Picosecond dynamics of bleaching and spin splitting in InP revealed by the photoinduced magneto-optical Kerr effect near the spin-orbit split-off exciton transition

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A time-resolved study of the photoinduced magneto-optical response of InP in the spectral range nearby the spin-orbit split-off exciton transition shows both bleaching of the split-off exciton transition and splitting of the exciton spin sublevels. The bleaching relaxes with a decay of 10 ps due to energy redistribution of the photoexcited carriers. The splitting of the spin sublevels shows a two-exponential decay with characteristic times of 20 ps and 200 ps, respectively. These times are related to the relaxation of the spin polarization of the electrons in the conduction band.

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I. INTRODUCTION

In recent years ultrafast spin dynamics in semiconductors have attracted intense research interest, due to the importance of the newly developing area of spintronics.¹ Most studies of spin dynamics in semiconductors have been done in the spectral range around the direct transition from the top of the heavy-hole valence band to the bottom of the conduction band.^{2,3} In contrast, only little is known about the dynamics of a photoexcitation from the split-off spin-orbit valence band. Linear optical studies of such excitation are hampered by strong absorption due to transitions from the heavy-hole band. A recent clear observation of the spin-orbit split-off (SO) exciton in a four-wave-mixing experiment has shown that nonlinear optical methods are very effective to study the properties of semiconductors in the spectral range far above the band gap.⁴ The observation of the SO exciton in Ref. 4 was based on the fact that excitons strongly influence the coherent response of the semiconductor through the interference of exciton and continuum polarization sources.

Here we report on the ultrafast nonlinear magneto-optical response of InP nearby the spin-orbit split-off exciton transition and show that this exciton also reveals itself in the incoherent response of InP. Our study is based on pump and probe measurements of the photoinduced magneto-optical Kerr effect. The spectral analysis showed that the photoexcitation resulted in both bleaching of the spin-orbit split-off exciton transition and a splitting of the exciton spin sublevels. After the photoexcitation the bleaching relaxed due to energy redistribution of carriers with a decay of 10 ps. The splitting of the spin sublevels exhibited a two-exponential decay with characteristic times of 20 ps and 200 ps, respectively. These are interpreted as spin-relaxation times of the electrons in the conduction band.

II. EXPERIMENTAL DETAILS

For the present study, we used an all-optical method in which an intense pump beam excites a spin polarization in the semiconductor due to the phenomenon of optical orien-

tation and a less intense probe beam monitors this photoexcited spin polarization through the magneto-optical Kerr effect. Phenomenologically, optical orientation is a manifestation of the inverse Faraday effect, which yields the magnetization of media under the influence of circularly polarized light.⁵ Microscopically, optical orientation is explained as optical excitation of carriers with preferential spin orientation because of the selection rules for the transitions induced by the circularly polarized photons.⁶ In a direct band-gap semiconductor with a zinc-blende crystallographic structure such as InP, optical transitions take place around the Γ point of the first Brillouin zone.⁷ An essential feature of the band diagram is the splitting of the p -type valence band into a degenerate Γ_8 band: one subband for heavy-hole (HH) states, the other for light-hole (LH) states, and a spin-orbit split-off Γ_7 band that lies 0.11 eV below the Γ_8 band. For photon energies equal or slightly superior to the direct band-gap energy ($E_g = 1.42$ eV), transitions can take place between the top of the degenerate Γ_8 valence band and the bottom of the Γ_6 conduction band. For higher photon energies, optical transitions are also possible between the top of the split-off Γ_7 band and the bottom of the Γ_6 band (Fig. 1). The exciton transition from the SO valence band is expected around 1.52 eV.

The measurements were performed in the spectral range between 1.47 eV and 1.57 eV in a pump and probe configuration, using a Ti:sapphire laser with a pulse duration of ≈ 100 fs and a repetition rate of 82 MHz. The setup for time-resolved magneto-optical Kerr effect measurements was described in detail elsewhere.⁸ The polarization of the circularly polarized pump beam was modulated between left-handed and right-handed helicities, while the probe was linearly S polarized. Both beams were focused on the sample to a spot diameter of 100 μm for the pump and slightly less for the probe. The pump fluence on the sample was about 10 $\mu\text{J}/\text{cm}^2$ and produced an electron-hole pair density around 10^{18} cm^{-3} . The Kerr rotation and ellipticity experienced by the probe beam were measured as a function of time delay between pump and probe pulses. The sample was

