Two-Photon Absorption in GaAs Quantum Wires

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We have investigated the polarization-dependent two-photon absorption in GaAs/AlGaAs quantum wires. The anisotropic selection rules of the multiphoton absorption process are exploited to study the one-dimensional $2p$ exciton states and the transitions between quantum wire subbands of different quantum numbers ($\Delta n_y \neq 0$ selection rule). The deviations from the selection rules derived for the strict one-dimensional case are discussed, and depend on the actual quasi-one-dimensional character of the excitonic wave functions.

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The nonlinear optical properties of low-dimensional semiconductors have attracted great interest in the last few years. In particular, two-photon absorption (TPA) has been investigated in detail in two-dimensional (2D) semiconductor quantum wells, both experimentally and theoretically, to elucidate the impact of the reduced dimensionality on the nonlinear absorption processes [1]. Currently, interest has turned to systems with even lower dimensionality, like 1D quantum wires and 0D quantum dots (for a review see Ref. [2]). The optical properties of undoped low-dimensional systems are strongly governed by excitonic effects. An interesting aspect of TPA is that it gives direct access to excited excitonic states having a significantly larger excitonic radius ($2p$ states). It is therefore expected that the influence of lateral confinement is stronger on these excited $2p$ states than on the excitonic ground state, i.e., the $1s$ exciton. A special situation should occur when the width of the lateral confinement is in between the extension of the $1s$ and $2p$ states. In this Letter we report an experimental study of TPA in GaAs/AlGaAs quantum wires. We find a strong quantum confinement effect on the $2p$ exciton states, significantly enhanced with respect to that on the $1s$ states observed in conventional photoluminescence excitation spectroscopy (PLE). The TPA curves show rich excitation spectra with a strong polarization dependence, in agreement with theoretical predictions [3,4]. In fact, we find that optical transitions between quantum wire states with the same quantum numbers ($\Delta n_y = 0$) are allowed when the polarization direction of the exciting beam (ϵ) is perpendicular to the wire quantization direction (y), whereas $\Delta n_y = \pm 1$ transitions are dominant in the $\epsilon \| y$ geometry. These peculiar selection rules allow us to discriminate between true one-dimensional and quasi-two-dimensional excitonic states which are involved as the final states of different linear and nonlinear absorption processes.

The investigated quantum wire array has been fabricated by holographic patterning and subsequent reactive ion etching of a molecular-beam-epitaxy-grown multiple-quantum-well heterostructure. The quantum well structure consisted of 25 GaAs wells of width $L_z = 103$ Å and $Al_{0.34}Ga_{0.66}As$ barriers of width $L_{bz} = 148$ Å grown on top of a 1-μm-thick $Al_{0.34}Ga_{0.66}As$ cladding layer providing optical confinement of the luminescence (throughout the paper the indices z and y indicate the confinement directions of the quantum well and of the quantum wire, respectively). The processed quantum well structure resulted in a regular array of quantum wires with 280-nm periodicity and a crystalline wire width $L_y = 60 \pm 5$ nm as determined by the analysis of high-resolution x-ray diffraction spectra. Details on the structural and linear optical properties of the sample are reported in Ref. [5] and Refs. [6,7], respectively. The nonlinear absorption has been studied by measuring the two-photon-absorption-induced photoluminescence excitation spectra (TPA-PLE) at 10 K for different polarization directions of the exciting laser beam. The detection energy was set at the fundamental quantum wire exciton state ($E_{11}^{1s} = 1.554$ eV), independently measured by linear PLE spectroscopy, and by scanning the exciting photon energy in the transparency region of the crystal. The laser source was a two-stage amplified dye laser pumped by the second harmonic of a Nd:YAG laser, operating at 10 Hz repetition frequency and with 9 nsec pulse duration. The dye-laser radiation was frequency converted in the infrared by a low-pressure H_2 Raman cell. The output beam could be tuned in the spectral range $0.75\ \text{eV} < \hbar\omega < 0.82\ \text{eV}$ with tuning accuracy of the laser dye of about 2 Å and with maximum power density of the or-

