Exciton dynamics in GaAs/$Al_xGa_{1-x}As$ quantum wells

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The changes induced in the optical absorption spectrum of a GaAs/$Al_xGa_{1-x}As$ multiple quantum well due to a photoexcited carrier distribution are reexamined. We use a femtosecond pump-probe technique to excite excitons and free electron-hole pairs. We find that for densities up to 10^{11} cm^{-2} the saturation of exciton absorption is caused both by a reduction of the oscillator strength and by a broadening of the exciton resonance, which exhibit quite different temporal evolutions. The restoring of the initial value of the reduced oscillator strength is determined by the free-carrier lifetime, which we have measured to be 65 ps, whereas the temporal evolution of the broadening is due to the exciton-exciton interaction and determined by the exciton lifetime, which is found to be 410 ps. We have measured the oscillator strength saturation densities and shown that at low temperature the influence of free electron-hole pairs on the exciton oscillator strength is twice as effective as the influence of other excitons. [S0163-1829(99)11011-7]

I. INTRODUCTION

The confinement of electrons and holes in semiconductor quantum wells leads to the strong enhancement of excitonic optical transitions. This provides strong optical nonlinearities that can be used for all-optical or electro-optical switching devices.[1] One fundamental nonlinearity is the absorption saturation of the exciton by photoexcited free and bound electron-hole pairs.[2] Understanding the basic physics of this saturation is essential for optimizing the performance of electroabsorptive photonics devices.[3]

The linear exciton absorption is described by three parameters: the oscillator strength f_x, the linewidth Γ_x and the resonance energy E_x. The nonlinearity arises when one or more of these quantities is changed by the influence of photoexcited free and bound electron-hole pairs. A change of E_x shifts the exciton line. A reduction of the oscillator strength (ROS) and a broadening decrease the initial exciton absorption,[4] that was denoted as exciton bleaching in the previous works and we shall keep this term throughout the paper. The carrier induced modification of the exciton absorption, or exciton bleaching, has been experimentally investigated by spectrally resolved nonlinear transmission measurements such as pump-probe experiment or four-wave mixing. However, there were some problems with distinguishing the separate influences on each of the parameters, because they simultaneously affect the exciton absorption.

Many experimental and theoretical works have been carried out to understand the relative importance of carrier-induced ROS and broadening on the exciton bleaching. In earlier works,[5,6] in which the multiple quantum wells (MQW) with a large contribution of inhomogeneous broadening were investigated, no changes in exciton absorption linewidths with increasing photoexcited quasiparticle density were detected and the exciton bleaching was attributed exclusively to ROS. The inhomogeneous broadening has a negligible carrier density dependence.[7] So in Refs. 5 and 6 the changes of homogeneous broadening, which should lead to the exciton bleaching, were hidden by the inhomogeneous broadening background. Nevertheless, direct measurements of broadening in bulk GaAs (Ref. 8) and GaAs/$Al_xGa_{1-x}As$ MQW (Ref. 9) by four-wave mixing showed that broadening of exciton lines has to play an important role. This result was proved by the authors of another experimental work,[10] in which a high-quality MWQ sample was investigated by a pump-probe experiment. The authors of Ref. 10 registered the significant contribution of broadening and found that for their experimental conditions broadening may be even the only reason of the exciton bleaching.[10] To separate the role of each exciton parameter in Ref. 10 the line shape of the exciton absorption spectrum was approximated by a Lorentzian function. Besides the fact that for a more accurate description of exciton absorption one should use a Gauss-Lorentzian function,[7,11] this approximation is very sensitive to the absorption background, which strongly depends on the fluctuations of the intensity of the exciting laser radiation. Theoretical calculations predict that ROS is detectable even at low photoexcited densities.[12,13] This, however, was not experimentally revealed in Ref. 10. Recent experimental studies employing a pump-probe experimental technique,[14] in which an integral determination of the OS was used, have shown that both ROS and broadening are clearly observed

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even at low carrier density and that pure broadening may be dominant only at very low carrier densities, while the contribution of ROS to the exciton bleaching becomes more important at higher densities.