FIG. 1. (a) Schematic electronic band structure of InP near Γ point and (b) allowed electric dipole transitions for circularly polarized light.⁷

placed in helium atmosphere and its temperature was varied between 20 K and 300 K. The accumulated static heating of the sample was estimated to be below 10 K.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Experimentally, a linear dependence of the photoinduced Kerr effect on the pump intensity was found for fluences ranging from 1 to 10 $\mu\text{J}/\text{cm}^2$. A typical example of the relaxation dynamics of the Kerr rotation in InP is shown in Fig. 2 for several photon energies. A clear variation of the temporal dependence with photon energy is observed. Obviously such a behavior results from the competition of several contributions with different spectral and temporal dependence. In the following we will analyze only the incoherent regime of the relaxation which occurs ≈ 1 ps after the photoexcitation.⁴ For delay times $\tau_d > 1$ ps our data can be fitted by the sum of three relaxation components:

FIG. 2. Dynamics of the Kerr rotation θ_K in InP for different photon energies.

FIG. 3. Experimental spectral dependencies of the three contributions to the Kerr rotation θ_K (solid circles) and ellipticity ε_K (open circles) in InP: (a) amplitudes A_1 and A'_1 of the contribution with decay time 10 ps, (b) amplitudes A_2 and A'_2 of the contribution with decay time 20 ps, (c) amplitudes A_3 and A'_3 of the contribution with decay time 200 ps. Lines are guides to the eye.

$$\theta_K = \sum_{i=1}^3 A_i \exp\left(-\frac{\tau_d}{\tau_i}\right), \quad (1)$$

with the amplitudes A_i and the relaxation times τ_i . A relation similar to Eq. (1) holds for the Kerr ellipticity ε_K , with amplitudes denoted by A'_i . We found that the best fit of the data was obtained for $\tau_1 = 10 \pm 2$ ps, $\tau_2 = 20 \pm 5$ ps, and $\tau_3 = 200 \pm 20$ ps. Note that the variation of the pump fluence between 1 and 10 $\mu\text{J}/\text{cm}^2$ did not result in any significant change of the observed decay times. The amplitudes A_i and A'_i resulting from the fit are shown in Fig. 3 as a function of photon energy. From the figure one can see the similarity between the spectral dependencies of the amplitudes A_2 , A_3 and A'_2 , A'_3 , respectively. In this case the rotation changes sign and the ellipticity reaches an extremum at the energy of the SO exciton transition. In contrast, the amplitudes A_1 and A'_1 show the opposite spectral behavior—ellipticity changes sign and the rotation reaches a maximum at the energy of the SO exciton transition. For the interpretation of these experimental results a theoretical analysis is necessary.

The photoinduced magneto-optical Kerr effect can be described by an expression⁹

$$\varepsilon_K + i\theta_K = \frac{C(n)}{I_0} \int dt (P^{(+)} - P^{(-)}) E_0^*, \quad (2)$$

where E_0 is the electric-field amplitude of the probe pulse, I_0 is the probe pulse intensity, and $P^{+}(P^{-})$ is the third-order electric polarization induced in the material by the right circularly polarized pump pulse and by the right (left) circularly polarized probe pulse, respectively. $C(n)$ is a function of the complex refractive index n . The explicit form of this function is not essential here. Even at the SO band edge (~ 1.53 eV) $\text{Im}(n) \ll \text{Re}(n)$ (Ref. 10) and the coefficient $C(n)$ may be considered as real. This means that the Kerr rotation θ_K and ellipticity ε_K are determined by the imaginary and real parts of the third-order polarization in Eq. (2), respectively.

FIG. 4. Optical transition scheme and simulated spectra of (a) the phase-space filling term of the photoinduced magneto-optical Kerr effect and (b) the spin-dependent energy renormalization term of the magneto-optical Kerr effect around a resonance at $h\nu_o$ (Ref. 15).

The photoinduced macroscopic polarizations $P^{(\pm)}$ are related to the interband elements of the density matrix, which should be numerically calculated from the semiconductor Bloch equations (SBE).² In the incoherent regime two sources of nonlinearity in the SBE should be taken into account.

