Influence of light holes on the heavy-hole excitonic optical Stark effect

P. Brick, C. Ell, S. Chatterjee, G. Khitrova, and H. M. Gibbs
Optical Sciences Center, University of Arizona, Tucson, Arizona 85721

T. Meier, C. Sieh, and S. W. Koch
Department of Physics and Material Sciences Center, Philipps University, Renthof 5, D-35032 Marburg, Germany
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Pump-probe experiments on high-quality $In_xGa_{1-x}As$ quantum wells are used to investigate the influence of light-hole excitons on the optical Stark effect. For anticircular polarization of pump and probe pulses and a moderate negative detuning of the pump energy, a redshift of the heavy-hole resonance is observed. However, with increasingly negative detuning a transition from this redshift to a blueshift is found. Microscopic calculations that include both heavy holes and light holes reproduce the experimental results. The theoretical analysis shows that the observation of the redshift depends very sensitively on the detuning of the pump pulses and the heavy-hole to light-hole splitting.

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I. INTRODUCTION

The first observations of the excitonic optical Stark effect in semiconductors[1–3] reported the blueshift of an exciton transition in GaAs quantum wells and bulk Cu_2O when pumping below those resonances. The direction of the Stark shift was such that the resonance shifted away from the pump pulse energy. In contrast, a subsequent experiment in a thin film of CuCl displayed for colinearly polarized pulses also a redshift when the pump laser was tuned to a very narrow spectral region slightly below the exciton to biexciton transition.[4] The latter effect was found to be caused by bound two-exciton states.[5] Only recently[6] it has been shown that in $In_xGa_{1-x}As$ quantum wells a redshift can be observed even for detunings well below the exciton and exciton-to-biexciton transitions when pump and probe pulses are anticircularly polarized. This effect, very much unlike the Stark shift in atoms, was analyzed by including in the microscopic calculations not only the Hartree-Fock terms but also higher-order Coulomb correlations. In the case of anticircularly polarized pulses where the Hartree-Fock contributions, i.e., the first-order Coulomb and the Pauli-blocking terms, to the optical response vanish, the higher-order Coulomb term becomes the leading contribution giving rise to a redshift. The theoretical analysis revealed that this redshift is partly due to Coulomb memory effects and partly due to the biexciton resonance.

In this paper we investigate experimentally and theoretically the influence of the light-hole transitions on the optical Stark effect of the heavy-hole exciton. In particular, for anticircular polarization of the pump and probe pulses, it is shown that the direction of the optical Stark shift and the occurrence of the redshift sensitively depend on the pump detuning and the heavy-hole to light-hole splitting. In Sec. II the experimental results are presented. The microscopic theory that includes Coulomb many-body correlations is briefly summarized in Sec. III, where also the detuning dependence of the individual contributions to the signal are evaluated. In Sec. IV numerical results are presented and compared to the experimental data. The most important conclusions are summarized in Sec. V.

II. EXPERIMENTAL RESULTS

Standard pump-probe experiments were performed in transmission geometry at 4 K. An actively mode-locked Ti:sapphire laser provided probe pulses of 100 fs at a repetition rate of 80 MHz. Part of the laser beam was split off and propagated through an external pulse shaper resulting in spectrally narrow pump pulses of 2.7 ps duration and 1 meV width. The pump pulses with a pulse energy of 110 pJ were then focused down to a spot of 100 μm diameter on the sample. In order to avoid averaging over regions of different excitation intensity, only the central part of the pump spot was probed. The pump-probe delay was set to zero.

Measurements were done on a structure containing thirty 8.5 nm thick $In_{0.04}Ga_{0.96}As$ quantum wells spaced by GaAs barriers. It was grown by molecular beam epitaxy on a semi-insulating GaAs substrate. The excitonic linewidth as reported in Ref. 7 is 0.56 meV, the αL peak height is 5.8, and the heavy-hole (hh) to light-hole (lh) splitting amounts to 12 meV. In this study, the pump pulse is tuned energetically well below the 1s-heavy-hole resonance and the exciton to biexciton transition. In particular, also detunings much larger than the ones in Refs. 6 and 8 are used. In the following, the detunings Δ_{hh} and Δ_{lh} are defined by the difference between the central pump energy and the heavy-hole and light-hole transition energies, respectively, minus the central pump energy.

