Spectrum of radiation diffracted by laser-induced gratings in GaAs/AlGaAs quantum wells

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A study has been made of the effect of microscopic roughness on the shape of the spectrum of radiation diffracted by laser-induced gratings in GaAs/AlGaAs quantum wells. The line profile was analyzed by using the composite complex permittivity profile that takes into account fluctuations in the resonance energy of the excited states. It is found from detailed comparison of the theory with the experimental results that the interface of the quantum well grown by molecular-beam epitaxy has "islands" of height equal to 1 monolayer (2.83 Å) with longitudinal dimensions much greater than the exciton diameter; they are covered by nano-size regions of roughness of diameter 80–90 Å.

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1. INTRODUCTION

The GaAs/AlGaAs quantum wells grown by molecular-beam epitaxy have low impurity and defect densities, and therefore states of free and localized excitons are dominant in their photoluminescence and excitation spectra. Because of size quantization, the positions E_x of the exciton levels depend strongly on the width of the quantum well. It was shown in Ref. 1 that the permittivity $\varepsilon(E)$ of a quantum well near an exciton resonance can be described by means of the following distribution, which takes into account both the homogeneous and the inhomogeneous broadening of the exciton levels:

$$\varepsilon(E)=\int_0^\infty \varepsilon_L(E)f(E_x)dE_x, \quad \varepsilon_L=\varepsilon_\infty+\frac{F}{E_x^2-E^2-iE\gamma} \tag{1}$$

where E is the photon energy, F is the exciton oscillator strength, and γ is the parameter of the inhomogeneous broadening of the exciton level, which is proportional to the phase relaxation time T_2. The real constant ε_∞ takes into account the contribution to the polarizability of states with energies greater than the exciton energy. The probability density $f(E_x)$ describes the inhomogeneous broadening that arises on account of the fluctuations of the resonance energy E_x near its mean value $\langle E_x\rangle$. The half-width σ of the inhomogeneously broadened exciton level is determined by scattering by lattice defects, and the fluctuations in the composition x of the material of the $Al_xGa_{1-x}As$ barrier. However, the greatest contribution to σ is made by scattering of excitons by nano-size regions of roughness on the quantum-well–barrier interfaces with a height of 1 monolayer, $\delta=2.83$ Å, and diameter less than the exciton radius.[2] For certain growth regimes, the fluctuations in the width of the quantum well lead to the formation of microscopic regions of roughness—"islands" of AlGaAs with a height of one or a few monolayers in the quantum well or "valleys" of GaAs in the barrier with a depth of one or a few monolayers with diameters appreciably greater than the exciton diameter D_x. In this case, the exciton level is split into several sublevels corresponding to different local widths of the quantum well: $W, W\pm\delta, W\pm 2\delta,\dots$. In the intermediate case, the profile of the exciton luminescence spectrum can consist of inhomogeneously broadened doublets or triplets[3] and depend on the distribution of the longitudinal length scales of the microscopic and nano-size regions of roughness in the region where an exciton with diameter D_x is localized.

Recent investigations of the nonlinear optical properties of quantum wells have used the methods of four-wave mixing[4] and diffraction of (sub)picosecond laser radiation by a laser-induced grating.[5] Analysis of the kinetics and angular dependence of the intensity of the radiation diffracted by such a grating makes it possible to determine in a self-consistent manner the exciton lifetime and diffusion coefficient in the quantum well.[2] It was shown in Ref. 6 that not only the intensity but also the profile of the diffracted radiation spectra contains information about the parameters of the homogeneous and inhomogeneous broadening of the exciton states. In Ref. 7, a model was proposed for describing the profile of the spectrum of an inhomogeneously broadened exciton state in terms of the probability of finding within the exciton diameter D_x a fluctuation in the width of the quantum well measuring $d \ll D_x$. In the present paper, this model is used to consider the interrelationship between the spectra of radiation diffracted by a laser-induced grating and the sizes and spatial distributions of the islands, valleys, and microscopic and nano-size regions of roughness of the quantum-well–barrier interfaces (see Fig. 3 below). In contrast to the luminescence spectra considered in Ref. 7, which are the result of recombination of thermalized quasiparticles, the spectra of radiation diffracted by a laser-induced grating

