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Spin polarization of exciton luminescence from ordered $Ga_{0.5}In_{0.5}P$

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Circularly polarized excitation light produces spin-polarized excitons in long-range ordered $Ga_{0.5}In_{0.5}P$ because of a splitting at the valence-band maximum. We observed the spin-relaxation process in ordered $Ga_{0.5}In_{0.5}P$ under resonant excitation of heavy-hole excitons. The circularly polarized exciton luminescence shows a maximum anisotropy of about 53%. The decay profile of the polarized luminescence is described by two components: the rapid decay by exciton-relaxation processes and the slower exciton recombination. The relaxation of the exciton-spin polarization obeys a decay with a time constant of 105 ps. [S0163-1829(98)51424-5]

Spontaneous CuPt-like ordering of $Ga_xIn_{1-x}P$ has been widely observed in vapor-phase epitaxy on a lattice matched GaAs (001) substrate.[1] This ordering has a periodic stack of the column-III sublattice along the [111] or [$\bar{1}\bar{1}\bar{1}$], the two $CuPt_B$ subvariants. All forms of deviations from perfect randomness profoundly affect the material properties especially for the electronic band structure. The presence of a uniaxis generated by the ordering reduces the symmetry from $F\bar{4}3m$ to $R3m$. Then the band structure in the zinc-blende Brillouin zone folds into the reduced superlattice-Brillouin zone. This type of ordering was predicted to cause a splitting at the valence-band maximum (VBM) and a reduction of the fundamental band gap relative to the random alloy.[2–5] With the inclusion of spin-orbit interaction, the valence band split into three twofold degenerate levels, because of the superlattice crystal field. The Γ_{8v} VBM of the random alloy splits into the $\Gamma_{4v,5v}$ and the Γ_{6v} states. The basis functions of heavy holes ($\Gamma_{4v,5v}$) indicate that the Γ_{4v} and Γ_{5v} are degenerate due to time-reversal symmetry.[6] Two Γ_{6v} appear and mix together to give the final states of the light hole and the spin split off band. The splitting of the VBM induces an anisotropy in the intensities of the transition between these split VBM and the conduction-band minimum (CBM).[5] The anisotropic transitions for linear polarization have been observed in many measurements such as photoluminescence (PL),[7–10] PL excitation (PLE),[9,11] electroreflectance,[12] piezoreflectance,[13] photocurrent,[14] and reflectance-difference spectroscopy.[15] Moreover, due to the splitting at the VBM, i.e., $\pm 3/2$-heavy-hole (HH) and $\pm 1/2$-light-hole (LH) splitting, it is possible to excite electrons selectively from the split VBM to the CBM. If this is done under optical pumping conditions with right circularly polarized, say, σ_+ light ($\delta m_j = +1$), only one transition from the $-3/2$HH state to the $-1/2$E state is allowed because of angular momentum conservation. Therefore, if the splitting at the VBM is large enough to allow optical pumping only from the VBM, both the electron and the hole can be 100% spin polarized along the excitation axis at the instant of their creation. According to calculations by Wei and Zunger,[5] photoelectrons generated from the $\Gamma_{4v,5v}$ and the Γ_{6v} states of a single variant crystal of ordered $Ga_{0.5}In_{0.5}P$ alloy are both fully polarized. Since a coupling between the two Γ_{6v} states depends on the degree of ordering, a spin transition intensity $(I_+ - I_-)_{6v}$ is a function of the splitting energy at the VBM, whereas a spin transition intensity $(I_+ - I_-)_{4v,5v}$ does not depend on the degree of ordering.[5] Here, I_+ and I_- are the transition intensities for the spinors parallel and antiparallel to the ordering vector, respectively.