(i) Phase-space filling (PSF) arising from the oscillator strength dependence on electron and hole distribution functions. When the semiconductor is pumped by a circularly polarized light pulse, the selectively excited transition does not respond to the following probe pulse because the upper state is already occupied. The relaxation of the PSF term gives information about the time-dependent thermalization of carriers.¹¹

(ii) Spin-dependent energy renormalization, which arises from the repulsive interaction among the carriers with equal spins. After the semiconductor has been optically excited with circularly polarized light, the spin system is no longer in equilibrium and spin splitting of the energy levels arises, before decreasing again during the process of spin relaxation.^{12–15}

Both these mechanisms are due to the Pauli exclusion principle and reflect the occupation of single-particle states. We have performed qualitative simulations of the spectral behavior of the photoinduced magneto-optical Kerr effect for a two-level system separately for the PSF (i) and the spin-dependent energy renormalization (ii) contributions. The results of the simulations are presented in Fig. 4 in arbitrary units, because the model used cannot supply a quantitative estimate of the photoinduced magneto-optical Kerr effect. Such quantitative simulations require a routine solution of the Maxwell equations for the reflected light as well as taking into account an inhomogeneous distribution of the photoexcited carriers along the penetration depth, diffusion of the carriers, and many other parameters, which make this task well beyond the scope of this paper.

From the comparison of the theoretical and experimental results nearby the energy of the SO exciton transition (in the range between 1.51 eV and 1.57 eV) one can observe a good qualitative agreement between calculated PSF spectra and the spectral behavior of the amplitudes A_1 and A'_1 . Therefore

the relaxation of this component is not related to spin scattering but to the processes of energy redistribution of the photoexcited carriers. Notice that together with the SO exciton the pump pulse creates free electrons and holes due to transitions from the HH and LH valence bands to the conduction band with an excess energy ~ 0.11 eV. This initially excited high-energy nonequilibrium electron population also contributes to the nonlinear optical response through the PSF effect and can mask the SO exciton similar to the situation in linear optical absorption. However, due to the fast (during ~ 1 ps) emission of optical phonons these electrons rapidly loose their energy, which diminishes the bleaching of the transitions from HH and LH bands and reveals the SO exciton.

In contrast to the fundamental exciton the ionization of the SO exciton is expected to be faster than 10 ps. Photoexcited holes created near the Γ point in SO band rapidly scatter due to the emission of LO phonons to the LH band and then rapidly loose their spin orientation.^{18,19} Because of the fast inelastic SO $\rightarrow$ LH hole scattering the SO excitons rapidly ionize leaving behind an electron-density distribution $n(\mathbf{k}) \sim |\phi(\mathbf{k})|^2$, where $\phi(\mathbf{k})$ is the exciton wave function in momentum space, localized near the bottom of the conduction band. This exciton-associated electron density results in a bleaching of the SO exciton transition due to the PSF effect. The bleaching relaxes with the decay time $\tau_1 = 10$ ps, which is attributed to the energy redistribution of the exciton-associated electron density. This observation is in very good agreement with simulations of nonequilibrium photoluminescence from the first principles.¹⁶ These simulations show that the energy redistribution of electrons in the conduction-band results in a relaxation of an exciton photoluminescence within 28 ps. The similar redistribution of the electrons manifests itself in our experiment as the relaxation of the bleaching of the exciton transition.

From the comparison of the theoretical and experimental results one can find a qualitative agreement between the spectral behavior of the calculated spin-dependent energy renormalization term and the amplitudes A_2, A_3 with an extremum of the Kerr ellipticity and sign change of the Kerr rotation at the energy of the SO exciton transition. Therefore the components with amplitudes A_2, A'_2 and A_3, A'_3 are assumed to be due to the splitting of the spin sublevels of the SO exciton and the relaxation of this component is thus related to spin dynamics. A similar effect of energy renormalization of the SO exciton in the coherent regime was observed in GaAs,¹⁷ but due to the linear polarization of the pump the photoexcitation did not result in the splitting of exciton spin sublevels in that case. A comparison of the Kerr effect induced by light and an external magnetic field shows that a photoexcitation of about 0.1 GW/cm² results in a splitting of the spin sublevels equivalent to that caused by an external magnetic field of about 1 T.¹⁰ This clearly demonstrates that the splitting of spin sublevels observed in our experiment cannot be caused by the magnetic field of the photoaligned spins, which is about 1 μ T, but has quantum origin being due to the strong exchange interaction among the carriers.

FIG. 5. The temperature dependencies of the spin related relaxation times τ_2 (open circles) and τ_3 (solid circles) measured at photon energy 1.52 eV. Dashed line is the function T^{-3} . Rhomb shows the value of the electron-spin relaxation in InP reported elsewhere (Ref. 20).

It is remarkable that the decay times τ_1 and τ_2 operate in the same temporal window. We do not exclude that both these decays could be related to the same physical mechanism, which could be a cooling of the electrons via radiation of LO phonon slowed down by the hot phonon effect.² Nevertheless, it is clear that the processes with decay times τ_1 and τ_2 manifest themselves differently in our experiment, so that they can be assigned to PSF effects and the spin-dependent energy renormalization, respectively.