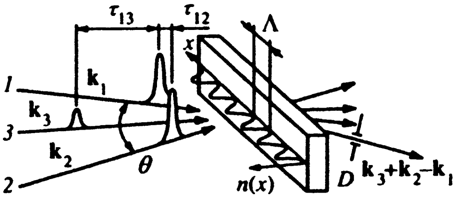

FIG. 1. Basic arrangement of the three-beam experiment on laser-induced gratings.

make it possible to investigate in greater detail the nonlinear optical properties of the ground and excited energy states of heavy and light excitons in a GaAs/AlGaAs quantum well.

2. DESCRIPTION OF THE EXPERIMENT AND RESULTS

The sample investigated in the present work was grown by molecular-beam epitaxy on a GaAs substrate oriented along the [001] axis that was then removed by selective chemical etching. The sample was a multilayer quantum structure consisting of 20 periods of GaAs layers of thickness 116 Å, between which there were $Al_{0.3}Ga_{0.7}As$ barriers of thickness 150 Å.

The source of exciting radiation was a titanium–sapphire laser with pulse duration 150 fs and repetition rate 76 MHz; the maximum of the spectrum of the laser radiation of width 40 meV coincided with the energy position 1.551 eV of the heavy exciton. The measurements were made in liquid-helium vapor at a temperature of about 5 K.

The laser radiation was divided into three beams, two of which, which had approximately the same intensity, were focused on the sample at angle θ in a spot with a diameter of about 200 μm. The interference pattern created by the coherent beams 1 and 2 (Fig. 1) led to the formation of a periodic distribution in the density of electron–hole pairs, and by virtue of the nonlinear properties of the medium this gave rise to an amplitude–phase diffraction grating with period $\Lambda=\lambda/2\sin(\theta/2)$, where λ is the wavelength of the laser radiation. The induced diffraction grating was tested by a third beam incident on the sample at a time τ_{13} after the first pulse. The radiation diffracted by the laser-induced grating in the direction $\mathbf{k}_3+(\mathbf{k}_2-\mathbf{k}_1)$ was filtered by a stop focused on the entrance slit of a spectrometer with resolution 0.1 meV and was detected by the multichannel optical detector OMA-4.

Figure 2 shows the characteristic profile of the spectrum of the diffracted beam and the change in the intensity of each of the four peaks as a function of the delay time τ_{13}. Studying the decay of each of the four peaks (Fig. 2), allows the lifetime and diffusion coefficients of the excitons in the corresponding energy states to be determined. The two most intense peaks correspond to a heavy exciton in a quantum well whose width fluctuates near the nominal value by an amount equal to the height of one monolayer, which led to a splitting of the state by 1.16 meV. The heavy-exciton lifetime in the "broad" sections of the quantum well with resonance energy 1.55062 eV was $T_1=560$ ps, and the diffusion coefficient was $D=3.1$ cm^2/s. For the heavy exciton in the "narrow" sections of the well with resonance energy 1.55181 eV, we have $T_1=450$ ps and $D=3.5$ cm^2/s. In both sections, the exciton binding energy was about 10 meV. The two other energy states correspond to light excitons split by 1.5 meV by the fluctuations in the width of the well. In this paper, we shall not consider these states on account of the low intensity of the diffracted radiation corresponding to them.