The next important question about the exciton dynamics was what quasiparticles—excitons or free electron-hole pairs—are dominant in the exciton broadening and ROS. The influence of free and bound carriers on the broadening was extensively investigated by four-wave mixing in the works[8,9] for low and intermediate carrier densities and in the works[15,16] for high carrier density. It was found that for low and intermediate densities the broadening exhibits a linear dependence upon the density of free or bound electron-hole pairs and that the efficiency of free carriers to produce exciton broadening is one order of magnitude higher than the efficiency of excitons. The density dependence of the broadening due to exciton-exciton interaction remains linear for even high carrier densities, whereas the broadening induced by free electron-hole pairs demonstrates a square-root density dependence for high carrier densities.

The investigation of the behavior of ROS was more intricate. The first attempts of finding the cause of ROS are undertaken in the works,[5,6] in which the influence of broadening on the exciton absorption was completely ignored. The authors of these works concluded that the influence of excitons on ROS was at least twice as effective as the influence of thermalized free electron-hole pairs. However, Schmitt-Rink *et al.*[12] predicted theoretically the completely opposite situation, that in quasi-two-dimensional media cold electron-hole pairs should produce ROS more effectively. In another theoretical investigation Zimmermann[13] obtained that at low temperature excitons should produce stronger ROS than free carriers, whereas at temperatures higher than 25 K the efficiency of both free and bound quasiparticles should be practically the same. In the experimental work,[14] in which the authors register the simultaneous influence of both ROS and broadening on the exciton bleaching, the OS was determined as a sum value for heavy- and light-hole excitons and for free electron-hole pairs which made it impossible to investigate each observed resonance separately. Thus, studying the literature, it is obvious neither theoretically nor experimentally what the main reason is for ROS.

The use of a high-quality sample with quite a large energy distance between heavy- and light-hole excitons and applying a moment analysis (see below) to process the absorption spectra, allow us to investigate the origin of the ROS of heavy-hole excitons. In this paper we experimentally measure the saturation density of the heavy-hole exciton OS and show explicitly that at low temperature the influence of free electron-hole pairs on the ROS is twice as strong as the influence of other excitons. Also we show the specific difference of each exciton parameter in the modification of the exciton absorption.

II. METHODS AND ANALYSIS

The MQW under investigation was grown by molecular-beam epitaxy and consisted of 20 periods of 80-Å-wide GaAs layers and 100-Å-wide $Al_xGa_{1-x}As$ layers. In all experiments the sample was placed in a helium optical cryostat and its temperature was maintained close to 5 K. The laser source was a tunable Ti-sapphire laser with 120-fs pulses and a repetition frequency of 76 MHz. As an experimental method of the investigation of the exciton dynamics we used the pump-probe technique.[14]

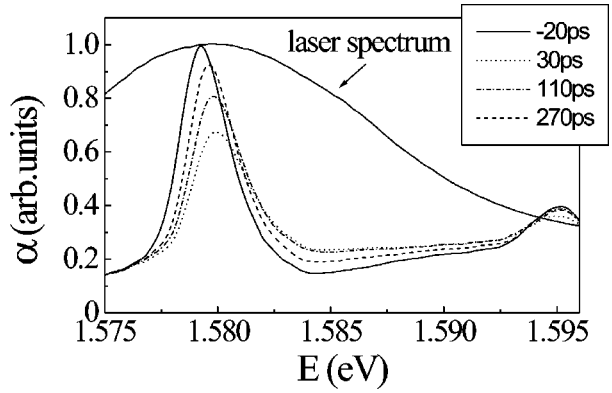

FIG. 1. Change in shape of the exciton absorption spectrum as a function of the time delay τ for the resonant excitation. The narrow solid line shows the spectrum of the exciting laser radiation.

