Femtosecond coherent polariton dynamics in the layered III-VI semiconductor InSe

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We report on the observation of the time-resolved free-induction decay (FID) in a layered III-VI semiconductor. The coherent propagation of the 1*s*-exciton polariton in InSe leads to severe distortions of the transmitted laser pulse shape and aperiodic modulation of the FID on a femtosecond time scale. This coherent polariton dynamic becomes operative even in optical thin samples. The characteristic dependence of the propagation beats as a function of sample thickness and excitation energy is demonstrated. The major features of the polariton dynamics are well described by coupled Maxwell and semiconductor-Bloch equations for linear pulse propagation. The scattering of polaritons by carriers and phonons is studied at different excitation densities and lattice temperatures. [S0163-1829(97)01207-1]

I. INTRODUCTION

Transient spectroscopy with femtosecond time resolution is a powerful method to study the ultrafast coherent dynamics of electronic excitations in bulk semiconductors and semiconductor heterostructures. The dynamics at the semiconductor band edge observed with ultrashort laser pulses is dominated by excitonic effects.[1,2] This is due to the strong enhancement of excitonic resonances and the fast dephasing of the continuum states.[3] Excitonic dynamics are even more pronounced in systems of low-dimensional structure such as two-dimensional (2D) quantum-well systems.[4,5] This is also the case in some naturally layered bulk semiconductors[6] such as those of the III-VI compounds.[7,8] The large optical nonlinearities of these materials can be used for possible future applications in optical switching.[9]

The coherent excitation of an optical interband transition with an ultrashort laser pulse is followed by the subsequent reradiation of the induced polarization, known as free-induction decay (FID).[10] The coherent dynamics and the loss of coherence of the material excitation due to scattering can be temporally resolved if the duration of the applied laser pulse is shorter than the dephasing time T_2 of the system.[2] Moreover, a coherent superposition of two closely spaced interband transitions can be excited if the laser spectrum is broader than their splitting $\Delta\nu$. In this case the coherence of the induced polarizations is directly reflected by the subsequent beating of the excited transitions.[11]

Beat phenomena in semiconductors of various physical origin have been investigated with several measurement techniques in the past few years.[4,5,11] A particular case of quantum beats results from the *coherent real-space dynamics of polariton wave packets*[12,13] compared to the usual *quantum beats of optical transitions*.[14,15] The explanation of real-space dynamics of wave packets in general has to take into account the dispersion of the optical excitation and cannot be understood within simple few level models.[11,16,17] In this paper we present a comprehensive study of *coherent propagation of exciton polaritons in bulk material* by time-resolved FID. Polaritons from the upper (UPB) and the lower (LPB) polariton branch of the polariton dispersion in InSe are excited by a laser pulse, whose spectrum is broader than the longitudinal-transversal (LT) split ω_{LT} of the polariton dispersion. As the laser pulse duration is shorter than the dephasing time T_2 of the exciton polariton, the coherent propagation dynamic of the polariton is resolved. The transmitted pulse form is severely distorted and aperiodic beats, known as polariton beats,[12,13,17–19] stem from the drastic changes in the polariton dispersion near the exciton resonance.

The investigation of coherent propagation effects is of interest for several reasons. As polariton formation is, in principle, connected with every optical excitation of an isolated material resonance,[20–22] propagation effects are a rather common and manifold phenomenon in semiconductors.[23] Investigating the FID gives valuable information about the spectral properties of a semiconductor for optical excitation and the coupling strength of the excitonic material polarization and the exciting light field.[12] In semiconductor heterostructures, in which translation symmetry is perturbed, the transmitted pulse shapes are sensitive to the detailed geometry of the system as well.[24] Most of all propagation effects may have significant influence on the detected signal shape even for nonlinear time-integrated experiments.[25,26] Never-

theless, the interpretation of experiments mostly neglects the effects of spatial dispersion and propagation, arguing with the use of optical thin samples. However, the critical investigation of propagation effects is most clearly done by studying the time-resolved FID of a system.[18]

Previous FID experiments were performed on materials with an extremely small LT split (some μeV in Cu_2O) compared to their large exciton binding energy (more than 50 meV).[12,13] The T_2 times are large, and rather slow polariton beats on a time scale of several hundreds of picoseconds are observed in thick bulk samples (several hundreds of micrometers in Cu_2O and ZnP_2).

In this paper we demonstrate ultrafast coherent dynamics of exciton polaritons in a bulk semiconductor. The coherent propagation effects become relevant even for optical thin samples and are resolved on a femtosecond time scale. Ultrafast polariton dynamics have been investigated previously by time-resolved studies of FID only in 2D $GaAs/Al_xGa_{1-x}As$ quantum wells (QW's).[17,18] Here we study the dynamics of the $1s$-exciton polariton in the layered III-VI bulk semiconductor InSe, which to our knowledge has not been examined previously. The particularity of the aperiodic polariton beats is demonstrated by studying in detail their dependence on sample thickness and excitation energy. The experimental data are quantitatively analyzed within the polariton concept by using the coupled Maxwell and semiconductor-Bloch-equations (MSBE's) for linear pulse propagation. This yields the dispersion of the $1s$-exciton polariton. Furthermore, we investigate the influence of increased scattering of carriers and phonons on the coherent propagation of the $1s$-exciton polariton by varying excitation density and sample temperature. We discuss the limitations of linear theory and of the introduction of phenomenological damping constants.

