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Effect of dimensionality and morphology on polarized photoluminescence in quantum dot-chain structures

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Change of the photoluminescence (PL) polarization is studied by changing the excitation intensity and temperature for aligned In(Ga)As quantum dot (QD) structures with varying inter-dot distances grown by molecular beam epitaxy on semi-insulating GaAs (100) substrates. An unusual increase of the polarization ratio is observed by increasing the temperature and/or excitation intensity throughout a low temperature (T < 70 K) and low intensity (Iex < 1 W/cm²) range. This increase as well as the general behavior of the polarized PL are the results of the exciton dynamics and the peculiarities of the system morphology. They are due to the varying inter-dot distances which change the system from zero-dimensional comprised of isolated QDs to one-dimensional comprised of wire-like structures. © 2012 American Institute of Physics.

I. INTRODUCTION

Semiconductor structures of reduced dimensionality such as quantum wells (QWs), quantum dots (QDs), quantum wires (QWRs), as well as nanorods, nanopillars, nanotubes, etc. have been the focus of both fundamental research and practical application development for several decades. Increasing the degree of quantum confinement generally leads to enhanced properties. For example, in zero-dimensional (0D) QDs, studies have found: long dephasing time and spin lifetimes, predictable sources of single-photons, strong exciton–photon coupling, and low threshold lasing.1,2 One-dimensional (1D), QWR structures have also demonstrated enhanced properties such as higher electron mobility, reduced nonradiative recombination, and lower threshold lasing which makes them suitable to drive the development of new devices such as nanowire solar cells,3 infrared photodetectors,4 nanowire injection lasers, and field-effect transistors.5,6 Recently, highly versatile structures have been demonstrated which exploit these systems of low dimensionality both together and separately. A QD-molecule system has been demonstrated in which the dots are tunnel coupled via connected QWRs.7 The more efficient tunnel coupling in this integrated QD-QWR system results in a hybridization of both electron and hole states, yielding direct-real-space excitonic molecules. Another GaAs/AlGaAs QWR-QD semiconductor system has been realized by metal organic vapor-phase epitaxy of site-controlled, self-assembled nanostructures in inverted tetrahedral pyramids.8 By systematically changing the length of the quantum wires, a continuous transition between the QD regime and the QWR regime can be realized. In addition to the QD-QWR coupling, interesting physical effects have been predicted in strained segmented nanowires involving different combinations of binary compounds considered as a function of segment length.9 Here, wires which could demonstrate spontaneous charge separation and wires which would preferentially generate polarized exciton gases were identified. As compared with a single layer of QDs, vertically stacked QDs exhibit quasi-one-dimensional, wire-like behavior induced by vertical electronic coupling which is controlled by changing the spacer thickness.10,11 A transformation of the polarization of the emission from TE to TM by increasing the number of layers and increasing the vertical coupling was found and attributed to the evolution of the system from predominantly 0D into predominantly 1D structures.

Recently, laterally aligned QDs were found to create long dot-chain structures with 12,13 and without vertical coupling.14–16 These (In, Ga)As/GaAs quantum dot-chain structures consist of elongated QDs aligned along the chain direction. The distances between neighboring QDs and their sizes are easily controlled by growth conditions, such that at very large dot to dot separations the ensemble acts like a collection of separate 0D QDs, and at very small separations where the dots are nearly touching, it acts more like a collection of coherent 1D nanostructures. Evidence for this 0D to 1D transition can be found in polarization-resolved photoluminescence (PL) spectroscopy, where strong linearly polarized emission along the chain direction will accompany the appearance of 1D structures. In other words, it is possible to transform the localized states of the QD exciton into propagating states of a 1D exciton in QWRs. In this paper, we investigate the PL polarization change in (In,Ga)As/GaAs quantum dot-chain structures with different inter-dot distances by varying temperature and excitation density. Such investigations allow for the distinguishing of the various mechanisms which cause linear polarization of the PL and

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assigning them to specifics of exciton dynamics and structural morphology.

