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Photoluminescence of Single Quantum Wires and Quantum Dots

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Abstract—The results of a study into the photoluminescence spectra of a set of quantum dots based on GaAs enclosed in AlGaAs nanowires are presented. The steady state and time resolved spectra of photoluminescence under optical excitation both from an array of quantum wires/dots and a single quantum wire/dot have been measured. In the photoluminescence spectra of single quantum dots, emission lines of excitons, biexcitons and tritons have been found. The binding energy of the biexciton in the studied structures was deduced to be 8 meV.

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INTRODUCTION

Freely arranged semiconductor nanowires (nanowhiskers) are one-dimensional nano-objects, which show promise for application in nanoelectronics [1, 3] and nanophotonics [2], as well as for fundamental research [4–10]. The production technology of such nano-objects is being actively developed [11–16]. Recently, the possibility of including one or several quantum dots in such a nanowire was demonstrated. Modern methods of epitaxial growth allow, in principle, control over the sizes and position of nanowires on the nanoscale, which is very attractive for their practical application.

One of the main issues in this field is the production of a uniform ensemble of nanowires that have a small scatter of sizes and well determined spatial position, necessary for utilization of these objects in nanoelectronics. Many aspects connected with the growth of such structures still remain unclear despite great progress in nanowire production. In particular, it is important to determine the growth parameters (temperature, flows etc.), allowing the production of uniform nanowire ensembles.

Another important issue is the investigation of new physical phenomena in such nano-objects which are unique one-dimensional structures. The lack of a wetting layer results in a significant rise in the emission quantum yield of up to 100%, which favorably differs such quantum dots from dots obtained by the Stranski–Krastanov method and makes study of their photoluminescence spectra under very low excitation intensities possible.

EXPERIMENTAL

In the experiments, AlGaAs nanowires with embedded GaAs quantum dots (QD) were used. Nanowires were grown on semi-insulating GaAs (111)B substrates in a EP1203 molecular beam epitaxy (MBE) system. The substrate surface was deoxidized at a temperature of 630°C, then a GaAs 100 nm buffer layer was grown at a temperature of 600°C to obtain an atomically smooth surface. An Au layer equivalent to 1 nm was deposited at a temperature of 550°C, and then there was a 1 min pause to achieve better uniformity of the gold droplets. The substrate temperature was fixed equal to 550 or 580°Cc. This procedure ended with the formation of Au containing droplets with a size equal to 40–50 nm. The growth of nanowires started with the simultaneous opening of Al and Ga sources. The nominal speed of growth (i.e. growth on a pure surface) was chosen equal to 1 monolayer per second for GaAs and 0.4 of monolayers per second for AlAs. Thus, the total speed of $Al_xGa_{1-x}As$ growth was 1.4 monolayers per second, which correlates with the Al content in the solid solution ($x = 0.285$) for a planar layer. For formation of the AlGaAs/GaAs/AlGaAs QD, GaAs growth began after 15 min of growth of the AlGaAs and then the AL source was closed for 5 s to

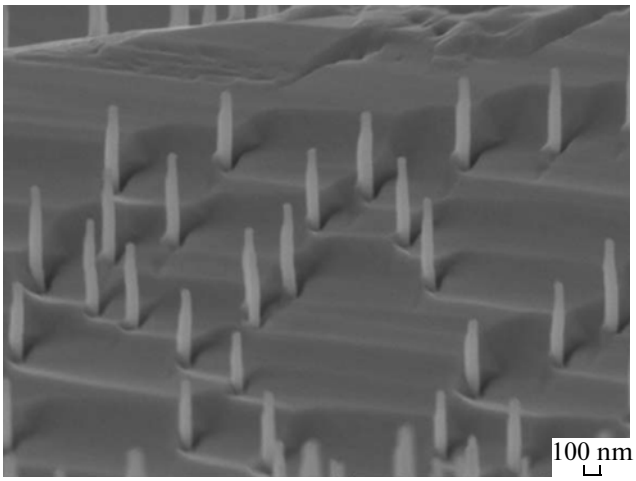

Fig. 1. SEM image of a typical structure containing nanowire, WD 5 mm.