The paper is organized in the following way. In Sec. II we give a summary of the theoretical approach. In Sec. III we describe the material system under investigation and the measurement setup for femtosecond time resolution of the FID. In Sec. IV we present a comprehensive study of polariton dynamics in InSe. Experimental data on different sample thicknesses and under various excitation conditions are given and discussed. Conclusions are made in Sec. V.

II. LINEAR PULSE PROPAGATION AND POLARITON BEATS

The coupled propagation dynamics of the exciting laser light field and the reradiated coherent polarization in a semiconductor can be described by simultaneous solution of the MSBE's. Exciton polaritons are excited at the front of the sample by the short laser pulse, subsequently propagate through the crystal, and are transformed into light again at the rear of the sample. This pulse propagation dynamics in a strongly dispersive medium is properly described by the Fourier-transform method.[19,12] The time evolution of the transmitted electric field $E_S(t)$ at the end of the sample is given by

$$E_S(t)=\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}e^{-i\omega t}\widetilde{E}_S(\omega),$$

$$\widetilde{E}_S(\omega)=E_0(\omega)\sum_{j=1}^{2}a_j(\omega)e^{ik_j(\omega)d}.$$

$E_0(\omega)$ is the Gaussian shape of the incoming pulse. The spectral weights $a_j(\omega)$ $(j=1,2)$ are assigned to the occupation of the lightlike portion of the polariton in the LPB and the UPB. From microscopic theory in real space these can be derived from the boundary conditions for the exciton envelope at the borders of the sample.[27,28] The propagation of the polariton is determined by its k vector $k(\omega)$ and is strongly dispersive near resonance. As the electric light field at the rear of the sample with thickness d is a superposition of the light fields from the LPB and the UPB, the detected intensity is critically dependent on the phase factors $e^{ik_j(\omega)d}$ $(j=1,2)$. The interference of polaritons from the LPB and the UPB at the end of the sample results in characteristic beat phenomena if the polaritons phase is preserved during propagation. The temporal resolution of the experiment is given by the pulse shape $E_P(t-\tau)$ of the laser pulses, as the emitted electric field $E_S(t)$ is upconverted, and the second harmonic is detected in a photomultiplier

$$S(\tau)=\left|\int_{-\infty}^{+\infty}dt\,E_P(t-\tau)E_S(t)\right|^2.$$

The main features of the detected signal $S(\tau)$ are sensitively dependent on the intrinsic material properties. The sample acts like a spectral filter whose shape is determined by the exciton polariton dispersion $\omega(k)$. It is defined by the Fourier transform of the wave equation and the linear SBE (Ref. 1)

$$\left(\frac{k}{\omega}\right)^2-1=\chi(\omega)\quad\text{with}\quad\chi(\omega)=\frac{A}{1+Bk^2-\omega-i\Gamma}.$$

For simplicity ω, Γ, and k are normalized, respectively, to the resonance frequency of the $1s$ exciton in InSe, $\hbar\omega_0=1.331$ eV, and the corresponding k vector of light in matter, $k_0=\omega_0 n/c=2.06\times10^7$ m^{-1}, with refractive index $n=3.05$. The linear susceptibility $\chi(\omega)$ describes the spectral dependence of the material response. Γ is introduced as a phenomenological damping constant with equal value for all k vectors. A and B are related to microscopic material parameters derived from density-matrix theory:[1]

$$A=\frac{\omega_{\mathrm{LT}}}{\omega_0}=\frac{1}{\omega_0}\frac{(m_0\Phi_0)^2}{\hbar\epsilon_0\epsilon_b},\qquad B=\frac{k_0^2}{\omega_0}\frac{\hbar}{2M}.$$

The LT split $\omega_{\mathrm{LT}}=\omega_{\mathrm{UPB}}(k=0)-\omega_0$ is a measure for the coupling strength of the light field and the material polarization at the exciton resonance. It is connected with the microscopic parameters of the interband transition dipole m_0 and as well with the amplitude of the exciton envelope function Φ_0. ϵ_0 and ϵ_b stand for the vacuum permittivity and the dielectric background constant, respectively. ω_{LT} determines the splitting Δ_0 of the LPB and the UPB at resonance (ω_0,k_0) by $\Delta_0=\sqrt{2A}\omega_0$.[29] The dispersion parameter B accounts for the center-of-gravity motion of the exciton. Its value is determined by the exciton center of gravity mass

