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Reversal of spin polarization direction in excitonic photoluminescence of AlGaAs

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Abstract – An anomalous reversal of the direction of exciton spin polarization is found in an optical-orientation study of $Al_xGa_{1-x}As$ alloys. A negative degree of circular polarization of excitonic photoluminescence is observed in direct-gap AlGaAs under excitation with circularly polarized light with the photon energy lying in a narrow range just above the free-exciton transition. The resonant switching of the polarization direction is attributed to the effect of the minor residual strain in the epitaxial film that results in lifting of the degeneracy of the light- and heavy-hole sub-bands. The electron g-factor is estimated from combined time-resolved and Hanle-effect data and amounts to 0.056–0.086 for $x = 15\%$ alloy.

The optical orientation of carrier spins in semiconductors [1] is increasingly used in the studies of spin relaxation and transport phenomena since its development until these days (see, *e.g.*, refs. [2,3]). By monitoring the dynamics of the degree of circular polarization of the photoluminescence (PL) after a pulsed-laser excitation, or as a function of a magnetic field rotating the spins in a *cw* experiment, one is able to track the relaxation kinetics and investigate the mechanisms of spin transfer. In general, along their relaxation path, the non-equilibrium carrier spins gradually lose the orientation transferred from the excitation photons to finally return to a non-polarized state. In a bulk material this leads to a gradual decrease in the degree of PL polarization, while the direction of polarization is preserved, provided carriers were created near the fundamental band gap without participation of the higher-lying spin-orbit split hole band [4]. In contrast to this "normal" behaviour, in this letter we report the observation of a resonant reversal of the polarization direction in the excitonic PL of uniform AlGaAs alloys, manifesting the need to consider finer details of the excitonic energy structure and spin transfer paths. As we will show, the direction of excitonic PL polarization in direct-gap AlGaAs alloys switches to the opposite upon excitation in a narrow range of wavelengths close to the excitonic transition. This polarization direction switching displays the need to take into account even the minor lattice strain present in this almost-lattice matched material system that leads to a significant modification of the structure of excitonic levels.

The samples studied in this work were double heterostructures grown on (100) GaAs by molecular beam epitaxy, with 2.5 μm high-purity layers of $Al_xGa_{1-x}As$ (x_{AlAs} up to 15%) sandwiched between 25 nm layers of AlAs. The details of sample growth and characterization were reported in ref. [5]. The high quality of the layers is established by a linear dependence of the free-exciton PL intensity on the excitation density in a wide range of excitation power of more than six orders of magnitude, and by the ratios of excitonic to shallow impurity-related transitions intensities ranging from 100 : 1 to 1000 : 1 at 4.2 K [5]. Steady-state PL was excited by circularly polarized radiation of a tuneable $Ti:Al_2O_3$ or discrete

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wavelength lasers and detected by a photomultiplier. To study the Hanle effect, a magnetic field was applied in the Voigt geometry so that it lay in the sample plane and the laser excitation and PL beam directions were parallel to the film growth axis and perpendicular to the field direction. To avoid nuclear magnetization, the direction of circular polarization of the laser beam was modulated between the two orthogonal directions by a photoelastic modulator. Additionally the beam was mechanically chopped. Using two lock-in amplifiers, this allowed for the recording of both the sum $I_\Sigma = (\mathrm{I}_{\sigma+} + I_{\sigma-})$ and difference $I_\Delta = (\mathrm{I}_{\sigma+} - I_{\sigma-})$ of the intensities of the PL components polarized in the same (σ^+) and orthogonal (σ^-) directions with respect to the laser beam. In what follows, to characterize the PL polarization state we will use either the PL polarization degree $\rho = I_\Delta/I_\Sigma$, or directly the registered I_Δ, since ρ is prone to experimental errors if small PL intensities are considered. Time-resolved PL measurements were carried out using a mode-locked $\mathrm{Ti:Al_2O_3}$ laser (≈ 1 meV spectral width, 2 ps pulses) and a Hamamatsu synchroscan streak camera.