II. SAMPLES AND EXPERIMENTAL DETAILS

The (In,Ga)As/GaAs quantum dot-chain samples were grown by molecular beam epitaxy (MBE) on semi-insulating GaAs (100) substrates consisting of 15 periods of (InxGa1−xAs QD layer/60 monolayer (ML) GaAs spacer). For the QD layers, InxGa1−xAs films of thicknesses 5.7, 8.5, and 15.5 MLs for x equal to 0.5 (sample C1), 0.4 (sample C2), and 0.3 (sample C3), respectively, were deposited at a temperature of 540°C.\textsuperscript{14} This guarantees ~25% excess above the critical thickness for 2D to 3D transition for each composition as determined by \textit{in situ} reflection high-energy electron diffraction. All samples were terminated with a final layer of uncapped QDs grown with the same composition and coverage as the underlying layers to examine the configuration by atomic force microscopy (AFM). Each of these three samples represents a different interdot coupling regime within the chains. Sample C1 (x = 0.5) contains QDs which are significantly smaller than the interdot distances (weak coupling regime). Sample C2 (x = 0.4) contains QDs which are only slightly smaller than their spacing (intermediate coupling regime). And, sample C3 (x = 0.3) contains QWR structures where the QDs within a chain have merged (strong coupling regime). Topographic, AFM images of these samples are shown in Fig. 1. Associated with each image is a wide, asymmetric QWR lobe in the FFT (Fig. 1(a)) transform into sharp features (Fig. 1(c)) testifying to the ordering of the chains/wires in the [0–11] crystallographic direction. Figure 1 reveals long QD chains laterally separated from each other by ~100 nm. At the same time, the average distance between the centers of QDs in the [0–11] crystalographic direction decreases from 65 nm in sample C1 to 44 nm in sample C2. The QD sizes in sample C1 are larger than those in sample C2 and depend on the InxGa1−xAs layer thickness and composition x. Typically, the QD shape is slightly elongated along the chain direction of [0–11].\textsuperscript{12,13} Such unique growth morphologies are mediated by the asymmetric surface diffusivities on the GaAs (001) surface. The multilayered growth produces well-ordered QD chains where the spacing between dots and between chains can be controlled by changing the specific growth conditions.\textsuperscript{14}

PL measurements were carried out at variable temperature and excitation intensity in order to better understand the effect of dimensionality and morphology on the optical properties of dot-chain structures. Excitation was with the 532 nm line of a frequency doubled Nd:YAG laser, focused to a diameter of ~30 µm at the sample. It is assumed in all that follows that using this high energy deep in-band, excitation leaves no polarization memory of the excitation in the emissions. The samples were mounted in a variable temperature 8–300 K closed-cycle helium cryostat, and the PL signal from the sample was dispersed by a monochromator and detected by a liquid nitrogen cooled OMAV: InGaAs photodiode detector array. In order to eliminate the effect of polarization dependence of the optics and spectrometer, a quartz depolarizer was inserted along the optical axis immediately following the analyzer during polarized PL measurements.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

Figure 2 shows the PL spectra from our samples measured at low temperature and low excitation intensity \(I_{ex} = 4 \times 10^{-3} I_0\), \(I_0 = 1000\) W/cm\(^2\), for two orthogonal polarizations: parallel to the chain direction ([0–11]) and orthogonal to it (parallel to [011]). We introduce here, the linear polarization degree, \(P = (I_{0–11} - I_{011})/(I_{0–11} + I_{011})\), with \(I_{0–11}\) and \(I_{011}\) being the intensities of the luminescence...
properties of asymmetric QDs are caused by the microscopic polarization degrees are understood that QD ensemble PL linewidth is directly related to the results of the AFM analysis. Indeed, we do find that \( P_1 > P_2 \). The highest polarization degree, however, \( P_3 = 25.9\% \), for this series of samples is observed in the wire-like sample C3 due to the 1D character of its excitonic states.

The linear polarization degree \( P \) is also found to be energy dependent, \( P(E) \), within the spectral range of a PL band as can be seen from Fig. 2. It gradually grows from 6.8\% at the high energy tail of the PL band to 13.9\% at the low-energy tail in sample C1, from 6.6\% to 14\% in sample C2 and from 13.2\% to 30\% in sample C3. This is understood for the QD samples C1 and C2 as being due to the above mentioned growth of inhomogeneous strain and the strength of piezoelectric fields around larger QDs. This substantially lowers the local QD symmetry, increases the valence band mixing, and hence, increases the polarization anisotropy. Therefore, since we expect larger (more elongated) QDs to contribute to the PL band on the low energy side, we would expect the polarization to increase here too.