Figure 1 shows the experimental differential absorption around the 1s-heavy-hole exciton for various detunings of the pump pulse below the excitonic resonance. For cocircular polarization, see Fig. 1(a), and a detuning Δ_{hh} of 6.2 meV below the exciton, a blueshift of the resonance is observed. A further increase of the pump detuning leads, as expected, to a reduction of the magnitude of the differential absorption, but the signal remains a blueshift. On the other hand, for anticircular polarization and the smallest Δ_{hh} of 6.2 meV a redshift is found [Fig. 1(b)], in agreement with Ref. 6. However, for this particular polarization geometry an increase of the detuning to 9.7 meV leaves little if any shift. This reduction is much more pronounced than the reduction of the blueshift for cocircular polarization. For the relatively large detuning

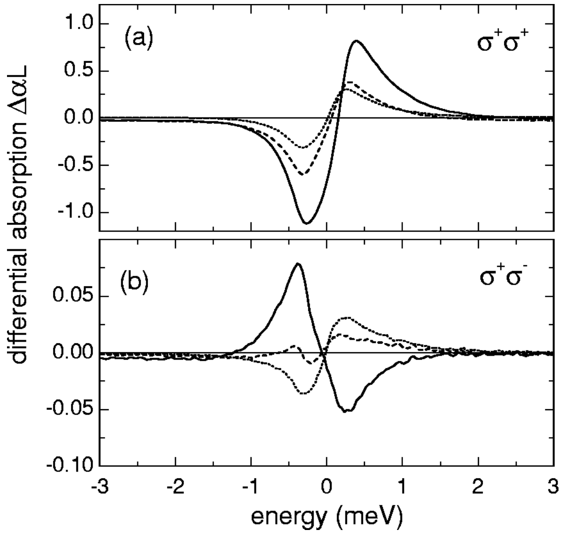

FIG. 1. Experimental differential absorption spectra of $In_{0.04}Ga_{0.96}As$ quantum wells at zero time delay for excitation below the $1s$ heavy-hole (hh)-exciton resonance and Δ_{hh} detunings of the pump pulse of 6.2 meV (solid line), 9.7 meV (dashed line), and 13.3 meV (dotted line). (a) Cocircularly and (b) anticircularly polarized pump and probe pulses. The origin of the energy scale corresponds to the position of the $1s$ hh exciton in the linear absorption.

of 13.3 meV below the exciton, the signal recovers but it has now changed into a blueshift.

III. THEORY

The redshift for anticircular polarization observed for moderate detunings is well explained by the approach outlined in Ref. 9, which took only the heavy-hole transition into account. This approximation is justified as long as the detuning Δ_{hh} is much smaller than the heavy-hole to light-hole splitting. However, if the pump pulse is tuned far below both the heavy-hole-exciton and light-hole-exciton resonances, the influence of light holes has to be included in the theory. For this purpose, we use our microscopic theory that includes all Coulomb many-particle correlations within the coherent $\chi^{(3)}$ limit, i.e., up to third order in the external fields.[6,9] The inclusion of light-hole transitions is straightforward and has been discussed in Refs. 10 and 11; for an extended review of the theory and also the model system see Ref. 12. The complete set of equations of motion is therefore not repeated here.

