Coherent exciton interactions: Dependence on excitation fluence and polarization

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The coherent interaction between excitons is revisited by examining the influence of coherent photo-excited excitons on the absorption spectra of ZnSe/ZnMgSSe multiple quantum wells for various polarization configurations. For purely resonant excitation at the heavy-hole exciton, a blueshift of the exciton absorption is found. This distinct blueshift shows a linear dependence on excitation fluence, but with different slopes for different polarization configurations. In the coherent regime the smallest shift is found for an orthogonal linear polarization of the pump and probe, noticeably smaller than the shift for same linear polarization. The strongest shift is found for the same circular polarization. Furthermore, we demonstrate experimentally that pump and probe and four-wave mixing spectra contain equivalent information about the shift and broadening of the exciton line. A linear correlation between the blueshift and the homogeneous linewidth is found in both pump and probe and four-wave mixing.

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I. INTRODUCTION

The coherent interaction of light and the electronic states of semiconductors at the fundamental band gap has been a very active research area since the development of femtosecond laser systems in the late 1980s. Two particularly well-suited and established techniques to investigate these interaction processes are pump and probe (P&P)[1–9] as well as four-wave mixing (FWM) spectroscopies.[1,9–18]

Among the first experiments on coherent light-matter interactions in semiconductors were measurements of the optical Stark effect.[19–21] To investigate the Stark effect a strong pump pulse tuned to the band-gap region excites the system, and a weak test pulse probes the shift at the resonance. The optical Stark effect is a nonresonant but coherent effect by definition, and is well known from atomic physics. As a consequence the initial models for the explanation of excitonic Stark shifts were based on the dressed-atom picture, and properly predicted a linear dependence of the shift on the excitation intensity.[22] The polarization dependence of the effect, and in particular the origin of the difference between same linear and orthogonal linear polarized excitation pulses, was debated intensely.[20,21] The description of the resonant pumping case however was out of reach because of the divergence in the models for zero detuning.[20,22,23]

In the case of resonant excitation, Coulomb correlations are important[23] and the significance of Coulomb effects for the nonlinear response of semiconductors and their dependence on the polarization of the incoming light fields has been explored in numerous publications.[1–6,9,11–13,15–18,24–29] Typically the polarization configurations of the exciting light fields are split between the two most established measurement techniques. The FWM technique was often used for comparisons between linear polarization cases,[10–13,15–18] while the P&P experiments mainly examined the circular polarizations.[2–5,30] This is at least partly based on the lack of a FWM signal for excitation beams with opposite circular polarizations.[17,31] By examining the difference between the linear polarization configurations, the importance of Coulomb correlations such as bound and unbound biexcitons or higher correlations was demonstrated, for example, by means of quantum beats,[11–13] spectral weights of signal-components,[15,17] or phase shifts[16,18] in the spectrally and/or temporally resolved FWM signal.

In this paper we show several results which bear on the interpretation of pump and probe data. First, we show that information from P&P and FWM measurements, when performed under the same conditions, reproduce the same features in terms of broadening and energy shifts. Second, we show that the energy shift and homogeneous linewidth have a mutual dependency. Finally, we show that in pump and probe measurements for linearly polarized light fields a strong influence of the relative orientation of the polarization can be observed. This observation has important consequences for the validation of microscopic theories of Coulomb correlations.

In order to keep the interactions and the description of coherent experiments as transparent as possible, it is desirable to excite only one single resonance. In material systems with a small exciton binding energy and small heavy-hole (hh)–light-hole (lh) splitting, it is difficult to resonantly excite one resonance with 130-fs laser pulses exclusively. This is, for example, the case for GaAs-based heterostructures which are often used for these studies because they are theoretically well understood and available in very high quality. When the exciting laser pulse is centered at the hh exciton, one always has to deal with continuum excitations and/or hh-lh interactions which makes the description and discrimination of influences from free carriers and excitons even more difficult. For example, lh excitons have a strong influence on the optical Stark effect at the hh exciton[22] in $In_xGa_{1-x}As$,[29] and higher-order Coulomb correlations are very important for the interpretation of such experiments. For off-resonant excitation a competition between red and