We measured the spin polarization of a band-to-band (''exciton'') recombination in ordered $Ga_{0.5}In_{0.5}P$. The dynamic process of spin relaxation in semiconductors has been extensively studied by static luminescence polarization measurements.[16–18] Such measurements have informed us of spin-relaxation mechanisms of carriers in semiconductors. On the other hand, today, we can directly observe the time-resolved polarization by means of ultrafast laser spectroscopy.[19–26] There are many reports about time-resolved polarization measurements applied to semiconductors, especially for quantum well structures because of enough splitting of the valence bands into HH and LH states in the wells.[19–21,23–26] We observed a spin-relaxation process of exciton luminescence by utilizing a streak camera. The experimental results give unique information about the relaxation of the spin-polarized excitons in ordered $Ga_{0.5}In_{0.5}P$. The decay profile of the polarized luminescence is described by two components: the rapid decay by exciton-relaxation processes and the slower exciton recombination. The relaxation of the exciton-spin polarization obeys a decay with a time constant of 105 ps.

The sample used in this study is a [$\bar{1}\bar{1}\bar{1}$] ordered $Ga_{0.5}In_{0.5}P$ alloy grown by low-pressure metalorganic vapor-phase epitaxy.[9] The degree of ordering was controlled by varying the growth temperature. The sample was grown on a (001) GaAs substrate misoriented 6° off towards $[111]_B$ at 690 °C. The growth rate was 2.0 μm/h at an input gas-flow ratio of $f(\mathrm{V})/f(\mathrm{III})$ of 240. The layer thickness is 2 μm. The order parameter can be estimated by the valence-band splitting energy because the spin-orbit splitting and the crystal-

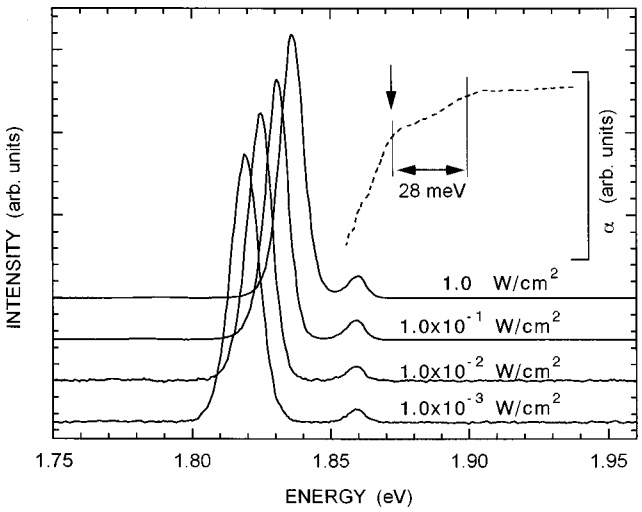

FIG. 1. PL spectra measured at various excitation powers of the Ar-ion laser. PLE of the low-energy PL line is plotted by the dashed line. The arrow indicates the excitation energy in our time-resolved measurements.

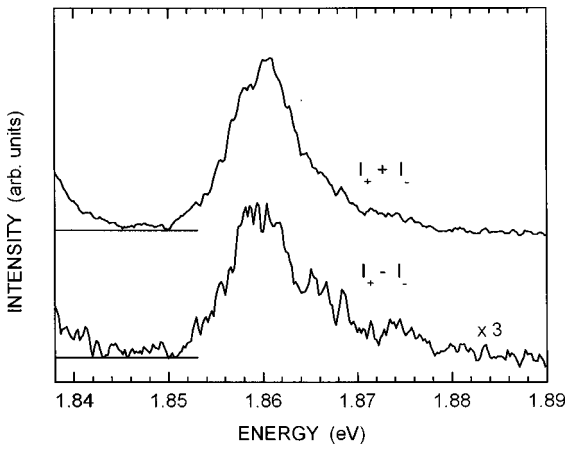

FIG. 2. Time integrated spectra of the depolarized PL $I_+ + I_-$ and the non-normalized degree of spin polarization $I_+ - I_-$ when excited with σ_+ light.

