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LOW-DIMENSIONAL SYSTEMS

Optical Detection of Asymmetric Quantum-Dot Molecules in Double-Layer InAs/GaAs Structures

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Abstract—Self-assembled quantum dots (QDs) in double-layer InAs/GaAs structures are studied by resonant photoluminescence and photoluminescence excitation spectroscopy. A weakly correlated (50%) double-layer system with an array of vertically coupled QDs (asymmetric quantum-dot molecules) was formed in a structure consisting of the 1.8-monolayer-thick first and the 2.4-monolayer-thick second InAs layers separated by 50 monolayers of GaAs. The nature of discrete quantum states in this system was studied and resonances corresponding to vertically coupled QDs were clearly observed for the first time.

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1. INTRODUCTION

Two quantum dots (QDs) so closely spaced that wave functions of the charge carriers localized in the dots overlap form a so-called "artificial molecule", which may be considered, in particular, as a quantum bit (*qubit*) in optical computers [1–3]. In these kinds of structures, coupling originates from the tunneling of electrons and holes between adjacent QDs separated by a thin barrier [4, 5]. In real systems, securing the conditions for resonance quantum-mechanical tunneling is problematic due to the inevitable spread in QD size, composition, and stress distribution present in the structures after molecular-beam epitaxy (MBE). For example, self-assembled QDs obtained by growing highly strained (In,Ga)As epitaxial layers on GaAs (100) substrates are typically characterized by a size variance of ~10% [6–9]. New possibilities for creating coupled quantum structures are offered by the effect of vertical alignment of nanoislands, which was recently discovered in multilayer systems [10, 11]. The elastic-stress fields induced by QDs from lower-lying layers cause vertical alignment. An increase in the QD size in the upper layer and a reduction of the effective critical thickness for the 2D–3D transition are related to the accumulation of elastic energy in this layer. Recent advances in the technology of growing the correlated QD systems make possible the fabrication of InAs/GaAs QD molecules with quantum-mechanical coupling energy on the order of tens of millielectronvolts, which is comparable to the value of inhomogeneous broadening [5].

A more detailed understanding of the quantum-mechanical coupling of QDs into QD molecules can be gained from the studies of double-layer InAs/GaAs structures that incorporate QDs of different sizes in the first (seed) and in the second layers separated by a GaAs spacer of variable width [12–14]. In such a structure, the density and size distribution of QDs in the second layer can be controlled independently.

In this paper, we report the results of a systematic study of the conditions for the formation of asymmetric quantum-dot molecules (i.e., artificial molecules composed from two QDs differing in size) in weakly correlated QD systems that can be formed in double-layer structures with sufficiently thick GaAs spacers. Channels for energy transfer in asymmetric quantum-dot molecules from small QDs in the first layer to large QDs in the second layer are not yet adequately understood [12, 15]. To identify these channels unambiguously, one needs to reliably distinguish the QD-molecule states among the complex set of discrete states manifesting themselves in photoluminescence (PL) or the photoluminescence excitation (PLE) spectra of weakly correlated double-layer InAs/GaAs QD systems. For this purpose, the additional confinement of charge carriers by AlGaAs barriers is introduced, which prevents carrier diffusion from the QD region into the GaAs substrate and the cap layer and favors efficient population of QD states under optical excitation. Fur-

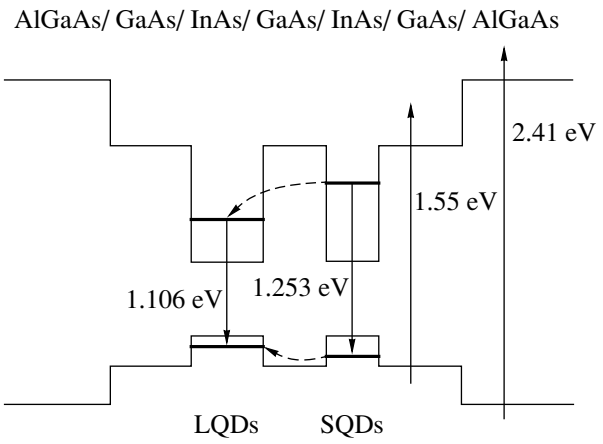

Fig. 1. Energy diagram of quantum-dot states in double-layer InAs/GaAs structures.

thermore, it is reasonable to expect that, in a structure of this configuration, the number of optically excited charge carriers above and below the QD region will be approximately the same.

