Carrier temperature relaxation probed by femtosecond transient grating experiments in CdS$_{x}$Se$_{1-x}$ semiconductors

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The relaxation of the electron temperature in CdS$_{x}$Se$_{1-x}$ was measured by transmission and transient grating experiments using fs light pulses. As compared with ordinary ambipolar diffusion, at high excitation a pronounced delayed rise and a faster decrease of the diffraction signal was observed. A theoretical model that includes one- and two-photon excitation, gap shrinkage, diffusion, and temperature relaxation explains these experimental findings as a result of temperature relaxation with a characteristic time constant of $1\pm0.2$ ps.

I. INTRODUCTION

The dynamics of nonequilibrium carriers in semiconductors, such as their relaxation and transport mechanisms, can be studied favorably by means of ultrashort light pulses, (see, e.g., Refs. 1–4). With the production of carriers with energies larger than the band-gap energy the first processes after excitation include thermalization of electrons and holes on a sub-picosecond time scale due to carrier-carrier interaction and a following slower temperature relaxation due to electron-phonon interaction. While the first mechanism leads to a carrier-temperature relaxation time of about $10^{-13}$ s at room temperature and can be observed at rather low excitations, the second mechanism that is responsible for the temperature relaxation toward an equilibrium value defined by the lattice. This relaxation of the energy of the excited carriers was the subject of various theoretical studies, and recently similar phenomena have been described in metals. At room temperature the direct measurement of temperature relaxation requires subpicosecond time resolution and mostly rests on the observation of the transient absorption spectra after excitation yielding information on the actual carrier distribution and energy. There are extensive experimental studies on carrier recombination including surface recombination and diffusion proceeding on a longer time scale; see, e.g., Ref. 10.

In the first part of this paper we describe fs transmission and grating experiments on II-VI semiconductors for the determination of temperature relaxation. These semiconductors turned out to exhibit large optical nonlinearities around the band gap caused by many-body effects as screening of the exciton absorption and gap shrinkage. The onset of the nonlinearities can be faster than what can be expected from ordinary ambipolar diffusion. At higher excitations a pronounced delayed increase of the diffraction signal after excitation occurs followed by a decrease that is considerably faster than what can be expected from ambipolar diffusion.

In the second part of this paper we present a theoretical model to simulate the experimental findings. Starting with the equation for a parabolic direct semiconductor, including one- and two-photon absorption, gap shrinkage, diffusion, and temperature relaxation we calculate the periodic modulation of the complex dielectric constant, its temporal evolution, as well as the transient diffraction. By fitting the experimental curves we obtained a temperature relaxation time of about $1\pm0.2$ ps.

II. EXPERIMENT

Pulses with a duration of 100 fs, an energy of 100 μJ, at a wavelength of $\lambda_p = 618$ nm were derived from a CPM dye laser followed by a four-stage amplifier pumped by an excimer laser. These pulses are split into two parts (1) and (2) serving as probe and pump channel, respectively (see Fig. 1). The pump pulse is then divided into two partial pulses (2.1) and (2.2), which are incident on the sample under a certain angle. The delay between these pulses is adjusted for their exact temporal overlap. The delayed probe pulse is focused by means of lens $L_1$ onto the excited area where its spot size is about half of the pump spot. With the detectors 3, 4 the probe light diffracted into the diffraction orders 1, 0, 1, 2 could be measured simultaneously. By blocking one of the pump beams the signal recorded with $D_1$ corresponds to the transient sample transmission. The samples were two thin...
platelets of CdS$_{0.52}$Se$_{0.48}$ (sample I) and CdS$_{0.45}$Se$_{0.57}$ (sample II) with the $c$ axes parallel to the polarization direction of the light. Since the mean photon energy is below the gap energy—133.9 meV in the case of sample I and 67.6 meV in the case of sample II—the initial absorption is mainly due to two-photon absorption. Figure 2(a) shows the time dependence of the first order of diffraction for two different excitations of sample I and Fig. 2(b) represents the corresponding transmission curves. At low excitation energies the diffraction efficiency does not decay on a time scale of 10 ps. Additional measurements revealed (not shown here) that the diffraction decreases on a scale of several 100 ps. The same behavior can be observed for the absorption. At larger excitation densities the diffraction efficiency not only increases but also shows a different temporal behavior. The maximum occurs about 2 ps after the excitation and is followed by a fast partial decay (several ps). The transmission shows a complicated behavior during the first ps but remains almost constant where the diffraction efficiency exhibits the rapid drop.

