Optical orientation of excitons induced by an energy-level anticrossing and by cross-relaxation in type-I GaAs/AlAs superlattices

P. G. Baranov, V. A. Vetrov, B. R. Namozov, and N. G. Romanov
A. F. Ioffe Physicotechnical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

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An optical orientation of excitons has been observed in type-I GaAs/AlAs superlattices in a narrow interval of magnetic fields during nonresonant excitation by unpolarized light. The magnetic field strength at which the optical orientation occurs depends sharply on the direction of the field with respect to the plane of the superlattice. The optical orientation is determined from the circular polarization of luminescence. It reaches a magnitude of 10%. It is attributed to an anticrossing of exciton energy levels. Some additional optical-orientation peaks which are observed are caused by a cross-relaxation. © *1995 American Institute of Physics.*

The excitation of semiconductors by circularly polarized light often results in an orientation of the spins of carriers and excitons (see a review[1] and the bibliography there). This optical orientation has proved to be a powerful tool for studying several properties of excitons and carriers, e.g., lifetimes and spin–lattice relaxation times. During optical orientation of electrons in semiconductors, one observes a dynamic polarization of lattice nuclei, which in turn affects the spin polarization of the electrons. This "feedback" between the polarizations of the electron subsystem and the nuclear subsystem has made it possible to detect nuclear magnetic resonance on the basis of the degree of circular polarization of luminescence.[2,3] Optical orientation has been achieved by applying circularly polarized light. This orientation is the result of spin-memory effects in the course of the excitation, since the lifetime of the excited state, τ, is shorter than the spin–lattice relaxation time τ_s. The excited system is thus in a nonequilibrium state. The optical orientation of excitons by circularly polarized light may occur either because the carriers retain their polarization upon the formation of excitons, or it may occur directly during resonant pumping of certain exciton levels.

A measure of the spin polarization of carriers and excitons is the degree of circular polarization of the luminescence: $P=(I_+-I_-)/(I_++I_-)$, where I_+ and I_- are the intensities of the right- and left-hand circularly polarized components, respectively. The polarization P can be written $P=P_0(1+\tau/\tau_s)^{-1}$, where the constant P_0 is proportional to the average spin of the system at the time of the optical excitation, and τ_s is the spin-relaxation time of the electrons. The spin-relaxation time of holes is usually extremely short (on the order of picoseconds), because of the strong spin–orbit coupling.

A nonequilibrium state of the electron subsystem can also be excitated by unpolar-

ized light, by virtue of an anticrossing of exciton energy levels in a magnetic field. In the present experiments we have observed an optical orientation of excitons in a narrow interval of magnetic fields during nonresonant excitation of type-I GaAs/AlAs superlattices by unpolarized light. This optical orientation is induced by a level anticrossing. Under certain conditions, the efficiency of the optical orientation is also influenced by cross-relaxation effects.

A level anticrossing and a cross-relaxation for triplet states were first detected from changes in the phosphorescence of organic materials[4] and ionic crystals (see Ref. 5 and the papers cited there). These effects have also been studied in semiconductors, e.g., in Refs. 6–9. Preliminary results on an anticrossing of exciton energy levels with a heavy hole in a type-I superlattice were reported in Refs. 10 and 11. An exciton is formed in a type I GaAs/AlAs superlattice from a Γ electron and a Γ hole, which are localized in a common GaAs layer. In a type-II superlattice, on the other hand, the hole is localized in a GaAs layer, which is a well (the Γ maximum of the valence band), while the electron is in an AlAs layer, which is a barrier (the X minimum of the conduction band). The radiative lifetime of excitons is on the order of nanoseconds in type-I superlattice and microseconds in type-II superlattices.

The superlattices were grown by molecular beam epitaxy on a semi-insulating substrate with an n-type GaAs buffer layer 1500 nm thick. The superlattice used in the present experiments had 100 periods, each consisting of a layer of GaAs (2.27 nm) and one of AlAs (1.13 nm). The top of the superlattice was coated with a GaAs layer 5 nm thick.

The optical orientation of the excitons was detected at a temperature of 1.6–4.2 K from the circular polarization of luminescence excited by the beam from an argon laser (488 or 514.5 nm, 10–100 mW/cm^2). The orientation was detected in the direction of the static magnetic field. In most cases, the luminescence was excited and detected along the superlattice growth direction (the z axis or [001] direction), i.e., along the perpendicular to the (001) plane of the superlattice. In the experiments on the angular distribution, there was provision for rotating the sample around the $\langle 110 \rangle$ vertical axis.

