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Individual neutral and charged $In_xGa_{1-x}As$-GaAs quantum dots with strong in-plane optical anisotropy

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We report neutral (X^0) and charged excitons (X^+) with high degrees of linear polarization ($\geqslant 70\%$) in charge-tunable InGaAs quantum dots (QDs). The QD emission exhibits a small Zeeman splitting for magnetic fields parallel to the growth direction, in contrast to "conventional" dots in the same sample, and remains predominantly linearly polarized up to 5 T. With the aid of in-plane field measurements, the observations are explained in terms of heavy–light-hole mixing due to QD anisotropy. This results in elliptical polarization of the QD emission and strong reduction of the exciton g factor. A combination of the data obtained for magnetic fields in Faraday and Voigt configurations allows a full determination of the electronic properties.

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The spin excitations of single quantum dots (QDs) have considerable potential for use as quantum bits[1] and for long-lived memory devices.[2] However, in real QDs, the ground eigenstates are not pure spin eigenstates, since the confinement potential does not possess axial symmetry. Due to electron-hole exchange, linearly polarized, mixed spin eigenstates[3] with splitting in the range $\sim 10-800$ μeV are formed.[4,5] As a result, exciton-based spin memories at zero magnetic field can only be achieved in charged dots where the exchange interaction is quenched.[2]

On the other hand, linearly polarized emission from QDs is desirable for a variety of applications, including single-photon sources with determined polarization. In-plane polarized emission has been found for arrays of arrowlike InAs dots deposited on (311) GaAs (Ref. 6) and for vertically coupled QD stacks.[7] Free-standing InP nanowires[8] and CdSe quantum rods[9] with up to 90% linearly polarized emission have been reported. A linear polarization degree of $\rho_l \sim 30\%$ of the trion in CdSe/ZnSe QDs was explained by heavy-hole (HH)–light-hole (LH) mixing.[10] Very recently, Favero *et al.*[11] observed InAs QDs with large ranges of ρ_l up to a maximum of 82%, a result suggested to be an intrinsic feature of dilute quantum-dot arrays.[12] Apart from InP nanowires, where the high linear polarization is explained in terms of band-structure properties,[13] in much of this work the nature of the electronic states giving rise to the linear polarization remains unexplored.

In this paper we report high degrees of linear polarization of $\geqslant 70\%$ for both neutral and positively charged excitons in dilute InGaAs/GaAs QD arrays (<10 QDs/μm^2). The behavior is investigated by applying magnetic fields parallel (Faraday) and perpendicular (Voigt geometry) to the growth axis. The strongly linearly polarized emission exhibits relatively small Zeeman splitting in the Faraday geometry, in contrast to "conventional" dots with small polarization in the same samples. The linear polarization is attributed to heavy–light-hole mixing caused by in-plane QD anisotropy, which also results in a strong reduction of the hole and, thus, the exciton g factor in the growth direction. The electron and hole g factors are evaluated from a combination of the data in magnetic field in Voigt and Faraday geometries.

The sample structure is described in Refs. 14 and 15. The diode structures [Fig. 1(a)] enable controlled hole charging of the dot from the p contact. Single dots were isolated through 500–800 nm apertures. Photoluminescence (PL) was recorded at $T=5$ K using linearly polarized laser excitation at ~ 100 meV above the QD ground state. QDs with ρ_l from 0% up to 80% were observed.

Figure 1(b) shows the emission of strongly linearly polarized dot A, consisting of two peaks. Spectra are recorded at a reverse bias $V=0.6$ V for two orthogonal linear polarizations π_x, π_y with directions close to $[1\bar{1}0]$ (x) and $[110]$ (y), respectively, and $\rho_l = I(\pi_x)\text{-}I(\pi_y)/I(\pi_x)+I(\pi_y)$ of both peaks of