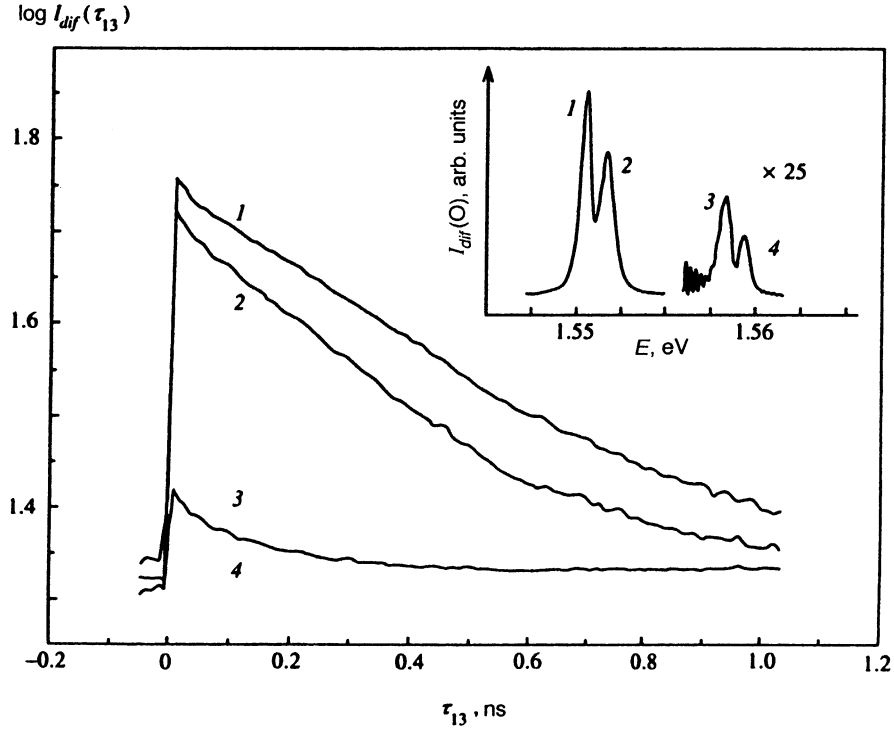

FIG. 2. Change in the intensity of the four peaks observed in the spectrum of the diffracted radiation (shown in the inset) versus the delay time τ_{13} for $\Lambda=30$ μm.

3. THEORETICAL MODEL

In the sample excited by the beams 1 and 2, there can exist, after damping of the coherent processes, a periodic distribution in the density of the excited states. Because the permittivity depends on the exciton density, such a system will exhibit the properties of an amplitude–phase diffraction grating with period Λ. The maximum phase difference that arises through the dependence of the refractive index near the exciton resonance on the density of the electron–hole pairs is much less than 2π in samples with thickness less than 200 nm, and therefore the effect of the phase modulation on the formation of the diffraction pattern can be ignored.

The nonlinearity of the permittivity is determined by the dependence on the exciton density n of the three following parameters: the energy position $E_x(n)$ of the excitons, the damping $\gamma(n)$, and the oscillator strength $F(n)$. With increasing delay τ_{13}, the amplitude and shape of the periodic distribution of the exciton density $n(x)$ along the transverse coordinate x change as a result of diffusion and the finite lifetime of the excited states; however, the grating period Λ remains unchanged. Therefore, with decreasing density of excitons the peaks in the spectrum of the diffracted radiation are slightly displaced and become narrower. However, the experimentally observed shifts and changes in the shape of the diffracted radiation were found to be slight compared with the change in the intensity of the radiation at all delays τ_{13}. Therefore, we assume that the dependence of the permittivity (1) on the exciton density n is mainly determined under the conditions of our experiment by the form of the dependence $F(n)$. The half-width of the spectrum of the laser radiation incident on the sample was much greater than the half-width of the energy states that we investigated, and therefore the dependence of the intensity of the diffracted radiation on the photon energy E has the form

$$I(E)\sim\alpha_0^2(E)\sim E^2\varepsilon_2^2(E), \tag{2}$$

where $\alpha_0(E)$ is the linear absorption coefficient, and ε_2 is the imaginary part of the permittivity (1). The damping γ in the expression (1) is determined by the uncertainty relation $\gamma T_2\sim\hbar$, where T_2 is the time of loss of phase coherence, or, equivalently, the exciton lifetime in one quantum state. For the sample we use, $T_2\sim 10$ ps, and therefore $\gamma\approx 0.4$ meV. This value is a few times less than the half-width of the observed lines and confirms the need to use the composite distribution (1) to describe the spectrum. However, the experimentally observed spectrum of the diffracted radiation is not only inhomogeneously broadened but also has a clear doublet structure. To understand what determines the value of σ and the profile of the heavy-exciton spectrum consisting of two lines, it is necessary to consider the structure of real GaAs/GaAlAs quantum wells.