Figure 3 shows the dependence of the diffraction decay time (fast component) on the grating period which was adjusted by varying the intersection angle of the two pump pulses. Obviously there is a dependence on the grating period. Simultaneous measurements of the zeroth-, first-, and second-order of diffraction of sample II are represented in Fig. 4. The complicated behavior of the transmission signal ($m=0$) is due to the interplay of the various diffraction orders. Note there is a region where the intensity of the first-order diffraction signal ($m=1$) exceeds the transmitted intensity.

III. THEORETICAL MODEL AND DISCUSSION

For simplicity, we assume a spatial dependence only with respect to the $x$ coordinate (intersection line of plane...
where $I_t$ is the intensity of the short test pulse and $T_m$ are the Fourier coefficients of the complex transmission function of the grating,

$$ T(x,t) = \sum_{m=0}^{\infty} T_m(t) \cos \left( \frac{2\pi}{\lambda} mx \right), \tag{2} $$

which can be determined from

$$ T_m(t) = \frac{2}{\lambda} \int_0^\lambda dx \ T(x,t) \cos \left( \frac{2\pi}{\lambda} mx \right). \tag{3} $$

The so-called grating period $\lambda = \lambda_p/2 \sin(\theta/2)$ is determined by the wavelength of pump light $\lambda_p$ and the angle $\theta$ between the two incident pump pulses. The theoretical simulation of the diffraction dynamics requires the knowledge of the space- and time-dependent optical transmission $T(x,t)$ of the sample which is closely connected to the optical susceptibility $\chi(x,t)$. If we assume Fabry–Perot effects to be small because of the high absorption of the semiconductor we can evaluate $T(x,t)$ by $\chi(x,t)$ using the relations

$$ T(x,t) = \exp(ikd) \tag{4} $$

and

$$ k^2 = (4\pi^2/\lambda_p^2) [1 + \chi(x,t)]. $$

$k$ can be approximated by

$$ k \approx (2\pi/\lambda_{sem}) [1 + (\lambda_{sem}/\lambda_p)/2] \delta \chi, $$

where

$$ \delta \chi = -\text{Re}(\chi_0), \quad \lambda_{sem} = \frac{\lambda_p}{\sqrt{1 + \text{Re}(\chi_0)}}. \tag{5} $$

The quantity $T_0$ is responsible for a constant phase shift and $\alpha$ corresponds to an effective optical thickness of the sample.

Obviously we need only the optical susceptibility $\chi(x,t)$ which in turn can be determined by the characteristic equation of the semiconductor. Our discussion of $\chi(x,t)$ is to start after excitation by the two 100 fs light pulses. Accordingly we do not consider the temporal behavior of the absorption process, but we lump the result of it into the initial values of the electron ($n$) and hole ($p$) densities and energies $U$. With “after excitation” we also mean that the carriers generated with different energies (e.g., by single- and two-photon absorption) have already been mixed and thermalized due to carrier-carrier interaction and can be characterized by one temperature $T$ differing from the temperature $T_f$ of the lattice (phonon system) and one quasi-equilibrium distribution of electrons and holes. This is about 200 fs after zero delay when our simulation starts. Moreover our time scale of interest is assumed to be larger as compared with the quasimomentum orientational relaxation time $\tau_\theta$ and phase relaxation time $\tau_\phi$. These time constants are typically shorter than 100 fs. We used a value of 75 fs for our simulations. If we take into account the influence of excitons the optical susceptibility of a parabolic semiconductor is determined by the energies and enhancement factors of all excitonic states which are defined by solutions of the hydrogen problem.