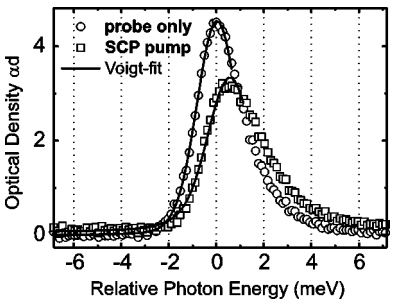

FIG. 1. Optical density of the hh-exciton resonance for probe only, i.e., linear absorption (circles), and with a SCP pump pulse (squares). The solid lines are Voigt profiles (Ref. 33).

blue shifting terms was observed. The shift in the experiment thus strongly depends not only on the polarization of P&P beams but also on both hh-lh splitting and the exact excitation energy.[20,22,23,30]

The situation is different for wide-gap II-VI semiconductor nanostructures. ZnSe-based heterostructures are an ideal model system to meet the requirements in the investigation of coherent exciton interaction in semiconductors beyond the weak-coupling regime.[8,9,15–18,27–29] The high exciton binding energy in these samples allows a resonant excitation of the sharp and well-separated hh resonance with short laser pulses, without exciting any continuum or lh states. In addition, the large nonlinear coefficients allow experiments with very low excitation intensities. $Zn_yCd_{1-y}Se$ multiple quantum wells (MQW's), for example, show a strongly polarization-dependent blueshift dynamics which could be explained by spin-flip and biexciton-formation processes.[32] Because of the strong influence of localization in $Zn_yCd_{1-y}Se$, and the high excitation fluences needed to generate biexcitons, the dephasing times in these experiments are too short to examine coherent exciton interactions.

The binary ZnSe quantum wells investigated here have long dephasing times of the order of 10 ps,[9,27,29] which provides us with a large time window for the discrimination of coherent effects from incoherent effects. We are also able to exclusively excite the $1s$ hh exciton resonantly. Therefore, we can exclude influences from Stark effect, spin-flip, continuum and/or lh excitations, as well as density effects, and can measure in the fully coherent regime.[9] As has already been noted in previous publications,[28,29] coherent exciton interactions are much stronger than incoherent ones (in ZnSe up to four times as strong).

II. SAMPLES AND EXPERIMENT

The investigated samples are ZnSe/ZnMgSSe MQW's with ten periods of 8 or 5-nm well width and 11-nm barrier width grown by molecular-beam epitaxy. The samples show a very small Stokes shift between photoluminescence (PL) and PL excitation spectra, and from this and temperature-dependent PL measurements we can conclude that localization plays only a minor role in these samples. Since all results are qualitatively similar in both samples, we will focus in the following on the 8-nm MQW in which the hh-lh splitting amounts to 22 meV and the exciton binding energy is

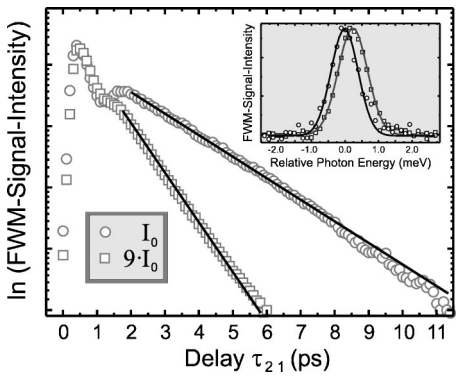

FIG. 2. FWM delay-time traces for two different intensities of $\vec{k}_1$. The lines are exponential fits to the data. The inset shows the corresponding FWM energy spectra. The decrease of T_2, i.e., the increase in the homogeneous linewidth, goes along with a blueshift.

about 21 meV which allows the resonant excitation of the hh exciton exclusively.