The exciton energy E_x in the GaAs well depends on the width of the well. If the boundaries between the GaAs and GaAlAs layers were absolutely smooth, the distribution $f(E_x)$ describing the inhomogeneous broadening [see the expression (1)] would take the form of a δ function. In such a case, we should observe in the spectrum of the diffracted signal just a single line corresponding to the energy position of the heavy excitons in a quantum well with definite width. The half-width of this line would be determined solely by the magnitude γ of the inhomogeneous broadening.

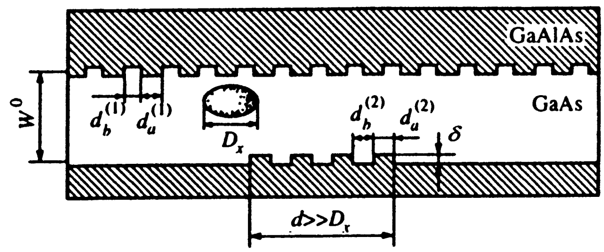

FIG. 3. Proposed model of the structure of the quantum well.

Molecular-beam epitaxy, by means of which the structure we investigated was fabricated, makes it possible to control the mean (over the substrate plane) number of grown GaAs and GaAlAs monolayers, but it is not possible by means of the method to create flat interfaces between these layers. At the GaAs–AlGaAs interface, steps with a height of one or two monolayers are formed in the process of growth,[3] and create islands of GaAlAs in the quantum well or valleys of GaAs in the barrier. The fluctuations in the local width of the quantum well change the resonance energy E_x of the excited states. If the longitudinal dimensions of the islands are much greater than the exciton diameter ($D_x=250$ Å), then the spectrum of the exciton energy states will consist of several narrow peaks, the position of which is determined by the local width of the quantum well in the region of the island or in the region of the valley. It was shown in Ref. 8 that decrease in the width ($W=116$ Å) of the GaAs quantum well by one monolayer leads to an increase in the heavy-exciton energy by ≈ 1.16 meV, which is very close to the splitting that we observed. Therefore, to describe the investigated doublet structure of the spectrum of the diffracted radiation, we propose to use a model in which one of the well–barrier interfaces is a smooth surface (valley) covered by islands with a height of one monolayer and having longitudinal dimensions much greater than the exciton diameter.

To explain the half-width of the observed lines, we also used the model of nano-size regions of roughness described in Ref. 7. This model makes it possible to take into account defects on the surface of the valleys and islands that arise on account of the local microscopic inclusions of GaAs in the barrier layer and of GaAlAs in the well layer. In their shape, the nano-size regions of roughness resemble the structure of the valleys and islands except that their longitudinal dimensions must be at least a few times less than the longitudinal dimensions of the exciton. In the presence of such nano-size regions of roughness, the energy of the quasiparticles reflects the average width of the quantum well in the region occupied by the exciton. Bearing in mind that GaAs grows much more nonuniformly on AlGaAs than does GaAlAs on GaAs, we represented the final form of our model as in Fig. 3. The upper boundary of the quantum well is completely covered with "protuberances" and "dips" (it is assumed that the quantum well grew from the top to the bottom). At the lower boundary, the defects are grouped into islands. We ignore the presence of nano-size regions of roughness between the islands.

For such a model of the quantum well, the separation between the peaks of the energy states corresponding to the positions of excitons in the region of an island and in the region of a valley will be determined by the effective height of an island, which depends on the longitudinal dimensions of the protuberances, dips, and the distance between them. Varying the mean values of these quantities, we can obtain an effective change in the width of the quantum well from zero to one monolayer. Thus, the half-width of the exciton line will be determined by the probability for finding dips and protuberances of different diameters in the region occupied by the exciton.