$$ \chi(\omega,x,t) = \sigma \sum_{k=1}^{m-1} \frac{1}{k^2} \delta \left( \frac{E_R}{k^2}, n \right) + \frac{\sigma}{2E_R} \int_{E_G-(E_R/m^2)}^{E_G} dE \delta(E,n) $$

$$ + \frac{\sigma}{4E_R} \int_{E_G}^\infty dE \frac{e^{\pi \sqrt{E_R/(E-E_G)}}}{\sinh[\pi \sqrt{E_R/(E-E_G)}]} \delta(E,n), $$

$\delta(E,n)$ contains all information about the actual state of the carriers $f_n/E$ are the distribution functions for electrons and holes, $E_G$ is the material dependent Rydberg energy (for our samples $\approx 20.5$ meV), and $\sigma$ is proportional to the geometrical thickness of the sample and $\text{Re}(\chi_0)$ has a value of approximately 12. $\delta \chi$ includes the full imaginary part of $\chi$ as well as the pump-induced change of $\chi$. Hence $T(x,t)$ can be written as

$$ T(x,t) = T_0 \exp \left[ i \alpha \delta \chi(x,t) \right], $$

with

$$ \alpha = (\pi/\lambda_{sem}) \frac{\lambda_p^2}{d}, $$

$$ T_0 = \exp \left[ 2\pi i (d/\lambda_{sem}) \right]. $$

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The quantity $T_0$ is responsible for a constant phase shift and $\alpha$ corresponds to an effective optical thickness of the sample.
averaged squared dipole matrix element. In Eq. (6) we summed up over the first $m$ bound excitons (first term) and summarized the rest in the second term. Scattered states are represented by the third term.

For the band-gap energy we have accordingly $^{20}$

$$E_{G}(n) = E_g^0 - \beta n^{1/3},$$

where $E_g^0$ is the gap energy in the unperturbed semiconductor. As to be described later we estimated $\beta$ with $(100 \pm 10) \times 10^{-6}$ meV cm. In the nondegenerated case for a quasithermal equilibrium the distribution functions $f_{n/p}$ are Boltzmann-like and can be written in the form

$$f_n(k,x,t) = \frac{1}{2} \left( \frac{2\pi \hbar^2}{m_e k_B T(x,t)} \right)^{3/2} n(x,t) e^{-\hbar^2 k^2 / 2m_e k_B T(x,t)},$$

where $T$ is the actual electron temperature, $n$ the electron density, and $m_e$ the effective electron mass. At first we want to deal with the transmission behavior only and neglect the spatial modulation of the sample properties. In the range of interest the total density of excited carriers remains constant and the only temporal dependence to be considered is that of the carrier temperature. To describe its relaxation quantitatively towards the lattice temperature we introduce a characteristic temperature relaxation time $\tau_w$ and assume

$$T(t) = (T_0 - T_f) e^{-(t-t_0)/\tau_w} + T_f,$$

where $T_f$ is the lattice temperature, $T_0$ is the initial carrier temperature, and $t_0$ is the moment at which carrier thermalization is completed. If we assume that thermalization does not change the mean carrier energy then $T_0$ can be determined from the kinetic energy of the carriers at excitation. Let $n_{SPA}$, $n_{TPA}$ be the number of carriers excited by two-photon absorption (TPA) and single-photon absorption (SPA), respectively. $T_0$ can be written as

$$T_0 = \frac{2}{3K_B} \left[ \hbar \omega - E_G(n) \right] n_{SPA} + \frac{2}{3K_B} \left[ 2\hbar \omega - E_G(n) \right] n_{TPA},$$

where $\Theta$ is the Heaviside function. The transmission behavior is determined by $\gamma$, the imaginary part of $\gamma$ which has to be computed numerically. It can be obtained by inserting Eqs. (7), (8), and (9) into (6) and taking into account $\delta_p$, the imaginary part of $\delta$ only,

$$\delta_p(E,n) = \frac{\hbar^2}{\tau_2} \left[ E + E_G(n) - \hbar \omega \right]^2 + (\hbar/\tau_2)^2.$$

By means of Eqs. (5) and (11) we are now able to simulate the transient sample transmissions and to compare them with the experimental results to be seen in Fig. 2(b).