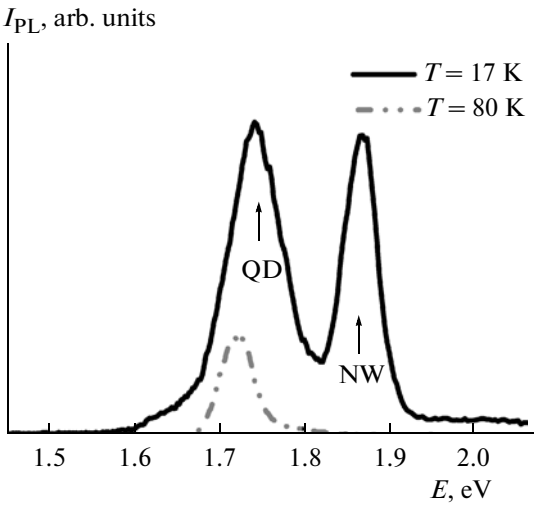

Fig. 2. Photoluminescence spectrum of a structure containing a nanowire. The solid line is that obtained at a temperature of 10 K; the dotted line is that obtained at a temperature of 77 K.

produce the GaAs quantum dot in each nanowire. Thereupon growth of the AlGaAs was resumed to complete formation of the structure. The growth was completed by the deposition of GaAs for two minutes at a temperature of 530°C to prevent the possible oxidation of AlGaAs. Growth was not interrupted at the phase interfaces. The diameter of the nanowire in the samples was 30–60 nm.

Characterization of the obtained samples was performed by scanning electron microscopy (SEM) and tunnel electron microscopy (TEM) methods before optical measurements.

The photoluminescence spectra under steady-state optical excitation and the time-resolved spectra for the sample series were measured in the temperature range 5–250 K and in a wide range of optical excitation densities. The samples were excited by the second harmonics of a Nd:YAG laser or a Ti:sapphire laser. A Jobin-Yvon monochromator (30 cm) equipped with an avalanche photodiode or a Hamamatsu streak camera was used for recording of the spectra.

RESULTS AND DISCUSSION

An SEM image of the sample grown at a temperature of 560°C is shown in Fig. 1. High homogeneity of the nanowire array in terms of length and diameter can be seen (variation in the nanowire diameters does not exceed 15%); moreover, the diameter did not change according to the length of the nanowire.

The photoluminescence spectrum of this structure taken at a temperature of 10 K is presented in Fig. 2 by a solid line. Two intense lines at energies of 1.73 and 1.87 eV dominate in this spectrum. We assign these lines to the recombination of carriers in quantum dots and nanowires respectively.

Actually, the material of the nanowire $Al_{0.25}Ga_{0.75}As$ has an energy gap width of 1.8–1.9 eV, and the energy gap width of the material of the quantum dot is equal to 1.519 eV at the temperature of liquid helium. Consequently, the pholuminescence line of quantum dots can have an energy from 1.519 eV in very large dots to 1.8 eV in very small dots. Alternatively, the photoluminescence line of the nanowire cannot be less in energy than the energy gap width of $Al_{0.25}Ga_{0.75}As$, i.e. cannot be less than 1.8 eV.

The difference in the nature of these two lines Fig. 2 is supported by the temperature dependence of the luminescence spectrum. The photoluminescence spectrum taken at a temperature of 77 K is shown in Fig. 2 by a dotted line. It can be seen that, contrary to QDs, the photoluminescence line of the nanowire disappears entirely with increasing temperature. This indicates the strong localization of charge carriers in quantum dots as opposed to carriers in nanowires, which can freely transfer to centres of non-radiative recombination.

An optical study of single QDs and single nanowires was carried out. For this aim a highly enlarged image of a sample surface was projected onto the spectrometer slit. Only the signal from a small number of nanowires was recorded. The spatial resolution of the setup was about 1.5 μm and the average distance between two nanowires was about 0.8 μm. Consequently, the photodetector could record the signal from several nanowires containing quantum dots that were close in size.

The emission spectra of three nanowires of close sizes depending on the intensity of optical excitation are shown in Fig. 3. For the given sample, the Al content in the nanowires was slightly below the usual content of 15%.