2. EXPERIMENTAL

The samples were grown by MBE in a chamber connected to an ultrahigh-vacuum scanning tunneling microscope (STM). These samples represented weakly correlated systems consisting of the following layers (listed from the surface towards the substrate):

(i) a 20-nm GaAs cap layer;

(ii) a 28-nm $Al_{0.3}Ga_{0.7}As$ layer;

(iii) a 57-nm GaAs layer;

(iv) 2.4 monolayers (MLs) of InAs (large-QD layer);

(v) a 50-ML GaAs spacer;

(vi) 1.8 MLs of InAs (small-QD layer);

(vii) a 57-nm GaAs layer; and

(viii) a 28-nm $Al_{0.3}Ga_{0.7}As$ layer on the GaAs (001) substrate.

Structural studies were carried out by an STM in the plan-view mode and by a transmission electron microscopy (TEM). The PL was excited by Ar^+, Ti:sapphire, or He–Ne lasers, depending on the spectral region under study. The PLE spectra were measured in a liquid-helium cryostat using a tungsten lamp, whose light was dispersed by a 0.27-m double grating monochromator that served as a tunable low-intensity (<0.02 W/cm^2) source of PL excitation; the PL itself was detected by a cooled germanium diode placed at the exit of a 0.30-m monochromator. The TEM data confirm that there is a weak (50%) correlation between adjacent QD layers in the structures under study. Statistical analysis of the STM images yields the following average characteristics of the QDs in the first layer: the height, (4 ± 1.5) nm; base size, (20 ± 3) nm; and density, 4.5×10^{10} cm^{-2}. The density of QDs in the second layer is about 2×10^{10} cm^{-2}. The volume of a QD in the second layer is twice as large as that in the first layer, which is caused both by larger amount of deposited InAs and by the effect of elastic-stress fields induced by the QDs of the first layer [13, 14].

3. RESULTS AND DISCUSSION

When studying the spectra of below-barrier states in these QD structures, we should take into account the presence of two different barriers (GaAs and AlGaAs), which confine the motion of charge carriers. The energy diagram of discrete states in double-layer InAs/GaAs structure is outlined schematically in Fig. 1. Ground-state levels in large QDs (the second layer) and in small QDs (the first layer) are shown by thick horizontal lines. Dashed lines represent the processes of the vertical transfer of charge carriers between tunneling-coupled QDs. The processes of above-barrier optical excitation are represented by upward vertical arrows, while the processes of radiative transitions between the QD ground states, by downward arrows. The PL spectra of a structure outlined in Fig. 1 are shown in Fig. 2a for three different values of the excitation photon energy E_{exc}: 2.41 eV (above the AlGaAs barrier), 1.55 eV (above the GaAs barrier), and 1.355 eV (below the wetting-layer absorption edge). Large variation in the shape of the spectra in Fig. 2a with the excitation photon energy is evident. For $E_{exc} = 2.41$ eV (excitation above the AlGaAs barrier), two bands are clearly seen in the PL spectrum; they are attributed to the emission from small QDs (SQDs) in the first layer at energies around 1.253 eV and to the emission from large QDs (LQDs) in the second layer at ~1.106 eV. When the excitation-photon energy is lower than the AlGaAs barrier energy ($E_{exc} = 1.55$ and 1.355 eV), the shape of the PL spectrum changes considerably. These changes represent the specific features of the charge-carrier relaxation both in the GaAs barrier and the InAs wetting layer. This conclusion follows from the analysis of the PLE spectra. Figure 2b shows two PLE spectra that correspond to the detection at the peak of the low- and high-energy bands observed in the PL spectra under excitation with $E_{exc} = 2.41$ eV (Fig. 2a). Strong absorption in the energy region above the small QD emission energy is evidence of the existence of long tails in the density of states of wetting InAs layer and excited states of the QDs both in the first and second layers. Note also that the intensity of the PL signal recorded in the energy region $E_{det} = 1.106$ eV, i.e., at the peak of the emission from large QDs (second layer), is fairly high when the excitation is tuned to the energy range of emission from small QDs (first layer). Thus, energy is efficiently transferred from this region to the one corresponding to the transitions between the ground states in large QDs. Such a transfer may take place, because of either intradot process, firstly, if after the absorption of a photon, a carrier remains in the initially excited state

