Signatures of High-Order Coulomb Correlations in Coherently Controlled Four- and Six-Wave-Mixing Experiments on a ZnSe Quantum Well

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Abstract. In this paper we present experimental and theoretical investigations of coherently controlled four- and six-wave-mixing experiments on a ZnSe single quantum well. In the signal which is mainly modulated with the exciton transition frequency we observe strong contributions of integer harmonics of this frequency if the experiment is carried out with co-linearly polarized excitation pulses. If the the polarization states are switched to a co-circular polarization state these harmonics are strongly suppressed but do not vanish completely. This polarization dependence of the harmonics suggests that biexcitons as well as two-exciton scattering states play an important role in the microscopic origin of this non-linear behavior. A theoretical approach based on the dynamics-controlled truncation scheme (DCT) is able to reproduce the experimental findings on a level on which six-point correlations are included. By selectively switching on and off parts of the full theory we clearly demonstrate that two-pair correlations, in particular biexcitons, crucially determine the occurrence and intensity of the observed additional frequencies.

1. Introduction

Coherent-control experiments have become very popular during the last few years because they provide the ultimate control over a quantum mechanical system with regard to both the amplitudes and the relative phases of different quantum mechanical excitations [1–6]. One possibility to achieve such a control is to excite the sample with two phase-locked optical pulses. When the relative phase of these two pulses is changed, the second pulse enhances or diminishes the optical polarization in the sample which has been induced by the first pulse. Since this control can be achieved on time scales being several orders of magnitude shorter than the recombination time of carriers in solid state systems the coherent control technique offers an outstanding potential to provide ultrafast optical switches which might be used in the field of quantum computing.

However, it has been shown in previous four- (FWM) and six-wave-mixing (SWM) experiments that higher Coulomb correlations affect the dynamics of the induced polarization even at low excitation intensities [7–9]. This also holds for the coherent control of excitonic polarization in semiconductor quantum wells where contributions of at least 5th order in the electric field, i.e., which are beyond the $\chi^{(3)}$-limit, have been experimentally observed and phenomenologically explained by solving the semiconductor Bloch equations supplemented with an excitation-induced dephasing of the excitonic polarization [10–12].

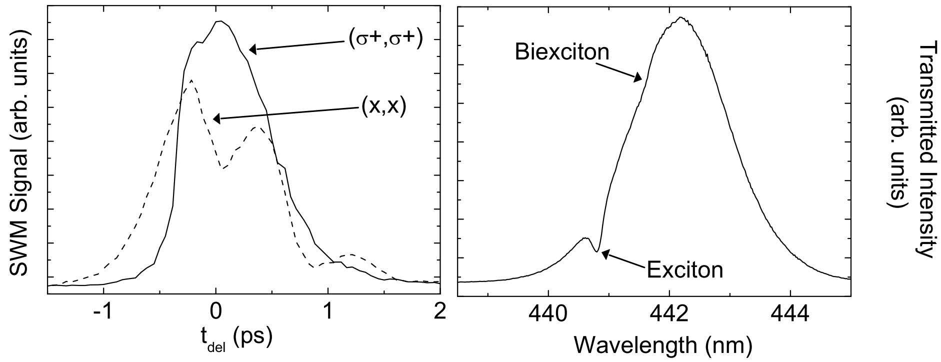

Figure 1. Left: SWM signal with (x,x) co-linearly (biexciton polarization enabled) and (σ^+,σ^+) co-circularly (biexciton polarization disabled) polarized excitation pulses. For the co-linear configuration the exciton-biexciton beating is clearly visible. Right: Corresponding spectrum of a transmitted laser pulse with absorption at the exciton and biexciton resonance.

Here, we report on the detailed experimental and theoretical investigation of signatures of higher Coulomb correlations in coherently controlled FWM and SWM experiments with different polarization states of the excitation pulses. The theoretical model based on the dynamics-controlled truncation scheme (DCT) reproduces the observed features and their dependence on the polarization states of the laser pulses very well. By selectively switching on and off parts of the full theory we are able to clearly demonstrate that two-pair correlations, in particular biexcitons, crucially determine the experimentally observed features.