To determine the required probability, we shall, as in Ref. 7, use statistical arguments (similar to those used by Lifshitz to find the local density of states in disordered alloys). We first consider the general case. It is assumed that the regions occupied by the dips, protuberances, and the planes between them are distributed randomly over the surface. The longitudinal dimensions of the smallest clusters associated with the protuberances, dips, and planes will be denoted by d_a, d_b, and d_c, respectively. The probability of finding fluctuation concentrations C_a, C_b, C_c in a region of linear dimensions D_x in the region of valleys is determined solely by the roughness of the upper boundary (see Fig. 3) and can be calculated in accordance with the expression

$$P^{(1)}(C_a^{(1)},C_b^{(1)},C_c^{(1)};D_x)=\exp\left[-\sum_{i=a,b,c}\frac{D_x^2}{d_i^{(1)2}}C_i^{(1)}\ln\left(\frac{C_i^{(1)}}{C_{0i}^{(1)}}\right)\right], \quad (3)$$

where $C_{0a}^{(1)}$, $C_{0b}^{(1)}$, $C_{0c}^{(1)}$ are the mean concentrations of the roughness of the upper interface between the GaAs and GaAlAs layers. The width of the quantum well in this region is determined by

$$W=W^0+\delta\ [(C_a^{(1)}-C_{0a}^{(1)})-(C_b^{(1)}-C_{0b}^{(1)})], \quad (4)$$

where W^0 is the mean width of the quantum well above the valleys ($W^0=116$ Å). Knowing the probability of a fluctuation in the width of the quantum well, we can obtain the probability distribution with respect to the energies of the exciton states and, thus, the explicit form of the dependence $f(E_x)$. To translate the width of the quantum well into the exciton energy, we used the results of Ref. 8. The experimental data are not sufficient to determine separately all the parameters d_i, and therefore we assume that

$$C_{0a}^{(1)}=C_{0b}^{(1)}=0.5, \quad C_c^{(1)}=0, \quad d_a^{(1)}=d_b^{(1)}. \quad (5)$$

Curve *1* in Fig. 4 shows the dependence on the longitudinal dimension $d_b^{(1)}$ of a dip of the half-width σ of the spectrum of the diffracted radiation $I_{\text{dif}}(E)$ calculated in this manner [see Eq. (2)] for the case of exciton excitation in the region of the valleys. The experimentally measured half-width of the corresponding state (1.55062 eV) for different periods of the lattice-induced grating was found to be 0.69 meV to accuracy 10%. It can be seen from Fig. 4 that the best agreement between theory and experiment is obtained for $d_a^{(1)}=82$ Å.

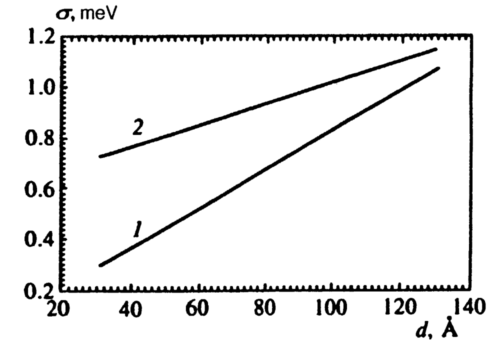

FIG. 4. Half-width σ of the energy states in the region of a valley versus the longitudinal dimensions of the microscopic regions of roughness $d_b^{(1)}$ (curve 1) and versus $d_b^{(2)}$ in the region of an island for $d_b^{(1)}=82$ Å (curve 2).

To determine the position and half-width of exciton energy states above islands, it is necessary to take into account the nano-size regions of roughness of the surface of the islands. As was shown in Ref. 7, in not-too-narrow quantum wells, with width $\geqslant 10$ monolayers, the microscopic regions of roughness of the upper and lower boundaries are not correlated. Then the probability of fluctuation of the parameters that determine the well size will be expressed by the product of the probabilities of the corresponding fluctuations of the two surfaces: $P^{(1)}P^{(2)}$, where $P^{(2)}$ can be calculated in the same way as $P^{(1)}$ if in the expression (3) we replace the parameters $C_i^{(1)}$ and $d_i^{(1)}$ by the parameters $C_i^{(2)}$ and $d_i^{(2)}$. Let

