Localized indirect excitons in a short-period GaAs/AlAs superlattice

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We have studied the optical properties of a short-period superlattice composed of 20.4-Å GaAs and 14.7-Å AlAs layers. The superlattice behaves as an indirect-gap material. A slow and nonexponential decay of the luminescence can be interpreted as the emission from the Λ indirect excitons localized at the GaAs/AlAs interfaces. The temperature dependence of the exciton decay time can be explained in terms of a transition by phonon-assisted tunneling, followed by a nonradiative transition.

Recent advances in crystal growth techniques, such as the molecular-beam-epitaxy (MBE) method, have made it possible to produce semiconductor crystals consisting of alternating layers of two different semiconductors, i.e., superlattice structures. These superlattices are expected to form a new device and have been extensively studied, especially the GaAs/AlAs system. A number of experiments on GaAs/AlAs superlattices of large layer thickness have been carried out and analyzed successfully by means of the Kronig-Penney model.[1] There have been a few works on ultrathin layered GaAs/AlAs superlattices. However, considerable confusion exists in the interpretation of the electronic structure of these superlattices, especially of the short-period superlattices with nearly equal GaAs and AlAs layer thickness and with periods ranging from ~ 10 to ~ 60 Å. Some groups have claimed that these short-period superlattices behave as indirect-gap materials,[2,3] while others have concluded that these materials are direct gap.[4–7] In order to clarify the electronic structure of short-period superlattices, we have studied the optical properties of a superlattice composed of 20.4-Å GaAs and 14.7-Å AlAs layers. Special attention is given to the dynamics of photoexcited carriers. The experimental results indicate that our sample behaves as an indirect-gap material and that the emission near the band edge is due to localized indirect excitons.

The GaAs/AlAs sample used in this study consisted of 250 periods of 20.4-Å GaAs/14.7-Å AlAs grown by MBE at 550 °C on a (100) semi-insulating GaAs substrate. The Al content in the sample was 42%, which is near the direct-indirect crossover value of $\sim 40\%$ in the $Al_xGa_{1-x}As$ alloy system.[8–10] In order to make absorption measurements, the GaAs substrate was removed by preferential etching over a region of 1×1 mm^2, leaving only the MBE-grown film. The cw photoluminescence and excitation measurements were made by a DCM dye laser pumped by an argon-ion laser. For time-resolved spectra, the sample was excited by a cavity-dumped mode-locked cw dye (DCM) laser synchronously pumped by a mode-locked argon-ion laser. The transmitted light and luminescence were monitored by a double monochromator equipped with a cooled photomultiplier. Lifetime measurements were made by the time-correlated single-photon counting technique. The sample was immersed in liquid helium. At elevated temperatures, it was exposed to an atmosphere of helium gas.

The absorption spectrum at 4.2 K is presented in Fig. 1. The absorption coefficient $\alpha(\hbar\omega)$ was obtained from the optical density by taking into account multiple reflections. The spectrum shows the characteristic behavior of indirect-gap materials: No feature due to discrete-state exciton absorption is observed and an absorption tail is seen at low energies. From the kink in the curve, the direct gap E_g is measured as ~ 1.975 eV. We estimated the indirect gap E_{gi} roughly as ~ 1.88 eV through the relation $\alpha(\hbar\omega)\propto(E_{gi}-\hbar\omega)^2$.[2] The luminescence and excitation spectra at 4.2 K are also shown in Fig. 1. A relatively sharp line, labeled by I_{ex}, is observed at ~ 1.873 eV and dominates the near-gap emission. On the basis of the spectral position and time dependence shown later, we believe that the I_{ex} line comes from the decay of localized indirect excitons. Although weak and broadband emissions are observed at lower energies, here we confine our attention to the I_{ex} line. The excitation spectrum monitored at the I_{ex} line shows a well-defined peak at the direct-gap energy. This also indicates that the sample is indirect gap. Further, this conclusion is borne out by the temporal behavior of the I_{ex} emission described below. These results support the conclusions of Refs. 2 and 3, while they are in conflict with those made in Refs. 4–7.

Figure 2 shows the time evolution of the I_{ex} emission at 4.2 K. The emission decays slowly and nonexponentially. This temporal behavior can be interpreted well in terms of the emission from indirect excitons localized by disorder

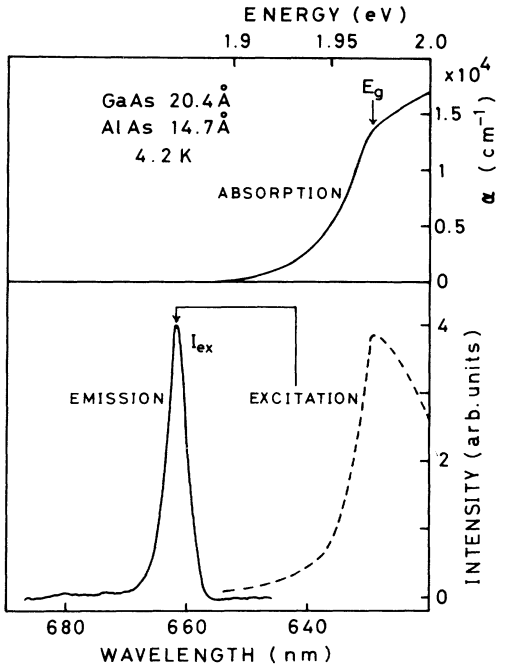

FIG. 1. Absorption, cw luminescence, and excitation spectra for the short-period GaAs/AlAs superlattice. The direct gap is denoted by E_g. The excitation spectrum was monitored at the I_{ex} line.