2. Experimental Setup

All FWM and SWM experiments are performed at a temperature $T = 4$ K with a 10 nm ZnSe single quantum well which is embedded in two 500 nm ZnSSe cladding layers. The phase-locked pulses in direction k_1 performing the coherent control are separated by a delay time $t_{\rm int}$ and are generated with an actively stabilized Michelson interferometer which provides a temporal resolution of 0.04 fs. A third pulse from a different direction k_2 formes a polarization grating with the coherent polarization induced by the first two pulses if its delay time $t_{\rm del}$ is shorter than the dephasing time of the polarization. The signal is observed in the background-free directions $2k_2 - k_1$ (FWM) or $3k_2 - 2k_1$ (SWM). All pulses have a temporal width of 120 fs FWHM (full width at half maximum) and are generated by the same self-mode-locked frequency-doubled Ti:sapphire laser running at a repetition rate of 83 MHz. The center wavelength of the pulses is tuned near to the heavy-hole biexciton resonance so that the wavelength profile extends over the exciton resonance (Fig. 1, right part). The small spectral width of 2 nm FWHM of the laser pulses allows us to exclusively excite these two resonances. The polarization states of the excitation pulses are controlled with two Pockels cells which allow to switch the polarization states between x-linear and σ^+-circular without mechanical adjustment of the experimental setup.

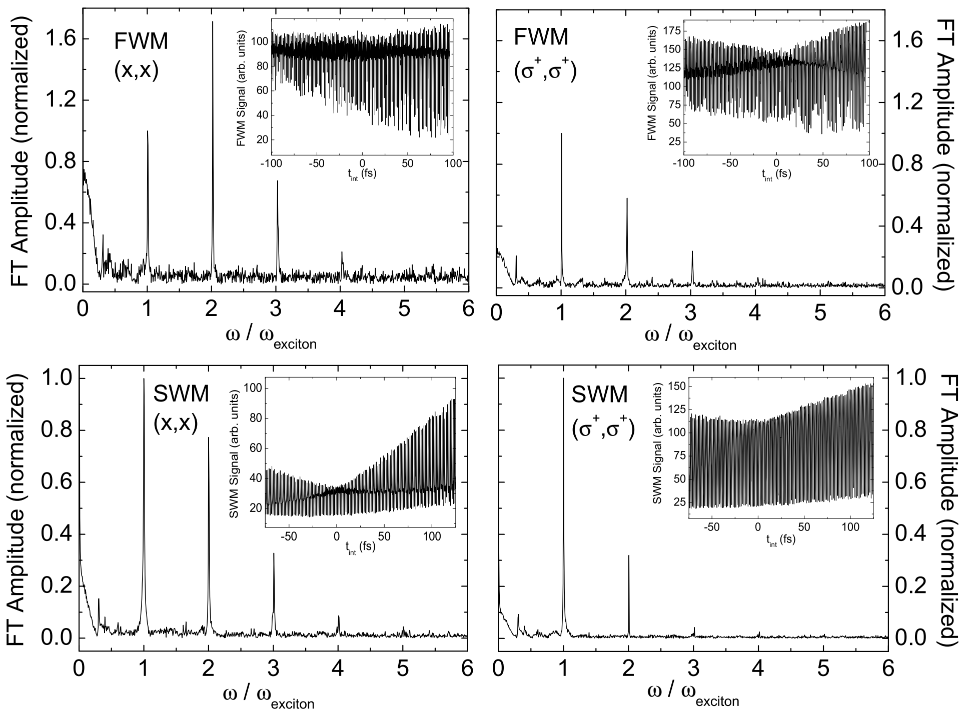

Figure 2. Fourier transforms of the coherently controlled FWM (top) and SWM (bottom) signal. Left: (x,x) co-linearly (biexciton polarization enabled), right: (σ^+,σ^+) co-circularly (biexciton polarization disabled) polarized excitation pulses. The measured coherently controlled wave-mixing signals from which the FTs are derived are shown as insets.

3. Experimental Results

A typical two-pulse SWM signal is depicted in Fig. 1 on the left side for (x,x) co-linearly and (σ^+,σ^+) co-circularly polarized excitation pulses. In the co-linear configuration the spin-selection rules [13] allow the creation of a bound-biexciton polarization which results in an exciton-biexciton beating. In the co-circular configuration where the formation of a bound-biexciton polarization is forbidden these beats are clearly absent. The signal has a larger overall amplitude but is less intense for negative delay times.

For the coherent-control experiments in the FWM as well as in the SWM configuration the delay time of the k_2 pulse is set to the fixed value $t_{\text{del}} = 600$ fs and the signal is recorded as a function of the delay time t_{int} between the two k_1 pulses generated by the interferometer. The signal mainly oscillates with the resonance frequency ω_{exciton} of the exciton polarization (see insets of Fig. 2). However, for co-linearly polarized excitation pulses additional frequencies being integer multiples of ω_{exciton} are clearly resolved in the corresponding Fourier transform spectra in both the FWM and SWM configuration (Fig. 2, left). In the coherently controlled excitonic signal (insets of Fig. 2, left) they manifest themselves by causing a fine structure around $t_{\text{int}} = 0$ fs. In the FWM configuration the higher harmonics of ω_{exciton} are more pronounced than in the SWM configuration, the peak at $2 \cdot \omega_{\text{exciton}}$ is even higher than that on the exciton transition frequency itself. In the SWM configuration the harmonics can be resolved with the highest corresponding to a frequency of $5 \cdot \omega_{\text{exciton}}$. A straightforward perturbational expansion of the induced macroscopic polarization shows that in the SWM direction $3k_2 - 2k_1$ this frequency is only generated by components which are of at least 9th order in the electric field. This means that the present experimental setup shows signatures of $\chi^{(9)}$ components in the coherent control of the exciton polarization with co-linearly polarized laser pulses.