In order to perform absorption measurements the samples were mounted on sapphire plates and the GaAs substrates chemomechanically removed. The samples were kept at $T=5$ K in a Helium flow cryostat and 130-fs-long, frequency-doubled pulses with a spectral full width at half maximum of 12 meV from a titanium sapphire laser with a repetition rate of 76 MHz were used as the excitation source. Both the pump-and-probe pulses were taken from the same initial laser pulse, and could be delayed with respect to each other via delay stages with 8-fs resolution. The pulses were focused onto the sample at a small angle. To ensure we probe a region of homogeneous excitation, we defocused the pump beam spot to double the probe spot ($\varnothing=50\ \mu$m). The polarization of both beams could be chosen independently to investigate several relative polarization configurations of P&P pulses, namely, the same circular polarization (SCP), opposite circular polarization (OCP), same linear polarization (SLP), and orthogonal linear polarization (OLP). The transmitted part of the probe or the FWM signal in direction $2\vec{k}_2-\vec{k}_1$ was spectrally resolved by a grating spectrometer and detected by a nitrogen-cooled CCD array. The exciting pump laser is tuned resonantly with the hh exciton, and the probe pulse is two orders of magnitude weaker than the weakest pump pulse. The delay between P&P pulses is chosen so as to maximize the strength of the blueshift and to ensure we are in the coherent regime (essentially zero delay).

In order to extract the changes of the absorption peak position and linewidth, we plotted the optical density αd, defined via the integrated form of Beer's law, $\alpha d=\ln(I_0/I_T)$, in the energy region of interest. I_0 denotes the incident laser intensity, I_T the transmitted intensity, α the absorption coefficient, and d the total MQW thickness.

III. RESULTS AND DISCUSSION

In Fig. 1 we show the optical density spectrum for the $1s$ hh exciton obtained from the P&P experiments. The linear spectrum has a linewidth of 2 meV. The absorption with pump pulse in the SCP configuration is bleached and blue-

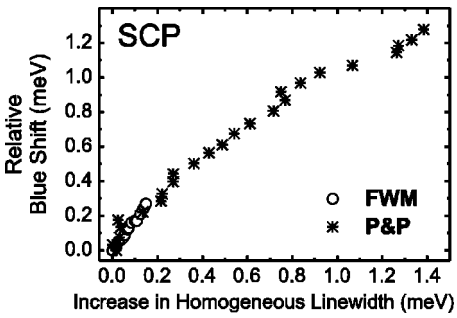

FIG. 3. Blueshift vs homogeneous linewidth for the SCP case deduced from both FWM and P&P measurements.

shifted. The solid lines in Fig. 1 are Voigt fits to the absorption lines. By choosing the Voigt line shape[34,35]

$$V(\omega)=V_0+A\frac{2\ln 2}{\pi^{3/2}}\frac{\Gamma}{\sigma^2}\times\int_{-\infty}^{\infty}\frac{e^{-t^2}}{\left(\sqrt{\ln 2}\,\frac{\Gamma}{\sigma}\right)^2+\left(\sqrt{4\ln 2}\,\frac{\omega-\omega_o}{\sigma}-t\right)^2}dt, \tag{1}$$

which is a convolution of Gaussian and Lorentzian line shape, it is possible to resolve the inhomogeneous (Gaussian), σ, and homogeneous (Lorentzian) parts, Γ of the absorption line.

The homogeneous linewidths[33] extracted from fitting the P&P results using Eq. (1) we found to be in excellent agreement with those extracted from the dephasing times we measured more directly in FWM (see Fig. 2) under the same excitation conditions [the same fluence of the pump beams ($\vec{k}_1$) and the same polarization configuration]. This is an important check of the accuracy of our fitting procedure, and ensures that reliable homogeneous linewidths can be extracted from P&P data. Thus it enables us to measure the enhancement of the homogeneous linewidth in the OCP case for which there is no FWM signal, and thereby compare it to the other polarization configurations. As a final check, we also extracted the blueshift of the hh-exciton resonance from FWM experiments; see the inset in Fig. 2, to compare it with the shifts in P&P measurements. In order to get rid of any uncertainty in the determination of the exact excitation intensity, and more importantly the excited density, we plotted the shift versus the homogeneous linewidth with both quantities determined from the same experimental data set. In Fig. 3 we show the results for the SCP configuration, which gives strong FWM signal intensities and the largest shifts (see below). We find a linear dependence over a wide range, which matches closely for both measurement techniques. The equivalence of the results for both techniques demonstrates that P&P and FWM spectra, taken under exactly the same conditions, contain the same information about shift and broadening of a resonance.