The typical absorption spectra of the sample are displayed in Figs. 1 and 2 for different time delays of the probe beam and different spectral positions of the laser radiation. The initial photoexcited quasiparticle density was approximately equal to 9×10^{10} cm^{-2} for both excitation conditions. The spectral positions of the exciting laser radiation are indicated in Figs. 1 and 2 by the thin solid curves. The lines of heavy-hole (1.579 eV) and light-hole (1.595 eV) excitons are clearly seen in the absorption spectra. In the present paper we shall examine the behavior of the heavy-hole excitons.

We analyzed the experimental results by the moment analysis method.[17,18] This method does not require knowledge of the exact shape of the absorption spectrum and consists of defining all excitonic parameters in terms of integrals. The computational results obtained for both excitation conditions are presented in Figs. 3 and 4. One can clearly see from these figures that the exciton bleaching is due to both ROS and broadening of the excitonic line. But these parameters explicitly exhibit different behavior in time.

In the case of resonant excitation (see Fig. 3) the ROS, the broadening and the shift of the exciton resonance occur at the time the pump pulse reaches the sample ($\tau=0$). The OS

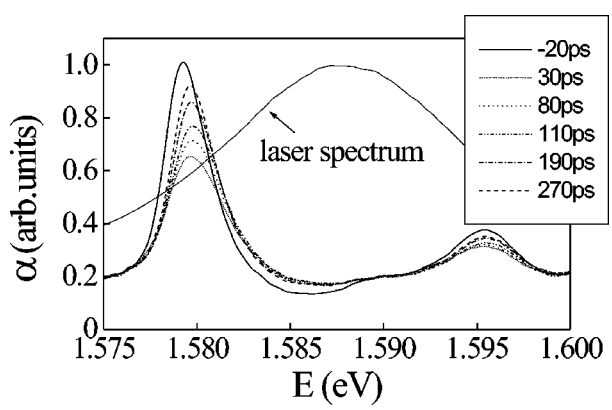

FIG. 2. Change in shape of the exciton absorption spectrum as a function of the time delay τ for the nonresonant excitation. The narrow solid line shows the spectrum of the exciting laser radiation.

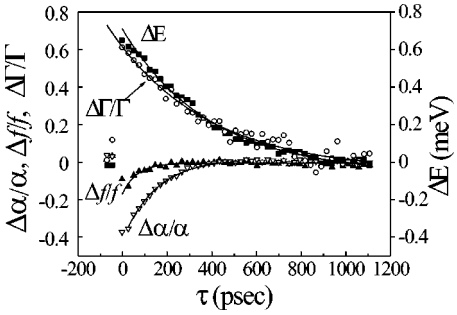

FIG. 3. Relative changes in the oscillator strength F, the maximum of the absorption coefficient α, the broadening Γ and energy shift ΔE of the exciton resonance as a function of the time delay τ for the resonant excitation (see Fig. 1).

exponentially restores its initial value with the characteristic time of 65 ps, whereas the time behavior of the broadening and the blueshift are described by a remarkably different characteristic time of 410 ps. As will be shown the ROS is basically due to the influence of free electron-hole pairs, which disappear from the interacting system during the first 100 ps forming the excitons.[15] The main reason for the broadening and the blueshift in this case is the exciton-exciton interaction, so the evolution of these parameters is determined by the exciton lifetime. In the case of nonresonant excitation (see Fig. 4) the ROS and the broadening exhibit almost the same behavior as for resonant excitation, but the relative change is twice as big for the same total photoexcited carrier density. The blueshift reaches its maximum only after 110–130 ps. This is caused, as was mentioned in, Ref. 10, by the influence of free carriers, which leads to an additional redshift of the excitonic lines. For larger time delays, more than 130 ps, the blueshift and the broadening decrease exponentially with the same characteristic time of 410 ps.