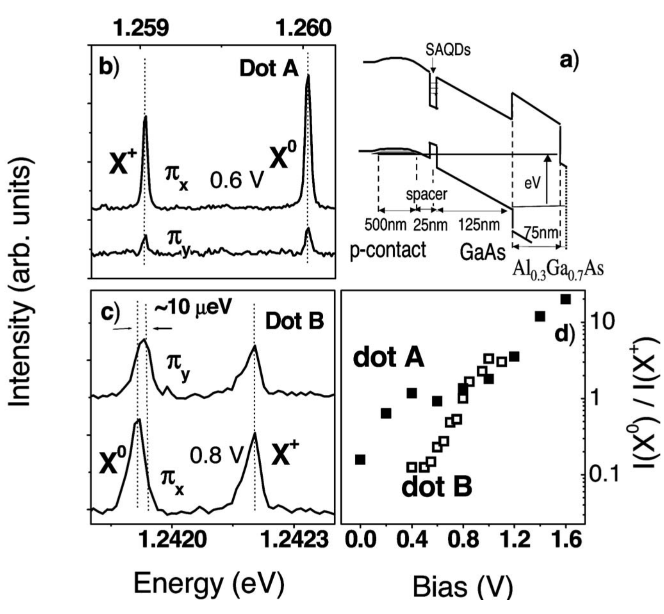


FIG. 1. (a) Schematic band diagram of Schottky photodiode. (b), (c) PL spectra of quantum dots A (b) and B (c) recorded for two orthogonal linear polarizations at applied biases of 0.6 and 0.8 V, respectively. Neutral and positively charged excitons, X^0 and X^+, are observed. (d) Ratio of the integrated intensities of X^0 and X^+ for B (open squares) and A (solid) QDs as a function of reverse bias.

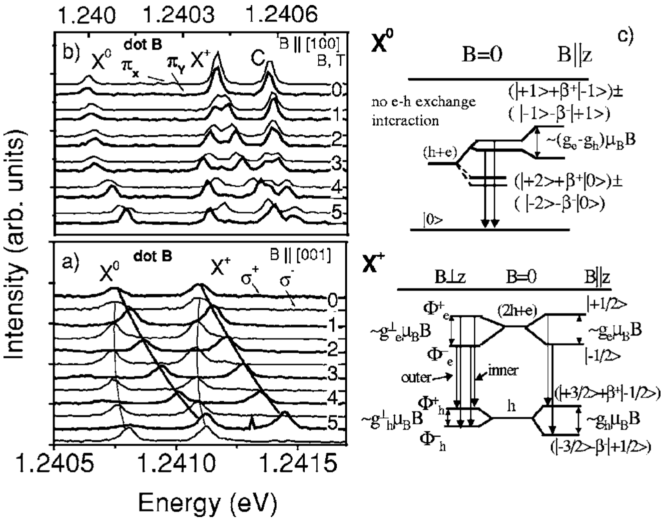
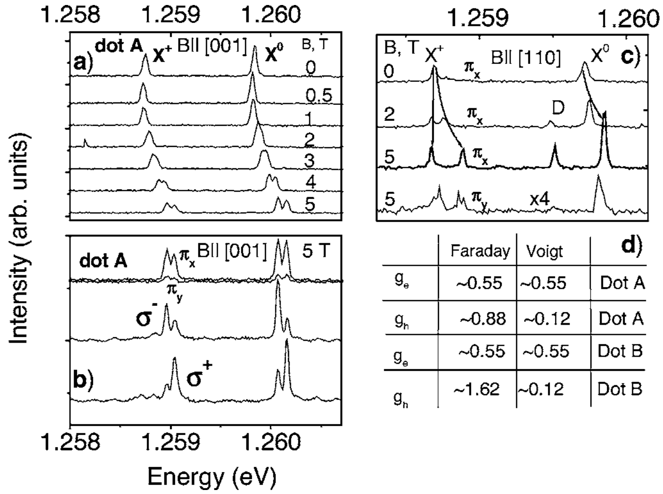


FIG. 2. (a) X^0 and X^+ PL spectra for QD B in magnetic field in the Faraday configuration ($B\|z$), for σ^+ and σ^- circular polarizations at reverse bias 0.8 V. (b) X^0 and X^+ PL spectra for QD B in magnetic field in the Voigt configuration ($B\|[100]$) for two crosslinear polarizations (π_x, π_y). The spectra were measured at 0.4 V, where emission (line C) attributed probably to X^{2+} is observed (see Ref. 15). (c) Schematic transitions of neutral and positively charged excitons.