der of 10 MW cm^{-2} after focusing. The emitted radiation was detected by a 0.5-m double monochromator equipped with a 60ER photomultiplier tube and the exciting beam was monitored by a fast-response photodiode. Both signals were sent to a digital oscilloscope for further processing. To reduce the effects of fluctuations in the input beam intensity, the ratio of the photomultiplier signal to the second power of the monitor signal was used. Moreover, the quadratic behavior of the detected luminescence signal versus excitation intensity was checked at each experimental point and several measurement runs were carried out at all exciting wavelengths. With the adopted experimental conditions the signal-to-noise ratio was of the order of 200:1. In Fig. 1 we show the typical intensity dependence of the nonlinear luminescence induced by the absorption of two photons of energy $\hbar\omega=0.795$ eV ($2\hbar\omega=1.590$ eV) and $\hbar\omega=0.805$ eV ($2\hbar\omega=1.610$ eV) versus the laser power density. Much care has been taken to reduce and test the influence of the inhomogeneity of the power distribution across the exciting laser section. The experimental points exhibit the expected quadratic behavior as shown by the comparison with the quadratic regression curves (solid lines in Fig. 1). The slope-2 curves fit the experimental data points within 3% at all wavelengths. The experimental TPA-PLE spectra at $T=10$ K in the $\epsilon\perp$y and $\epsilon\parallel$y configurations are shown in Fig. 2 together with the linear PLE.

Before entering into a detailed discussion of the TPA-PLE data we briefly summarize the results of the linear spectroscopy under low and high excitation intensity. As a result of the relatively large width of the investigated quantum wires, which amounts approximately to 4 times

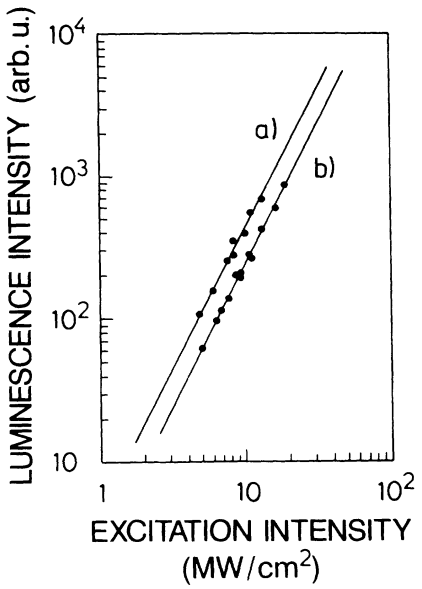

FIG. 1. Logarithmic plot of the intensity dependence of the photoluminescence induced by two-photon absorption at energy (a) $2\hbar\omega=1.590$ eV and (b) $2\hbar\omega=1.610$ eV, respectively, versus the laser power density in the $\epsilon\perp$z configuration. The straight lines are the best-fit quadratic regression curves to the experimental points. Curves fit all data points within $\pm 3\%$.