In order to understand which interaction mechanisms influence the ROS, the broadening, and the shift of the excitonic line we investigated the change of each excitonic parameter as a function of the quasiparticle density for a fixed

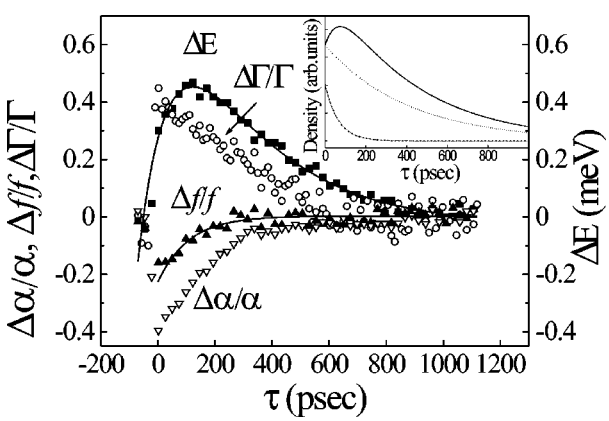

FIG. 4. Relative changes in the oscillator strength f, the maximum of the absorption coefficient α, the broadening Γ and energy shift ΔE of the exciton resonance as a function of the time delay τ for the nonresonant excitation (see Fig. 2). In the insert the temporal evolution of exciton density in the presence (solid line) and in the absence (dot line) and the decreasing of the free electron-hole pair density (dash line) are shown.

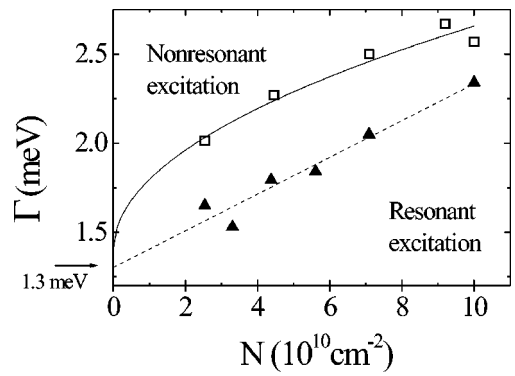

FIG. 5. Half-width of the heavy-hole exciton absorption line vs the electron-hole pair density for constant time delay $\tau \approx 20$ ps and for both excitation conditions.

time delay $\tau \approx 20$ ps. This time was chosen because all relaxation processes for both free and bound carriers should be completed after this time has elapsed.[15]

III. BROADENING OF THE EXCITON RESONANCE

The broadening of the excitonic line as a function of the quasiparticle density for a fixed time delay ($\tau \approx 20$ ps) is displayed in Fig. 5. As one can see from this figure, we obtained a clearly linear dependence for the case of resonant excitation (solid triangles). This confirms that exciton-exciton interaction is responsible for the broadening. Free electron-hole pairs produce a square-root dependence[16] as one can see for the case of nonresonant excitation (open squares).

Let us consider the resonant excitation. To describe the changing of the excitonic linewidth in this case, we employ the relation[8,9]

$$\Gamma_x(n) = \Gamma_x(0) + \gamma E_b a_b^2 N_x, \tag{1}$$

where a_b is the Bohr radius of the exciton in the MQW, N_x is the density of two-dimensional excitonic gas, and γ is a proportionality coefficient, or scattering efficiency.[8,9] To determine a_b we shall use the relation

$$E_{1S}^{(\alpha)} a_b^{(\alpha)} = \text{const}, \tag{2}$$

where $E_{1S}^{(\alpha)}$ and $a_b^{(\alpha)}$ are the binding energy and Bohr radius of an exciton in a medium of dimension α.[19] A variational calculation[20] also confirms the validity of relation (2). For our MQW it was found that $E_{1S}^{(\alpha)} = E_b \approx 10$ meV ($\alpha \approx 2.3$) and, consequently, $a_b^{(\alpha)} \approx 59$ Å. Substituting $a_b^{(\alpha)}$ for a_b in Eq. (1) and approximating the experimental results, we find that for resonant excitation $\gamma = 3.1 \pm 0.4$ and $\Gamma_x(0) = 1.30 \pm 0.08$. We shall now compare these values to the experimental results obtained in Ref. 9, where the broadening of the exciton resonance was determined as a function of the quasiparticle density in a four-wave mixing experiment with preexcitation of both free electron-hole pairs and excitons. Using relation (2), we get from Ref. 9 $\gamma = 2.8 \pm 0.56$ for the case of exciton-exciton scattering and $\gamma = 21.8 \pm 3.7$ for scattering by free carriers. The value of γ we obtained in our experiment confirms the conjecture that in the case of reso-