Figure 1 shows the spectra of excitonic PL intensity I_Σ (black solid line) and difference I_Δ (blue dashed and red dash-dotted lines) signals taken on a $x_{\mathrm{AlAs}} = 15\%$ sample. The spectra are dominated by the free-exciton transition (FX), while weaker lines below it are attributed to bound excitons. When the laser photon energy is tuned from above-bandgap excitation (1.7482 eV, upper traces) to 3 meV above the FX line position (1.7318 eV, lower trace), marked as $\hbar\omega_{\mathrm{neg}}$, a reversal in the sign of I_Δ is observed, which indicates that the overall direction of orientation of exciton spins is inverted without application of an external magnetic field. The reversal of the PL polarization direction occurs simultaneously for all the lines in the spectrum, free and bound, and is observed in a range of laser intensities spanning more than five orders of magnitude. The negative polarization degree of excitonic PL was found in all the layers with x_{AlAs} up to 15% (the study was limited to this concentration by the available excitation energy range). The inset to the figure shows the FX polarization degree *vs.* excitation photon energy taken on samples with $x_{\mathrm{AlAs}} = 3$, 9, and 15%. The reversal of the polarization direction takes place in narrow (less than 1.6 meV) ranges of excitation energies 2–4 meV above the FX transitions, which are indicated by arrows. Negative values of ρ as high as -12% are found in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ starting from $x_{\mathrm{AlAs}} = 1.5\%$, while in layers of GaAs ($x_{\mathrm{AlAs}} = 0$) the PL polarization degree remained positive. The resonant-like behaviour indicates that electronic states in a narrow range of energies close to the bandgap control the generation and relaxation of carrier spin in this system.

In the same ranges of photon energies where the reversal of polarization direction takes place for each excitation spectrum shown in the inset to fig. 1, an additional fine feature appears in the PL spectra taken under non-resonant above-bandgap excitation. Figure 2

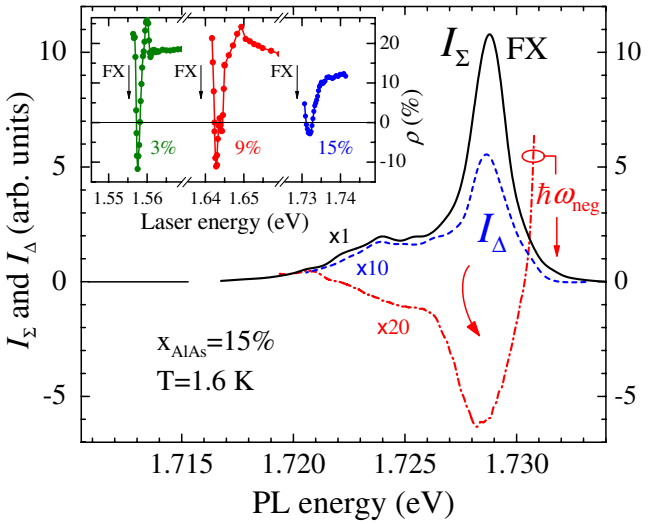

Fig. 1: (Colour on-line) Spectra of the sum I_Σ (PL intensity, solid line) and difference I_Δ (dashed and dash-dotted lines) signals taken on the $x_{\mathrm{AlAs}} = 15\%$ sample. Upper traces: excitation energy 1.7482 eV; lower trace (dash-dotted line, marked as $\hbar\omega_{\mathrm{neg}}$): 1.7318 eV. Excitation power density: 20 W/cm^2. Inset: spectra of the free-exciton (FX) polarization degree *vs.* excitation photon energy for samples with $x_{\mathrm{AlAs}} = 3\%$, 9%, and 15%.