To understand the relevance of the respective heavy- and light-hole contributions, we discuss the pump- and probe-polarization-dependent transitions schematically shown in Fig. 2. For cocircularly polarized pump and probe pulses [Fig. 2(a)] a σ^+-polarized pump couples the $m=-\frac{3}{2}$ heavy-hole state with the $m=-\frac{1}{2}$ electron state and the $m=-\frac{1}{2}$ light-hole state with the $m=\frac{1}{2}$ electron state. The σ^+-polarized probe pulse monitors the $m=-\frac{1}{2}$ electron spin system, i.e., the heavy-hole resonance, thus experiencing the regular Stark shift (blueshift) dominated by the Hartree-Fock contributions.[6,9]

For anticircular pump-probe polarizations, transitions

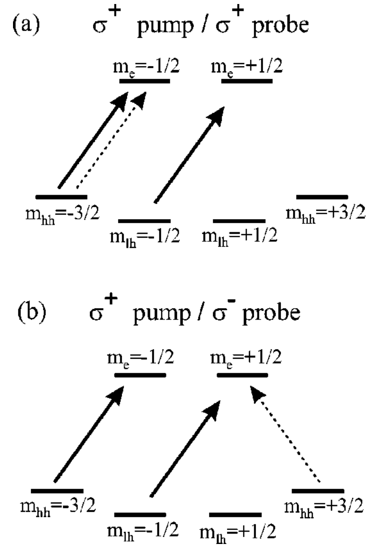

FIG. 2. Simplified electron, heavy-hole and light-hole level scheme, transitions induced by a σ^+-pump beam (solid arrows), and heavy-hole transitions probed (dashed arrows).

from heavy holes and light holes involve the same electron states; see Fig. 2(b). The σ^--polarized probe pulse monitors the $m=\frac{1}{2}$ electron spin system, thus experiencing from the heavy-hole transition only the influence of the higher-order Coulomb correlations and from the light-hole resonance also Hartree-Fock contributions. Within the coherent $\chi^{(3)}$ limit the higher-order correlations are represented by the contributions from bound and unbound two-exciton states to the optical signals. Light holes, heavy holes, and electrons are coupled by the higher-order Coulomb correlations.[11] The strong Coulomb interactions among heavy- and light-hole excitons result in distinct signatures due to the two-exciton manifold. As shown in Ref. 11, if neither the electrons nor the holes of the two interacting excitons share a common state (which means that the excitons are not coupled directly by the light field) a bound two-exciton complex is formed, which may consist of two heavy- or light-hole excitons for anticircular pump-probe polarizations, or it can be formed by one heavy-hole and one light-hole exciton for cocircular polarized excitation.

To get an idea about the microscopic origin of the intricate detuning dependence seen in the experimental results it is sufficient to go through some simplified analytical evaluations. If only a few-exciton and two-exciton states are relevant the full equations can be projected onto these levels.[9] For our purposes it is sufficient to consider the simplest case where the system consists of a ground state with zero energy, one single-exciton state with polarization p and energy ω_p, and one two-exciton state with amplitude B and energy ω_B. The reduced equations of motion representing this situation are[9]

$$-i\partial_t p=(-\omega_p+i\gamma_p)p+\mu^*E(1-bp^*p)-V_p p^*pp + V_B p^*B, \tag{1}$$

$$-i\partial_t B=(-\omega_B+i\gamma_B)B+pp, \tag{2}$$

where b, V_p, and V_B denote the strengths of the optical nonlinearities that are due to phase-space filling, first- and higher-order Coulomb contributions, respectively. μ is the optical dipole matrix element, and γ_p, γ_B are constant phenomenological dephasing rates.

In order to solve these equations analytically, the light fields are modeled as follows: For the pump field, we assume cw excitation with a frequency ω_L, and the probe field is taken as a δ pulse, i.e.,

$$E_{\text{pump}}(t)=\tilde{E}_{\text{pump}}e^{-i\omega_L t},\quad E_{\text{prob}}(t)=\tilde{E}_{\text{prob}}\delta(t). \tag{3}$$

As shown earlier,[13] cw excitation might result in a more pronounced shift compared to spectrally broad pulsed excitation.