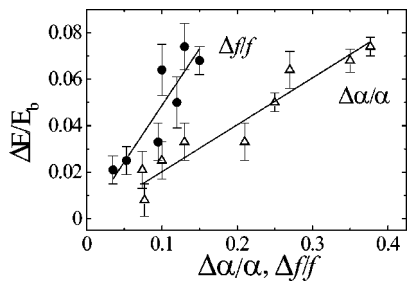

FIG. 6. Energy shift of the exciton resonance in units of the binding energy vs the relative change in oscillator strength and vs the relative change in the maximum of the absorption coefficient for a fixed time delay ($\tau \approx 20$ ps) and different electron-hole pair densities.

nant excitation the broadening of the excitonic lines is governed by the exciton-exciton interaction.

Let us now compare the experimental scattering efficiency γ with the calculations performed in Ref. 21. It was found there that the exciton-exciton scattering is described by the parameter $\gamma^{(2D)}=0.41$ for an ideal two-dimensional quantum well and by $\gamma^{(3D)}=56.6$ for an ideal bulk semiconductor. Our value $\gamma \approx 3.1$ falls between $\gamma^{(2D)}$ and γ^{3D}, attesting to the fact that we are dealing with an intermediate quasi-two-dimensional case.

The nonresonant excitation case is shown in Fig. 5 by squares. The appearance in the interacting system of an additional free carrier density changed the dependence of the broadening on the quasiparticle density. One can see from Fig. 5 that the broadening is now proportional to the square-root carrier density. This dependence is in a very good agreement with the experimental results of Ref. 16 in which the authors directly measured the exciton broadening caused by free electron-hole pairs and found

$$\Gamma_x(N)=\Gamma_x(0)+kN^{1/2}. \tag{3}$$

The best fit to our data is found for $\Gamma(0)=1.40 \pm 0.18$ meV and $k=(0.39\pm 0.07)\times 10^{-10}$ meV cm^2 and is shown in Fig. 5 by the solid curve. It should be mentioned that the magnitudes of $\Gamma_x(0)$ coincide for both excitation conditions, so the models we have used to describe the influence of both free and bound quasiparticles on exciton broadening are compatible. For a relatively short time delay, it is expected for nonresonant excitation that the broadening of the exciton line is caused by scattering with free electron-hole pairs. After 110–130 ps, when all free carriers have formed the excitons, the density dependence of the broadening again becomes linear due to exciton-exciton scattering.

IV. EXCITON OSCILLATOR STRENGTH

Figure 6 displays the shift of the excitonic line normalized to the binding energy $\Delta E/E_b$ versus the relative change $\Delta f/f_b$ in the OS and versus the relative change $\Delta\alpha/\alpha_b$ in the maximum of the absorption coefficient in the case of resonant excitation. Each point of these curves corresponds to a certain value of the photoexcited quasiparticle density at the same time delay $\tau \approx 20$ ps. To describe these results we used the formulas[12]

$$\frac{\Delta E}{E_b}=C^{(f)}\frac{\Delta f}{f_{1S}}, \tag{4}$$

$$\frac{\Delta E}{E_b}=C^{(\alpha)}\frac{\Delta \alpha}{\alpha_{1S}}, \tag{5}$$

where E_b, f_{1S}, and α are the experimental values of the binding energy, OS, and maximum absorption coefficient of the unperturbed excitonic state, respectively, and $C^{(f,\alpha)}$ are proportionality coefficients. From Fig. 6 we obtained $C^{(\alpha)}=0.20\pm 0.02$ and $C^{(f)}=0.49\pm 0.06$. The value of $C^{(\alpha)}$ agrees very well with previous results,[22] but the value of $C^{(f)}$ differs significantly. This proves the assumption employed in Ref. 22 that the exciton bleaching is determined exclusively by a decrease of the OS to be incorrect. Our value of $C^{(f)}$ is the first experimental determination of this quantity, so that it can be compared only with the results of theoretical calculations. Using the result of Ref. 12, we find for a real quantum well that in the case when cold excitons are responsible for the change in OS the proportionality coefficient $C^{(f)}$ equals