in the sample. In the presence of potential fluctuations, no-phonon radiative transitions of indirect excitons are allowed because these fluctuations break the **k**-selection rule. Reflecting the random nature of the scattering potential, the radiative decay rate is not well defined, but has a probability distribution. The no-phonon transitions of indirect excitons in such a case have been studied by

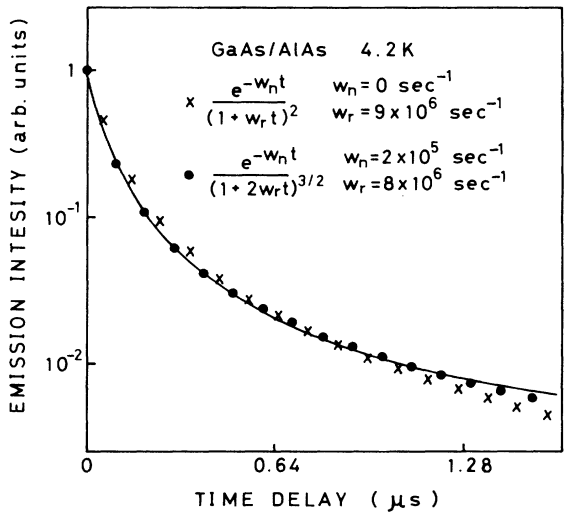

FIG. 2. Temporal evolution of the I_{ex} emission at 4.2 K. The solid line shows the experimental curve. Crosses represent Eq. (1) with $w_r=9\times10^6$ sec^{-1} and $w_n=0$ sec^{-1}. Circles represent Eq. (2) with $w_r=8\times10^6$ sec^{-1} and $w_n=2\times10^5$ sec^{-1}.

Klein *et al.*[11] According to their theory,[11,12] the time response of the emission from localized indirect excitons is given as

$$I(t)\propto e^{-w_n t}/(1+w_r t)^2 , \tag{1}$$

for indirect excitons not at the zone boundary, or

$$I(t)\propto e^{-w_n t}/(1+2w_r t)^{3/2} , \tag{2}$$

for indirect excitons at the zone boundary, where w_r is the average radiative decay rate due to random potential scattering, and w_n is the total decay rate due to all other processes. The experimental data fit the time dependence predicted from this model. Theoretical curves derived from Eqs. (1) and (2) are also shown in Fig. 2. The best fits were obtained for $w_r=9\times10^6$ sec^{-1} and $w_n=0$ sec^{-1} in the case of Eq. (1), and for $w_r=8\times10^6$ sec^{-1} and $w_n=0.2\times10^6$ sec^{-1} in the case of Eq. (2).

A better fit is obtained to Eq. (2). From this result, however, one cannot conclude immediately that the I_{ex} emission is due to the zone-boundary excitons, because the theory of Klein *et al.* was developed for indirect excitons in an alloy system where scatterers are uniformly distributed. In superlattices potential fluctuations are considered to come from the inhomogeneity of the interface and scatterers to be distributed in the interface planes. In such a case, the summation in Eq. (6) of Ref. 11 must be performed over the position vectors **L** of the scatterers in the interfaces spanned by the exciton. The exciton in the present system encompasses only a few interface planes. The phase factor $e^{i\mathbf{k}\cdot\mathbf{L}}$ in this equation then does not have many different values for indirect excitons at wave vector **k** normal to the interface (i.e., on the Λ line). The theory should thus be modified for these excitons since the assumption of the uniform distribution of phase, used in Ref. 11, is no longer a good approximation. By modifying the model of Klein *et al.*, it can be deduced that the decay of indirect excitons on the Λ line follows Eq. (2) rather than Eq. (1).[13] In the superlattices, therefore, excitons both at the zone boundary and on the Λ line should decay as predicted by Eq. (2).

To determine the origin of the I_{ex} emission, it is necessary to know which points of the Brillouin zone are likely candidates for positions of the band extrema. Since it is well established that the topmost of the valence bands lies at Γ, we will search for the location of the conduction-band minimum. Following Ref. 3, we take the $Al_{0.42}Ga_{0.58}As$ alloy bands as unperturbed states and treat the superlattice potential as a perturbation. In the parent alloy the minimum of the conduction band occurs at the X point.[10] In the superlattices, the $X(001)$ point is folded back to the Γ point due to the superlattice potential, and through the coupling with other Γ states, it is lower than the unmixed $X(100)$ and $X(010)$ points. This point is then expected to become the lowest state of the conduction band. Actually, in the alloy the conduction-band minimum is not at X, but displaced from it by the "camel's-back" effect. This camel's-back structure would be reflected in the band structure of the superlattice. The conduction-band minimum is then expected to lie on the

Λ line.[3] On the basis of this consideration and the time dependence obeying Eq. (2), we think that the I_{ex} emission is attributable to indirect excitons on the Λ line, i.e., the camel's-back excitons near the Γ point.