The differential absorption $\delta\alpha$ is approximately given by the imaginary part of the third-order polarization δp in pump-probe geometry,[9] which is calculated as

$$\begin{aligned}\delta p(\omega)=\frac{1}{2\pi}(\mu^*\tilde{E}_{\text{pump}})(\mu\tilde{E}^*_{\text{pump}})(\mu^*\tilde{E}_{\text{prob}})\Bigg[&-b\frac{1}{(\omega_p-\omega_L)+i\gamma_p}\frac{1}{[(\omega-\omega_p)+i\gamma_p]^2}+b\frac{1}{(\omega_p-\omega_L)^2+\gamma_p^2}\frac{1}{(\omega-\omega_p)+i\gamma_p}\\&-V_p\frac{1}{(\omega_p-\omega_L)^2+\gamma_p^2}\frac{1}{[(\omega-\omega_p)+i\gamma_p]^2}+V_B\frac{1}{(\omega_p-\omega_L)^2+\gamma_p^2}\frac{1}{[(\omega-\omega_p)+i\gamma_p]^2}\frac{1}{(\omega_B-\omega_p-\omega_L)-i(\gamma_B-\gamma_p)}\\&+V_B\frac{1}{(\omega_p-\omega_L)^2+\gamma_p^2}\frac{1}{(\omega-\omega_p)+i\gamma_p}\frac{1}{[(\omega_B-\omega_p-\omega_L)-i(\gamma_B-\gamma_p)]^2}\\&-V_B\frac{1}{(\omega_p-\omega_L)^2+\gamma_p^2}\frac{1}{[\omega-(\omega_B-\omega_L)]+i\gamma_p}\frac{1}{[(\omega_B-\omega_p-\omega_L)-i(\gamma_B-\gamma_p)]^2}\Bigg].\end{aligned} \tag{4}$$

In Eq. (4), the first two terms are due to Pauli blocking, and the third one is the first-order Coulomb contribution. The sum of these three terms defines the Hartree-Fock approximation. The remaining three terms are introduced by transitions to two excitons and are thus caused by Coulomb correlations. In terms of the detuning $\Delta\equiv\omega_p-\omega_L$, which is taken to be much larger than the decay constants γ, the leading contributions to each of these nonlinearities are given by the first, third, and fourth terms, which show a detuning dependence of Δ^{-1}, Δ^{-2}, and Δ^{-3} (since $\omega_B\approx 2\omega_p$), respectively. The imaginary part of the dominant contributions yields the shift of the exciton line

$$\delta\alpha(\omega)\propto\frac{2\gamma_p(\omega-\omega_p)}{[(\omega-\omega_p)^2+\gamma_p^2]^2}. \tag{5}$$

IV. NUMERICAL RESULTS AND COMPARISON WITH EXPERIMENT

In this section we present numerical evaluations of the full third-order equations as discussed in Refs. 10–12. We take the splitting between the heavy- and light-hole excitons as 12 meV and assume that the pump pulse has a Gaussian envelope with a duration that corresponds to the spectral width used in the experiments. For the oscillator strength of the heavy- and light-hole transitions a ratio of 4:1 is used.

Figures 3 and 4 show the calculated differential absorption for the parameters of the InGaAs quantum well. For cocircular polarization of pump and probe pulses, the calculations reproduce the experimental observations, i.e., a reduction of the blueshift with increasing detuning; see Fig. 3(a). The results for anticircular polarization are displayed in Fig. 4(a). The redshift, that is found for $\Delta_{hh}=6.2$ meV, decreases drastically for larger detunings. At $\Delta_{hh}=13.3$ meV one finds a blueshift, in good agreement with the experiment. Deviations of the measured differential absorption from the symmetric shape are due to disorder present in the sample. The absorption spectrum of the heavy-hole exciton resonance is weakly inhomogeneously broadened and displays an asymmetric line shape.[7]