$$C_{exc}^{(f)}=0.45\frac{E_b^{(2D)}}{E_b}, \tag{6}$$

and in the case when the cold plasma is responsible,

$$C_{pl}^{(f)}=0.22\frac{E_b^{(2D)}}{E_b}, \tag{7}$$

where $E_b^{(2D)}$ is the binding energy of an ideal two-dimensional exciton. It should be mentioned that in Ref. 12 the influence of the electron-hole plasma was completely ignored in the calculation of the quasiparticle-density dependence of the magnitude of the blueshift, which we employed to obtain expressions (6) and (7). As a result of the quite wide spectrum of the laser radiation, free charge carriers are generated under our experimental conditions. However, their effect on the energy position of the excitonic line is very small. This effect is expressed as a small deviation of the time dependence of the blueshift from an exponential for small value of time delay τ (see Fig. 4) and leads to an error in the determination of ΔE of only a few percent. Substituting into expressions (6) and (7) the value $E_b \approx 10$ meV, we obtained for our MQW $C_{exc}^{(f)}=0.76$ and $C_{pl}^{(f)}=0.37$. Comparing these values with experimental value $C^{(f)} \approx 0.49$, one can conclude that despite their relatively low density the free carriers have a decisive influence on the ROS. This result confirms the prediction by Schmitt-Rink that the influence of free carriers on OS is more effective than that of excitons.[12]

As one can see from Fig. 4, a change in the OS is observed during first 120 ps, while the broadening and shift of the exciton line, which are caused by the exciton-exciton interaction, continue for several hundreds of picoseconds. Such a rapid return of the OS to its initial value is due to the vanishing of the free carriers from the system of interacting particles. Using an exponential function to describe the behavior of the OS, we were able to measure directly the lifetime of free electron-hole pairs. It was found that this time remains practically unchanged right up to quasiparticle density 10^{11} cm^{-2} and equals $T_{1,eh}=(65\pm 8)$ ps.

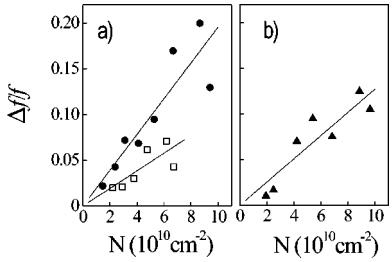

FIG. 7. Relative changes in the exciton oscillator strength as a function of electron-hole pairs density for nonresonant excitation: solid circles ($\tau \approx 20$ ps) and solid squares ($\tau \approx 135$ ps); for resonant excitation: open triangles ($\tau \approx 20$ ps).

In order to determine the contribution of free and bound quasiparticle to the ROS, we simultaneously investigated the relative changing of the OS ($\Delta f/f$) as a function of quasiparticle density for both excitation conditions and fixed time delay ($\tau \approx 20$ ps). The case of resonant excitation is shown in Fig. 7(b) by solid triangle, and the other excitation condition in Fig. 7(a) by solid circles. For fitting the experimental results we used the following linear dependences:

$$\frac{\Delta f^{(res)}}{f} = C^{(res)} \cdot N^{(res)} = \frac{N_{exc}^{(res)}}{N_{s,exc}} + \frac{N_{pl}^{(res)}}{N_{s,pl}},$$
$$\frac{\Delta f^{(non)}}{f} = C^{(non)} \cdot N^{(non)} = \frac{N_{exc}^{(non)}}{N_{s,exc}} + \frac{N_{pl}^{(non)}}{N_{s,pl}}, \tag{8}$$
$$N^i = N_{exc}^{(i)} + N_{pl}^{(i)} \quad i = \text{res, non},$$