Note that in our experiment the electrons are excited from three different valence bands and the excitation results in a highly nonequilibrium distribution of the electrons in the conduction band. The processes of spin scattering of these electrons will be accompanied by their thermalization. Thus spin relaxation of the electrons in the conduction band is a complicated process which cannot be described by one-exponential decay. To our opinion the decay times τ_2 and τ_3 observed in our experiment can be assigned to the electron-spin relaxation in the conduction band.

The observed decay time $\tau_3 = 200$ ps lies in the temporal window that is typical for final stage of cooling of the electrons in the conduction band.² Nevertheless, we should note that this slow process cannot only be attributed to cooling, since the measured magneto-optical response approaches zero following the exponential decay with a characteristic time $\tau_3 = 200$ ps. Electron cooling itself, without spin relaxation, is unable to result in a vanishing of the Kerr effect.

The temperature dependencies of the spin related relaxation times τ_2 and τ_3 measured at the energy of the exciton transition 1.52 eV are shown in Fig. 5. The decay time τ_2 almost does not depend on temperature in the range between 20 K and 160 K. The decay time τ_3 does not show any

temperature dependence between 20 K and 60 K, but above 60 K the relaxation time strongly decreases with temperature. At room temperature we distinguish only one-exponential decay of the spin sublevels splitting with a characteristic time of 25 ps.

In contrast to the PSF, the photoinduced spin splitting depends on the electron distribution over the entire phase space² so that the incomplete cooling leads to a faster relaxation of the spin-splitting. Therefore, the fast spin-relaxation time $\tau_2 = 20$ ps observed in our experiment can be associated with the hot electrons and should be determined by their temperature. If the temperature of the photoexcited carriers is higher than that of the phonons, spin relaxation of hot carriers should not be sensitive to the variation of the lattice temperature. This can explain why the spin-relaxation time τ_2 does not depend on temperature in our experiment.

The longer decay ($\tau_3 = 200$ ps) of the spin polarization can be assigned to the carriers almost cooled to the lattice temperature. In that case τ_3 should be sensitive to the variation of the lattice temperature. The observed temperature dependence of τ_3 cannot be fitted by a function T^{-3} (Fig. 5) as in the case of a Boltzmann equilibrium electron distribution and the Dyakonov-Perel spin-relaxation mechanism due to spin splitting of the conduction band. In Fig. 5 we have also plotted the electron-spin relaxation time in InP at 77 K, as reported elsewhere.²⁰ This value is about one order of magnitude higher than the one observed by us. Such a large discrepancy can be due to much higher excitation densities and an increased role of the exchange interaction between electrons and holes in the spin relaxation (Bir-Aronov-Pikus mechanism).⁶ It is important to note that the spin-orbit splitting of the conduction band in InP, which is responsible for the Dyakonov-Perel spin-relaxation mechanism, is three times smaller than in GaAs.²¹ This results in a domination of the Bir-Aronov-Pikus mechanism in InP at lower excitation densities in comparison with GaAs. We should also note that in InP the electron gas at a density $\sim 10^{18} \text{ cm}^{-3}$ and a temperature $T = 20$ K is degenerate at equilibrium. Though full equilibrium cannot be reached during the observation time of ~ 140 ps, the electron gas can be nearly degenerate, which leads to the weak sensitivity of the spin lifetime to temperature changes. Due to the Pauli exclusion principle the spin relaxation in such gases can only happen near the Fermi level, which only weakly depends on temperature. It is worthwhile to note that the degeneracy of a two-dimensional (2D) electron gas can lead to an increase of the Dyakonov-Perel spin-relaxation rate.²² However such behavior cannot occur in a 3D electron gas due to its relatively low mobility.

IV. CONCLUSION

In summary, using time-resolved nonlinear magneto-optical spectroscopy we observed a clear signature of the split-off spin-orbit exciton in the incoherent response of InP. Based on a theoretical analysis it was found that the photoinduced magneto-optical Kerr effect originates from both bleaching of the split-off exciton transition and a splitting of the exciton spin sublevels. The bleaching was found to relax with a decay of 10 ps due to an energy redistribution of the

photoexcited carriers. The splitting of the spin sublevels showed a two-exponential decay with characteristic times of 20 ps and 200 ps, respectively. These can be shown to be related to the relaxation of the spin polarization of the electrons in the conduction band.

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