can be measured independently using FWM by selecting the polarization of the excitation beams: the relaxation of the total population is given by the line width of FWM response when the excitation beams are linearly copolarized and the relaxation of the net population transfer is measured when the excitation beams are linearly cross polarized. With application of a magnetic field, the FWM line shape becomes a combination of the two normal decay modes, leading to a dip or spike in the FWM response for linearly cross-polarized fields.

The data were obtained at 4 K using the light-hole (lh) exciton in a GaAs thin film of thickness 200 nm grown at 630 °C by molecular-beam epitaxy on a semi-insulating (100) GaAs substrate. At zero magnetic field, the lh-hh degeneracy is lifted by axial stress. The lh-hh splitting is of order 3 meV with the lh exciton as the lowest energy state. At 6 T (field normal to the sample surface), the Zeeman splitting (sublinear in the applied field) is comparable to the full linear absorption linewidth (0.3 meV). More detailed sample characterization was given in an earlier work.[6]

The nonlinear optical measurement is based on the backward FWM configuration using two frequency stabilized cw dye lasers (Fig. 1). Two nearly degenerate linearly polarized fields, $\mathbf{E}_i(\mathbf{k}_i,\omega_i)$ ($i=1,2$ and $|\omega_2-\omega_1|=|\delta|\ll\omega_i$; spectral resolution is ~ 5 neV), intersect in the sample, producing a traveling-wave modulation of absorption and dispersion proportional to $\mathbf{E}_1\cdot\mathbf{E}_2^*$. The resulting grating is probed by a third beam, $\mathbf{E}_3(-\mathbf{k}_1,\omega_3)$, giving rise to a coherent signal propagating in the direction $-\mathbf{k}_2$. (The contribution due to $\mathbf{E}_3\cdot\mathbf{E}_2^*$ is negligible because exciton diffusion washes out the small grating spacing.) The measured FWM signal is proportional to the modulus squared of the complex third-order susceptibility.

Decay dynamics are determined from the FWM line shape as a function of δ.[7] This FWMδ response is measured as a function of incident field polarization, the excitation energy, and magnetic field. When $\mathbf{E}_1\|\mathbf{E}_2$ (FWM$_\|$), the profile is independent of the polarization of the back beam $\mathbf{E}_3$. For all applied magnetic fields and excitation energies, the

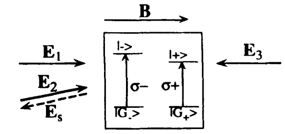

FIG. 1. Backward FWM geometry and energy level diagram for semiconductor GaAs.

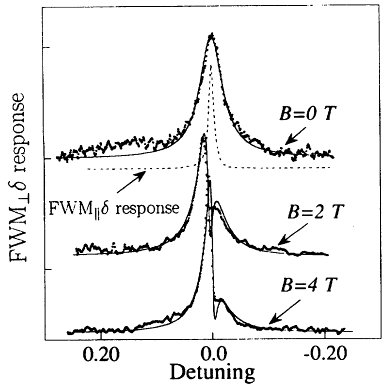

FIG. 2. FWM$_\perp\delta$ response measured at $B=0,2,4$ T. FWM$_\parallel\delta$ response (dotted curve) is also shown for reference. Solid curves: fit based on the theory discussed in the text.

FWM$_\parallel\delta$ response is a single Lorentzian (Fig. 2) with a linewidth of $(0.85\ \mathrm{ns})^{-1}$ identified as the exciton recombination rate. This rate, as well as the dephasing rate [given by the linewidth of the linear absorption, $\gamma_\pm \approx (4\ \mathrm{ps})^{-1}$] for σ^+ and σ^- excitons, respectively, remained nearly constant for all values of magnetic field.