Figure 1a shows a luminescence line of the superlattice (in the inset) and a plot of the circular polarization of the luminescence versus the magnetic field. These results were measured for various values of the angle θ, between the magnetic field and the [001] axis of the superlattice. The curve of the circular polarization of the luminescence versus the magnetic field has a resonant shape. The change in the degree of circular polarization of the luminescence reaches 10% at the maximum of the curve. The smallest width of the curve (0.3 T) is observed when the magnetic field is along the [001] axis of the superlattice ($\theta=0$). In this case, the optical orientation occurs at a minimal magnetic field (1.16 T). As the angle θ is raised, the magnetic field in which optical orientation occurs increases, as does the width of the resonant peak. At $\theta=68°$, the magnetic field corresponding to the maximum circular polarization of the luminescence is 3.2 T, and the width of the resonant peak is about 3 times that in the case $\theta=0°$.

Taking the heavy and light holes into account, we can describe the exciton energy-level scheme by a common spin Hamiltonian with a hole angular momentum of 3/2 and an electron spin of 1/2. In short-period superlattices, the splitting of the heavy-hole level

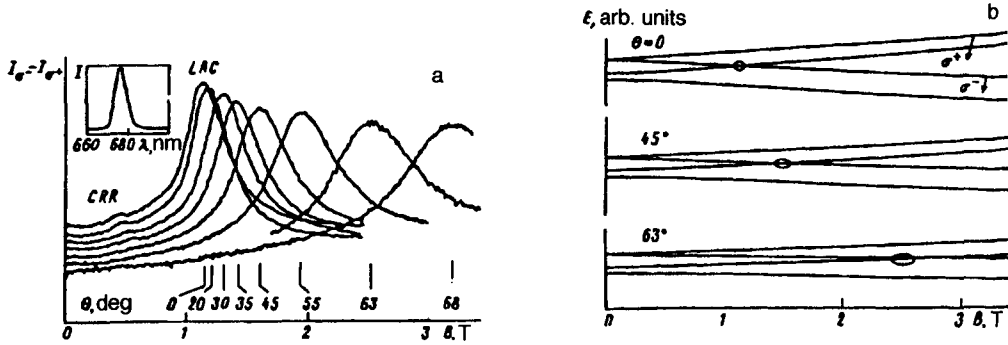

FIG. 1. a: Signal representing the circular polarization of the luminescence versus the magnetic field for a type-I GaAs/AlAs superlattice for various values of the angle (θ) between the magnetic field and the [001] axis. The inset shows a luminescence line of the superlattice. $T=1.6$ K. LAC—Signal representing the level anticrossing; CRR—signal representing the cross-relaxation resonance. b: Exciton energy levels calculated for three values of the angle θ through the use of spin Hamiltonian (1).

($\pm 3/2$) from the light-hole level ($\pm 1/2$) reaches several tens of meV, or much more than the exchange splitting of excitons and also much more than the Zeeman energy in the magnetic fields used. At low temperatures only the heavy-hole levels are filled, so it is a good approximation to restrict the discussion to the energy levels of only those excitons which have a heavy hole (heavy-hole excitons) and to use the effective spin of a heavy hole, $S_h=1/2$. The spin Hamiltonian of an exciton can then be written

$$H=\beta B g_e S_e+\beta B g_h S_h+S_h c S_e, \tag{1}$$

where the first two terms are the Zeeman energy of an electron and a heavy hole in the magnetic field, and the tensors g_e and g_h characterize the g-factors of the electron and the heavy hole. The third term describes the exchange interaction between the electrons and a heavy hole with a tensor exchange interaction tensor c; β is the Bohr magneton.