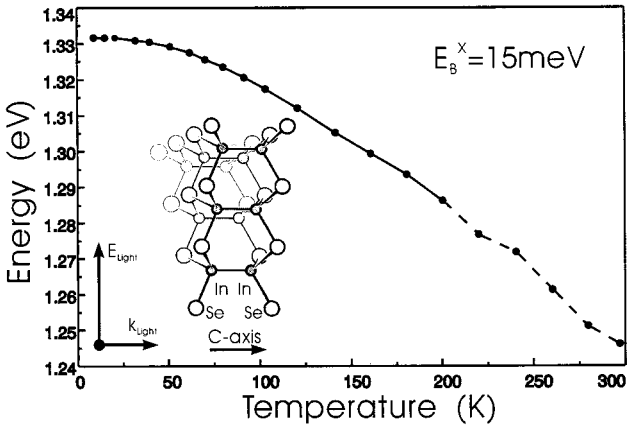

FIG. 1. Spectral position of the excitonic band gap (exciton binding energy $E_B^X = 15$ meV) in InSe determined by linear absorption measurements, as a function of temperature. Above $T = 180$ K the excitonic absorption peak vanishes due to thermal dissociation of the exciton. The observed onset of the continuum absorption is depicted as a dashed line above $T = 180$ K. The inset shows the layer structure of InSe. The layers are oriented perpendicular to the crystal's **c** axis. As indicated, this direction coincides with the direction of polariton propagation in the experiment.

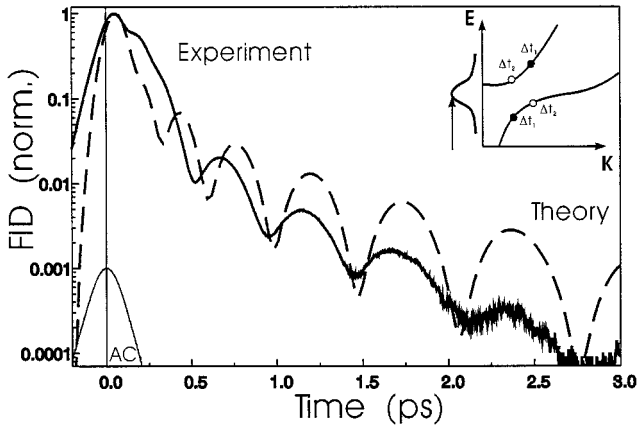

FIG. 2. FID signal at the $1s$-exciton resonance (at a temperature of 10 K) in InSe shown as a solid line and the autocorrelation trace (AC) as a reference. The transient is taken at a low excitation density of 10^{16} cm^{-3}. The strong modulation of the decaying FID signal is clearly aperiodic and the duration of the beats increases with increasing time delay. The theoretical model of the data is plotted as a dashed line. The graph in the inset illustrates the excitation and the propagation dynamics of the exciton polariton.

$M = 1.68 m_e$ along the InSe crystal's **c** axis[30] to $B = 7.2 \times 10^{-6}$. A and Γ are the only fit parameters in this model.

The original polariton picture is adequate for small damping.[27] In this case the near resonance splitting of the polariton branches, determined by Re$[k(\omega)]$, is only rarely affected by the value of Γ. Due to the polariton's mixed character of light and exciton polarization, the values of the occupied, near resonance polariton k vectors can greatly exceed those of light k vectors of the same frequency. The resonant polariton components propagate with a strongly reduced group velocity, as can be seen from the derivation of the polariton dispersion $v_g = \partial\omega/\partial k$. Moreover, the extinction of the propagating polariton frequencies Im$[k(\omega)]$ is small for the lightlike part of the polariton, but drastically enhanced for resonant polariton frequencies. In the measurement, therefore, a high contrast ratio is required to observe these near-resonance polariton components.

III. EXPERIMENT AND MATERIAL SYSTEM

InSe is a III-VI class direct-band-gap semiconductor[31,32] with a layered crystal structure and therefore represents a naturally grown quasi-two-dimensional system.[8] The layer structure of InSe is depicted in Fig. 1. The intralayer bonding of the Se-In-In-Se atoms is of covalent character, but the layers themselves are bound only by weak van der Waals forces. The crystal therefore exhibits various molecularlike properties,[30] e.g., a rich spectrum of phonon modes and strong anisotropic properties of the phonon and electron system. InSe is an outstanding candidate for investigations of excitonic properties on an ultrashort time scale because of its large exciton binding energy ($E_B^X = 15$ meV) compared to the excitonic gap ($\hbar\omega_0 = 1.331$ eV). In linear absorption measurements we observe the resonance of the $1s$ exciton up to temperatures of 180 K (Fig. 1). Above 80–100 K the homogeneous linewidth of the exciton line is effectively broadened due to the efficient occupation of optical phonons.[7] Above 200 K the exciton is thermally dissociated. We investigate several thin, high-quality β-InSe crystals grown by the Bridgman method.[33] By carefully cleaving the samples perpendicular to the crystal's **c** axis, thin slabs with optical flat surfaces and thicknesses of 1–6 μm are prepared. The slabs are strain-free attached to a sapphire substrate and mounted in a closed-cycle cryostat.