Unlike the above FWM$_\parallel\delta$ response, when $\mathbf{E}_1 \perp \mathbf{E}_2$ (FWM$_\perp\delta$) the profile depends sensitively on the intensity of the applied magnetic field and the excitation energy. The FWM$_\perp\delta$ response measured at zero magnetic field is a single Lorentzian with a width corresponding to a lifetime ~ 125 ps. The exciton recombination component (the narrower Lorentzian observed in the FWM$_\parallel\delta$ response) is absent. In the presence of a magnetic field ($\omega_{1,2}$ tuned to the zero-field resonance), more than one resonance is seen (lower two curves in Fig. 2); i.e., *the presence of a magnetic field induces a new resonance* observed as a dip. The data shown were obtained at $B=0$, 2, and 4 T using σ^+ circularly polarized $\mathbf{E}_3$. For $\mathbf{E}_3$ polarized σ^-, the line shape is similar but reflected about $\delta=0$. When the exciting beams are tuned off resonance, the FWM$_\perp\delta$ profiles become similar to the FWM$_\parallel\delta$ profiles.

To understand the above behavior, we examine the nonlinear optical response of excitons in GaAs in a magnetic field. When the lh-hh degeneracy in GaAs is removed such as through the application of axial stress or confinement, the appropriate exciton energy diagram consists of two separate two-level systems (Fig. 1). For each subsystem, the ground state $|G_\pm\rangle$ corresponds to the state of no excitons, and the state $|\pm\rangle$ corresponds to an excitonic state prepared with either σ^+ or σ^- polarized light. (Although the biexcitonic states have been observed in this system,[8] the contributions due to the ground-state–biexciton coherence as well as the so-called dark states are not important for the present qualitative discussion.[9]) In the absence of spin flips, creation of population for a given spin state (say $|+\rangle$) can be generated directly only by the applied optical fields with the appropriate polarization (σ^+). However, in the presence of spin relaxation, that same state may be excited by either direct creation or excitation transfer from the state of opposite spin (state $|-\rangle$). The FWM response originates from both direct excitation and excitation transfer, characterized by different decay rates, leading to complex spectral structure in general.

The nonlinear optical effects of excitons in semiconductors can be described by the effective semiconductor Bloch equations,[10] which are formally identical to the density matrix equations. For the system under study, the equations of motion for the exciton population and optical polarization are (in the rotating-wave approximation):

$$\partial N_\pm/\partial t = -\gamma_{sp} N_\pm - \Gamma_\pm N_\pm + \Gamma_\mp N_\mp + i[(\boldsymbol{\mu}\cdot\mathbf{E}/2\hbar)p - \mathrm{c.c.}]_{(\pm)}, \tag{1}$$

$$\partial p_\pm/\partial t = (i\omega_\pm - \gamma_\pm)p_\pm - i(\boldsymbol{\mu}^*\cdot\mathbf{E}^*/2\hbar)[N_0/2 - 2N_\pm]. \tag{2}$$

The radiation field is of the form $\mathbf{E}(t) = \frac{1}{2}\Sigma \mathbf{E}_i \exp[i(\mathbf{k}_i\cdot\mathbf{x}_i - \omega_i t)] + \mathrm{c.c.}$ All other notations used in Eqs. (1) and (2) have the conventional meaning with the subscript $+$ ($-$) indicating σ^+ (σ^-) spin orientation of the excitons. The recombination rate γ_{sp} is assumed to be independent of the spin orientation of excitons.

For semiconductor systems, the resonant frequencies and decay rates are generally excitation density dependent due to Coulomb correlation.[11] As will be shown, however, the origin of the induced resonance is not due to the inclusion of the excitation dependence of decay rates and resonant frequencies. Hence, we first solve for the third-order nonlinear polarization by ignoring this effect and then expand the concerned parameters in a power series of excitation density. Using perturbation theory to second order in the electromagnetic fields, the exciton distributions are given by