The linearity of the result may be a coincidence, because both the shift and the broadening are linear in the excitation

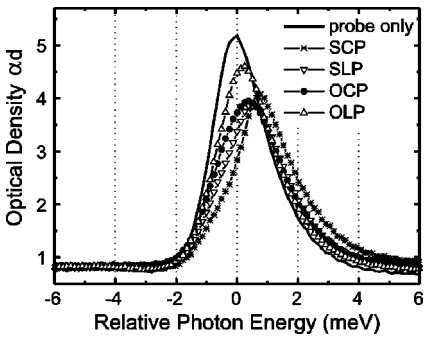

FIG. 4. Optical density of the hh-exciton resonance for probe only (linear absorption) and different relative polarizations of pump and probe pulses with a pump fluence of 0.5 μJ cm^{-2}.

fluence. But since both quantities are part of the exciton self-energy[36] this could also be an intrinsic relationship. Up to now these two quantities have largely been examined independently both in experiment and in theory, where the approximations necessary to be able to calculate each of the two quantities are often different. However, as we find that the ratio of the blueshift to homogenous linewidth is constant and does not require a precise determination of the excitation density, it is an obvious candidate for a comparison with microscopic models.

In the following we will focus on the polarization dependence of the shifts and particularly on the differences between the SLP and OLP. In Fig. 4 P&P spectra corresponding to the four different polarization configurations are shown along with the linear absorption. The smallest shift is seen in the OLP configuration, followed by OCP, SLP, and finally SCP configurations. In these spectra we have subtracted the biexciton contribution, which appears in the OCP and SLP cases, in order to make the comparison more meaningful. We have measured the intensity dependence of the exciton shift in all four polarization configurations up to pump powers of 1 μJ/cm^2. The results are shown in Fig. 5. We find in all cases a good linear behavior with the slope increasing from OLP, to OCP, to SLP and finally SCP. The overall linearity has been predicted[23,36] and experimentally shown[5,35] but a systematic investigation of the polarization dependence of the slopes has not been published until now, to our knowledge.

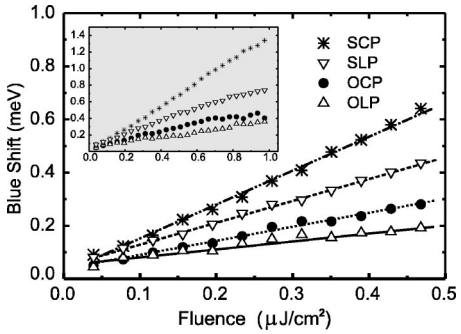

FIG. 5. Polarization dependence of the blueshift vs pump fluence in P&P measurements. The lines are just guides to the eye, highlighting the linearity. The inset shows the same set of data for a wider range of fluences.

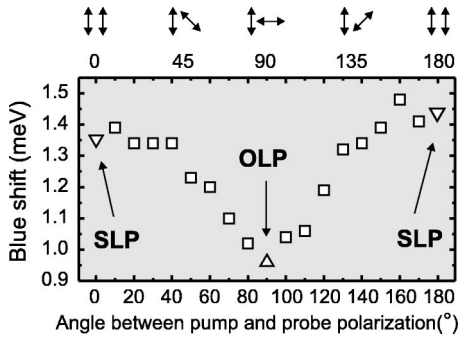

FIG. 6. Angle dependence of the shift for linear polarizations going from SLP to OLP configurations and back to a SLP configuration. Experimentally the polarization direction was changed by turning a polarizer while the excitation fluence was kept constant.