	Faraday	Voigt	
g_e	~0.55	~0.55	Dot A
g_h	~0.88	~0.12	Dot A
g_e	~0.55	~0.55	Dot B
g_h	~1.62	~0.12	Dot B

FIG. 3. (a) X^0 and X^+ of PL spectra for QD A in magnetic field in the Faraday configuration ($B\|z$) at 0.8 V. (b) PL spectra for X^0 and X^+ recorded for crosslinear (π_x, π_y) and circular (σ^+, σ^-) polarizations at reverse bias 0.8 V at 5 T. (c) X^0 and X^+ PL spectra for QD A in the Voigt configuration ($B \perp z$) for two crosslinear polarizations (π_x, π_y) for $B\|[110]$ at 0.7 V. (d) Summary of electron and hole g factors for $B\|z$ and $B \perp z$ for QD A and QD B.

~70%. The intensities of both peaks vary linearly with excitation power. The polarization of the PL was found to be independent of the plane of polarization of the excitation. The ratio of the higher- to the lower-energy peaks grows monotonically with V, as shown in Fig. 1(d) (solid squares). This identifies the higher- and lower-energy peaks as neutral (X^0) and singly (X^+) charged states: at low V the probability of occupancy from the contact is high, favoring X^+, whereas at high bias the dot will be empty, favoring X^0 (Ref. 15).

Figure 1(c) shows spectra for a weakly linearly polarized QD B. The spectra again consist of two peaks. The ratio of the lower- and higher-energy peaks decreases monotonically with decreasing V, similarly to the dependence of X^0 and X^+ for QD A. We thus attribute the lower- and higher-energy peaks to X^0 and X^+ (Ref. 16). X^0 in this case consists of a doublet with components linearly polarized along $[1\bar{1}0]$ (x) and $[110]$ (y), with a splitting of ~10 μeV determined by the anisotropic exchange interaction [Fig. 2(c)]. By contrast X^+ exhibits no splitting, as expected for positively charged excitons where the exchange interaction is quenched since the two holes have antiparallel spin. The polarization axes of QD B cannot be determined precisely due to the small linear polarization degree of both peaks of ~15–20 %. The X^0 and X^+ identifications for both QDs A and B are further confirmed from the studies in magnetic field.

The high degree of linear polarization for dot A and similar dots arises from heavy-hole–light-hole mixing[10] due to QD in-plane anisotropy. The hole wave function for a QD in low ($x \neq y, C_2$) symmetry is of the form $\psi_h^{\pm} = |\pm 3/2\rangle \pm \beta^{\pm} |\mp 1/2\rangle$, with the mixing coefficient $\beta^{\pm} \sim \gamma^{\pm} \exp(\pm i2\Theta)/\Delta E_{l\text{-}h}$. $\Delta E_{l\text{-}h}$ is the splitting between heavy- and light-hole ground states and $\gamma^{\pm} \exp(\pm i2\Theta)$ is determined by shear strain, confinement, and chemical composition.[10] The QD transition becomes elliptically polarized with axes determined by the in-plane QD symmetry. The X^+ and X^0 states in the presence of HH–LH mixing are depicted in Fig. 2(c). The isotropic and anisotropic spin exchange interactions lift the degeneracy of the pure spin states, resulting in fine-structure splitting (FFS) of the X^0 quartet into bright and "dark" states, which are linear superpositions $(|+1\rangle + \beta^+| -1\rangle) \pm (|-1\rangle - \beta^- |+1\rangle)$ and $(|+2\rangle + \beta^+ |0\rangle) \pm (|-2\rangle - \beta^- |0\rangle)$, respectively. QDs in our sample exhibit FFS in the range from 0 to 30 μeV. We did not observe any fine-structure splitting of X^0 for QD A, indicating small anisotropic exchange.[17] From the experimental polarization degree, using the expression in Ref. 10 we estimate $\beta \sim 0.7$ for QD A. Although the polarization degree of QD B is small, indicating β less than 0.2, the HH–LH mixing is finite as shown below.