$$N_\pm = \sum_{\alpha,\beta} \frac{N_0 e^{-i(\omega_\beta-\omega_\alpha)t}}{2(2\hbar)^2[\gamma_{sp} - i(\omega_\beta-\omega_\alpha)]} \left\{ \left[1 - \frac{\Gamma_\pm}{\gamma_{sp}+\Gamma_+ + \Gamma_- - i(\omega_\beta-\omega_\alpha)} \right] \frac{(\boldsymbol{\mu}\cdot\mathbf{E}_\beta \boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(\pm)}}{\gamma_\pm + i(\omega_\alpha-\omega_\pm)} \right.$$
$$\left. + \frac{\Gamma_\mp}{\gamma_{sp}+\Gamma_+ + \Gamma_- - i(\omega_\beta-\omega_\alpha)} \frac{(\boldsymbol{\mu}\cdot\mathbf{E}_\beta \boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(\mp)}}{\gamma_\mp + i(\omega_\alpha-\omega_\mp)} \right\} + \mathrm{c.c.}, \tag{3}$$

where the indices α and β run across all possible combinations of incident fields. Each combination of $\{\alpha, \beta\}$ represents a possible interaction sequence which generates N_- excitons, with spin orientation σ^-, and N_+ excitons with spin orientation σ^+.

This equation, however, may be reformulated in terms of variables, $N_s(\mathbf{E}_\beta,\mathbf{E}_\alpha^*)$ and $N_d(\mathbf{E}_\beta,\mathbf{E}_\alpha^*)$, which represent the sum of the appropriate Fourier components of two oppositely spin-oriented exciton population gratings for N_s and the grating produced by the net population transfer between the two states $|\pm\rangle$ for N_d:

$$N_\pm=\sum_{\alpha,\beta} e^{-i(\omega_\beta-\omega_\alpha)t}\left\{\frac{\Gamma_\mp}{\Gamma_++\Gamma_-}\frac{\gamma_{sp}N_s(\mathbf{E}_\beta,\mathbf{E}_\alpha^*)}{[\gamma_{sp}-i(\omega_\beta-\omega_\alpha)]}\pm\frac{\gamma_{sp}N_d(\mathbf{E}_\beta,\mathbf{E}_\alpha^*)}{\gamma_{sp}+\Gamma_++\Gamma_--i(\omega_\beta-\omega_\alpha)}\right\}+\text{c.c.} \tag{4}$$

with

$$N_s(\mathbf{E}_\beta,\mathbf{E}_\alpha^*)=\frac{N_0}{2(2\hbar)^2\gamma_{sp}}\left[\frac{(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(+)}}{\gamma_++i(\omega_\alpha-\omega_+)}+\frac{(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(-)}}{\gamma_-+i(\omega_\alpha-\omega_-)}\right] \tag{5}$$

and

$$N_d(\mathbf{E}_\beta,\mathbf{E}_\alpha^*)=\frac{N_0}{2(2\hbar)^2(\Gamma_++\Gamma_-)\gamma_{sp}}\left[\frac{\Gamma_+(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(+)}}{\gamma_++i(\omega_\alpha-\omega_+)}-\frac{\Gamma_-(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(-)}}{\gamma_-+i(\omega_\alpha-\omega_-)}\right]. \tag{6}$$

In the above expressions, the Fourier component of the population grating of a given state (say, $|+\rangle$) is written as a linear combination of N_s and N_d. These variables can be viewed as normal decay modes of the problem. The total population grating decays with the recombination rate as seen in the width of the resonance $[\gamma_{sp}-i(\omega_\beta-\omega_\alpha)]^{-1}$ and the net excitation transfer grating decays with a rate primarily determined by the spin-flip rate. The amplitudes of these gratings are critically related to the *balance of excitation transfer* depending on the spin-flip rates, the field polarizations, and the resonant conditions.

For the FWM$_\parallel\delta$ response, the selection rules result in $(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(+)}=(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(-)}$, leading to a complete cancellation of the net excitation transfer in Eq. (6) for $\Gamma_+=\Gamma_-$. The FWM$_\parallel\delta$ response shows therefore a *narrow* resonance (determined by recombination) only. If the excitation transfer between the two states is unequal (e.g., by applying a magnetic field or using circularly polarized $\mathbf{E}_\alpha$ and $\mathbf{E}_\beta$), the effective population for a given state becomes a function of both recombination and spin-flip processes leading to the presence of an additional (broad) resonance. [For semiconductors, however, *spin-independent* effects, such as excitation-induced dephasing (EID), often overwhelm the spin-dependent nonlinearity and this effect will not be observed in the FWM$_\parallel\delta$ response, as discussed below. The spin-flip process is then nearly undetectable using copolarized excitation beams.]