of a large QD and subsequently undergoes nonradiative relaxation to the ground state of the same QD, or secondly, if a photogenerated carrier appears in the ground state of a small QD and then makes a transition to the ground state of an adjacent tunneling-coupled large QD. The possibility of the latter process is an indication of the vertical coupling of QDs located in adjacent layers; such a coupling results in the formation of new type of quantum-mechanical objects, which we call asymmetric QD molecules. To gain a more detailed understanding of the interlayer coupling between QDs, we studied the dependence of the PL spectra on the excitation photon energy in the below-barrier region.

A broad low-energy band peaked at ~0.96 eV is seen in the PL spectrum of QDs (Fig. 2a) under excitation with photon energy in the region corresponding to the transitions within the QDs of the first layer (E_{exc} = 1.355 eV); apparently, this band originates from the defect centers inevitably existing in MBE-grown samples. An additional band that appears in the PL spectrum of the QDs in the energy region ~1.36 eV under the excitation below the AlGaAs barrier (E_{exc} = 1.55 eV) is very intense and can be attributed to localized states in the InAs wetting layer related to fluctuations in its width. A broad unresolved band seen in the PL spectrum at 1.46–1.49 eV under excitation with E_{exc} = 1.55 eV arises from the recombination of electrons with light and heavy holes in the InAs wetting layer.

Comparing the three PL spectra corresponding to different E_{exc} (Fig. 2a), we see that the features of relaxation processes and the efficiency of the PL depend strongly on the excitation photon energy. Taking into account the TEM data, one can expect that, both vertically coupled QDs (asymmetric quantum-dot molecules) and QDs that are not correlated by interlayer interaction (isolated QDs), will simultaneously manifest themselves in the PL spectra. In order to determine the density of the states within the GaAs band gap, a wide range of PL-excitation photon energies were scanned, with the most careful consideration being given to the spectra of resonance PL (i.e., PL excited by the radiation tuned into the region of optical transitions in the QDs). Figure 3a shows a series of PL spectra corresponding to the excitation photon energies varied from 1.658 to 1.476 eV with ~15-meV increments. One can see that, with decreasing E_{exc}, spectral features designated as *A*, *B*, *C*, and *D* shift proportionally to lower energies. These features are attributed to (*A*) optical transitions in large QDs, (*B*) optical transitions in small QDs, (*C*) transitions between localized states in InAs wetting layer, and (*D*) to electron–hole transitions in the wetting layer. As E_{exc} approaches the energy corresponding to the GaAs absorption edge, PL bands originating from the transitions in the wetting layer and in small QDs are intensified considerably, which indicates that carriers optically excited in the GaAs barrier are more efficiently captured in the wetting layer and the QD region. With a further decrease in the excitation photon energy from 1.473 eV (extended states in the wetting layer) to the peak of the PL band at 1.355 eV (localized states in the wetting layer), contribution of the wetting layer to the PL virtually disappears (see Fig. 3b) and the integrated intensity of the PL from QDs becomes significantly reduced. In this case, the QD states are occupied via the tails of the states density of the InAs wetting layer and via the excited states of QDs in the course of the phonon-assisted relaxation. Since no distinguishable phonon structure can be found in the PL spectra as the excitation energy approaches the region around ~1.355 eV, the *C* band cannot be assigned to the QD states, which are characterized by δ-function distribution of the density of states; rather, this band is associated with quasi-continuous localized states in the wetting layer. Nevertheless, the appearance of a fairly strong QD PL signal upon below-barrier excitation tuned outside the region of QD states (i.e., beyond resonance) verifies that the GaAs band gap is