To better understand the above behavior, magnetic-field studies were performed. Figure 2(a) shows spectra for X^0 and X^+ of weakly polarized dot B from 0 to 5 T in the Faraday configuration ($B\|z$). Both X^0 and X^+ split into two oppositely circularly polarized (σ^+, σ^-) lines, with identical Zeeman splitting of 0.32 meV at $B=5$ T. The identical Zeeman behavior arises since X^0 consists of one electron and one hole in the initial state, and X^+ of one electron in the initial state (the total hole spin is zero), and one hole in the final state [see Fig. 2(c)]:[4] in both cases the observed splitting is given by the difference of the electron and hole splittings $\Delta E = \mu_B (g_e\text{-}g_h) B$ (g_e and g_h are electron and hole g values).

The behavior of the strongly polarized QD A for $B\|z$ is very different. Both X^0 and X^+ again show very similar behavior [Fig. 3(a)]. However, the Zeeman splitting is only 0.08 meV at 5 T, a factor of 4 smaller than for dot B. Spectra at 5 T are shown in Fig. 3(b) for π_x, π_y linear and σ^+, σ^- circular polarizations. Even at high field, the doublet remains ~70% linearly polarized. For circularly polarized detection, ρ_c of ~50% is found [Fig. 3(b)]. The strong reduction of the

Zeeman splitting in $B \parallel z$ relative to weakly polarized dots is a general observation. For example, we observed emission of three QDs with $\rho_l > 50\%$ with Zeeman splitting less than 0.2 meV at 5 T, in contrast to dots with $\rho_l < 30\%$, which exhibited splittings at 5 T in the range 0.3–0.4 meV.

Experiments in the Voigt geometry ($B \perp z$) provide complementary information, and enable full determination of the g values and key features of the electronic structure. Figure 2(b) shows PL spectra for X^0 and X^+ of weakly polarized dot B for $B \perp z$ from 0 to 5 T. X^0 splits into a doublet with small energy splitting between the components of 0.04 meV at 5 T. By contrast X^+ splits into a quadruplet with splitting between the outer and inner lines of 0.21 and 0.14 meV, respectively. The Zeeman pattern had nearly isotropic character for rotation of the sample about the growth direction: the lower (higher)-energy component of the X^0 doublet and the inner (outer) components of the X^+ quadruplet were found to emit in crosslinear polarizations rotated by 20°–30° from [110], $[1\bar{1}0]$.

The behavior of the strongly polarized QD A for $B \perp z$ is very similar. As seen from Fig. 3(c), X^+ and X^0 split into a quadruplet and doublet for $B \parallel [110]$. The splitting between the inner (outer) lines of the X^+ quadruplet and between the components of the X^0 doublet is 0.21 (0.14) and 0.033 meV at 5 T, respectively, nearly the same as for QD B. However, in this case, the X^+ quadruplet has polarization axes which depend on the direction of the field; for instance, the inner and outer lines are crosslinearly and colinearly polarized for $B \parallel [110]$ and $B \parallel [100]$, respectively, with polarization axes close to the [110], $[1\bar{1}0]$ directions.

To analyze the results in magnetic field, we consider the Hamiltonian of the electron and hole, $H_B = a\vec{S}_e.\vec{J}_h + b\vec{S}_e.\vec{J}_h^3 + \mu_B g_0(\vec{S}_e.\vec{B} + \kappa \vec{J}_h.\vec{B} + q\vec{J}_h^3.\vec{B})$, where $\vec{S}_e, \vec{J}_h$ are electron, hole angular momentum operators, the first two terms arise from e-h exchange, and κ and q are Luttinger parameters for the Zeeman splitting of the valence band. For $B \perp z$, the electron and hole spin states precess around the in-plane magnetic field, with the result that the states are no longer pure spin eigenstates. For X^+, where there is no exchange splitting, this results in splitting into a quadruplet since all four Zeeman transitions are now allowed, in contrast to $B \parallel z$, as shown in Fig. 2(c). This further confirms the identification of the X^+ line, and enables g_e and g_h to be determined separately. The splitting for $B \perp z$ between the outer and inner components of X^+ is given by $\mu_0 B(g_e^\perp + g_h^\perp)$ and $\mu_0 B(g_e^\perp - g_h^\perp)$, allowing the g values in Fig. 3(d) to be deduced. The $g_e^\perp$, $g_h^\perp$ values are very similar for dots A and B as expected from the similar $B \perp z$ splittings.