where $N_{exc}^{(i)}$, $N_{pl}^{(i)}$ and the densities of excitons and free electron-hole pairs for resonant ($i=$res) and nonresonant ($i=$non) excitation, respectively. $N_{s,exc}$ and $N_{s,pl}$ are the saturation densities of the OS, when only excitons and only free carriers influence the OS.[12] $C^{(i)}$ ($i=$res, non) are the experimentally measured proportionality coefficients. We obtained that $C^{(res)} = 1.2 \times 10^{-12}$ cm^2 and $C^{(non)} = 2 \times 10^{-12}$ cm^2.

In order to estimate the value of $N_{s,exc}$ and $N_{s,pl}$ we have to know what is the contribution of free carriers and excitons to the total quasiparticle density. The photoexcited carrier density is proportional to the integral of the product of the absorption coefficient of these quasi-particles and the intensity of the exciting laser. The spectra of the laser radiation and the theoretical absorption spectra of free and bound electron-hole pair states, which was obtained by the general Eliott formula,[19] are shown in Fig. 8. In this way we estimated that $N_{exc}^{(res)}/N_{pl}^{(res)} \approx 10.5$ and $N_{exc}^{(non)}/N_{pl}^{(non)} \approx 1.7$. Now, using the values of $C^{(res)}$ and $C^{(non)}$, the magnitudes of the OS saturation densities from Eq. (8) are found to be $N_{s,exc} = 1 \times 10^{12}$ cm^{-2} and $N_{s,pl} = 2.5 \times 10^{11}$ cm^{-2}. The absolute values of the OS saturation densities are one order of magnitude higher than predicted by the authors of the theoretical works Refs. 12 and 13. However, their ratio is in a very good agreement with the result of Ref. 12.

The density of free carriers becomes negligibly small after 130 ps (see Fig. 4), therefore the ROS has to be due to the only excitons. Let us estimate $N_{s,exc}$ from the dependence of $\Delta f/f$ on the carrier density for $\tau = 135$ ps. The exciton density for this time delay can be obtained from the following equations:

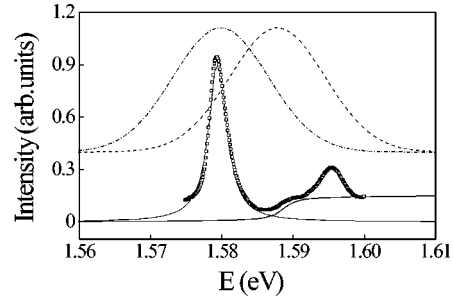

FIG. 8. Experimental (squares) and theoretical (solid lines) heavy-hole exciton and free carriers absorption spectra. The dash and dash-dot lines represent the spectra of the exciting laser radiation.

$$\frac{dN_{exc}}{dt} = -\frac{N_{exc}}{T_{1,exc}} + \frac{N_{pl}}{T_{1,pl}},$$
$$\frac{dN_{pl}}{dt} = -\frac{N_{pl}}{T_{pl}}, \tag{9}$$

where $T_{1,exc} = 410$ ps and $T_{1,pl} = 65$ ps are the lifetimes of excitons and free electron-hole pairs, respectively. The initial values of exciton and free carrier density $N_{pl}(0)$ and $N_{exc}(0)$ are obtained using their ratio (see above) and the value of the total pair density. For $\tau \to 0$ the density of both types of quasiparticles tend to zero. The solutions of Eq. (8) are presented on the inset to Fig. 4 by solid line. For comparison the temporal evolution of free electron-hole pair density (dash line) and the density of excitons in the absence of free carriers (dot line) are shown on the same insert.

Figure 7(a) shows the dependence of the relative ROS on the exciton density for $\tau = 135$ ps (open square). The best linear simulation takes place for proportionality coefficient equaled $C = 9.6 \times 10^{-13}$ cm^2 and indicates in Fig. 7(b) by the solid line. Inserting the value of C in Eq. (9) one obtains $N_{s,exc} = 1 \times 10^{12}$ cm^{-2} in agreement with the result above. Considering a 10% accuracy of determination of the real experimental carrier density, this confirms the validity of the model used.