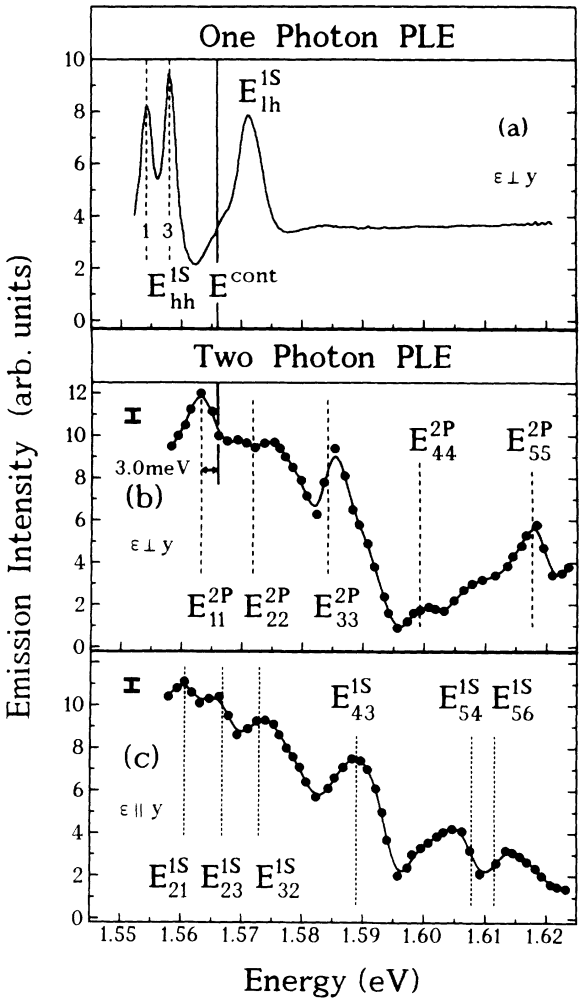

FIG. 2. (a) Linear photoluminescence excitation (PLE) spectrum of the quantum wire array at $T=10$ K. The detection energy was set at the low-energy tail of the fundamental quantum wire exciton state (around 1.552 eV). E^{cont} indicates the energy of the exciton continuum. (b) Two-photon-absorption-induced photoluminescence excitation spectrum (TPA-PLE) of the quantum wires measured in the $\epsilon\perp$y configuration at $T=10$ K. The vertical lines indicate the calculated energy positions of the $2p$ exciton states associated with $\Delta n_y=0$ transitions ($E^{2p}_{n_y,n_y}$ transitions). The 3.0-meV splitting indicates the estimated binding energy of the $2p$ excited state of the $n_y=1$ exciton in the quantum wire. The vertical error bar deriving from signal-to-noise ratio and slope-2 regression on experimental points is indicated. The horizontal error bar coincides with the solid dots (the tuning accuracy was 2 Å). (c) The same as (b) but in the $\epsilon\parallel$y configuration. The vertical lines indicate the calculated energy positions of the $1s$ exciton states associated with $\Delta n_y=\pm 1$ transitions ($E^{1s}_{n_y,n_y\pm 1}$). For both TPA spectra the detection energy was set at around 1.554 eV.

the bulk exciton Bohr radius (a_0), the quantum effects induced by the lateral confinement (y direction) are expected to be different under different excitation conditions. It has been shown that 1D intersubband transitions can be clearly resolved under high-excitation conditions in stationary [6] and time-resolved photoluminescence experiments [7]. Comparison of PL spectra with Kohn-Luttinger-type calculations taking into account the complex valence-band structure in the wires [8] allowed us to identify intersubband recombination processes involving quantum wire subbands with quantum numbers as high as $n_y=5$ and the selection rule $\Delta n_y=0$ [6,7]. In particular, the confinement energies of the 1D valence subbands were taken from Ref. [8] for a GaAs wire with $L_y=60$ nm and $L_z=10$ nm, whereas the confinement energies of the conduction subbands were calculated by the square-well model assuming infinite height of the potential barrier. However, when the optical properties are dominated by excitonic effects, as is the case under low-excitation conditions, the quantum size effects induced by the lateral confinement appear to be different. It was pointed out in Ref. [9] that in the rather wide quantum wires of our experiment the lateral confinement leads to a quantization of the excitonic center-of-mass motion, which leaves the internal structure of the exciton (i.e., the wave function of the *relative* motion of the electron and hole) unchanged and therefore 2D-like. The center-of-mass motion quantization manifests itself in the split heavy-hole exciton peaks of Fig. 2(a), which are quantized states of the translational motion of the $1s$ heavy-hole exciton. Therefore the linear PLE profile of Fig. 2(a) still maintains some 2D character (including a sharp light-hole exciton resonance around 1.574 eV) and does not exhibit pure 1D states. The results of these experiments demonstrated that quantum size effects are differently pronounced under different experimental conditions. In fact, depending on the wire width L_y one can find one-dimensional excitons if $L_y < a_0$ and two-dimensional excitons if $L_y \gg a_0$. In our sample we have roughly $L_y \simeq 4a_0$; hence, $1s$ excitons behave almost like 2D excitons, while $2p$ excitons, having a larger extent of the relative motion, should exhibit an almost 1D character (with transition energies close to the 1D intersubband transitions observed in the high-excitation-intensity photoluminescence spectra of Refs. [6,7]).

Due to the complementary selection rules, two-photon spectroscopy offers the unique possibility to access directly $2p$ excitonic states. Since the diameter of the $2p$ excited states is enlarged by a factor of 4 (valid for 3D excitons) in comparison with $1s$ excitons, these states should be effectively quantized and therefore exhibit a true one-dimensional character. This should enable us to observe 1D excitonic states in the TPA-PLE spectra. In order to use the familiar classification of 2D excitons for 1D excitons also, we will label states of even parity by "s" and of odd parity by "p" [10]. This allows us to classify one- and two-dimensional excitonic states without changing notation. Furthermore, it leads to identical selection rules for 1D and 2D systems with respect to the confined and unconfined directions. Inspection of experimental TPA spectra in the $\epsilon \perp \mathbf{y}$ and $\epsilon \| \mathbf{y}$ configurations reveals a clear difference with respect to the linear PLE spectrum [Fig. 2(a)]. In addition, the TPA-PLE line shapes are different depending on the relative orientation of the laser polarization vector with respect to the quantum wire lateral quantization direction. Such an anisotropy is theoretically predicted by the theory for quasi-one-dimensional systems [3,4]. In particular, in the $\epsilon \perp \mathbf{y}$ geometry it is expected that the matrix elements of the TPA process do not vanish for transitions involving quantum wire states with the same quantum numbers (selection rule $\Delta n_y=0$). Conversely, in the $\epsilon \| \mathbf{y}$ geometry only intersubband transitions with $\Delta n_y = \pm 1, \pm 3, \ldots$ are allowed. In addition, if excitons are included in the 1D theory of two-photon absorption [4], it results that only $2p$ exciton states related to $\Delta n_y=0$ transitions are allowed as final states of the TPA process in the $\epsilon \perp \mathbf{y}$ configuration. On the contrary, $1s$ exciton states are allowed in the $\epsilon \| \mathbf{y}$ geometry for $\Delta n_y \neq 0$ transitions. These additional constraints result from the requirements of parity conservation in the two-photon-absorption process. In the light of these theoretical arguments we can interpret the experimental spectra of Figs. 2(b) and 2(c).