In the presence of HH–LH mixing, the κ term of the hole Zeeman Hamiltonian leads to first-order magnetic coupling between hole states for $B \perp z$, resulting in QD PL emission polarized parallel and perpendicular to the QD symmetry axes. This is the case for the weakly polarized dot B where the polarization axes are not associated with the direction of the magnetic field, demonstrating the importance of HH–LH mixing even for the weakly polarized dots.[10] By contrast, for the case of strongly reduced QD symmetry (dot A), an additional correction to the J^3 term of Hamiltonian H_B arises.[18] The q parameter, which arises from spin-orbit coupling, may also be modified due to changes in the spatial wave functions.[21] We suspect that for the strongly polarized dot A both the κ and anisotropic J^3 terms play a significant role, introducing coupling within the HH subspace, with the result that the relative polarization of the outer and inner components of the X^+ quadruplet depends on the in-plane field direction.

For X^0, the effect of $B \perp z$ is much smaller than for X^+, since the e-h states are coupled by isotropic exchange into bright and dark states [Fig. 2(c)], with splitting larger than the Zeeman perturbation. The transverse field mixes the bright and dark exciton states, resulting in bright exciton splitting into a doublet and the dark exciton emission being allowed due to admixture with the bright exciton.[19] We observed the appearance of the dark exciton emission for QD A [line D, Fig. 3(c)] at an energy ~0.33 meV below X^0 for $B > 1$ T. By contrast we did not observe the dark exciton for the weakly polarized QD B, indicating that the bright–dark exciton splitting is significantly larger than the Zeeman perturbation (~0.21 meV). In dot A the observed weaker e-h exchange interaction energy (both isotropic and anisotropic), which is inversely proportional to the sum of electron and hole confinement lengths squared,[20] may arise from the expansion of the hole (electron) spatial wave function in an elongated dot. In addition, the hole confinement length will increase due to strong admixture of the light hole, which has smaller effective mass, again decreasing the exchange interaction in dot A. It will also be reduced by the reduction of the total hole angular momentum due to the light hole admixture.

In the Faraday geometry for X^0, the magnetic field quickly removes the mixing due to the anisotropic exchange interaction, resulting in pure $(|+1\rangle + \beta^+|-1\rangle)$ and $(|-1\rangle - \beta^-|+1\rangle)$ excitonic spin eigenstates [Fig. 2(c)]. These are expected to be σ^+ and σ^- polarized with $\rho_c = (1 - 1/3\beta^2)/(1 + 1/3\beta^2)$ of $>95\%$ and ~70% for QDs B and A, respectively, close to ρ_c observed in the experiment. Since at $B = 5$ T the upper (lower) component of the exciton doublet split by magnetic field is σ^+ (σ^-) polarized for both QDs A and B [Figs. 2(a) and 3(b)], we conclude that g_X has the same sign for both dots. Assuming the electron g factor is isotropic with the same value ~0.55 for $B \parallel z$ and $B \perp z$, g_h in the z direction can then be deduced to be ~0.88 and ~1.62 for QDs A and B, respectively [Fig. 3(d)]. The marked reduction of g_h for the strongly linearly polarized QD A has a natural explanation in terms of the greater HH–LH mixing (as for the linear polarization), and arises both due to the decrease of the total hole angular momentum and to the decrease of the Luttinger parameter κ, which is strongly dependent on the chemical composition and confinement.[21,22] Although the greater HH–LH mixing should lead to higher in-plane $g_h^\perp$ in QD A than in QD B (Ref. 10), the observed very similar $g_h^\perp$ values in both QDs could also be explained by a smaller Luttinger parameter κ in QD A than in QD B.

In conclusion, we report the observation of both neutral and charged excitons in self-assembled QDs with strong degrees of linear polarization. The identification of neutral and charged excitons is supported by electric- and magnetic-field studies. A combination of the data for magnetic fields parallel and perpendicular to the growth direction allows a full determination of the electronic properties of the QDs. Finally, we note that elongation of In(Ga)As dots along $[1\bar{1}0]$ directions grown at low density, as we find here, has been reported in structural studies,[12] the elongation arising from fundamental anisotropies of the surface during growth.

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