The FID experiments are performed with 100-fs pulses. The pulses are derived from a Kerr-lens mode-locked Ti:sapphire laser, with a repetition rate of 76 MHz and tunable from 900 nm to 1000 nm. One weak laser pulse is transmitted through the sample and a second variable delayed pulse is used for up-converting the transmitted pulse in a nonlinear crystal (β-barium borate). This yields a time resolution of the FID signal given by the duration of the laser pulse. The sum frequency signal is detected with a GaAs photomultiplier. The pulse intensity is adjusted for low excitation densities of 10^{16} cm^{-3}. A third laser pulse can be used to excite additional carrier densities up to 10^{18} cm^{-3}.

IV. RESULTS AND DISCUSSION

A. Coherent polariton propagation in FID

Figure 2 depicts the normalized FID signal for resonant excitation of the $1s$-exciton polariton in InSe on a logarithmic scale. The experimental data are plotted as a solid line with the autocorrelation trace as a reference. The time shift Δt of the FID signals maximum is defined by the thickness $d = \Delta t(c/n)$ of the sample. The rise time of the signal is limited by time resolution, whereas its decay exceeds the pulse length by at least 3 ps and is resolved over at least four orders of magnitude. This behavior reflects the reemitted coherent polarization induced by the exciting laser pulse. The strong aperiodic modulation of the FID is due to the coherent propagation of the $1s$-exciton polariton through the InSe

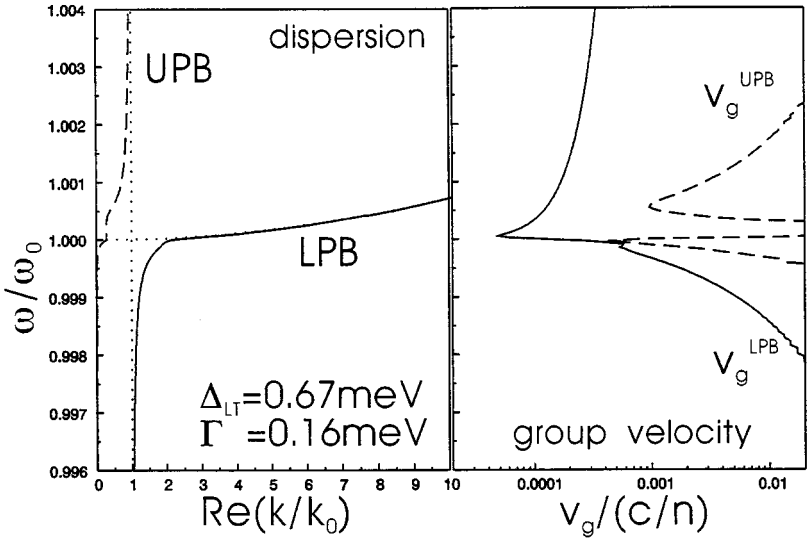

FIG. 3. Dispersion of the $1s$-exciton polariton in InSe and corresponding group velocity as determined by fitting the FID transients. The LT split ω_{LT} and the phenomenological damping constant Γ are the only fit parameters in this model.

sample along the crystal's **c** axis. The interference of the polaritons from the LPB and the UPB at the rear of the sample results in the characteristic dependence of the polariton beats on time delay. The duration of the polariton beats is short at early time delays and increases gradually with increasing time delay. The resolution of the fast beats at early time delays can be improved by using shorter laser pulses.

The time evolution of the measured signal can be understood in a simple picture[19,12] as illustrated in the inset of Fig. 2. The exciton-polariton dispersion and the excitation on the $1s$-exciton resonance are shown. The broad spectrum of the laser pulse covers the LT split of the polariton dispersion and polariton wave packets are excited at the front of the sample, which then propagate along the InSe crystal's **c** axis. The propagation is strongly dispersive near resonance and the group velocity v_g is strongly reduced. At the rear of the sample only polaritons can interfere, which arrive there at the same time, and therefore these must have the same group velocity. However, polaritons of the same group velocity from the LPB and the UPB have slightly different frequency. The inserted points mark the different spectral positions of two pairs of polaritons with equal group velocity of the polaritons from the LPB and the UPB, respectively. Those labeled with Δt_1 have a larger group velocity and therefore arrive at earlier times than those labeled with Δt_2. According to the polariton dispersion, the early arriving polaritons have larger energy splittings than those arriving at later times. As a consequence, the observed characteristic beating behavior reflects the energy splitting of these polaritons and the oscillation period of the polariton beats increases in time.