In the wire-like sample C3, the polarization degree reaches a maximum of \( P_3 = 25.9\% \). In order to understand the nature of the polarization anisotropy in a QWR system, many sources have to be taken into account. There exists two basic sources of anisotropy for the optical transitions in a QWR: (i) a strong valence band mixing (due to spatial confinement and strain)\(^{27-33}\) and (ii) a discontinuity in the dielectric constant between the wire and its surroundings (due to optical confinement)\(^{34-39}\). It is shown within the framework of a \( k \cdot p \) approximation that the maximum degree of linear polarization produced by mixing the bulk \( \Gamma_8 \) states is \( \sim 60\% \) for the ground state \( h_l-e_1 \) transition, independent of wire orientation, diameter, or composition.\(^{27,40}\) The optical confinement caused by the difference in dielectric constants, \( \varepsilon \), for QWRs and their surroundings, e.g., \( \varepsilon = 14.6 \) for an InAs semiconductor wire versus \( \varepsilon = 1 \) for air, leads to even higher \( P \) values. For isolated GaN nanorods\(^{39}\) or InP nanowires,\(^{46}\) size dependent polarization anisotropy was observed above 90\%.

![Image of PL spectra](image_url)

**FIG. 2.** Low temperature PL spectra measured in two orthogonal polarization planes parallel to the [0–11] direction (red curves) and perpendicular to it ([011] direction; black curves), at low excitation intensity \( I_{\text{exc}} = 4 \times 10^4 \text{W/cm}^2 \) in sample C1 (a), sample C2 (b), and sample C3 (c). Linear polarization degree \( P = (I_{011} - I_{011})/(I_{011} + I_{011}) \) refers to the right ordinate axis. A polar plot of the polarized emission is shown to the right of the PL spectrum in each sample. Here, 90° corresponds to the [01–1] direction.
Taking into account that the QWRs in sample C3 are sufficiently wide (~100 nm), we can expect polarization contributions from both valence band mixing and dielectric discontinuities. Indeed, valence band mixing is essential in comparatively thin QWRs. For thin (several tens of nm) In$_{0.5}$Ga$_{0.5}$As/GaAs wires, the extension of the exciton wave function (double Bohr radius $2\alpha_B \approx 26$ nm) becomes comparable to the wire width and pronounced confinement effects are expected to develop. If the wire width significantly exceeds this Bohr radius, the quantization energy decreases and will approach the thermal energy or the level broadening, where the size quantization is no longer observable. Here, polarization due to the discontinuity in the dielectric constant can dominate. In order to estimate the contribution of optical confinement to $P$ in sample C3, we can use the dipole approximation for the luminescence strength which is proportional to the scalar product of the local electric field $\mathbf{E}$ and the inter-band dipole moment, $\mathbf{d}$, averaged over the full electron-hole wave function. Then one gets$^{33,35}$

$$P = \frac{|d^e_{cv}|^2 - \delta^2|d^v_{cv}|^2}{|d^e_{cv}|^2 + \delta^2|d^v_{cv}|^2}, \quad (1)$$

where $d^*_{cv} = \langle \Psi^v_d | d^* | \Psi^e_i \rangle$ is the dipole matrix element between conduction ($c$) and valence ($v$) bands for the $I = ||, \perp$ components of the dipole moment parallel and perpendicular to the wire axis. In Eq. (1),

$$\delta = \frac{\epsilon_s}{\epsilon_s + \epsilon_w}, \quad (2)$$

with $\epsilon_s$ and $\epsilon_w$ being the dielectric constants of the surrounding material and the wire, respectively. We take $\epsilon_s = 13.1$ for GaAs and $\epsilon_w = 13.55$ for In$_{0.5}$Ga$_{0.5}$As, which was derived by linear extrapolation from $\epsilon = 14.6$ for InAs, and make a simplifying approximation of $|d^e_{cv}| = |d^v_{cv}|$, i.e., no anisotropy in the dipole moment. Then we can use Eq. (1) to estimate the contribution to $P$ from the dielectric discontinuity alone to be equal 1.7% only. Thus, we conclude that the main contribution to $P$ in our samples must be from the anisotropy of the dipole matrix elements in Eq. (1), which is determined by the valence band mixing. This mixing in QWRs is not only due to lateral confinement, but can also be affected by high-index confinement,$^{40}$ anisotropic strain,$^{41}$ and anisotropic interface roughness.$^{42}$ For example, it has been shown$^{41}$ that for compressively strained, InAsP QWRs, large in-plane polarization anisotropy is caused by the broken symmetry due to the effective triaxial anisotropic strain alone. As a result, the net effect on the valence band mixing becomes rather complicated.