$$C_{0a}^{(2)}=0.5+z, \quad C_{0b}^{(2)}=0.5-z,$$

$$\frac{C_{0a}^{(2)}}{C_{0b}^{(2)}}=\frac{d_a^{(2)2}}{d_b^{(2)2}}, \quad C_c^{(2)}=0, \quad (6)$$

where z is a small parameter by means of which we adjusted ratio of the longitudinal dimensions of the dips and protuberances. This enabled us to obtain for the splitting a value corresponding to the one observed experimentally (for our data, we found $z=-0.018$). Then for the width of the quantum well, we have

$$W_1=W_1^0+\delta(C_a^{(1)}+C_a^{(2)}-1-z). \quad (7)$$

We use $P^{(1)}$ and $P^{(2)}$ to denote the probabilities that the independent parameters $C_i^{(1)}$ and $C_i^{(2)}$ have certain values. As can be seen from (7), the well width depends on the sum of $C_a^{(1)}$ and $C_a^{(2)}$, and therefore we obtain for the probability $P(W)$

$$P(W)=\begin{cases}\displaystyle\int_0^C P^{(1)}(x)P^{(2)}(C-x)dx, & C<1,\\[2ex] \displaystyle\int_{C-1}^1 P^{(1)}(x)P^{(2)}(C-x)dx, & C>1,\end{cases} \quad (8)$$

where $C=C_a^{(1)}+C_a^{(2)}$.

The dependence of the half-width of the energy spectrum of the diffracted signal on $d_b^{(2)}$ (for $d_a^{(1)}=d_b^{(1)}=82$ Å) obtained from (2), (7), and (8) is shown by curve 2 in Fig. 4, from which we find that the experimental results (σ=0.97

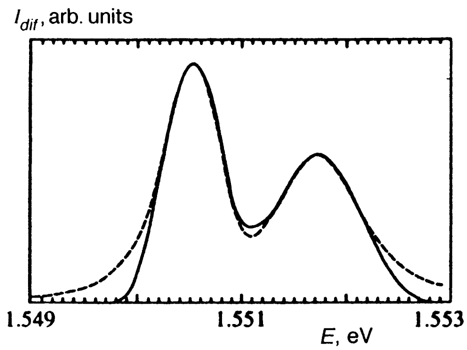

FIG. 5. Comparison of the experimental (dashed curve) and theoretical (solid curve) profiles of the spectrum of the diffracted radiation.

meV for the peak with maximum 1.55181 eV) are best described for $d_b^{(2)}=90$ Å. The protuberance diameter is found from (6) to be $d_a^{(2)}=93$ Å.

It follows from the ratio of the intensities of the experimental lines with energies 1.55062 and 1.55181 eV that islands occupy about 30% of the surface of one of the walls of the quantum well. The experimental profile of the spectrum of the diffracted radiation is compared with the theoretical profile obtained for the parameters $d_i^{(j)}$ that we found in Fig. 5. The intensities of the peaks of the theoretical spectrum are normalized to the corresponding experimental peaks.

4. CONCLUSIONS

From analysis of the kinetics and angular dependence of the intensity of radiation diffracted by a laser-induced grating we have determined the diffusion coefficients $D_1=3.1$ cm^2/s and $D_2=3.5$ cm^2/s of excitons propagating, respectively, between and above islands of height 2.83 Å with diameter much greater than the exciton diameter. We have also determined the lifetimes of such excitons: $T_1=560$ ps and $T_2=450$ ps. Their quantitative difference is due to the additional localization of the excitons in the valleys between the islands. From the spectral position of the exciton resonances, we have determined the mean width of the quantum well to be 116 Å, and from the amount of the splitting we have determined the fluctuations of the well width. Using the ratio of the intensities of the doublet, we have estimated the specific area occupied by islands and valleys. From the profile of the spectrum of the radiation diffracted by the laser-induced grating we have determined the mean diameters of the dips and protuberances of the nano-size regions of roughness at the boundaries of the quantum well and also the mean separation between them.

Thus, analysis of the radiation diffracted by a laser-induced grating makes it possible to obtain information about the kinetic parameters of the exciton states and the geometrical characteristics of the quantum-well–barrier interfaces in the nanometer range.

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