If the polarization states of the excitation pulses are changed to a (σ^+, σ^+) co-circular configuration where the generation of a bound-biexciton polarization is forbidden the situation will become completely different. As can be seen in the right part of Fig. 2 the harmonics of ω_{exciton} are strongly suppressed and contributions of $4 \cdot \omega_{\text{exciton}}$ and $5 \cdot \omega_{\text{exciton}}$ are almost no longer visible neither in the FWM nor in the SWM configuration. Simultaneously, the amplitude of the coherently controlled signal around $t_{\text{int}} = 0$ fs increases, a fact which is more distinct in the SWM configuration. This result clearly shows that the fine structure produced by the higher harmonics of ω_{exciton} as well as the shape of the whole signal strongly depend on the polarization states of the excitation pulses. This suggests that bound-biexciton states and two-exciton scattering states play an important role in the microscopic origin of the fine structure and have to be included in an appropriate theory which models the experimental results on a microscopic level.

4. Comparison between experiment and theory

Since especially SWM experiments being free of $\chi^{(3)}$-contributions are a very sensitive tool to investigate the influence of higher Coulomb correlations a rather advanced level of the theory is needed to reproduce and explain the main features of the experimentally observed signals. The calculations presented here are carried out by using a microscopic density matrix description which is based on the DCT concept. A detailed description of this approach is given in [9]. The dynamics of four types of density matrices are taken into account. Of these, Y as the single-pair transitions and $\underline{B}$ as the correlated two-pair coherences describe the coherent parts of the dynamics, whereas $\underline{N}$ accounts for fluctuations of the coherent amplitude Y, and $\underline{Z}$ describes correlated transitions towards two-pair states induced by these fluctuations. These four density matrices evolve according to a set of coupled nonlinear equations which are numerically integrated. By using an implementation which represents $\underline{B}$ and $\underline{Z}$ by a memory kernel [9] we are able to selective study the contributions resulting from transitions to biexcitons or to two-exciton scattering states because these contributions can be selectively removed from the memory kernel.

In order to compare the experimental results with the theoretical simulation a range of -3 fs$\leq t_{\text{int}} \leq +3$ fs is shown in Fig. 3 for the FWM and SWM configuration at co-linear as well as co-circular polarization of the excitation pulses. The fine structure is clearly visible as a 'dip' in the signal at the position where the sine oscillation would normally have its maximum. This 'dip' is obviously produced by the oscillation with frequency $2 \cdot \omega_{\text{exciton}}$ which is resolved in the Fourier transform spectra in Fig. 2 and which is phase-shifted by $\frac{\pi}{2}$ with respect to the fundamental frequency. It can also be seen that the relative amplitude of the 'dip' decreases when the polarization state is changed from (x,x) to (σ^+, σ^+), in the SWM configuration the 'dip' vanishes completely.

This behavior is excellently reproduced by the numerical simulation, but only if the full theory is applied (Fig. 3, right, solid line). For the (x,x) configuration the simulation reproduces the oscillation with frequency $2 \cdot \omega_{\text{exciton}}$ phase shifted by $\frac{\pi}{2}$ compared with the fundamental frequency which is clearly showing up as a 'dip' at the maxima of the fundamental oscillation. When the polarization is switched to (σ^+, σ^+) the relative amplitude of the 'dip' becomes lower. However, even for the SWM configuration the signal still is not