This behavior is quantitatively described by theory (dashed line in Fig. 2). The model yields a fit of the $1s$-exciton polariton dispersion with the parameters for the LT split $\omega_{LT}=0.67$ meV and the damping constant $\Gamma=0.16$ meV. All other parameters for exciton mass, sample thickness, spectral width, and tuning of the laser are known from experiment and the literature. The value of ω_{LT} determines the resonant splitting to $\Delta_0=42$ meV. The corresponding time period agrees well with the observed fastest beats at early time delays.

The real part of the exciton polariton dispersion $\mathrm{Re}[k(\omega)]$ and the corresponding frequency-dependent group velocity $v_g(\omega)$ of the polariton are shown in Fig. 3. The imaginary part of $k(\omega)$ (not shown here) describes the resonant-enhanced extinction of the polarization, as the polariton propagates through the sample. However, damping affects the real part of the dispersion as well. As a result, the true LT split is reduced slightly and the UPB develops even below ω_0. The additional low-frequency dip of the group velocity as well reflects this behavior. However, these polariton frequencies are not observed in the experiment, as this part of the UPB is strongly damped.

Linear theory within the polariton concept fits the major features of the signal well. Multiple reflection on the sample boundaries have no influence on the FID profiles. The relative contribution compared to the FID signal is much too small, as calculated from the reflection and absorption coefficients of InSe. Moreover, the transit times of a sequence of multiple reflected laser pulses are clearly too short in the samples under investigation to be temporally resolved. Besides this, one would expect a periodic modulation of the FID from multiple reflected laser pulses, which is clearly not observed in the experiment. The slight deviations of experiment and theory are mainly connected with the introduction of a phenomenological damping constant. First, the amplitude of the modulation due to the polariton beats diminishes faster in the experiment than proposed by the theory. As this can be accounted for by introducing two separate damping constants for the LPB and the UPB, this indicates the existence of different scattering channels for the two branches. Second, for early time delays the FID signal decays faster than for later time delays. This is not reproduced by theory. In addition, one observes slight deviations of the measured beat period at early time delays in experiment from those proposed by theory. We attribute these deviations mainly to the increased polariton scattering at early time delays and the disturbance of the $1s$-exciton polariton dispersion by energetically higher polariton resonances. The excitation of higher resonances and continuum states, though of minor oscillator strength, is possible because the spectral width of the laser pulse is in the range of the exciton binding energy.

To model the decay of the observed FID more accurately, one has to take into account the dispersion of energetically higher polariton states and the increased scattering rates of the energetically higher $1s$-exciton polariton frequencies. In general one must consider the damping constant as a function of energy, i.e., k vector.[20] However to model the functional quantity of $\Gamma(k)$, a microscopic theory of the scattering processes should be applied.[34]

The excitons finite center of gravity mass $M=1.68m_e$ along the InSe crystal's **c** direction clearly affects the time evolution of the FID and cannot be neglected to model the data. However, the kinetic energy of the polariton at resonance, $(\hbar k_0)^2/2M=0.01$ meV, is small compared to the LT split $\omega_{\mathrm{LT}}=0.67$ meV. Because the large k vectors of the LPB have small group velocities, the effect of the exciton dispersion in the FID is observable only on a long time scale ($\sim$40 ps). In the experiment the dephasing of the signal is too fast to see the very small group velocities. One can estimate the lower limit of v_g observed in the experiment from the duration of the slow polariton beats in the FID to approximately $5\times10^{-3}c/n$.

The LT split determined here from the analysis of the FID experiments for the uniaxial layered semiconductor InSe can be compared with values of ω_{LT} known from cw measurements in cubic semiconductors. The observed value of ω_{LT} for InSe is somewhat smaller than the LT split known, e.g., for the polariton of the A-exciton resonance in CdSe ($\omega_{\mathrm{LT}}=0.9$ meV), which has nearly the same exciton binding energy as InSe.[35] However, CdSe has a higher absorption coefficient than InSe, and our finding of ω_{LT} in InSe appears to be overestimated compared to the value of ω_{LT} one would expect from a linear relationship between LT split and excitonic absorption coefficient. We assign the rather large LT split in InSe to a partly 2D character of the exciton in this layered material and the corresponding enhancement of the exciton envelope function Φ_0. Nevertheless, there is some uncertainty about this. Due to the strong extinction of the polariton frequencies near resonance only the larger frequency splittings with moderate damping show up in the FID. Therefore, the theoretically fitted LT split results from an extrapolation of the polariton frequencies observed in the experiment to frequencies closer to resonance according to the theoretical model for the dispersion. In addition, there may be some slight deviation in the polariton dispersion due to a small uncertainty in the damping constant Γ. From the FID experiment the dephasing time cannot be assigned to either homogeneous or inhomogeneous broadening of the exciton resonance. Here the fitted phenomenological damping constant is taken to be homogeneous.