In the $\epsilon \perp \mathbf{y}$ configuration [Fig. 2(b)] we observe five structures whose relative energy splittings are the same as those observed for the $\Delta n_y=0$ transitions in the time-resolved [6] and in the stationary luminescence spectra [7] under the high-photoexcitation condition, i.e., they directly reflect the 1D subband spacing. Furthermore, the first resonance of the TPA-PLE spectrum in Fig. 2(b) is about 3 meV below the onset of the continuum in Fig. 2(a). This confirms that indeed $2p$ excitonic states are observed. The estimated binding energy of 3 meV for $2p$ state is additionally in good agreement with that predicted for a perfectly quantized 1D exciton. In fact, such a $2p$ exciton follows the normal 3D hydrogenic series [10] that gives for its binding energy a value of about 4 meV in GaAs quantum wires. The spectral positions of the higher-energy transitions [which are indicated by dashed lines in Fig. 2(b)] have been obtained by adding the 1D subband spacing of Ref. [6] to the transitions labeled E_{11}^{2p}. The coincidence strongly supports our assumption that the observed resonances are related to one-dimensional excitonic transitions. Therefore, we conclude that the TPA-PLE peaks of Fig. 2(b) are related to $2p$ excited states of the 1D excitons associated with $\Delta n_y=0$ transitions, in agreement with the theoretical expectations.

In the $\epsilon \| \mathbf{y}$ configuration the TPA-PLE spectrum appears to be even more structured, and exhibits several transitions. The vertical lines of Fig. 2(c) represent the energy positions of the $1s$ exciton states associated with $\Delta n_y = \pm 1$ transitions in the quantum wire. These are calculated by adding the net confinement energies of the

1D conduction and valence subbands to the E_{11}^{1s} exciton energy (i.e., the detection energy). Comparison of these calculated transitions with the experimental data points reveals reasonable agreement. This demonstrates that in this configuration $1s$ states with $\Delta n_y = \pm 1$ are allowed, in agreement with the theory. However, the TPA-PLE spectrum of Fig. 2(c) exhibits rather broad peaks in this configuration, which requires further comment. In fact, the TPA selection rules employed for the interpretation of the presented nonlinear absorption data have been derived by a strict one-dimensional model [3,4], which obviously should be modified to treat the intermediate regime of quasi-one-dimensional excitons where $L_y \simeq 4a_0$. Under these conditions departures from the selection rules valid for the strictly 1D systems are expected due to the residual 2D character of the exciton wave functions. In particular, one should consider either pure 1D exciton states (both $1s$ and $2p$ states) or 2D excitons with quantized center-of-mass motion as final states of the multiphoton absorption process. A linear combination of the wave functions of these two different excitons can be used to represent a realistic form of the quasi-1D exciton wave function. Additional broadenings are expected in the TPA spectra due to this superposition, which qualitatively explains the observed broad structures in the TPA-PLE spectrum of Fig. 2(c). A detailed discussion of these preliminary theoretical results will be reported elsewhere.

In conclusion, we have reported the first study of two-photon absorption in GaAs quantum wires. Our experiment provides evidence for the strongly anisotropic selection rules of the interband two-photon absorption related to the reduced dimensionality of the system. Further, we have observed the excited $2p$ excitonic states and several $\Delta n_y = \pm 1$ transitions in these one-dimensional structures. Finally, the mixed 1D and 2D confinements of the excitonic states in wide quantum wires have been discussed with relevance to the peculiar nonlinear absorption selection rules.

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