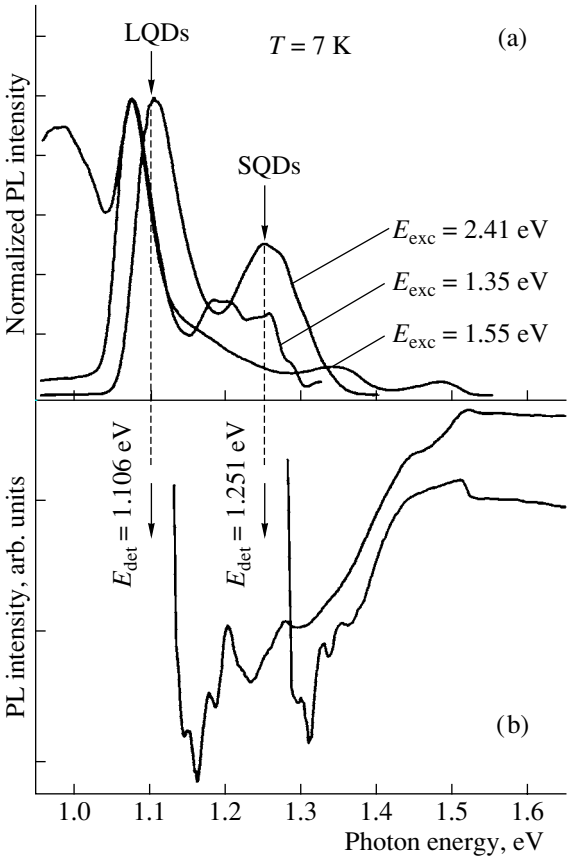

Fig. 2. (a) Normalized spectra of low-temperature (T = 7 K) photoluminescence recorded for three different values of the excitation photon energy E_{exc} = 2.41 eV (above the AlGaAs barrier), 1.55 eV (above the GaAs barrier), and 1.35 eV (below the wetting-layer absorption edge). Arrows mark the photoluminescence peaks related to the emission from large and small QDs in the spectrum corresponding to E_{exc} = 2.41 eV. (b) Two photoluminescence excitation spectra corresponding to the detection at the peaks of emission from large and small QDs (see arrows).