The curves in Fig. 1a can be explained with the help of the exciton energy-level diagram shown in Fig. 1b for three values of the angle θ: 0°, 45°, and 68°. The energy levels were calculated through a direct diagonalization of spin Hamiltonian (1) with the help of Grachev's "R-spectr" program. The following parameter values were used for the spin Hamiltonian: the electron g-factor was taken to be $g_e=0.4$ and to be isotropic; the g-factors of the holes were taken to be $g_{h\parallel}=2.45$ for the parallel orientation of the magnetic field and $g_{h\perp}=0$ for the perpendicular orientation. The exchange splittings were $c_x=-67$ meV, $c_y=67$ meV, and $c_z=-340$ meV. Spectroscopy studies of superlattices ordinarily use an exchange-splitting parameter $\Delta=c_z/2$ and splittings $\delta_1=(c_x-c_y)/2$ and $\delta_2=(c_x+c_y)/2$ for radiative and nonradiative levels, respectively.

The parameters of the spin Hamiltonian were determined from measurements of the level anticrossing and from measurements of some additional resonances due to cross-relaxation. We also made use of information on the heavy-hole g-factor in type-II superlattices. The value of $g_{h\parallel}$ was taken from the experimental dependence of the heavy-hole g-factor on the width of the GaAs layer in the type-II superlattice: $g_{h\perp}=0$ (Refs. 12–14).

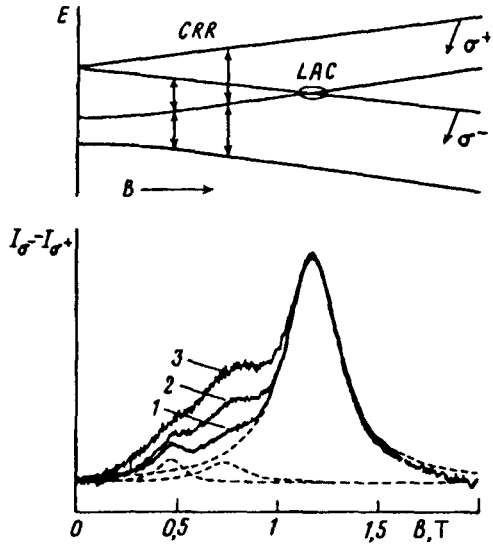

FIG. 2. Normalized curves of the signal representing the circular polarization of the luminescence versus the magnetic field, measured at various intensities of the exciting light. *1*—3; *2*—30; *3*—100 meV. For curve *1* we show the results of a decomposition into three Lorentzians. Shown at the top of the figure is the exciton energy-level scheme, with the level anticrossing (LAC) and the cross-relaxation resonance (CRR). The corresponding curves are shown by the open circles and the dashed lines, respectively.

We assumed that no radiative exciton levels were split in the type-I superlattice, because of the symmetry of the excitons, D_{2h}. The splitting of the radiative levels was thus taken to be zero ($\delta_1 = 0$).

In this superlattice we also observed some weak resonances on the low-field side of the signal corresponding to the level anticrossing (Fig. 1a). The relative intensity and width of these resonances depend strongly on the intensity of the optical excitation of the luminescence. Figure 2 shows curves of the circular polarization of the luminescence versus the magnetic field measured at $\theta = 0°$ and at various levels of the optical excitation. Also shown here is the result of a decomposition of experimental curve *1* into three Lorentzians. Some additional resonances of the same type had been observed in our previous studies of type-II superlattices[11,12] and were attributed to cross-relaxation resonances.

Cross-relaxation resonances can arise in certain magnetic fields at which the Zeeman splitting of the exciton levels coincides with the splitting of the levels of another exciton, of an electron, or of a hole. In this case there is a mutual flip of the spins of the different paramagnetic centers with coincident Zeeman energies. Transitions which are possible during a cross-relaxation resonance in the case of the interaction of two identical excitons are shown at the top of Fig. 2. Here there is a fundamental distinction between the level anticrossing and the cross-relaxation resonance which are induced by a magnetic field. In the case of the level anticrossing, all effects stem from internal properties of an individual paramagnetic center (a certain system of energy levels), while in the case of the cross-relaxation resonance the involvement of a second entity (a partner) is necessary. An

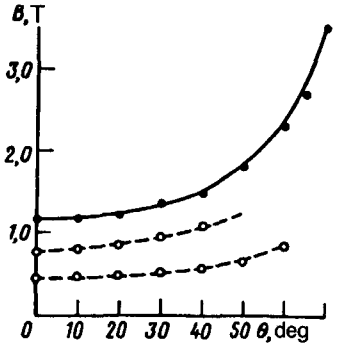

FIG. 3. Experimental (solid circles) and theoretical (solid line) angular dependencies of the level anticrossing signals. For cross-relaxation resonances these dependences are represented by open circles and dashed lines, respectively.

increase in the relative intensity of the cross-relaxation resonance with increasing intensity of the optical excitation is attributed to an increase in the concentration of excitons and carriers and thus to a decrease in the distance between them. This decrease should enhance the efficiency of the cross-relaxation resonance.