field splitting are functions of the order parameter.[2–4] The valence-band splitting measured by PLE measurements yields an order parameter of 0.51 for this sample. Conventional PL and PLE were measured by a lock-in detection system under excitation by the 488-nm line of an Ar-ion laser and a monochromatic light of a Xe lamp, respectively. Ultrafast laser spectroscopy was performed using 250-fs pulses from a doubler of an optical parametric oscillator excited by a mode-locked Ti:sapphire laser with a repetition rate of 80 MHz. The laser wavelength is 663 nm, which corresponds to the absorption edge of the HH exciton. The excitation density is 2.0 μJ/cm^2 corresponding to the sheet carrier density of 6.8×10^{12} cm^{-2}. Here we used σ_+ light for the excitation. The incident direction of the laser light was normal to the (001) surface, and the emitted luminescence was detected at the same direction. The PL is dispersed in a 0.25-m spectromator and detected with a spatial and temporal resolution of 0.15 nm and about 5 ps due to dispersion in the collection optics, respectively, by a streak camera with two-dimensional readout. The samples were mounted in a He-gas-flow cryostat. The sample temperature was 3.6 K.

Figure 1 shows depolarized PL spectra measured at various excitation powers of the Ar-ion laser. The ordered $Ga_{0.5}In_{0.5}P$ shows two separated peaks that will be referred to as the ''high-energy'' and ''low-energy'' PL lines. The high-energy PL line does not show any excitation density dependence. Whereas the high-energy PL line exhibits all features typical for band-to-band recombination, the low-energy PL line shifts to higher emission energies with increasing excitation intensity. In addition, the temporal decay dynamics is nonexponential.[9] All these properties of the low-energy PL line can be consistently explained by assuming spatially indirect recombination. The dashed line in Fig. 1 plots the PLE spectrum of the low-energy PL line. Two excitonic absorption maxima are resolved that correspond to the valence-band splitting. The splitting is 28 meV, which gives an order parameter of 0.51. Comparison of the PL and PLE spectra points to the fact that the high-energy PL line represents the excitonic band-to-band recombination with the Stokes shift of about 10 meV.

Spin-polarization properties of the high-energy PL line, i.e., exciton PL, were investigated by the ultrafast laser spectroscopy. The excitation energy of the pulse laser was set at the HH-exciton energy as indicated by an arrow in Fig. 1. Figure 2 plots time integrated spectra of a depolarized PL, $I_+ + I_-$, and a non-normalized degree of spin polarization, $I_+ - I_-$, when excited with σ_+ light. Here I_+ and I_- are the intensity of right and left circularly polarized PL, respectively. The polarization peak is attributed to the HH-exciton PL. The maximum polarization at the peak position is about 53%. The linewidth of the polarized PL is almost the same as one of the depolarized PL. In this experiment, a relative shift between the polarized and depolarized spectra was not observed. If the sample is excited by a sufficient density, the excitons will have an energy distribution because of the exclusion principle acting separately on the electrons and the holes.[25] The $|+1\rangle$ exciton experiences a repulsion, while the $|-1\rangle$ exciton an attraction. The energy difference depends on the density of excitons. Our results, which show no energy splitting, indicate that the exciton-spin polarization is not influenced by many-body effects under our excitation condition. Since the lifetime of the low-energy PL line is on the order of a μs,[9] and the maximum time window of our streak camera is 2 ns, detailed polarization analysis for the streak-camera data about the low-energy PL line is not performed. A static spin polarization obtained by PLE of the low-energy PL line, on the other hand, does not show any structure around the HH and LH excitonic transitions. This result indicates that the spin memory is completely lost during a localization of photoexcited carriers in a fluctuated bad gap of the $Ga_{0.5}In_{0.5}P$ film.

Figure 3 shows time resolved I_+ (solid line) and I_- (dashed line) of the exciton PL. The inset of this figure plots a depolarized PL decay. We observed a rapid decay of the depolarized PL, which can be attributed to a rapid capture of excitons into some nonradiative deep centers. For $t>150$ ps, one gets in the curve a simple exponential decay with a time constant of 220 ps. The observation of the rapid decay is a typical feature of exciton PL under resonant excitation.[27] A similar trend can be observed in the I_+ and I_- decays. For the I_+, the rapid decay is caused by a loss of the spin memory together with the rapid capture process of excitons.