When the exciting beams are linearly cross polarized ($\mathbf{E}_\alpha\perp\mathbf{E}_\beta$), the selection rules of Fig. 1 result in $(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(-)}=-(\boldsymbol{\mu}\cdot\mathbf{E}_\beta\boldsymbol{\mu}^*\cdot\mathbf{E}_\alpha^*)_{(+)}$. Under this condition, the broad resonance $[\gamma_{sp}+\Gamma_\pm+\Gamma_\mp-i(\omega_\beta-\omega_\alpha)]^{-1}$, associated with the decay of the net population transfer grating, is the primary resonance in the FWM$_\perp\delta$ response. The amplitude of the narrow resonance is zero due to the vanishing of the total population grating when the excitonic states are degenerate. Once the symmetry of the system is broken, leading to a net population grating, the narrow resonance appears. The line shape of the response is determined by the magnitude of the total population grating which is a function of the magnetic fields via the parameters $\{\omega_\pm,\gamma_\pm\}$.[12] We use the absolute value of $\beta_\pm(\omega_\alpha)=[\gamma_\pm-i(\omega_\alpha-\omega_\pm)]/[\gamma_\mp-i(\omega_\alpha-\omega_\mp)]$ to characterize the line shape.

To illustrate quantitatively the relative importance of the two resonances, we restrict our discussion to the FWM$_\perp\delta$ experiment described above, and we consider explicitly the case with $\mathbf{E}_3$ polarized σ^+. The nonlinear susceptibility measured at $\omega_s=\omega_1-\omega_2+\omega_3$ in the $-\mathbf{k}_2$ direction is

$$\chi^{(3)}=\frac{-iN_0|\mu|^4}{2(2\hbar)^3[\gamma_+-i(\omega_s-\omega_+)]}\times\left[\frac{1}{\gamma_+-i(\omega_1-\omega_+)}+\frac{1}{\gamma_++i(\omega_2-\omega_+)}\right]\times\left\{\frac{\Gamma_++\Gamma_-\beta_+(\omega_1)\beta_+^*(\omega_2)\frac{2\gamma_-+i\delta}{2\gamma_++i\delta}}{(\Gamma_++\Gamma_-)(\gamma_{sp}+\Gamma_++\Gamma_-+i\delta)}+\frac{\Gamma_-}{(\Gamma_++\Gamma_-)}\frac{\left[1-\beta_+(\omega_1)\beta_+^*(\omega_2)\frac{2\gamma_-+i\delta}{2\gamma_++i\delta}\right]}{(\gamma_{sp}+i\delta)}\right\}. \tag{7}$$

In the limit that $\gamma_\pm\gg\Gamma_\pm$, γ_{sp}, all the factors outside the curly brackets remain virtually constant for the FWM$_\perp\delta$ measurement with the tuning range $\delta_{\max}\ll\gamma_\pm$. Within this approximation, expression (7) becomes a sum of two resonances, a broad resonance dominated by the spin-flip process and a narrow resonance associated with direct recombination.