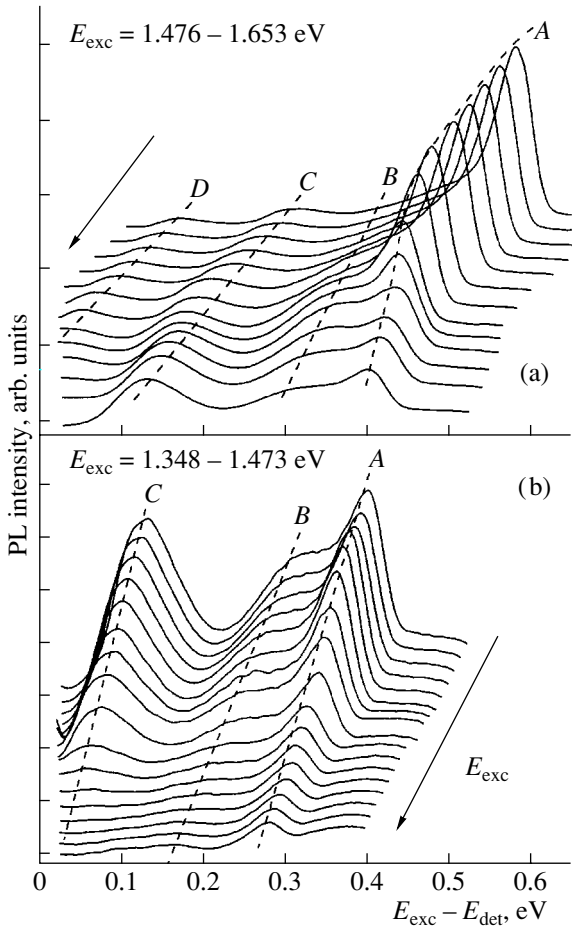

Fig. 3. (a) Low-temperature (T = 7 K) photoluminescence spectra corresponding to the excitation below AlGaAs barrier; E_{exc} varies from 1.476 to 1.653 eV in ~15-meV increments. (b) Low-temperature (T = 7 K) photoluminescence spectra corresponding to the excitation below GaAs barrier; E_{exc} varies from 1.348 to 1.473 eV in ~8-meV increments. Spectral features designated as *A* and *B* correspond to the emission from the large and small quantum dots, respectively; *C* and *D* correspond, respectively, to the transitions between localized states and electron–hole transitions in the InAs wetting layer.

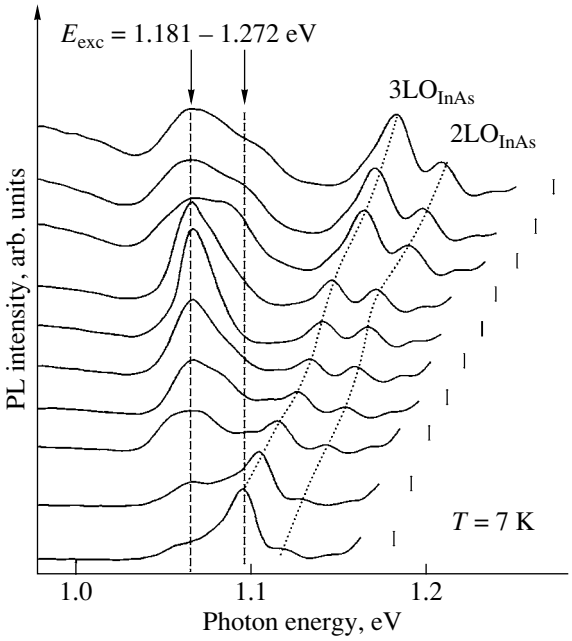

Fig. 4. Spectra of low-temperature (T = 7 K) resonant photoluminescence recorded for different values of E_{exc} varying in the range of optical transitions in small QDs. A two-component structure of the large-QD PL band is clearly visible. Selective narrowing and enhancement of the PL band corresponding to asymmetric quantum-dot molecules is observed. Vertical dashed lines indicate the positions of the band peaks in the photoluminescence spectra recorded under the excitation with photon energies of 1.55 and 2.41 eV (see Fig. 2a). Dotted lines represent the shifts of $3LO_{InAs}$ and $2LO_{InAs}$ phonon replicas.

fairly tightly filled by tails of the density of states from the wetting layer, localized and impurity-related states.

The most interesting behavior of the PL spectra is observed when the excitation photon energy scans the region corresponding to the transitions between the QD ground states (Fig. 4). Typical LO-phonon modulation of the lineshape of the PL from small QDs is observed. Sharp peaks correspond to 3LO and 2LO phonon replicas. Experimental data demonstrate that the replicas correspond to the energies 3×29 meV and 2×31 meV and, thus, are related to InAs LO phonons. The 3LO band has a higher amplitude than the 2LO band, and its dependence on the excitation photon energy in the spectral range of small-QD emission reproduces the lineshape of the PL from small QDs observed under 2.41-eV excitation (Fig. 2a).