Using a first order approximation for the valence band mixing,$^{38}$ the optical anisotropy that results from lateral confinement and anisotropic strain can be reduced to the following simple relation:$^{43}$

$$\left[ \frac{\gamma_2 h^2 \pi^2}{m_0 \rho} + b(e_\perp - e_{||}) \right] \left( E_{hh} - E_{ih} \right). \quad (3)$$

Here, $\gamma_2$ is the Luttinger parameter for the hole effective mass, $b$ is the shear deformation potential ($b > 0$), $e_{||}$ and $e_\perp$ are the strain components along and perpendicular to the wire axis, and $l$ is the lateral extent of the hole wave function which is equal to the QWR width for sufficiently wide QWRs. From Eq. (3), it follows that the optical anisotropy becomes inversely proportional to the square of the QWR width. In turn, the biaxial strain in the wires leads only to an increase of the difference, $E_{hh} - E_{ih}$, which reduces the optical anisotropy again. In the case of triaxial strain, the optical anisotropy can be enhanced, because $(e_\perp - e_{||}) > 0$ due to strain in the normal direction. Thus, coming back to Fig. 2(c), we conclude that a large value of the polarization degree even in wide QWRs can be assigned to the lateral confinement enhanced due to the anisotropic strain in the vicinity of the QWR interfaces. Larger QWRs induce larger symmetry distortion, exhibit a significant piezoelectric field, have a larger degree of valence band mixing, and as a result increase the polarization degree. Therefore, the observed polarization in sample C3 grows as the energy decreases within the QWR PL band as shown in Fig. 2(c).

![Figure 3](image-url) 

**FIG. 3.** Low temperature PL spectra and linear polarization degree $P$ measured in polarizations parallel to the [0–11] direction and along the [011] direction at high excitation intensity ($I_{ex} = 10^4$ W/cm$^2$) in sample C1 (a), sample C2 (b), and sample C3 (c).
in sample C2, and $E_{\text{max}}^3 = 1.266 \text{ eV} \rightarrow 1.290 \text{ eV}$ and $\Gamma_3 = 35.8 \text{ meV} \rightarrow 61 \text{ meV}$ in sample C3. In addition, a higher energy PL band develops at $E_{\text{max}}^{2a} = 1.355 \text{ eV}$ with $\Gamma_{1a} = 77 \text{ meV}$ in sample C1, $E_{\text{max}}^{2b} = 1$. 355 eV with $\Gamma_{2a} = 69.5 \text{ meV}$ in sample C2, and at $E_{\text{max}}^{3a} = 1.323$ with $\Gamma_{3a} = 40 \text{ meV}$ in sample C3. The blue shift and broadening of the PL bands are attributed to the state filling effect that results in the appearance of the excited QD states. At high excitation intensity, such as in Fig. 3, we believe that the piezoelectric fields are fully screened by the photo-excited carriers both in the QD and QWR samples.

The higher energy peak for the QD samples, C1 and C2, has been assigned to a one-dimensional wetting layer (1D WL) state,\textsuperscript{12,43,45} in analogy to the 2D WL generally arising during Stranski-Krastanov growth of QDs. It has been shown,\textsuperscript{45} however, that this 1D WL develops after the dot formation, during overgrowth with GaAs creating a 1D channel between and connecting the dots in a line. As seen in Figs. 3(a) and 3(b) at the maximum of this PL band, the polarization degree reaches a comparatively high value of $P_{\text{max}} = 11\%$ which would be expected for 1D structures. In samples C1 and C2, the polarization degree of sample C3 is most likely, simply an excited state of the source of the additional band in the high intensity PL spectra of sample C3 as a function of excitation intensity. It can be seen that the solid line. Here, we see both the measured and calculated polarization degrees taken at the PL maximum for sample C3 as a function of excitation intensity. Calculated assuming perfect recombination, the density of excess carriers $n_{\text{exc}}$ and the excitation density $I_0$ were assumed to be proportional $n_{\text{exc}} \propto I_0$.