To demonstrate the influence of the total population grating, we calculate typical line shapes of the FWM$_\perp\delta$ response for various magnitudes of the total population grating using expression (7) and parameters close to those of GaAs. We have assumed $\Gamma_++\Gamma_-=0.04$ (all parameters in units of γ_+) for all applied magnetic fields and the ratio Γ_+/Γ_- to be proportional to the overlap between the σ^+ and σ^- resonance. (This form accounts for the known decrease in spin-flip rates with increasing magnetic field but does not affect the qualitative behavior of the resonance.) The normalized FWM$_\perp\delta$ profiles calculated for $|\beta|-1=-0.08,0,0.02,0.08,0.20$ are shown in Fig. 3 where $\gamma_{sp}=0.005$. The asymmetry in the spectra is mainly the result of the δ dependence of the total population grating. As the sign (phase) of the total population grating changes, the extra resonance changes from a spike to a dip around $\delta=0$. The amplitude of the extra

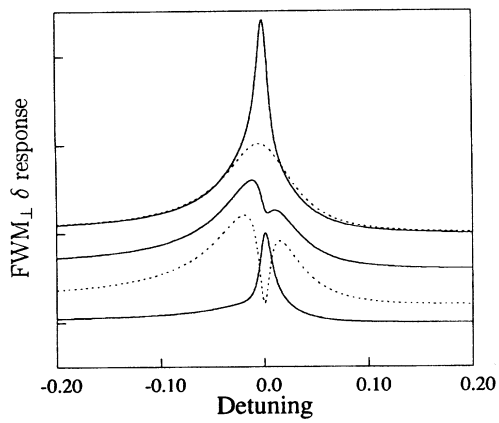

FIG. 3. FWM$_\perp$ δ line shapes calculated for various degrees of population balance. From top to bottom: $|\beta|-1=-0.08,0,0.02,0.08,0.20$.

resonance increases nearly linearly with the magnitude of the total population grating and finally becomes dominant.

For a given population distribution in a semiconductor, the amplitude of the narrow resonance is enhanced by inclusion of *spin-independent* effects such as Coulomb interactions, which result in excitation-induced dephasing and frequency shifts in Eqs. (1) and (2). For GaAs, the change in resonant frequencies is very small because of the cancellation between Coulomb screening and exchange effects,[11(a)] particularly for three-dimensional systems. The excitation-induced modification in dephasing rate, however, is experimentally and theoretically shown to be very important.[11(b)] Since the Coulomb interaction is independent of the spin characteristics of the excitons, the excitation-induced dephasing can be treated phenomenologically by expressing the dephasing rate in Eq. (2) as $\gamma_\pm \rightarrow \gamma_\pm(1+\Gamma_{SI}N_s/N_0)$ and applying the phase-matching condition. Correspondingly, the nonlinear susceptibility $\chi^{(3)}$, initially given by expression (7), has an additional contribution to the second term in the curly brackets, namely, $[\Gamma_-/(\Gamma_+ + \Gamma_-)] \rightarrow [\Gamma_-/(\Gamma_+ + \Gamma_-) + \Gamma_{SI}/2]$. For systems with large Γ_{SI} the broad resonance is easily concealed by the narrow recombination rate resonance for a small total population grating. Hence, in FWM$_\parallel$, the FWM$_\parallel$ δ profile is dominated by the recombination resonance, and the spin-flip process appears only as a small deviation at the wings. With inclusion of the EID effect, the experimental data shown in Fig. 2 are well fit by the model discussed above as shown by the solid curves.

In summary, this work demonstrates that the FWM line shapes of the excitonic nonlinear response in GaAs are determined by two normal decay modes. These modes can be independently excited by an appropriate choice of incident laser field polarizations. However, removal of spin degeneracy leads to a mixing of the decay modes as seen in the FWM spectra.

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[9] The biexcitonic effects consist of two terms in the third-order calculation: the coherent two-photon excitation $(\mu_{BX}E^*)p_{GB}$, and stepwise two-photon absorption $(\mu_{XB}E)N_\pm$. The contribution due to the first term is negligible because of the large dephasing rate of p_{GB}. For the second term, the interaction sequence for generating FWM response is the same as that for excitons except for the third-order perturbation. Hence, within the approximation that the dephasing rate is much larger than the recombination and spin-flip rates, the line-shape discussions made for FWMδ experiment stand for both the excitonic and biexcitonic nonlinear response. The above argument also applies to the dark state effect, which leads to modifications in the third-order calculation only and gives no qualitative changes in the theory.

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