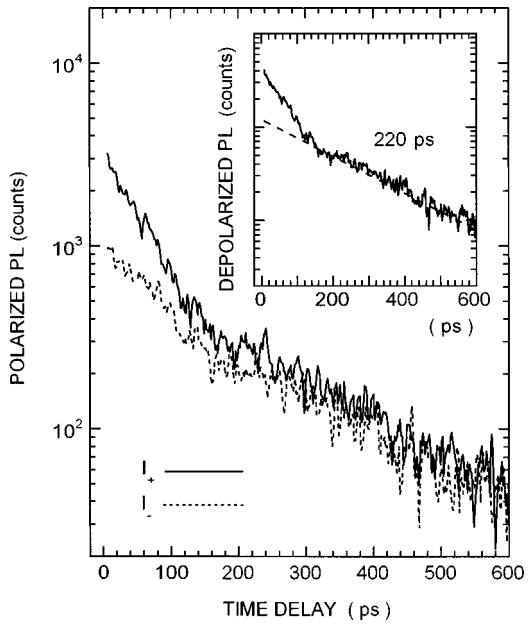

FIG. 3. Time resolved I_+ (solid line) and I_- (dashed line) of the high-energy PL line. The inset shows the depolarized PL decay.

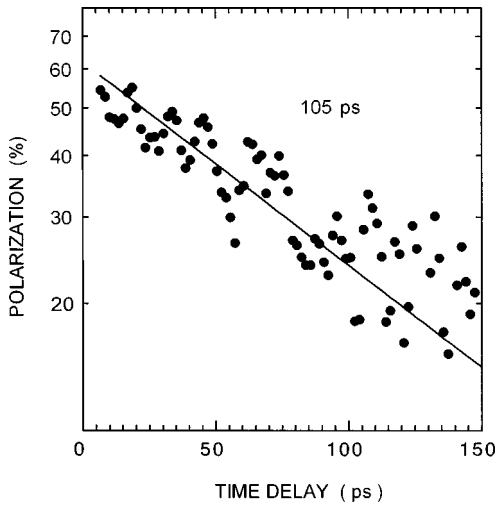

FIG. 4. Decay profile of the spin polarization defined by $(I_+ - I_-)/(I_+ + I_-)$. The relaxation of the exciton-spin polarization obeys a decay with a time constant of 105 ps.

On the other hand, at the beginning of the I_- decay the decrease is not so rapid because of a creation process of ''antiparallel-spin excitons.'' After 150 ps, both I_+ and I_- show the same decay. The dynamic anisotropy between I_+ and I_- indicates the exciton-spin relaxation process. The spin relaxation is only observed in the time region of the exciton-capture process, $t<150$ ps. A decay profile of a normalized spin polarization defined by $(I_+ - I_-)/(I_+ + I_-)$ is plotted in Fig. 4. The relaxation of the exciton-spin polarization obeys a decay with a time constant of 105 ps. Although the theoretical calculation suggests a full polarization for the fundamental transition at the instant of the carrier generation,[5] the measured degree of the polarization is reduced at the beginning of the PL decay. This may be explained by a rapid relaxation of the spin polarization during the Stokes shift and the deviation of the angular momentum of the excitation light from the ordering direction that is 55° from the [001] toward the [110]. A static spin polarization of the HH-exciton luminescence, in the present case of selective $-3/2\mathrm{HH}\rightarrow -1/2\mathrm{E}$ pumping, is given by $(1+\tau_r/\tau_{sp})^{-1}$, if we assume a simple decay with τ_r of the recombination time and τ_{sp} of the spin-relaxation time.[18] The evaluated τ_r and τ_{sp} from the time-resolved measurements give a static spin polarization of 55%. This value agrees with the degree of the exciton-spin polarization (about 53%) obtained from the time integrated spectra in Fig. 2.

In summary, we observed an exciton-spin relaxation process in ordered $Ga_{0.5}In_{0.5}P$ under resonant excitation of HH excitons. The circularly polarized excitonic PL spectrum shows a maximum anisotropy of about 53%. The polarization peak is attributed to the HH-exciton PL. The decay profile of the polarized PL is described by two components: the rapid decay by exciton-relaxation processes and the slower exciton recombination. The relaxation of the exciton-spin polarization obeys a decay with a time constant of 105 ps.

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