shows a magnified view of the high-energy tail of an I_Δ spectrum of the $x_{\mathrm{AlAs}} = 15\%$ sample taken with an above-bandgap excitation. It is seen that the resonant reversal of the polarization sign in the excitation spectrum (a) coincides with a minimum in the I_Δ spectrum (b). This minimum is observed in a wide range of excitation photon energies, ranging to at least 200 meV above the bandgap. Although the values of I_Δ are positive, the signal around the minimum arises from two contributions: a negative contribution of some transition similar to that described above, and a positive background due to the high-energy tail of the FX line, which red-shifts the minimum, and the strongly polarized laser stray light. This is corroborated by Hanle-effect data for PL energies corresponding to the minimum (see inset in fig. 2), which show that, contrary to the well-known behaviour, the application of magnetic field results not in quenching but in an increase in ρ. This demonstrates that in the same narrow range of PL energies the spin polarization is reversed under non-resonant excitation.

The dynamics of the excitonic transitions for the $x_{\mathrm{AlAs}} = 15\%$ sample under excitation resulting in negative ρ's, dubbed in the following as resonant excitation, is illustrated in fig. 3. The time-resolved measurement shows that switching from above-bandgap to resonant excitation leads to the reversal in the FX polarization direction, but does not result in significant changes in the excitonic lifetime and polarization decay time, which amount to 780 ps and 400 ps, respectively. The *cw* Hanle curves of the FX transition taken with both the above-bandgap and resonant excitations (see inset in fig. 3) show wide

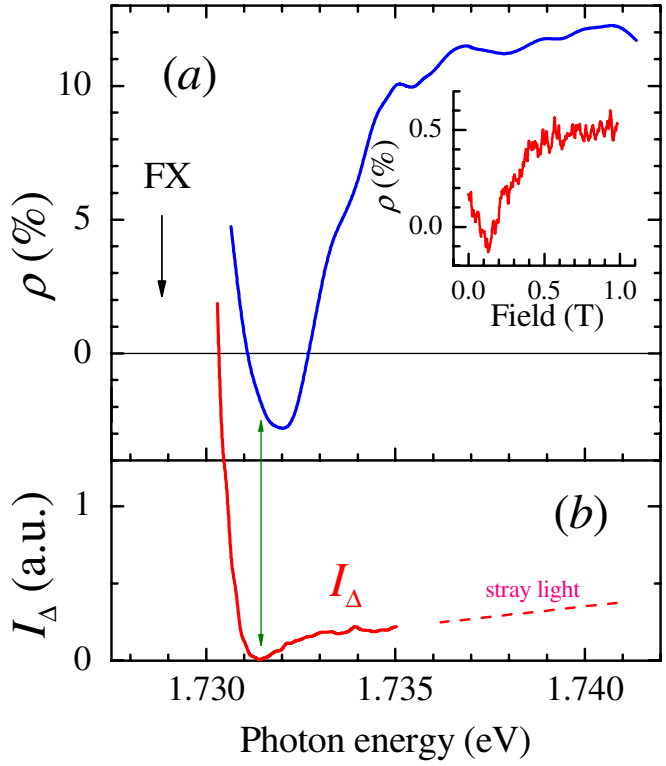

Fig. 2: (Colour on-line) Spectra of (a) free-exciton (FX) polarization degree *vs.* excitation photon energy, and (b) high-energy tail of the I_Δ signal *vs.* PL photon energy with above-bandgap excitation at 1.7482 eV, shown *vs.* a common abscissa axis for comparison. Sample: $x_{\mathrm{AlAs}} = 15\%$; temperature: 1.6 K. The arrow indicates the spectral position of the FX line maximum, where the excitation spectra shown in (a) is recorded. Inset: Hanle curve taken at a PL energy of 1.7313 eV under the above-bandgap excitation.