To analyze these results we now make use of the detuning dependence of the various nonlinearities discussed in the previous section. For cocircular polarization geometry, the $\Delta m=1$ heavy-hole transition is pumped, and the same heavy-hole transition is probed [Fig. 2(a)], and affected by three nonlinearities: Pauli blocking and first-order Coulomb terms produce a blueshift, whereas the higher-order Coulomb contributions induce a small redshift that is, however, overcompensated by the blueshift contributions.[6] The influence of the light-hole transition arises only due to the Coulomb interaction among the heavy holes, light holes, and electrons. This influence is negligibly small compared to the changes induced by pumping the heavy-hole transition, as can be seen by comparing Figs. 3(a) and 3(b) where the light-hole contribution has been omitted. Furthermore, looking at Fig. 3(c) where the higher-order Coulomb correlations have been ignored, we see that also these contributions affect the differential absorption for cocircular polarization only to a small extent. This conclusion is also supported by the normalized spectra shown in the lower panel of Fig. 3(a). Here the differential spectra have been multiplied by the detuning

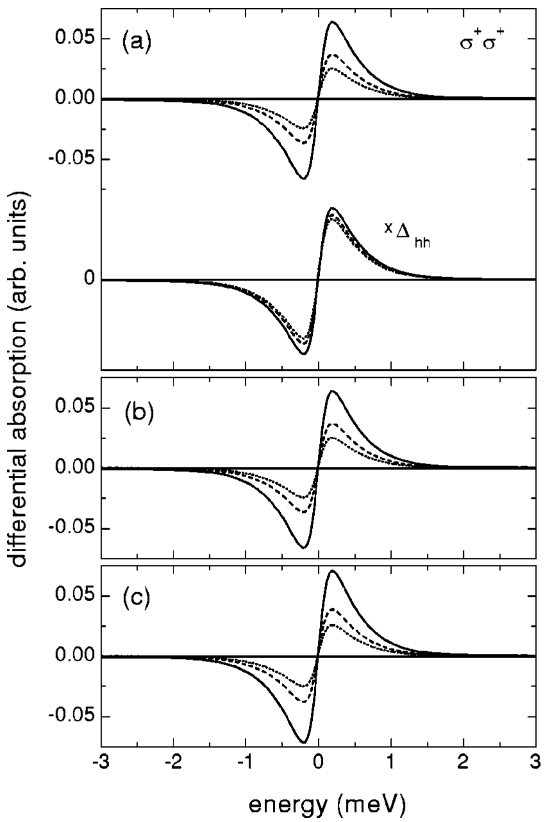

FIG. 3. Calculated differential absorption spectra at zero time delay for excitation below the $1s$ hh-exciton resonance, a heavy-hole to light-hole splitting of 12 meV, and Δ_{hh} detunings of the pump pulse of 6.2 meV (solid line), 9.7 meV (dashed line), and 13.3 meV (dotted line) for cocircularly polarized pump and probe pulses. (a) Full calculation (upper panel) and multiplied with Δ_{hh} (lower panel), (b) without the light-hole contribution, (c) without the higher-order Coulomb correlations. The origin of the energy scale corresponds to the position of the $1s$ hh exciton in the linear absorption.