Figure 5 shows the corresponding results for different excitation densities simulated with the characteristic constants of sample 1. We approximated the carrier density generated by SPA to grow linearly and that of TPA quadratically by increasing the excitation. At the highest excitation the relation between $n_{SPA}$ and $n_{TPA}$ reaches a value of 3 and the whole electron density can be approximated by $4 \times 10^{17}/\text{cm}^3$. The simulations showed that the initial decrease of the transmission by increasing excitation is a result of gap shrinkage, i.e., the coefficient for SPA becomes larger. Comparing the measured transmission change (see Fig. 2) with the theoretical simulations we found a value for the parameter $\beta$ of $(100 \pm 10) \times 10^{-6}$ meV cm. It turned out that the dynamics of the transmission increase is almost exclusively determined by the temperature relaxation time $\tau_w$. The best fit for our experimental data was obtained with $\tau_w = (1 \pm 0.2) \text{ ps}$. Now we want to turn to the transient grating experiments. The fact that in certain time ranges the intensity in the first diffraction order is larger than the transmitted intensity suggests the dominant influence of a phase grating. For its description we need to consider the time and space dependence of the real part of $\delta$. To avoid divergences we have to split $\delta_R$ in the following way:

$$\delta_R(E,n) = \frac{\tau_2}{\hbar} \left[ E_G(n) - \hbar \omega \right] \delta_p(E,n) + \delta_0(E) - \frac{2E [E_G(n) - E_g^0] (E + E_g^0 - \hbar \omega)}{\left( [E + E_G(n) - \hbar \omega]^2 + (\hbar/\tau_2)^2 \right) \left( [(E + E_G(n) - \hbar \omega)/\tau_2]^2 + (\hbar/\tau_2)^2 \right)}.$$
where $\theta_R^0$ is independent of the carrier density and lets us determine a constant real part of $\chi$ which is unimportant for our further treatment because it depends on the frequency only and causes a constant phase shift. By inserting Eq. (13) in Eq. (6) the first term of Eq. (13) effects a part of $\chi_0$ being proportional to $\chi_f$.

With an initial carrier and temperature distribution given and with Eqs. (3) and (5) we can determine the Fourier coefficients $T_{mn}$ which are necessary for the calculation of the diffraction signals. In addition to Eq. (10) which describes temperature relaxation, for the dynamics of the diffraction we have to consider diffusion processes. That the latter effect plays a role in our time range of interest is indicated by the strong dependence of the diffusion decay on the grating period; see Fig. 3. If we assume that only diffusion is responsible for the diffraction behavior we obtain an unrealistically large value for the ambipolar diffusion constant in the order of $D \approx 800$ cm$^2$/s, which is about 80 times larger than what is known for CdS$_2$Se$_1-x$ at room temperature.$^{21}$ As we will see later it is the interplay of temperature relaxation and diffusion that is responsible for the fast decay of the diffraction signal.

To describe this situation theoretically we start from the Boltzmann equation for the distribution functions $f_{n/p}$ for electrons and holes. For ambipolar diffusion, i.e., $n(x) = p(x)$, and without an external field this equation reads

$$\partial f_n(k,x,t)/\partial t + v_x(k) \partial f_n(k,x,t)/\partial x = S_{ep} + S_{ee},$$  

(14)

where $v_x(k) = \hbar k/m_e$ is the electron velocity, $S_{ee}$ is the term taking into account electron-electron interaction, and $S_{ep}$ is responsible for electron-phonon interaction. Because the energy and carrier densities are not changed by the Coulomb interaction we can neglect $S_{ee}$ in further calculations. In the frame of the so-called relaxation approximation,$^{22}$ $S_{ep}$ can be written as

$$S_{ep} = -S_{ep} \frac{1}{\hbar^2} \left( f_n(k,x,t) - \frac{1}{e^{[E(k)-E_f]/k_B T_f}+1} \right),$$  

