Coherent exciton-LO-phonon polarons in ZnSe quantum wells with strong confinement

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We observe coherent exciton-LO-phonon polarons in a 3 nm ZnSe single quantum well using four-wave mixing with 30 fs light pulses. The formation of these superposition states is attributed to a strong LO-phonon coupling by the Fröhlich interaction in a system in which the exciton binding exceeds the LO-phonon energy and the 2*s* exciton is nearly resonant with the LO-phonon energy. The rapid decay of these coherent quasiparticles is mainly attributed to the disintegration into zero-phonon excitons and free LO phonons as well as to the inhomogeneous broadening of LO-phonon energies due to disorder and **k**-space dispersion.

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INTRODUCTION

Relaxation and scattering processes of optically excited quantum states on a time scale of several tens of femtoseconds are fascinating physical phenomena that contribute to a deeper understanding of the underlying quantum mechanical description as well as have application potential, e.g., for quantum computation. In this time regime, scattering processes can no longer be treated as collisions between classical pointlike particles as described in Fermi's golden rule. For short times, the wavelike nature of interacting quasiparticles leads to mixed state coherences. As long as these coherences persist, scattering processes are not terminated but can be reversed or enhanced as demonstrated in bulk GaAs.[1] The finite duration of scattering events defines a "memory time" leading to a non-Markovian relaxation,[2] where the dynamics of quasiparticles becomes nonlocal in time. These memory effects are theoretically described by quantum kinetics.[3]

The nonlinear spectroscopic technique of degenerate four-wave mixing (FWM) using ultrashort (~10 fs) light pulses represents a powerful tool in studying quantum kinetic processes. Most of the investigations that include electron-LO-phonon quantum kinetics have been performed on III-V epilayers[1,4] and III-V nanostructures.[5] Fewer studies[6–8] have been performed on II-VI semiconductors which possess higher exciton binding energies as well as an enhanced electron-LO-phonon coupling (approximately seven times higher than in GaAs) due to the increased Fröhlich interaction. This enhanced LO-phonon coupling gives rise to simultaneous emission of multiple LO phonons in bulk ZnSe.[6] To date, no exciton-LO-phonon quantum kinetic investigations have been performed on II-VI quantum wells with strong confinement. Such nanostructures provide the unique opportunity of tuning the exciton binding energy from below to above the LO-phonon energy due to quantum confinement. While electron-phonon quantum kinetics is well understood in the weak coupling regime (appropriate for GaAs based samples), no theoretical description currently exists for intermediate phonon coupling in systems with strong confinement. Coherent exciton-LO-phonon polaron modes with finite dephasing times may result due to the enhanced phonon coupling in II-VI quantum wells (QWs), in particular, if the exciton+1LO phonon energy lies below the exciton continuum.

In this paper, we report on two-beam degenerate FWM experiments on a 3 nm thick ZnSe QW having an exciton binding energy that exceeds the LO-phonon energy. The FWM spectra reveal a coherent spectral feature that is separated by 31 meV from the heavy-hole exciton signal. The separation energy is equal to the energy of a LO phonon. Accordingly, the new FWM signal is attributed to an *exciton+1LO phonon quasiparticle*. Tuning the center pulse energy to even higher energies also enables the observation of an exciton+2LO polaron mode.

EXPERIMENTAL DETAILS

The single QW structure investigated here was pseudomorphically grown on (001) oriented GaAs substrate by molecular beam epitaxy. It consists of a 3 nm wide ZnSe QW sandwiched between two $Zn_{0.9}Mg_{0.10}S_{0.16}Se_{0.84}$ barriers of 30 nm thickness on the top and 60 nm at the bottom, with a 20 nm thick ZnSe buffer layer between the barrier and the substrate. A similar structure with 4 nm multiple QWs and the same barrier composition revealed a heavy-hole exciton binding energy of ~33 meV as derived from two-photon excitation (TPE) spectroscopy.[9] A homebuilt mode locked Ti: sapphire laser with two intracavity fused silica prisms used to control the dispersion inside the resonator provides 19 fs sech^2-shaped pulses around 1.45 eV center energy at a repetition rate of 80 MHz. The infrared pulses were frequency doubled by a 150 μm thick beta-barium borate crystal providing pulses with ~80 meV bandwidth [full width at half maximum (FWHM)]. The pulse duration of the frequency doubled pulses was determined to be 25±1 fs using two-photon absorption autocorrelation in a SiC photodiode. Glass elements in the FWM setup further increase the pulse duration to ~30 fs.[10] The two-beam FWM experiments involving excitation pulses $\mathbf{k}_1$ and $\mathbf{k}_2$ have been performed in backscattering geometry with the sample mounted in a helium cryostat at 30 K. The $1/e^2$ focus diameter of the pulses