B. Dependence of polariton beats on sample thickness and excitation energy

The time evolution of the polariton beats depends not solely on the LT split of the polariton dispersion, but also on sample thickness. This is a remarkable distinction to usual quantum beats of optical transitions. With the coherent propagation of the polariton, its spatial phase factor $e^{ik(\omega)x}$ evolves according to its dispersion relation and the interference at the rear of the sample at $x=d$ is dependent on the quantity of d as well. We demonstrate this dependence of the

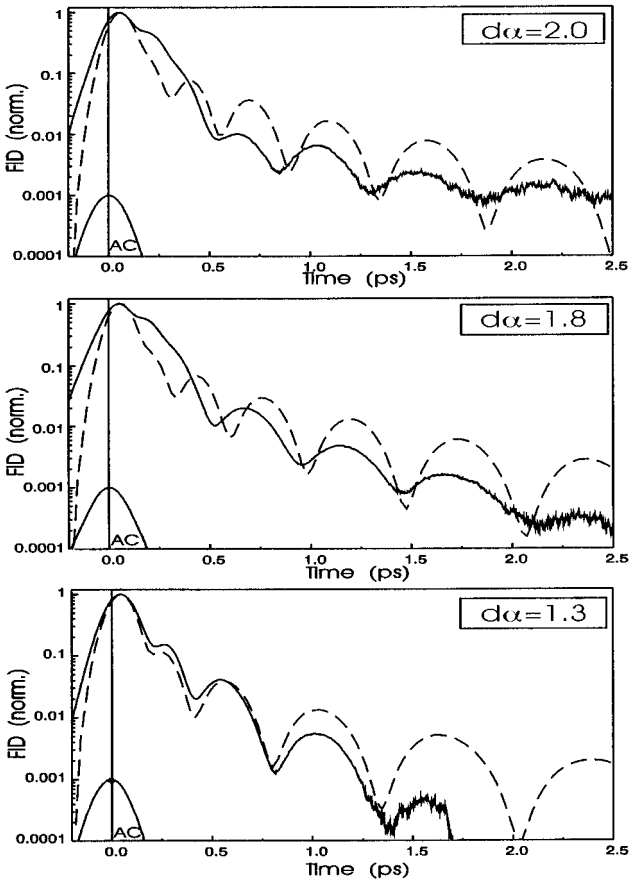

FIG. 4. Time transients of the FID taken on samples (at $T=10$ K) with thicknesses between one and two absorption lengths. The dashed lines show the theoretical calculation for each thickness with equal parameters for the polariton dispersion.

polariton beats on sample thickness by FID measurements on several samples with thicknesses between 1 and 6 μm. Figure 4 shows time transients for the FID taken for three sample thicknesses of $1.3\alpha^{-1}$, $1.8\alpha^{-1}$, and $2.0\alpha^{-1}$. The absorption length in InSe is $\alpha^{-1}=2.5$ μm. The results are well described by theory for the corresponding thickness with equal parameters for the dispersion of the polariton. Clearly, a slowing down of the polariton beats with decreasing sample thickness is observed, which can be understood again if one considers the strong spectral dependence of the group velocities on resonance. At a given fixed time delay, in a thicker sample those polaritons with a corresponding larger group velocity interfere and in a thinner sample those with a corresponding decreased group velocity interfere. As lower group velocities are connected with smaller energy splittings of the interfering polaritons, the oscillations in thinner samples are slower than in thicker samples. Although with decreasing sample thickness the pulse distortions are diminished, it is important to note that even at thicknesses below one absorption length we find the FID affected by coherent propagation effects. We conclude that the polariton propagation can be neglected only at very thin samples with thicknesses well below one absorption length. Similar results have been reported for 2D GaAs/Al_xGa_{1-x}As QW's.[17,18] These findings are even more important for resonant excitation and increasing coupling strength of the material resonance and the light field.