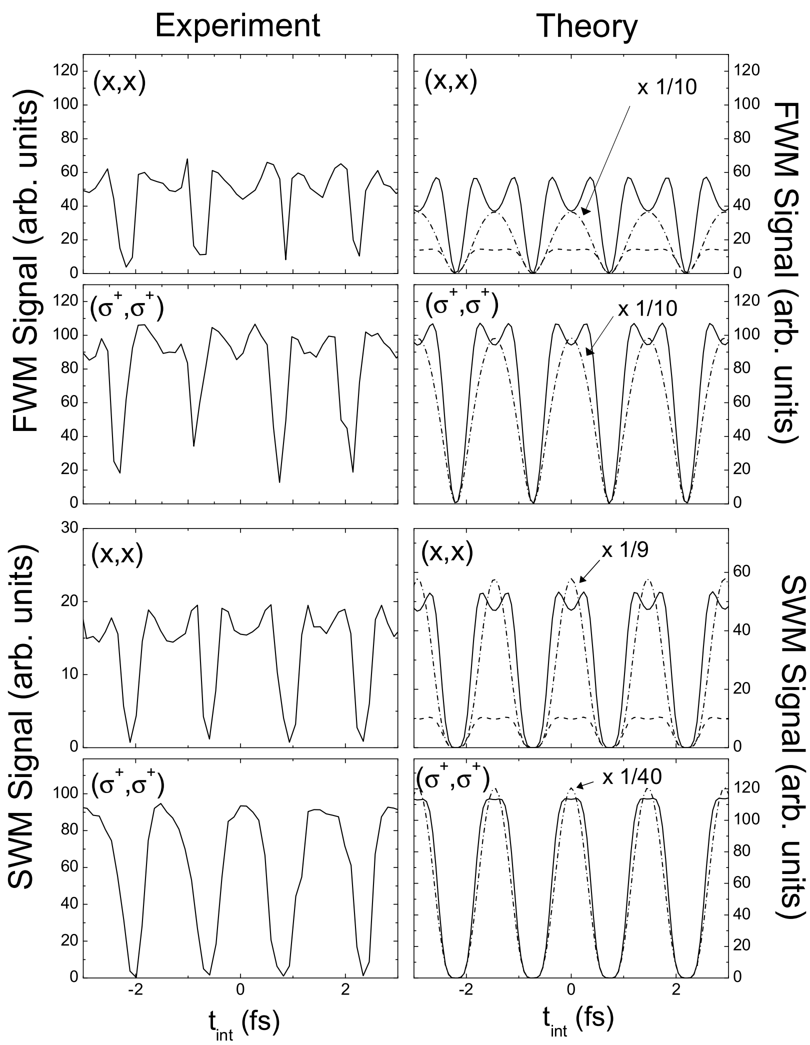

Figure 3. Comparison of the measured (left panel) and calculated coherently controlled wave-mixing signals (right panel) for the FWM (top) and SWM configuration (bottom). A residual background has been subtracted from the experimental data. The theoretical simulations are shown for the full theory (solid), full theory without biexcitonic contributions (dashed) and mean field theory scaled by the indicated factor (dot-dashed).

a perfect sine but remains slightly asymmetric with respect to its minima and maxima. In the frequency domain this corresponds to strongly suppressed but non vanishing frequency components at the positions of the harmonics of ω_{exciton}. All these theoretical results are in an excellent agreement with the experimental findings. For the FWM configuration the theory is also able to reproduce the relative amplitudes of the observed signals (note the same scales for experiment and theory). In the SWM configuration the strength of the signal for the (x,x) polarization state is overestimated. However, the trend of the amplitude to become smaller compared to the co-circular configuration is still well reproduced.

To investigate the influence of biexcitons and two-exciton scattering states on the harmonics calculations are performed on a mean-field level (dot-dashed lines in the right panel of Fig. 3) and on a level where the biexcitonic contributions are switched off but the mean-field parts as well as the correlated transition to two-pair scattering states are still included (dashed lines in the right panel of Fig. 3). It can be clearly seen that the mean-field calculation does not show the experimentally observed features of the coherently controlled excitonic polarization. Furthermore, it overestimates the overall signal strength (note the scaling in the figures), a fact which has been analyzed before in FWM signals [14]. Likewise, the calculations performed without biexcitonic contributions are not able to reproduce the experimental results. It is remarkable that the signal strength is now strongly underestimated and the sharp 'dip' produced by the harmonics is reduced to a rather weak modulation of the signal. This clearly demonstrates that higher Coulomb correlations and especially biexcitons are responsible for the fine structure that is observed in the coherent-control experiments.

It must be noted that in the theory a strong dependence of the fine structure on the laser pulse area that is used for the calculations is found. If the pulse area of all three pules is lowered the fine structure will become less pronounced and will quickly disappear. This behavior is also observed in the experiments which are all performed in a medium-density regime (about 1.5 pJ per pulse focused to a spot size of roughly 100 μm).

5. Summary

In conclusion, we have experimentally and theoretically analyzed the impact of higher Coulomb correlations on the coherently controlled excitonic FWM and SWM signal on a ZnSe single quantum well. It was found that apart from the fundamental exciton transition frequency also higher harmonics of this frequency strongly contribute to the observed signal which are generated by effects which are of up to 9th order in the electric field. It was found that the harmonics show a strong dependence on the polarization states of the excitation pulses. By comparing the experimental results with theoretical simulations based on the DCT it was demonstrated that biexcitons as well as two-exciton scattering states crucially determine the occurence of additional higher frequency components and cause the observed characteristic polarization dependence of the harmonics.

Acknowledgments

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