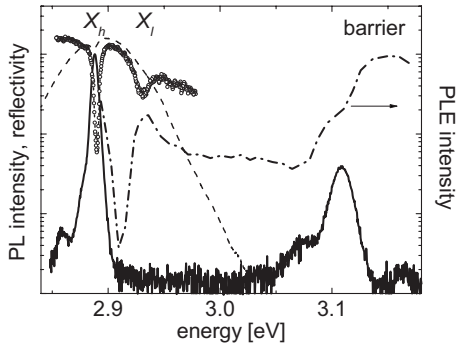

FIG. 1. Photoluminescence spectrum (full line) and photoluminescence excitation spectrum (dotted-dashed line) of a 3 nm ZnSe quantum well sample as described in the text. Also shown are reflectivity spectra using a Hg lamp (open circles) and a 30 fs light pulse (dashed line) used in the FWM experiments. X_h and X_l denote the heavy-hole and light-hole exciton transitions, respectively. All spectra were taken at 30 K.

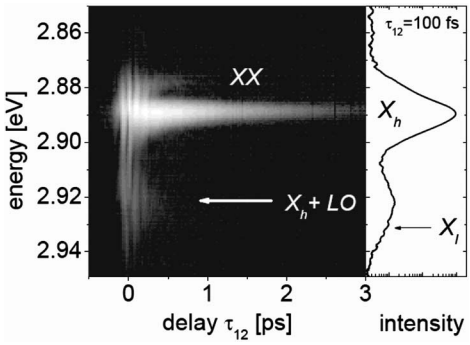

FIG. 2. Contour plot (logarithmic scale) of the FWM signal of a 3 nm ZnSe QW obtained in collinear (↑↑) configuration as a function of delay τ_{12} between pulses $\mathbf{k}_1$ and $\mathbf{k}_2$ with center energy of 2.9 eV. The total pulse intensity is 15 MW/cm^2. Also shown is the FWM spectrum at τ_{12}=100 fs on a logarithmic scale. *XX* and X_h +*LO* denote the biexciton induced transition and the exciton +1LO-phonon polaron transition, respectively.

on the sample was 150 μm. The polarizations of the incident pulses with mutual delay time τ_{12} varied by piezoelectric actuators have been adjusted to collinear (↑↑), cocircular ($\sigma^+\sigma^+$), and cross linear (↑→) using wave plates with sufficiently wide spectral bandwidth. The first (second) symbol in the parentheses indicates the polarization of the $\mathbf{k}_1$ ($\mathbf{k}_2$) pulse, respectively. The FWM signal in the reflected $2\mathbf{k}_2-\mathbf{k}_1$ direction was time integrated and spectrally resolved by a combination of a spectrometer and an optical multichannel analyzer. For optical characterization of the sample photoluminescence (PL), photoluminescence excitation (PLE) and reflection measurements were performed at 30 K. In the reflection measurements, the reflected light from a Hg lamp that was sent nearly parallel to the QW growth direction was analyzed.