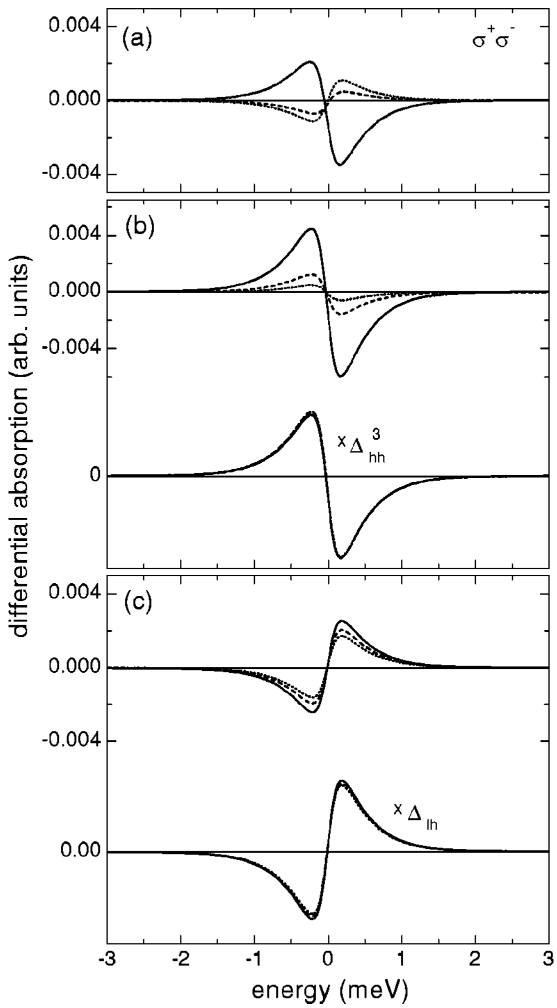

FIG. 4. Calculated differential absorption spectra at zero time delay for excitation below the $1s$-hh-exciton resonance, a heavy-hole to light-hole splitting of 12 meV, and Δ_{hh} detunings of the pump pulse of 6.2 meV (solid line), 9.7 meV (dashed line), and 13.3 meV (dotted line) for anticircularly polarized pump and probe pulses. (a) Full calculation, (b) without the light-hole contribution (upper panel) and multiplied with Δ_{hh}^3 (lower panel), (c) without the higher-order Coulomb correlations (upper panel) and multiplied with Δ_{lh} (lower panel). The origin of the energy scale corresponds to the position of the $1s$ hh exciton in the linear absorption.

Δ_{hh}. As shown earlier[14] and derived analytically in the previous section, the Pauli-blocking contributions to the Stark shift scale with $1/\Delta_{hh}$ and the first-order Coulomb correlations vanish like $1/\Delta_{hh}^2$. The higher-order Coulomb correlations exhibit the strongest detuning-dependent decay proportional to $1/\Delta_{hh}^3$. As a consequence, if the normalized spectra are almost invariant with respect to Δ_{hh}, the differential spectra display a $1/\Delta_{hh}$ dependence that is characteristic for the Pauli-blocking contribution.

If one neglects the light holes for the case of anticircular pump-probe polarizations, the $\Delta m=-1$ transition that is probed would involve only states different from those that are pumped. Thus the Hartree-Fock contributions, the first-order Coulomb and Pauli-blocking terms, do not affect the transition that is probed. The only remaining contribution in this case is the higher-order Coulomb term that results in a redshift of that transition energy for all detunings; see Fig. 4(b). As discussed above, this Coulomb correlation contribution decays with $1/\Delta_{hh}^3$ [see Fig. 4(b) lower panel] in excellent agreement with the analytical result.

On the other hand, if the light-hole states are properly included the pump couples the $m=-\frac{1}{2}$ light-hole states with the $m=\frac{1}{2}$ electron states shifting the latter to higher energies and thus contributing a blueshift of the $\Delta m=-1$ transition probed by the σ^- pulse. Neglecting the higher-order Coulomb correlations, i.e., the influence of the heavy-hole transition, we obtain a blueshift that falls off like $1/\Delta_{lh}$; see Fig. 4(c). For these large detunings Δ_{lh} studied here, both the first- and higher-order Coulomb correlations involving the light hole no longer contribute significantly.

This blueshift due to the light-hole coupling competes with the redshift due to the Coulomb correlations induced by pumping the $\Delta m=1$ heavy-hole transition. For small detunings and a large heavy-hole to light-hole splitting the blueshift is weaker than the redshift. However, for larger detunings both heavy-hole and light-hole transitions are pumped well off-resonance, and consequently the influence of the light-hole resonance increases relative to that of the heavy-hole resonance. The light-hole induced blueshift then overcompensates the Coulomb correlation induced redshift.