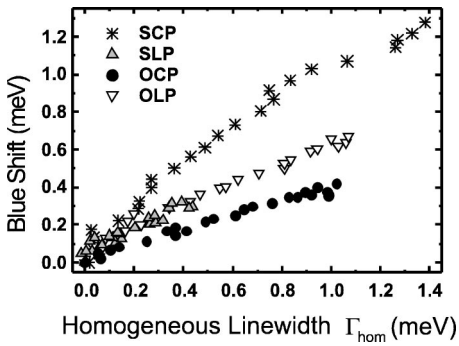

FIG. 7. Blueshift vs homogeneous linewidth extracted from the Voigt fits for different P&P polarization configurations.

The slope of SLP lies roughly midway between the SCP and OCP cases which, in an intuitive way, one would argue reflects the fact that SLP is just a superposition of the circular polarization cases SCP and OCP. However, intriguingly, the OLP case greatly differs from SLP and shows the weakest blueshifts.[37] This then has a significant implication, as the OLP is the natural configuration to use for best signal discrimination, but produces the smallest effect. The linear cases are often used in the literature without differentiating between them.[7,35] This may be of little consequence in the incoherent regime[32] but, as demonstrated above, not at all in the coherent regime. It is seductive to look at the different linear polarization configurations just as a superposition of the two circular polarizations, as one would do in a simplified level picture. In such a picture one would not expect any difference between SLP and OLP configurations, because in both cases the same amount of σ^+ and σ^- photons impinge on the sample. That the difference in the blueshifts for SLP and OLP configurations are not explainable in a simple level scheme is illustrated once more in Fig. 6, where the blueshifts of the absorption for different angles between the linearly polarized pump and probe pulses are shown. The shift is largest for the parallel configuration (0° and 180°) of the two linear beams (SLP) and is decreasing toward 90° (OLP). Nothing is changed except the phase between the two circular components of the exciting light fields, indicating the great importance of this phase in the coherent regime.

Shown in Fig. 7 is the blueshift versus the homogeneous linewidth extracted from the Voigt fits for the four different P&P polarization configurations. We see that for all four configurations there is an essentially linear behavior, although due to the small shifts in the OLP case the data are rather noisy. Plotted now in this way, the SLP and OLP results are rather similar and lie between the SCP and OCP data. It is reasonable to suppose that the physical origins of both the blueshift and the linewidth enhancement lie in exciton-exciton interactions (including the influence of the two-exciton pair continuum states). Were all four of these lines to lie on top of one another it would indicate a universal relationship between the scattering time and energy renormalization of excitons. As it stands, the differences between the curves reflects the strength of exchange interactions between the excitons. We emphasize that the OCP data shown here could not be obtained from FWM measurements because of the polarization selection rules.

IV. SUMMARY

We present a detailed systematic investigation of the polarization dependence of coherent exciton-exciton interactions. The blueshift of the hh-exciton absorption resonance in P&P pulses pulses is examined for various polarizations and excitation fluences. We observe a blueshift for *all* investigated polarization configurations under resonant excitation. The shift for the SCP configuration is approximately twice as large as for the OCP configuration and increases linearly with the the excitation intensity. We experimentally show the equivalence of P&P and FWM data concerning the shift and broadening of excitonic resonances. Furthermore, we highlight the close connection between these two quantities via the exciton self-energy. This and the difference between the OLP and SLP cases in pump and probe measurements, particularly provides important input for future experimental and theoretical developments.

The theoretical understanding of the coherent interaction processes in semiconductors has made enormous progress during the last decade and the combined effort of experiment and theory has led to a detailed understanding of many important physical phenomena.[1] In these experiments we have striven to avoid anything which could lead to complications in modeling these interactions, such as incoherent carrier densities, continuum excitations, or band mixing due to lh excitations. We hope that the presented data will be theoretically explained soon, leading to a deeper understanding of the fundamental coherent interactions in semiconductors.

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