EXPERIMENTAL RESULTS

The full line in Fig. 1 shows the PL spectrum of the 3 nm ZnSe QW structure at 30 K, revealing a strong 1*s* heavy-hole exciton X_h emission at 2.889 eV. The FWHM of the emission line is ~4 meV, indicating some inhomogeneous broadening due to width fluctuations and weak interdiffusion from the barriers. The weak signal at 2.858 eV is attributed to the X_h-1LO replica corresponding to a LO-phonon energy of ~31 meV, which is close to the value of 31.7 meV known from bulk ZnSe.[11] The additional broadening of the phonon replica with FWHM ~6 meV compared to the X_h PL line and the slight variation of the LO-phonon center energy compared to the bulk value are attributed to disorder in the QW caused by the quaternary barriers. The PL spectrum further shows the energetic positions of the barrier excitons at ~3.1 eV. These energies are 80 meV higher than found in the 4 nm QW sample used in earlier TPE measurements,[9] indicating a higher S and Mg concentration and hence a stronger quantum confinement as nominally specified. The dotted-dashed curve shows the PLE spectrum of the 3 nm QW sample. The PLE signal, which was detected at the maximum of the X_h PL peak, reveals a broad (~25 meV FWHM) light-hole exciton X_l transition at 2.931 eV that is separated by ~42 meV from the X_h emission. The large X_l bandwidth indicates broadening due to X_l coupling to the X_h continuum and due to stronger penetration of the X_l wave function into the disordered barriers. Additional broadening is caused by $n \geqslant 2$ X_h transitions on the low energy side of the X_l band. These assignments are further supported by reflection measurements using white light emission from a Hg lamp (open circles) and a 30 fs laser pulse (dashed line) that was used in the FWM experiments. Only a small Stokes shift (~1 meV) of the 1*s* X_h transition is observed, somewhat smaller than the 2.4 meV expected from the X_h linewidth of 4 meV.[12]

Figure 2 shows the contour plot of the FWM measurements in (↑↑) configuration on a logarithmic scale as a function of delay τ_{12} between pulses $\mathbf{k}_1$ and $\mathbf{k}_2$ with center energy of 2.9 eV. The total intensity of the pulses was ~15.0 MW/cm^2. Also shown in Fig. 2 is the FWM spectrum at τ_{12}=100 fs on a logarithmic scale. Besides the photon echo of the inhomogeneously broadened X_h transition at 2.889 eV and the biexciton induced transition *XX*, the FWM signal reveals a spectrally broad (~12 meV at FWHM) and fast decaying signal with maximum at 2.920 eV. The FWM spectrum does not exhibit a spectrally resolved light-hole exciton signal X_l that is separated by ~42 meV from the X_h transition. The weakness of the X_l FWM signal is attributed to a high scattering rate with electron-hole (*eh*) pairs within the X_h continuum. The use of cocircular ($\sigma^+\sigma^+$) pulses in the FWM experiments suppresses the biexciton induced transition *XX* but shows the same X_h photon echo and spectrally broad feature with maximum signal energy at 2.920 eV, as summarized in Fig. 3. The unexpected signal that is separated by ~31 meV from the X_h transition is attributed to an exciton+1LO quasiparticle that is caused by a mixed coherent polaron mode.

Our assignment is based on the following arguments: As mentioned earlier, a X_h exciton binding energy of E_{1s} =33 meV was observed in a similar 4 nm ZnSe QW sample. The value was deduced from the 1*s* to 2*p* exciton

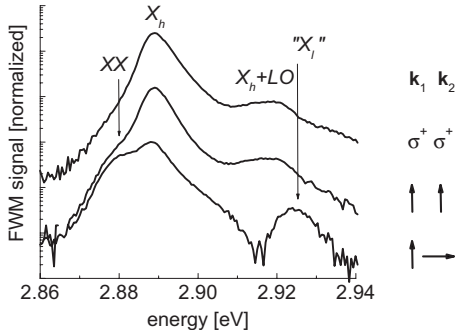

FIG. 3. Normalized FWM spectra obtained in cocircular ($\sigma^+\sigma^+$), collinear ($\uparrow\uparrow$), and cross-linear ($\uparrow\rightarrow$) configurations, as labeled, at pulse delay $\tau_{12}=0$ and pulse center energy of 2.9 eV. "X_l" denotes localized light-hole exciton transitions. The total pulse intensity is 15 MW/cm^2. The spectra are offset to each other for better comparison.

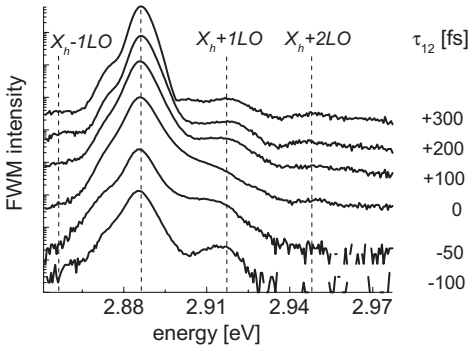

FIG. 4. Normalized FWM spectra obtained in collinear ($\uparrow\uparrow$) configuration at different delay τ_{12}, as labeled. The pulse center energy is 2.915 eV; the total pulse intensity is 35 MW/cm^2. The spectra are offset from each other for better comparison.