The results of these calculations are shown in Fig. 4 as the solid line. Here, we see both the measured and calculated polarization degrees taken at the PL maximum for sample C3 as a function of excitation intensity. It can be seen that the theoretical dependence, calculated assuming perfect rectangular QWRs and taking into account only the phase-space filling and electron-hole wave-function overlap, describes the high intensity side of the experimental data quite well. In the low intensity range, however, theory and experiment give very different results. This deviation between the

$$I(h\omega) \propto (h\omega)^2 \sum_{ij} \int dk_y \int dE_c \int dE_v \delta(E_c - E_v^i(k_y))$$

$$\times \delta(E_v - E_v^i(k_y)) |M_{ij}(k_y)|^2 \delta(E_c - E_v - h\omega)$$

$$\times F(E_c - E_v^i)F(E_v - E_v^i).$$

Now, we have the 1D density of states in the conduction and valence band given by

$$D^c(E) = \frac{2}{\pi} \int_{-\infty}^{\infty} dk_y \delta(E - E_v^i(k_y)),$$

where $E_v^i(k_y)$ are the energy dispersion branches for electrons and holes. The quasi-Fermi energies $E_F^c$ and $E_F^v$ of the conduction and valence bands are determined from the conditions

$$n = \int_{-\infty}^{\infty} D^c(E)F(E-E_F^c)dE = \int_{-\infty}^{\infty} D^v(E)F(E-E_F^v)dE.$$
theoretical dependence which reaches a maximum and the experimental, \( P \), dependence which falls quickly as the excitation power is reduced to a minimum, indicates that real QWR behavior at low excitation intensities does not correlate with the behavior of a pure 1D system. We believe the localization of 1D excitons into 0D states by wire imperfections or in-plane piezoelectric fields to be the source of the non-ideal nature of the 1D QWRs. At low power excitation and low temperature, 1D excitons can be trapped by fluctuations of the local potential. This localization reduces the 1D wire to an ensemble of effective 0D QDs significantly influencing the symmetry of the excitons and hence the polarization rules for the optical transitions. As a result, the polarization degree decreases at low excitation intensities. With an increase in hot carrier concentration in the QWR region due to an increase in excitation intensity or temperature, the excitons, which dominantly contribute to the PL, are from the unbound states where they are free to move in the 1D band. The degree of polarization thus grows with the excitation intensity, and the experimental points in Fig. 4 tend to the theoretical values calculated for the ideal 1D structure. Further increase in intensity begins to screen the internal fields and ultimately saturates the polarization. Before the full density of states of the QWRs become filled, thus populating the WL, we believe the biexciton-like states become populated which through the anisotropic exchange mechanism are polarized perpendicularly to the ground state excitons. The emitted light, being the sum of all mechanisms, would then lose some polarization value, which is observed experimentally. These observed characteristics of the polarization degree with increasing excitation intensity correlate with the behavior of the PL decay time in this QWR structure (sample C3) observed earlier by means of time resolved spectroscopy as a function of temperature.\(^{1,4}\)

A similar scenario is observed in the temperature dependences of the degree of polarization for our samples. Figure 5 shows the \( P(T) \) dependences measured at the maximum of the PL band in each of samples C1, C2, and C3. The low temperature growth of \( P(T) \) observed in the QD samples derives from thermally activated carrier transfer to larger QDs or QWRs that generate stronger piezoelectric fields at the interfaces. Due to stronger mixing of the valence bands caused by these stronger piezoelectric fields, the emitted light has a higher degree of polarization. Further temperature increase does not change the \( P(T) \) value in sample C1, slightly reduces it in sample C2, and drastically reduces it in sample C3 at a rate of 0.13%/K within the temperature range 50–200 K. The temperature induced reduction of the \( P(T) \) in comparatively wide QWRs and large QDs is attributed to effective population of the excited hole states with the increase in temperature. While the energy gap between the hole states decreases with the growth of lateral dimensions, the moderate temperature leads to a thermal population of the excited states. Emission from these states can be polarized perpendicular to the wire axis,\(^{42}\) resulting in the temperature induced depolarization of the emitted light. It should be noted that similar to the power dependent results, we have a low temperature range where everything acts like a QD due to localization effects, i.e., the polarization increases as

the “larger” structures are populated. However, at some point, the true QDs saturate due to the discrete nature of the energy spectrum. Finally, similar to the high excitation power case, the polarization at higher temperatures vanishes for the QWR-like structures.

IV. CONCLUSIONS

Concluding, the change of the PL polarization is studied by changing excitation intensity and temperature for aligned In(Ga)As QD structures with varying inter-dot distances grown by molecular beam epitaxy on semi-insulating GaAs (100) substrates. An unusual increase of the polarization ratio is observed with temperature and/or excitation intensity in the low temperature (\( T < 70 \) K) and low intensity (\( I_{ex} < 1 \) W/cm\(^2\)) ranges. This increase as well as the general behavior of the polarized PL in the structures under consideration is attributed to the exciton dynamics changing from 0D, QD-like, to 1D, wire-like and to the peculiarities of the resulting system morphologies.

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