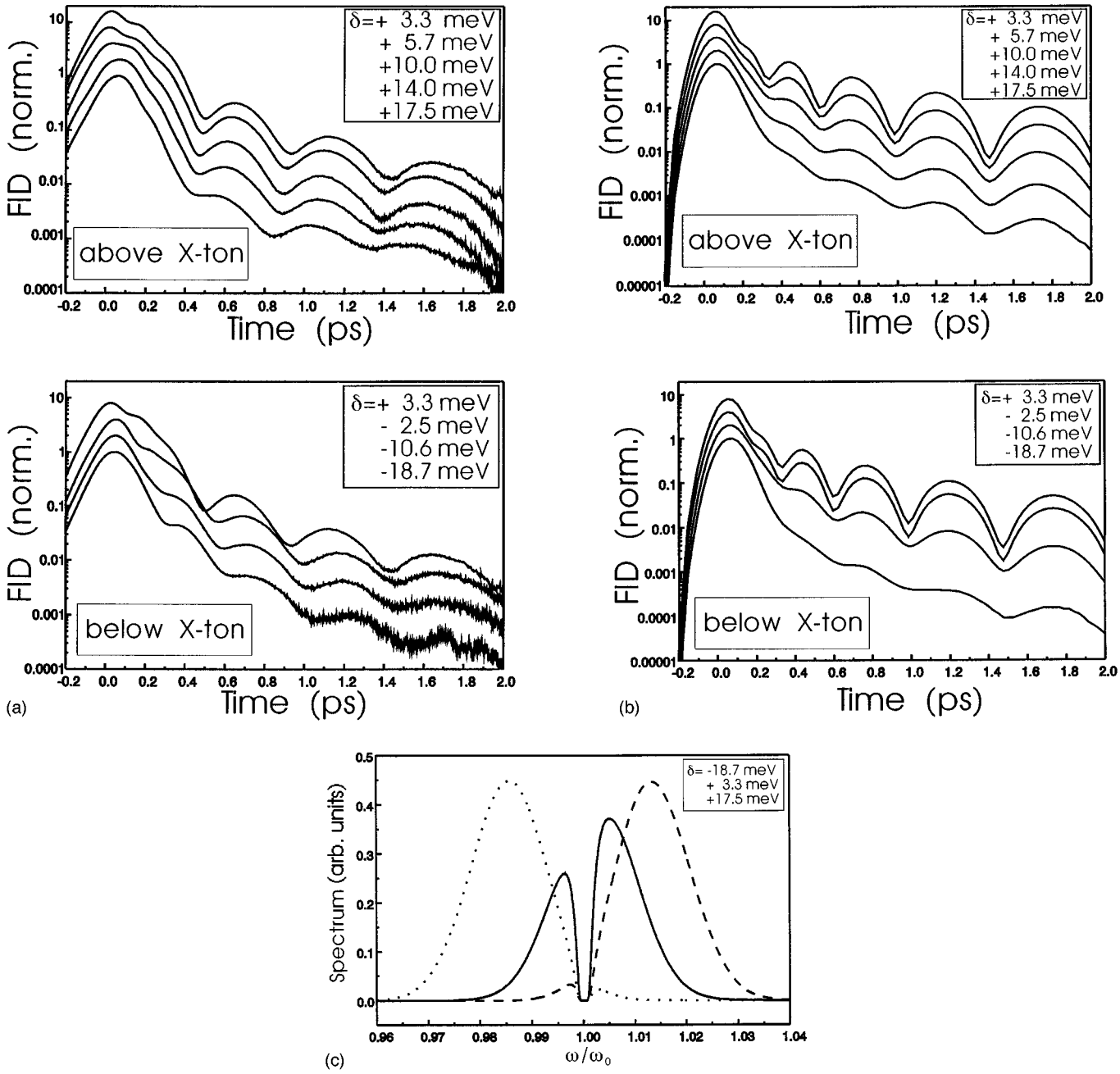

FIG. 5. FID transients for several detunings of the laser above (positive) and below (negative) the 1s exciton in InSe (at $T=10$ K). The quantity of the detuning of the laser is listed in the plots and corresponds to the transients from top to bottom. The data are normalized and shifted for clarity. (a) Experimental data. (b) Theoretical calculation. (c) Three calculated spectra exemplifying resonant (solid line) and off-resonant (dashed and dotted lines) excitation of the 1s-exciton polariton.

The spectral dependence of the polariton beats in the FID is measured by detuning the laser from the 1s-exciton resonance. The normalized experimental data [Fig. 5(a)] and theoretical predictions [Fig. 5(b)] for detuning of the laser within roughly one spectral width below (negative) and above (positive) the 1s-exciton resonance are depicted. For positive and negative detuning the amplitude of the FID signal and the modulation depth due to the polariton beats decrease. Moreover, the fast oscillations at early time delays disappear. All observations are explained well by theory. One can understand these findings from the transmitted pulse spectrum at the rear of the sample. Figure 5(c) shows three calculated spectra for resonant tuning and large detuning of the laser respectively. If the laser is tuned to resonance, the transmitted spectrum is strongly modified and split into two parts. This is due to the strong extinction of the near-resonance polaritons. The time evolution of the detected propagation beats is sensitive to the form of the transmitted spectrum and the spectral gap separating the two parts. However, with increased detuning of the laser, the occupation of the polariton frequencies near resonance, and thus the distortion of the transmitted spectrum, becomes smaller. There-

fore, the FID amplitude and the modulation of the FID signal are diminished. Moreover, with gradually increased detuning of the laser, faster propagating polaritons are excited on one side of the resonance, which do not have a corresponding counterpart of the same group velocity on the other side of the resonance. Therefore, there is no interference from these polaritons, which results in the observed vanishing of the fast polariton beats at early times.

The features described are predicted from theory to be roughly the same for positive and negative detunings of the laser [Fig. 5(b)]. However, in the experiment this is not exactly verified [Fig. 5(a)]. For positive detuning the polariton beats undergo a slight additional phase shift. We assign this effect to the modification of the 1*s*-exciton polariton dispersion due to higher exciton polaritons and continuum states. For large positive detuning of the laser these are increasingly excited, but as they are not included in the calculation of the dispersion, the model of the data does not account for the observed phase shift.