energy difference $\Delta_{1s\text{-}2s}=27$ meV obtained from TPE measurements.[9] Due to the smaller QW size (3 nm) and higher confinement of the sample investigated in these FWM studies, we expect an even higher X_h binding energy, namely, $E_{1s}\geqslant 33$ meV and a $1s$-$2s$ splitting energy of $\Delta_{1s\text{-}2s}\leqslant 29$ meV. The strong Fröhlich interaction with a constant of $\alpha=0.43$ (Ref. 6) and the high X_h binding energy exceeding the LO-phonon energy generates a strong coupling between the LO-phonon mode and the X_h exciton level. This strong coupling leads to the formation of a coherent mixed exciton+1LO polaron not unlike to the coherent vibronic progression of excitons in molecules[13] or in quantum dots[7] where the phonon coupling leads to sidebands in the FWM spectrum. Recent investigations have shown that stable mixed polarons can be realized in InAs quantum dots[14] where the energy splitting between electronic levels nearly matches the LO phonon energy. This stabilization process might also contribute in our QW sample where the expected $1s$-$2s$ X_h energy as well as transition energies of $n>2$ exciton states lie close to LO-phonon energy. In contrast to stable polarons, the coupling of higher order LO phonons to the X_h continuum leads to a decomposition of the exciton +1LO-phonon polaron into a "free" phonon and scattered zero-phonon exciton in our sample. In addition, the LO-phonon lifetime due to the decay into LA and TA phonons limits the stability of the exciton+1LO-phonon quasiparticle.[15–17]

Strong coupling between X_h excitons and LO phonons is further supported by FWM experiments that were performed with pulses at a center energy of 2.915 eV that cover the exciton+2LO energy. The total excitation intensity of the pulses was ~35 MW/cm^2 in this case. The resulting FWM spectra are shown in Fig. 4 at different delay times as labeled. In addition to the X_h exciton signal and mixed X_h +1LO sideband, faint structures at energies X_h+2LO and at X_h-1LO become visible for positive delay times. This observation is again attributed to the formation of coherent mixed polaron modes. It should be noted that no such LO sidebands have been observed in previous quantum kinetic studies using III-V bulk semiconductors[1,4] or QW structures.[5] Even in the case of bulk ZnSe (Ref. 6) or CdSe (Ref. 7) with a coupling constant of $\alpha\approx 0.4$, no coherent polaron satellites were observed in the FWM spectra. However, in none of those samples did the exciton binding energy exceed the LO phonon energy. The experimental result indicates that the energetic separation of the exciton +1LO-polaron mode from the exciton+0LO continuum states is important for the formation of coherent exciton-LO-phonon sidebands.

Due to non-Markovian exciton-LO-phonon scattering, clear phonon quantum beats between mixed states have been observed in all above-mentioned samples at the spectral position of the exciton transition shortly after the pulse excitation. Likewise, quantum beats are observed in our ZnSe single QW. The FWM traces at the spectral position of the X_h exciton and at the mixed polaron energies X_h+1LO and X_h +2LO of our QW sample are displayed in Fig. 5 on a logarithmic scale. Also shown in the inset are the Fourier transformed spectra at energy positions X_h and X_h+1LO, respectively. The Fourier transformed traces reveal weak (but well above noise level) signal maxima around 31 meV as expected from the FWM spectra and in agreement to studies in bulk ZnSe.[6] The fast Fourier transformed signal broadening

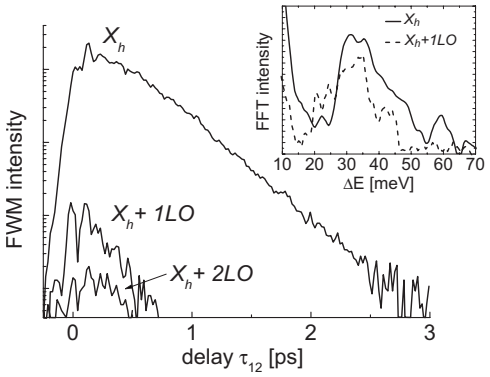

FIG. 5. FWM traces at the spectral position of the heavy-hole X_h exciton and at mixed polaron energies X_h+1LO and X_h+2LO on a logarithmic scale. The pulse center energy is 2.915 eV; the total pulse intensity is 35 MW/cm^2. The inset shows the Fourier transformed spectra of the FWM traces at positions X_h and X_h+1LO, respectively.