Because of the competition of the heavy-hole-induced redshift and the light-hole-induced blueshift, not only the detuning from the heavy-hole exciton resonance is crucial in determining direction and amplitude of the shift, but also the detuning with respect to the light-hole exciton resonance, and therefore the splitting between these two resonances. Since it is experimentally not possible to easily vary the heavy-hole to light-hole splitting in a continuous way, we additionally investigated a sample containing fifty 18 nm wide GaAs quantum wells with an excitonic linewidth of 1.4 meV, where the splitting is just 6.5 meV. In contrast to the observation in $In_xGa_{1-x}As$ quantum wells but in support of Refs. 2, 3, and 15, we found that for cocircular as well as for anticircular polarization a blueshift is induced for moderate detunings below the heavy-hole exciton.

Theoretically this case has been studied in Ref. 10, where the dependence of the Stark shift on the heavy-hole to light-hole splitting was analyzed. For a splitting of 12 meV the blueshift for cocircular and the redshift for anticircular polarization are reproduced. A reduction of the splitting resulted in hardly any change for cocircular polarization. However, for anticircularly polarized pulses the redshift is strongly affected by the reduced splitting. It decreases and finally, for small splitting, changes to a blueshift,[10] which is in agreement with the experiments performed on GaAs quantum wells.

V. CONCLUSIONS

In conclusion, we have shown experimentally and theoretically that the redshift for anticircular polarization of pump and probe pulses depends critically on the detuning of the pump pulse and the heavy-hole to light-hole splitting. It is only observable in samples with a large heavy-hole to light-hole splitting and only within a certain range of moderate detunings of the pump pulse. The most important features of the polarization-dependent absorption changes can be well described by microscopic numerical calculations that include Coulomb correlations. The underlying physics can be qualitatively understood on the basis of analytical results that show that the different contributions to the signal diminish with different power laws as function of the detuning between the pump and the excitons.

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1. D. Fröhlich, A. Nöthe, and K. Reimann, Phys. Rev. Lett. **55**, 1335 (1985).
2. A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoc, Phys. Rev. Lett. **56**, 2748 (1986).
3. A. Von Lehmen, D. S. Chemla, J. E. Zucker, and J. P. Heritage, Opt. Lett. **11**, 609 (1986).
4. D. Hulin and M. Joffre, Phys. Rev. Lett. **65**, 3425 (1990).
5. M. Combescot and R. Combescot, Phys. Rev. Lett. **61**, 117 (1988).
6. C. Sieh, T. Meier, F. Jahnke, A. Knorr, S. W. Koch, P. Brick, M. Hübner, C. Ell, J. Prineas, G. Khitrova, and H. M. Gibbs, Phys. Rev. Lett. **82**, 3112 (1999).
7. C. Ell, J. Prineas, T. R. Nelson, Jr., S. Park, H. M. Gibbs, G. Khitrova, S. W. Koch, and R. Houdré, Phys. Rev. Lett. **80**, 4795 (1998).
8. T. Meier, S. W. Koch, P. Brick, C. Ell, G. Khitrova, and H. M. Gibbs, Phys. Rev. B **62**, 4218 (2000).
9. C. Sieh, T. Meier, A. Knorr, F. Jahnke, P. Thomas, and S. W. Koch, Eur. Phys. J. B **11**, 407 (1999).
10. S. W. Koch, C. Sieh, T. Meier, F. Jahnke, A. Knorr, P. Brick, M. Hübner, C. Ell, J. Prineas, G. Khitrova, and H. M. Gibbs, J. Lumin. **83-84**, 1 (1999).
11. T. Meier, S. W. Koch, M. Phillips, and H. Wang, Phys. Rev. B **62**, 12 605 (2000).
12. T. Meier and S. W. Koch, in *Semiconductors and Semimetals*, edited by R. K. Willardson and E. R. Weber (Academic, San Diego, 2001), Vol. 67, pp. 231–313.
13. C. Ell, J. F. Müller, K. El Sayed, L. Bányai, and H. Haug, Phys. Status Solidi B **150**, 393 (1988).
14. C. Ell, J. F. Müller, K. El Sayed, and H. Haug, Phys. Rev. Lett. **62**, 304 (1989).
15. M. Joffre, D. Hulin, A. Migus, and M. Combescot, Phys. Rev. Lett. **62**, 74 (1989).