In order to separate the contribution of correlated QDs from that of uncorrelated QDs, it is important to examine the variation in the lineshape of the PL from large QDs as E_{exc} scans the region corresponding to optical transitions in small QDs. In this case, in addition to the phonon modulation of the PL spectrum of small QDs, considerable changes of the large-QD PL lineshape take place, which can be seen in Fig. 4. Thin vertical lines indicate the excitation energies within the PL band of first-layer QDs. Arrows show the positions of the PL bands in the spectra recorded under excitation with photon energies of 2.41 and 1.55 eV. As E_{exc} is scanned across the peak of the small-QD PL band, two separate bands clearly manifest themselves in the large-QD PL spectrum (see dashed vertical lines). The high-energy band decreases rapidly and disappears, while the low-energy band experiences pronounced narrowing, reaching FWHM of ~30 meV, and grows in amplitude. The PL lineshape was analyzed by fitting it with the sum of Gaussian curves, maintaining the smallest root-mean-square deviation. With the variation of the excitation photon energy, the high-energy band shifts slightly within a region of ~6 meV; however, these

shifts are irregular, while E_{exc} decreases by about 30 meV. Thus, the emerging band cannot be attributed to a phonon replica, since such a replica should be shifted by energy equal to that of the excitation light. We assign this band to the PL of uncorrelated large QDs whose excitation conditions are not satisfied due to energy relationships and the lack of suitable intermediate states. The low-energy PL band virtually does not shift with the variation in the excitation photon energy. This band can be much less attributed to a phonon replica, since energy relationships characteristic of low-order phonon resonances are not satisfied ($E_{exc} - E_{det} \gg 3\hbar\omega_{LO}$). The contribution of phonon processes of an order higher than 3LO might be assumed. Indeed, as the 5LO replica approaches the peak of the large-QD PL band, resonance enhancement of this band can be expected due to exit resonance. However, we do not observe any sign of a 5LO phonon replica in the "pre-resonance" region and, thus, we rule out the possibility that phonon processes contribute to the observed modification of the large-QD band upon scanning the excitation photon energy across the region of small-QD transitions. Thus, we have to conclude that one of the main factors causing a pronounced reduction in the linewidth of large-QD PL band and selective enhancement of a part of it is the effective transfer of charge carriers from QDs in the first layer to QDs in the second layer, which occurs in spite of the relatively thick spacer barrier. Due to phonon-assisted tunneling, the carriers optically excited in small QDs are transferred into large QDs and subsequently undergo radiative recombination. Actually, it is the presence of this type of phonon-assisted electronic coupling between two QDs of different sizes that defines the "asymmetric quantum-dot molecule". As the excitation photon energy is reduced further, so that the large-QD energy region is scanned, phonon replicas related to the energy states in these QDs are dominant in the spectra. The lineshape of the large-QD PL is modulated by 2LO and 3LO replicas. The dependence of the intensity of 3LO replica on the excitation photon energy in the range of optical transitions in large (second-layer) QDs well reproduces the shape of the PL band of these QDs observed under 1.55-eV excitation.

4. CONCLUSIONS

Thus, using PL, PLE, and resonant PL spectroscopy, we investigated energy states of self-organized QDs in double-layer InAs/GaAs structures. In a system with the first and second InAs layer thickness of 1.8 and 2.4 MLs and GaAs spacer thickness of 50 MLs, we determined the energy states of QDs in each layer. We have shown that, in spite of the complicated structure of the density of below-barrier states, discrete states in QDs can be reliably identified using the resonant PL excitation. States corresponding to vertically coupled QDs (asymmetric QD molecules) in weakly correlated systems are observed for the first time. These states are responsible for a pronounced narrowing of the PL line (down to FWHM of ~30 meV) and selective enhancement of the large-QD PL band that take place when the excitation energy is scanned across the region of optical transitions in small QDs. Resonances corresponding to the states of asymmetric QD molecules are selectively separated from the transitions in the uncorrelated QDs in double-layer InAs/GaAs structures with a weak correlation. This makes studies of both genealogy of asymmetric quantum-dot molecules and their properties possible.

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