on the high energy side might indicate the contribution of continuum-LO-phonon coupling with a renormalized beat frequency $\bar{\omega}_{LO}=\omega_{LO}(1+m_e/m_h)$. With $m_e=0.145m_0$ and $m_h=0.8m_0$,[18] the renormalized frequency amounts to an energy of $\hbar\bar{\omega}_{LO}\approx 37$ meV. Continuum-LO-phonon beats have been observed in GaAs bulk material[1,4] with a weak coupling constant $\alpha=0.06$ and indirectly in CdSe bulk samples where beat frequencies between the bulk and the renormalized value have been found.[7] Frequencies corresponding to energies larger than ~40 meV may originate from weak heavy-hole–light-hole X_h-X_l exciton beats. The contribution of the $2s$ exciton to the X_h+1LO polaron signal is estimated according relation $I_{FWM}\propto o_{1s}^2\cdot o_{2s}^2$, where o_{1s} and o_{2s} are the oscillator strength of the $1s$ and $2s$ excitons, respectively. Due to the small ratio of $1s/2s$ oscillator strength, that is, $o_{1s}/o_{2s}=1/8$ for bulk material and $o_{1s}/o_{2s}=1/27$ in pure two dimensional QWs, we expect a $2s$ X_h FWM signal more than 100 times lower than the $1s$ signal at $\tau_{12}=0$ in our QW sample. Accordingly, we anticipate that the X_h+1LO polaron signal exceeds the $2s$ exciton signal but $2s$ transitions may contribute to the broadening on the low energy side of the X_h+1LO signal (see Figs. 1–4).

The photon echo (PE) at position X_h reveals an exciton dephasing time of $T_{X_h}=1.3$ ps, which is predominantly limited by exciton-carrier scattering at applied high excitation densities. The FWM traces at the energy position X_h+1LO and X_h+2LO also show the character of a PE but reveal faster dephasing times of $T_{X_h+1LO}\approx 700$ fs and $T_{X_h+2LO}\approx 900$ fs. The enhanced dephasing of the exciton+1LO quasiparticle is attributed to the decay of the coherent polaron into a $1s$ exciton state and free LO phonons. Due to the vicinity of the $2s$ transition energy to the 1LO-phonon energy, a rapid decay of the X_h+1LO superposition state is expected. This nearly resonant condition is not fulfilled for the X_h+2LO quasiparticle, which might explain the slightly longer coherence time for this polaron mode. Also, the inhomogeneous broadening of LO-phonon energies due to disorder and their dispersion in **k** space as well as the decay of the LO phonon into acoustic and other phonons may contribute to the faster decay of the superposition states compared to the X_h transition. While the later case is probably of minor importance since most of the LO-phonon lifetimes in III-V (Refs. 14 and 15) or II-VI materials (Ref. 16) exceed several picoseconds, inhomogeneous broadening of LO phonons may affect the dephasing of superposition states due to destructive interference of LO-phonon wave packets involved.[19]

Figure 6 shows the contour plot of the FWM measurements for cross-linear (↑→) fields on a logarithmic scale as a function of delay τ_{12} between pulses $\mathbf{k}_1$ and $\mathbf{k}_2$ with center energy of 2.9 eV. The total intensity of the pulses was 15 MW/cm^2. Also shown in Fig. 6 is the FWM spectrum at $\tau_{12}=15$ fs on a logarithmic scale. As expected, the biexciton induced transition XX gains in importance due to the lack of excitation induced dephasing in this configuration.[20] The X_h FWM signal is reduced by a factor of ~8 compared to (↑↑) fields. Also, the X_h+1LO mixed polaron sideband is strongly reduced while a new band with maximum at 2.925 eV appears, which is shifted by ~5 meV with respect to the X_h