(15)

where $\hbar^2 k^2 S^1_{ep} = 0$. $E_f$ is the Fermi level of the actual carrier density cooled down to the lattice temperature $T_f$, and obeys the relation

$$\int d^3 k f_n(k,x,t) = \int d^3 k \frac{1}{e^{[E(k)-E_f]/k_B T_f}+1}.$$  

From Eq. (13) we can obtain corresponding kinetic equations for $n(x,t)$ and $T(x,t)$ if we represent them by means of $f_n$ as

$$n(x,t) = 2 \int \frac{d^3 k}{(2\pi)^3} f_n(k,x,t),$$  

(16)

$$U(x,t) = 3k_B T(x,t) n(x,t) = 2 \int \frac{d^3 k}{(2\pi)^3} \left( \frac{\hbar^2 k^2}{2m_e} f_n(k,x,t) \right.$$  

$$+ \frac{\hbar^2 k^2}{2m_p} f_p(k,x,t) \bigg).$$  

(18)

For convenience we will use the energy $U(x,t)$ rather than the temperature in what follows. Equations (14) and (15) together with Eqs. (9), (16), and (18) yield

$$\frac{\partial}{\partial t} n - \frac{1}{3} \frac{\mu}{e} \frac{\partial^2}{\partial x^2} U = 0,$$  

(19)

$$\frac{\partial}{\partial t} U - \frac{1}{\tau_w} (U - 3k_B T n) - \frac{5}{9} \frac{\mu}{e} \frac{\partial^2}{\partial x^2} \left( \frac{U^2}{n} \right) = 0,$$  

(20)

where $\mu$ is the ambipolar carrier mobility which can be approximated by the values of the electron and hole mobilities $\mu_n/\mu_p$.  

(21)

In our case we have $\mu = 73.5$ cm$^2$/V s. For a solution of Eqs. (19) and (20) we need the initial distributions $n(x,0)$ and $U(x,0)$ and with Eqs. (19) and (20) we need the initial values $n(x,0)$ and $U(x,0)$ determined by the actual excitation conditions.

Note that after the pump pulse excitation we not only have a periodic modulation of the carrier density but also a modulation of the carrier temperature and the band gap.

A great problem is to determine the exact profile of the initial carrier distributions, because the absorption efficiency is essentially influenced by the actual values of gap energy and light intensity which are both functions of time. Only an exact simulation of the initial absorption process would solve this problem. We can only make some assumptions by evaluating the experimental results. Usually, only SPA plays a part and for low excitations the carrier distribution is given by a cos function. In our case the situation is more complicated. The single-photon energy is below the band gap so that the initial carrier generation is through TPA and phonon-assisted SPA. At a certain carrier density, due the gap shrinkage a part of the pump pulse may be absorbed by SPA, too. Both effects, a reduction of absorption by saturation and an increase by gap shrinkage, have to be taken into account and determine the initial (spatial) carrier distribution. The ratio of $n_{SPA}$ and $n_{TPA}$ gives the initial values of the carrier temperature. To find an initial distribution we assumed a simple energy saturation and introduced an energy-dependent cross section $\alpha(E)$,

$$n_{SPA} = n_0 (1 - e^{-\alpha(E)/E_0}),$$  

(22)

$$\alpha(E) = \frac{1}{2} \left[ 1 + \tanh \left( \frac{E - E_S}{E_0} \right) \right],$$  

(23)

$$n_{TPA} = \gamma n_0 (E/E_0)^2,$$

where $\gamma$ determines the ratio of $n_{SPA}$ and $n_{TPA}$. The other constants $E_0$, $E_S$, and $\kappa$ are responsible for the simulated forms of the carrier distributions. Concerning the dynamical behavior of the diffraction our calculations showed that the results are rather independent from the initial spatial carrier distribution.

A numerical solution of Eqs. (19) and (20) together with the initial conditions (11), (22), and (23) now allows us to calculate the optical susceptibility [Eqs. (6)] and the diffraction efficiencies as a function of (delay) time. Before
discussing the numerical results let us recall Eq. (19), which by means of Eq. (17) can be written as

\[
\frac{\partial}{\partial t} \frac{K_{br}}{e} \frac{\partial^2}{\partial x^2} (T_n) = 0.
\] (24)

This equation turns into the ordinary equation of ambipolar diffusion with the diffusion coefficient \(D = (K_{br}/e)T\) if there is no temperature gradient, which means either the electrons do not have excess kinetic energy after excitation, e.g., \(n_{TPA} = 0\), or the temperature has already been recovered to its equilibrium value \((T_f)\). Transient grating experiments performed with ps light pulses referred to the latter case. The interpretation of our experiment, however, requires the consideration of \(T\) as a function of \(x\) and \(t\).