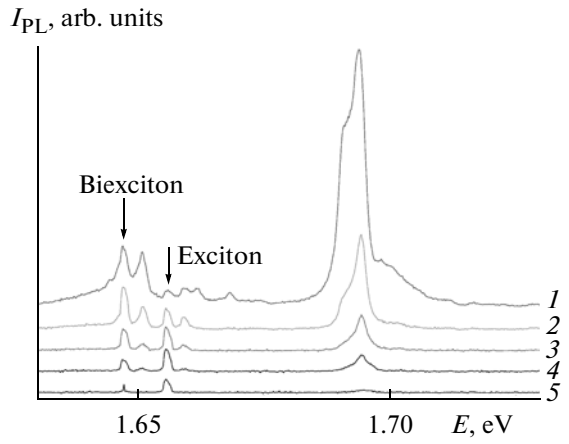

Fig. 3. Photoluminescence spectra from a single quantum dot and a single nanowire under various excitation intensities I_{exc}: (*1*) I_{exc} = 20 μW; (*2*) I_{exc} = 7.2 μW; (*3*) I_{exc} = 3.2 μW; (*4*) I_{exc} = 1.6 μW; and (*5*) 0.5 μW.

In these spectra, at the smallest excitation intensities, only one line of exciton emission is observed in a single quantum dot at the energy 1.6558 eV. This quantum dot is the nearest to the excitation spot's center and hence exhibits the greatest brightness. The photoluminescence intensity of this line flattens with rising excitation intensity and the biexciton emision line of the same dot appears in the long-wavelength range at an energy of 1.6471 eV. At a high photoexcitation intensity, the intensity of both the lines flattens out and the intensity of the biexciton line is set at the point of two intensities for the exciton emission line. This supports our interpretation of the emission line at 1.6471 eV as the biexciton emission line.

Other emission lines appear in the spectrum with increasing excitation intensity. The brightest line appears at an energy of 1.699 eV. The intensity of this line's photoluminescence increases linearly with increasing excitation intensity and does not saturate even at the highest intensities used in our experiments. We associate this line with photoluminescence from the nanofiber. Actually, the nanofiber's length is several microns and its density of states is high, which is why the intensity of its photoluminescence does not saturate. The energy position of this line corresponds to an Al content in the nanowires of ≈15%.

As well as this line, new lines appear at about 1.65 eV at rather high excitation intensities. These lines correspond to the exciton–biexiton doublet in a single QD. They are thought to belong to another quantum dot of smaller size situated on the periphery of the excitation spot. Calculation demonstrates that the binding energy of the biexciton in our quantum dots can even reach 10 meV [21].

Triton line emission can be observed in the long-wavelength range of the exciton emission line in other samples (as well as in another area of the same sample). The binding energy of the triton in such quantum dots is about 1 meV [21]. The intensity of the photoluminescence line increases with increasing excitation intensity. This line flattens out at the same level as the exciton emission line. Such behavior of the triton emission line is typical also for a quantum hole. One can expect the occurrence of chemical equilibrium in the system of excitons and tritons at sufficiently high photoexcitation levels [22].

Measurements of the photoluminescence spectra of quantum dots were also conducted in a magnetic field directed along the wire axis. All the lines in the region of 1.65 eV have been found to undergo approximately the same diamagnetic shift and the same Zeeman splitting. The diamagnetic shift of this line is 0.006 meV/T^2 and the *g*-factor is about 1. At the same time, the line of the nanowire at an energy of 1.69 eV behaves differently. The diamagnetic shift of this line is 0.009 meV/T^2, but the Zeeman splitting is small and corresponds to *g*-factors close to 0.

Measurements of the photoluminescence decay of a single quantum dot have shown that the time of decay is several nanoseconds. This value is close to data obtained on planar quantum dots.

CONCLUSIONS

Two doublets of narrow lines were observed in the photoluminescence spectra of a single nanowire at a very low intensity of optical excitation in the region of single quantum dot luminescence. These lines are believed to be explained by the recombination of an exciton and biexciton in two neighboring quantum dots. The energy of the biexciton binding was found to be about 8 meV in both cases. This interpretation is supported by the dependence of these spectra on the intensity of optical excitation. The intensity of the photoluminescence line for the biexciton increases superlinearly with increasing excitation intensity and all the lines flatten out with a further rise in intensity. The intensity of the biexciton line is set at the level of two exciton intensities. In a magnetic field, a diamagnetic shift of all lines in the spectrum and their Zeeman splitting, which is different for an exciton in a nanowire and in a quantum dot, are observed.

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