V. ENERGY SHIFT OF THE EXCITON RESONANCE

The energy shift ΔE of the excitonic line as a function of the photoexcited carrier density (at $\tau \approx$ 20 ps) is displayed in Fig. 9 for the resonant excitation. As it was expected for the exciton-exciton interaction, a clear linear dependence is ob-

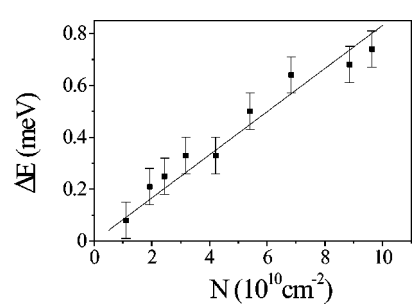

FIG. 9. The energy shift ΔE of excitonic line for the resonant excitation as a function of photoexcited carrier density N at $\tau \approx 20$ ps.

served. The proportionality coefficient between ΔE and N was found to be $C=0.083$, while for an ideal two-dimensional quantum well it should be $C=0.24$.[12] In our opinion, this discrepancy is due to the interaction between the excitons from neighboring quantum wells, as a result of which wells approach the three-dimensional limit and therefore the magnitude of the blueshift decreases. It follows from the linear dependence of ΔE on the quasiparticle density that the characteristic time of the exponential decrease of ΔE should equal the lifetime $T_{1,exc}$ of excitons (see Figs. 3 and 4). Therefore we have found that the exciton lifetime is $T_{1,exc}=(410\pm 14)$ ps and remains constant right up to densities of 10^{11} cm^{-2}. The fact that $T_{1,exc}$ is constant indicates that the range of the quasiparticle densities employed in our experiment is much smaller than the Mott density. This justifies using the low-density limit for describing the experimental results.

In the case of the nonresonant excitation the maximum blueshift is reached after all free electron-hole pairs have formed excitons. There are two reasons for the increasing the exciton blueshift. First, the blueshift is proportional to the exciton density (see above), so the higher the exciton density the bigger blueshift. Second, as it was experimentally shown in Ref. 10 free electron-hole pairs can lead to an additional redshift of exciton resonance. Unfortunately, we have not detected the dependence of the blueshift on the density of free carriers with sufficient precision in our experiment. The simultaneous influence of both free and bound quasiparticles on the energy shift of excitons needs a more detailed theoretical and more accurate experimental investigations.

VI. CONCLUSIONS

In conclusion, we investigate the temporal and density dependence of the exciton bleaching in GaAs/Al$_x$Ga$_{1-x}$As MQW. We distinguish the different contributions of exciton parameters to the exciton absorption using a moment analysis. We find that the ROS takes part in the exciton bleaching only during the first 110–130 ps and is mainly caused by the influence of free electron-hole pairs. We experimentally evaluate the OS saturation densities, which are found to be $N_{s,exc}=1\times 10^{12}$ cm^{-2}, when only excitons affect the OS, and $N_{s,pl}=2{,}5\times 10^{11}$ cm^{-2}, when only free electron-hole pairs are in the sample. We confirm the prediction of Schmitt-Rink *et al.* that cold free electron-hole pairs more effectively reduce the OS than other excitons do. From the temporal evolution of the magnitude of the OS we found the free electron-hole pair lifetime $T_{1,eh}=(65\pm 8)$ ps.

The broadening of the exciton resonance is observed during 500–600 ps and becomes the only source of the exciton bleaching after all free carriers disappeared from the interacting system having formed excitons. The lifetime of excitons is found to be $T_{1,exc}=(410\pm 14)$ ps. We have explained the measured blueshift of excitons by the exciton-exciton interaction and shown that the extra density of free charged carriers may lead to a compensating redshift.

ACKNOWLEDGMENTS

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