Using the model introduced above we simulated the diffraction behavior (see Fig. 6) of the experimental results from Fig. 4 on the basis of parameters of sample II. Obviously the main features of the diffraction zeroth, first, and second order are described satisfactorily. Deviations are mainly for delay times \(\approx 0.5\) ps. It is likely that our model of a quasiequilibrium state, defined, e.g., by one carrier temperature, is not fully valid in this short time domain after the excitation.

Figure 7 shows a simulation of the experiment depicted in Fig. 2(a). For comparison we also calculated the action of a pure phase grating neglecting \(\chi_f\). These results show the dominant role played by the phase grating in our experiment. This also explains why the transmission changes only slightly during the rapid drop of the diffraction efficiency in the experiment [see Figs. 2(a) and 2(b)]. The theoretical curves (compare Figs. 5 and 7) support this conclusion as well.

If we analyze the evolution of the pure phase grating we find that the maximum phase difference between high and low excited regions exceeds \(\pi\) at the beginning of the simulation by \(t\), e.g., 1 ps after the excitation the value of this phase difference is already \(1.7\pi\). During the further relaxation of the electron temperature this phase difference decreases and after 2 ps reaches approximately \(\pi\), i.e., in this moment the contrast of the phase grating has its highest value and the intensity of the first diffraction order reaches its maximum. After that the contrast of the phase grating is reduced by the decreasing phase difference within the grating.

As mentioned before, from ordinary ambipolar diffusion a decay of the diffraction signal in the order of some 100 ps is expected. The data from Fig. 3, however, suggest that diffusion-controlled processes proceed on a much shorter time scale. From our calculations, the following turn out to be responsible.

Diffusion processes are much accelerated by high carrier temperatures due to a high share of TPA. All deviations of a cos modulation of the carrier densities caused by the nonlinearities of SPA result in steeper edges and higher gradients and in connection with that in a stronger diffusion.

Diffusion of highly excited carriers can be amplified by nonlinearities as described in Eq. (20).

Self-diffraction of the two pump beams, which depends on the angle of incidence, could have influenced the resulting carrier and temperature distribution we have started with.

The phase and amplitude grating sensitively depends on the band gap. The initial modulation of the gap energy (renormalization) is washed out with the carrier diffusion, where according to Eq. (8) the gap shrinkage in the originally unpumped regions \([n(x,0) = 0]\) dominates over the gap recovery in the pumped regions. Mathematically, the strong influence of a small change of the carrier density on \(\chi\) can be seen by inspection of singularities of the quantity \(\partial \chi/\partial n\).

Figure 8 shows calculations of the time-dependent
first-order diffraction for different grating periods in order to illustrate the influence of diffusion. We used the ambipolar mobility known from Eq. (21) and reached diffusion determined changes in the decay times of some ps. Obviously the interplay of diffusion and the various nonlinearities is responsible for a fast component in the diffraction signal in addition to the transient which is attributed to the temperature relaxation. This diffusion-controlled process depends on the grating spacing as expected.

IV. SUMMARY

Theoretical and experimental studies on transient grating and transmission experiments have been performed to study temperature relaxation and diffusion processes in II-VI semiconductors. After excitation of CdS<sub>x</sub>Se<sub>1-x</sub> by two 100 fs light pulses being incident under a certain angle we have the following situation. A grating exists that represents a periodic modulation of free carriers excited by one- and two-photon absorption. This also leads to a modulation of the induced gap shrinkage and of the carrier temperature. These two effects result in a rapid change of the first-order diffraction signal during the first 10 ps after excitation which can not be explained with ordinary ambipolar diffusion. The delayed rise of the diffraction efficiency which was observed at high excitations is explained to be due to the combined action of temperature relaxation and gap shrinkage induced by carrier diffusion into regions originally unpumped.

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