Figure 3 shows experimental angular distributions of the positions of the anticrossing signals (the filled circles) and of the cross-relaxation resonances shown in Fig. 2 (the open circles) in a magnetic field. The solid and dashed curves show results calculated on the level anticrossing and the cross-relaxation resonance, respectively, through the use of spin Hamiltonian (1). We see a good agreement between the experimental and theoretical results. This is a primary argument in favor of the validity of the system of energy levels found for an exciton.

Here is the mechanism by which a nonequilibrium population of fine-structure energy levels or an optical orientation of excitons is arranged in the case of an anticrossing. During nonresonant optical excitation, the population of exciton levels is determined by an annihilation of excitons: Under the condition $\tau < \tau_s$, radiative levels have a smaller population than nonradiative levels. In the case of an anticrossing of radiative and nonradiative energy levels, the excess population of a nonradiative state or a state with a bottleneck, due to the continuous optical pumping, can be transferred to the radiative state, where it gives rise to an increase in the emission intensity. Optical excitation in a magnetic field corresponding to a level anticrossing is actually equivalent to the resonant pumping of one level by circularly polarized light. In this case, during steady-state optical pumping in magnetic fields corresponding to a level anticrossing, the degree of circular polarization of the luminescence, P, can be written in the form above for the case of optical orientation by circularly polarized light. The magnitude of the change in the circular polarization of the luminescence induced by the anticrossing is evidence that τ_s is comparable to τ, i.e., that this quantity is on the order of nanoseconds in type-I superlattices. We have also observed a circular polarization of luminescence induced by a level anticrossing in type-II superlattices, in which the time τ was on the order of microseconds, while the time τ_s was on the same order of magnitude, judging from the

magnitude of the level-anticrossing signal. The primary reason why τ_s is shorter in the type-I superlattice than in the type-II superlattice is apparently a sharp increase in the exchange interaction of the electron and the hole in an exciton. This interaction is considerably stronger in a type-I superlattice than in a type-II superlattice.[10]

It has been shown previously[7] that a change in the circular polarization of luminescence at the time of a level anticrossing is caused by a static interaction which mixes states. Such an interaction might be a hyperfine interaction with matrix nuclei (Ga, Al, and As); this interaction would have to be added to spin Hamiltonian (1). The trivial case in which a level anticrossing arises because of the presence of a transverse component of the static magnetic field in the case $\theta \neq 0$ follows directly from the solution of spin Hamiltonian (1).

We believe that the small width of the resonant line corresponding to the level anticrossing in Fig. 1a is evidence that the superlattice is of high quality. The width of this Lorentzian line corresponds to a time on the order of 120 ps, which is close to the lifetime of excitons in type-I superlattices (0.3–1 ns).

In some of the type-I superlattices, we detected level-anticrossing signals on the basis of the linear polarization of the luminescence, apparently because of an optical alignment of excitons.

The electron spin polarization induced by the level anticrossing may lead to a nuclear polarization by virtue of the hyperfine interaction. A nuclear magnetic resonance might thus be detected from a circular polarization of luminescence, as in the case of optical pumping by circularly polarized light.[1-3] In a magnetic field of 1.16 T, corresponding to the level anticrossing shown in Fig. 1a, a nuclear magnetic resonance should be observed at the following frequencies: 12.88 MHz (Al^{27}), 11.88 (Ga^{69}), 15.10 (Ga^{71}), and 8.49 MHz (As^{75}). There is a unique opportunity here to vary the magnetic field corresponding to the level anticrossing and therefore the NMR frequency over broad ranges simply by changing the orientation of the superlattice in the magnetic field. It would also be interesting to study nuclear quadrupole resonance, which looks particularly useful for studying nuclei at interfaces at which there are substantial gradients of the electric field. Studies in this direction are currently being carried out at the A. F. Ioffe Physicotechnical Institute, Russian Academy of Sciences.

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