C. Scattering of coherent propagating polaritons

To study the effect of increased scattering on the coherent polariton propagation, we performed FID experiments at increased excitation densities and at increased lattice temperatures. As already mentioned the influence of additionally excited electronic states shows up in the FID at early time delays. One observes a faster initial decay of the FID signal and a slight deviation of the observed beat period from the one expected solely due to the 1*s*-exciton polariton. This behavior cannot be accounted for perfectly by linear theory for one exciton resonance and one phenomenological damping constant.

We varied the intensity of the propagating laser pulse within two orders of magnitude and applied an additional strong pump pulse at negative time delay as well. Both experiments result in a reduction of the FID and a loss of the modulation. This demonstrates that additionally excited carriers reduce the coherent propagation of the polaritons due to scattering. However, the density range examined is limited and only a slight increase of the dephasing rate of the signal can be observed within this range.

The investigation of the FID signal in a temperature range between 10 and 300 K shows the gradually increasing influence of phonon scattering on the FID signal. Figure 6 shows the time transients for temperatures between 10 and 160 K. The laser is adjusted to the temperature shift of the 1*s*-exciton resonance at the corresponding temperature. The transients for 10, 80, and 160 K are labeled separately, as they show the significant changes in the FID very clearly.

The amplitude, modulation, and relaxation time of the FID signal are steadily reduced with increasing temperature. Above 180 K the transmitted pulse shape is nearly autocorrelationlike. Specifically, the dephasing rate reflects the increase of the homogeneous linewidth of the 1*s*-exciton resonance: below 40 K the reduction of the FID is rather weak. This is because at low temperatures the decay of the FID signal is mainly due to crystal imperfections and static disorder. The comparatively small increase of the dephasing rate of the FID signal with rising temperature is attributed to the increasing scattering from acoustic phonons. The slow

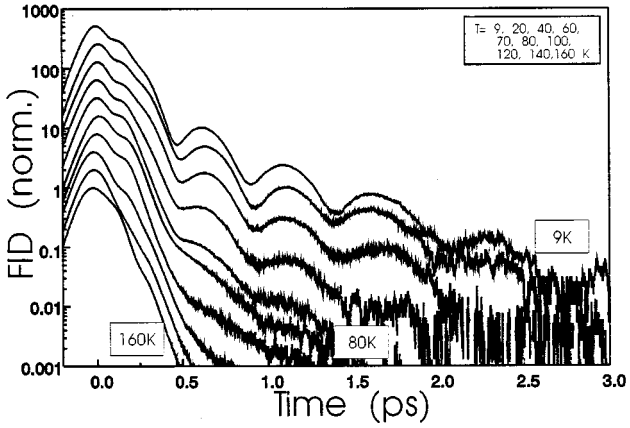

FIG. 6. FID measured at several lattice temperatures as shown in the inset. The listed temperatures correspond to the transients from top to bottom. The data are normalized and shifted for clarity.

polariton beats at later time delays of the FID are diminished already at low temperature because they are due to the small frequency splittings of the polariton branches. The fast polariton beats at early time delays remain more stable with increasing temperature, as they result from the large frequency splittings of the polariton branches. This is most clearly seen from the FID transient at 80 K, where the slow polariton beats are almost suppressed. In addition, this temperature coincides with the onset of the effective broadening of the 1*s*-exciton resonance due to the efficient occupation of optical phonons in InSe. The effective scattering due to optical phonons strongly reduces the FID at temperatures above 80 K and the transmitted signal shows nearly a total loss of FID above 180 K. The thermal energy equivalent of this temperature coincides well with the exciton binding energy and the energy of the strong $A_1^{(1)'}$ phonon mode in InSe. Therefore, the loss of the FID at 180 K can be attributed to the effective dissociation of the 1*s*-exciton in InSe at this temperature.

V. CONCLUSION

In conclusion, we demonstrate the time-resolved FID in a layered III-VI semiconductor. A comprehensive study of the ultrafast coherent dynamics of exciton polaritons at the 1*s*-exciton resonance in InSe is given. The detected beats result from the coherent propagation and interference of polaritons from the LPB and the UPB. The particular dependence of the aperiodic polariton beats on sample thickness and excitation energy distinguishes them from the usual quantum beats of optical transitions. Moreover, we find that propagation effects are important on a femtosecond time scale and even for sample thicknesses below one absorption length. Therefore, one has to be careful with the neglect of propagation effects even in thin samples. The major features of the experimental data are quantitatively described by solving the MSBE for linear pulse propagation. The limitations of theory are mainly due to the introduced simple phenomenological damping and the neglect of energetically higher polariton resonances. Propagation effects are enhanced with increasing sample thickness and resonant excitation of the

exciton polariton, whereas they are diminished due to increased scattering of the polaritons from carriers and phonons. The FID signal is less modulated and dephases faster at higher excitation densities and lattice temperatures. In particular we find the increased destruction of the FID signal due to the thermal dissociation of the exciton.

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