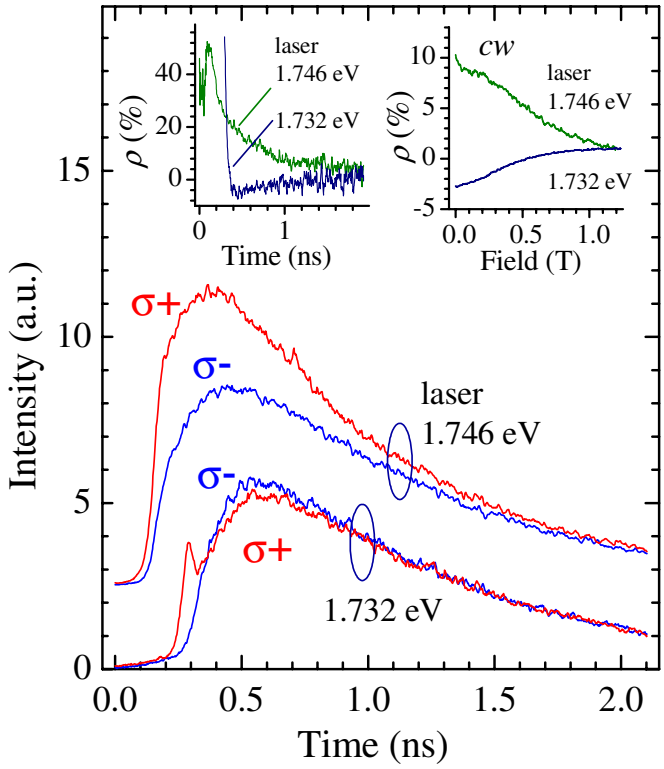

Fig. 3: (Colour on-line) Dynamics of the free-exciton transition under circularly polarized above-bandgap 1.746 eV (upper curves, shifted both vertically and horizontally for clarity) and resonant excitation (1.732 eV, lower curves). Temperature: 5 K; detection energy: 1.7288 eV; excitation density: 4 nJ/cm^2 per pulse. The insets show the corresponding decays of the PL polarization degree (left), and *cw* Hanle curves taken with the above-bandgap and resonant excitations at 1.6 K (right).

Lorentzian contours with a gT_S product of 15 ps and 23 ps, respectively, where g is the g-factor and T_S is the total spin lifetime. An additional weaker narrow contour with $gT_S = 275$ ps is observed at lowest fields under non-resonant excitation. The relative contribution of this contour increases with increasing excitation power. This additional contribution is attributed to hot carrier thermalisation, while the wide contours correspond to the final stage of spin relaxation. The electron g-factor, estimated from combined time-resolved and Hanle-effect data, is in the range 0.056–0.086 for the $x = 15\%$ alloy. Assuming that the light-hole (LH) and heavy-hole (HH) degeneracy is lifted (see model below) and that thus, similarly to 2D [6], the contribution to the in-plane g-factor of heavy holes is nearly zero, these values would correspond directly to the electron g-factor, in good agreement with the data of Weisbuch and Hermann for an $x_{\mathrm{AlAs}} = 15\%$ alloy [7].

Reversal of the spin orientation direction under near-bandgap excitation has not been reported so far for a comparable uniform bulk material. On the other hand, similar observations are known in low-dimensional structures like quantum wells [8–12], quantum dots [13] and microcavities [14]. In low-dimensional structures, size quantization lifts the degeneracy of the LH and HH levels, which results in the inversion of the spin orientation direction under excitation resonant with the LH exciton. The same origin can be attributed to our experimental observations in AlGaAs, if the LH and HH levels are no longer degenerate as they are in GaAs (fig. 4), and one takes into account the high spin relaxation rates of holes [1,15]. In the following, though the effective masses of holes in the sub-bands split by quantization or strain might not differ as strongly as they do in bulk GaAs around the zone center [16], we will still designate the sub-bands with hole angular-momentum projections of 1/2 and 3/2 as LH and HH, respectively. In order to illustrate the generation and recombination of both free carriers and excitons, fig. 4 sketches together excitonic and single-particle states, plotted against the electron momentum projection. A non-resonant above-bandgap polarized excitation (denoted as NR) produces free electrons with spins oriented predominantly in the direction defined by the HH channel, and the PL polarization direction of the dominating HH exciton corresponds