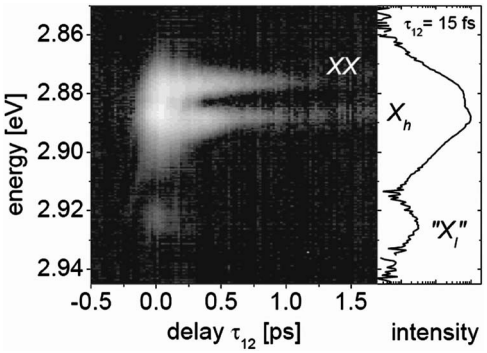

FIG. 6. Contour plot (logarithmic scale) of the FWM signal obtained in cross-linear (↑→) configuration as a function of delay τ_{12} between pulses $\mathbf{k}_1$ and $\mathbf{k}_2$ with center energy of 2.9 eV. "X_l" denotes localized light-hole exciton transitions. The total pulse intensity is 15 MW/cm^2. Also shown is the FWM spectrum at $\tau_{12}=15$ fs on a logarithmic scale.

+1LO maximum observed in (↑↑) configuration (see Fig. 3). This band is attributed to localized light-hole exciton transitions—denoted as "X_l" in what follows—that lie energetically below the X_h continuum and are therefore less affected by rapid scattering processes with *eh* pairs. This interpretation suggests a $1s$ X_h binding energy of $E_{1s}\approx 36$ meV and a $1s$-$2s$ splitting energy of $\Delta_{1s\text{-}2s}\approx 29$ meV using a fractional dimension approach.[21] These energies are close to values that we expect from the QW structure. Additional FWM measurements in (↑→) configurations with pulse center energy of 2.915 eV show the same dynamical behavior. In particular, no 1LO or 2LO sidebands are visible in the FWM spectrum.

Figure 7 shows the FWM traces in (↑→) configuration at the spectral position of the X_h and light-hole related "X_l" transition. As in the collinear (↑↑) configuration, the inhomogeneously broadened transition states show a PE decay. The X_h dephasing time results to $T_{X_h}\approx 1$ ps, which is slightly longer as the "X_l" dephasing that is determined to be $T_{\text{"}X_l\text{"}}\approx 800$ fs. The shorter dephasing time of the X_h transition in (↑→) compared to (↑↑) configuration is attributed to an ad-

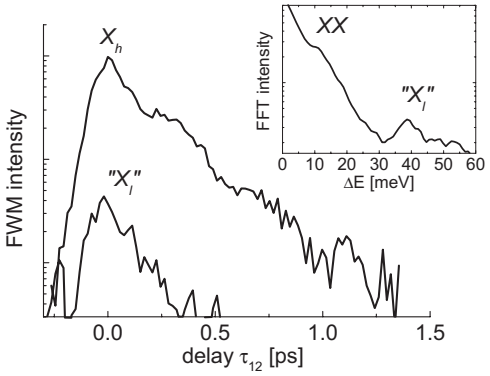

FIG. 7. FWM traces at the spectral position of the X_h exciton and at the light-hole related "X_l" transition on a logarithmic scale. The pulse center energy is 2.9 eV; the total pulse intensity is 15 MW/cm^2. The inset shows the Fourier transformed spectrum of the FWM trace at position X_h.

ditional decay that is caused by the inhomogeneous broadening of bound and unbound biexciton states.[22] The Fourier transformed signal at the X_h energy shown in the inset of Fig. 7 reveals a frequency of 11 meV due to a beating between biexciton induced XX and heavy-hole X_h transitions as well as a broad band with center frequency at ~39 meV, which is predominantly attributed to the X_h-"X_l" beating.

SUMMARY

In conclusion, we have observed coherent mixed exciton-LO-phonon polarons in a 3 nm ZnSe single QW in which the exciton binding energy exceeds the LO-phonon energy. The formation of these superposition states is assumed to be resonantly enhanced by the vicinity of 2*s* exciton states. The new quasiparticles decay faster than the zero-phonon X_h transition, which is attributed to an effective disintegration of the polaron into the 1*s* exciton state and into free LO phonons as well as to inhomogeneous broadening of LO-phonon energies, leading to a fast destructive interference of LO-phonon wave packets that couple to the X_h excitons. Further studies using ZnSe QWs that comprise 2 up to 6 nm wide wells shall provide more information about the importance of the exciton binding energy on the occurrence of exciton-LO-phonon sidebands in the FWM spectrum. Likewise, the influence of 2*s* exciton states on the formation of exciton-LO-phonon polarons will be subject of further studies.

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