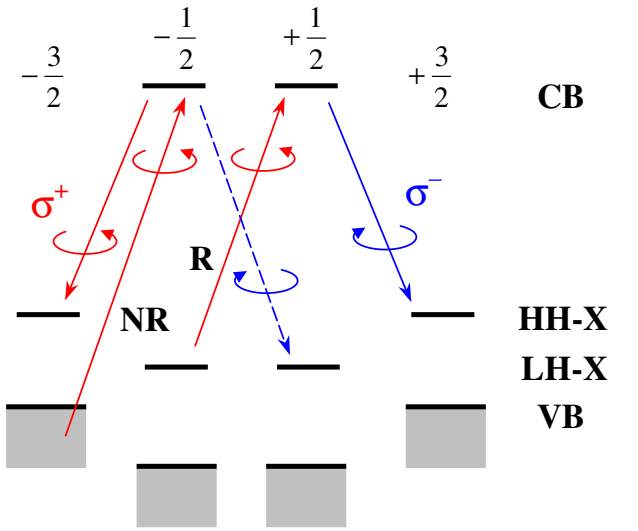

Fig. 4: (Colour on-line) Schematic diagram of transitions with split light- and heavy-hole states (not to scale). CB and VB: conduction and valence band continuum, respectively; HH-X and LH-X: heavy- and light-hole exciton levels, respectively; NR: non-resonant excitation; R: excitation in resonance with LH-X; σ^+ and σ^-: recombination transitions.

to that of the excitation (σ^+). Additionally, a weak σ^--polarized higher-energy LH exciton luminescence will be produced if a small amount of LH excitons will be generated during thermalisation of holes (the dashed arrow, cf. fig. 2). In contrast, with excitation tuned to the resonance with the LH exciton transition (denoted as R), the spins of electrons in the photogenerated excitons are reversed. After thermalisation of holes, which should include their conversion from LH to HH branches, this will result in σ^--polarized HH exciton PL. This breaking of the degeneracy between the HH and LH sub-bands explains the reversals of the PL polarization direction shown in figs. 1 and 2.

The origin of the splitting of the LH and HH levels considered in this model can be twofold. The first possibility, a splitting due to size quantization, seems quantitatively unlikely in the layers since the potential modulations due to alloy disorder effects are too small [17] to account for the experimental data. An alternative reason for the splitting is lattice strain in the epitaxial film. An elastically deformed AlGaAs film on a GaAs substrate is uniformly compressed in the film plane and expanded along the growth direction. In this case the HH exciton level is the ground state, which agrees with the model of fig. 4. We calculated the splitting of the HH and LH levels following the approach of ref. [18] for computation of the effective constants of exciton deformation potentials in a weakly strained crystal. The exciton wave functions were considered in the spherical model taking into account the valence band degeneracy. The alloy broadening that leads to increased linewidths in ternary films was not taken into account. The wave functions of the exciton ground state were used to calculate a renormalization coefficient applied to the constants of deformation potentials of free holes. By means of the effective constants thus found and using the alloy parameters of ref. [19] the HH-LH splitting of exciton levels was calculated to be 1.4 meV for $x_{\mathrm{AlAs}} = 15\%$. The splitting of the exciton absorption line into two components separated by approximately 2 meV observed recently in the 15% sample in optical transmission, and ascribed to the influence of strain [20], agrees with our PL data and the computation result. This evidences that the residual strain, often disregarded in the AlGaAs/GaAs material system, can result in a strong modification of the spin transfer paths. In turn, this property can possibly be applied to the creation of "spin-preserving" carrier transport elements for future quantum computer schemes.

In summary, a resonant reversal of the optical spin orientation direction has been found in direct-gap AlGaAs for excitation photon energy in a narrow range just above the free exciton transition. The switching of the polarization direction is attributed to the splitting of the heavy- and light-hole excitonic levels due to the lattice strain between the epitaxial film and the substrate.

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