The one-photon transition to the lowest Γ state of the conduction band is considered to be weakly allowed, since this Γ state is derived from the X point of the parent alloy and is only slightly modified by the superlattice potential.[3] Further, it can be shown that the decay of this "pseudodirect" exciton follows Eq. (2). Thus one may think that there is no camel's-back structure near the Γ point and the I_{ex} emission is due to the pseudodirect exciton consisting of a Γ hole and an electron at the Γ point which has the folded-X character of the $Al_{0.42}Ga_{0.58}As$ alloy. However, this is not our case. At the Γ point, the superlattice potential makes a more appreciable contribution to the radiative decay rate than the interface disorder does, and this nonstochastic contribution should be included in the decay rate w_n. This assignment then is not consistent with the result that w_n is much smaller than the average radiative decay rate w_r, which is a measure of the magnitude of interface disorder.

There is another possibility that the I_{ex} emission is due to excitons bound to neutral donors or acceptors. However, this assignment is very unlikely, because, by contribution to nonradiative Auger recombination, the bound excitons can decay exponentially with lifetimes less than 20 nsec even in indirect-gap materials.[12,14] Further evidence that the emission is not from excitons bound to impurities is provided by the temperature dependence of the emission. Since the bound exciton has a binding energy of a few meV, thermal dissociation of the bound exciton to the free indirect exciton should occur above ~20–30 K. The free-exciton emission should then dominate the bound-exciton emission at high temperatures. However, the experimental result shows that I_{ex} emission is predominant above 60 K. This possibility therefore is ruled out.

The temperature dependence of the decay of the I_{ex} line is shown in Fig. 3. There is a concomitant decrease in the emission intensity. At low temperatures, the emission shows a slow and nonexponential decay. With the increase of temperature, the decay becomes faster and approaches an exponential form. A changeover to exponential decay occurs above ~11 K. The temperature dependence of the decay time is shown in Fig. 4. Here we have defined the decay time as the slope of the exponential decay curve above ~11 K. In the figure we also show the temperature dependence of the steady-state luminescence intensity. The parallel decreases in the decay time and the emission intensity show that nonradiative processes are dominant above ~11 K. The decay time (or luminescence intensity) does not exhibit Arrhenius-type behavior. A reasonable fit is obtained to an expression of the form $\tau \propto e^{-T/T_0}$, with $T_0 \sim 10$ K. This temperature dependence is familiar in amorphous materials, such as As_2S_3 (Ref. 15) and a-Si:H (Ref. 16), and believed to be a characteristic of localized systems with a "mobility edge."[17] Further, this dependence has recently been observed in several mixed crystals and is considered to be associated with the mobility edge.[18] We thus think that the e^{-T/T_0} dependence in our sample indicates the existence of a mobility edge and excitons localized in the Anderson sense.[19] In this picture, the temperature dependence is explained as follows. At low temperatures only the processes which lower the energy are possible and the localization is fast and irreversible. The localized excitons then dominate the emission. They respond to the potential fluctuations and decay nonexponentially. The temperature dependence can be understood as a result of thermal delocalization of excitons. As the temperature is raised, the exciton mobility increases via phonon-assisted tunneling and the probability of reaching nonradiative recombination centers increases. As nonradiative processes are dominant, the decay becomes exponential and the radiative efficiency drops off.

Finally we make a short mention of the latest work on the same subject performed by Finkman *et al.*[20] From the temporal response of photoluminescence, they concluded that the lowest excited state of the short-period superlattices is an X-point (zone-boundary) exciton. On the

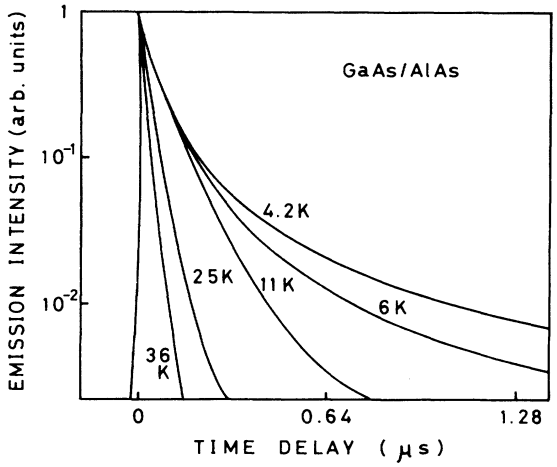

FIG. 3. Luminescence decay curves of the I_{ex} line for different temperatures.

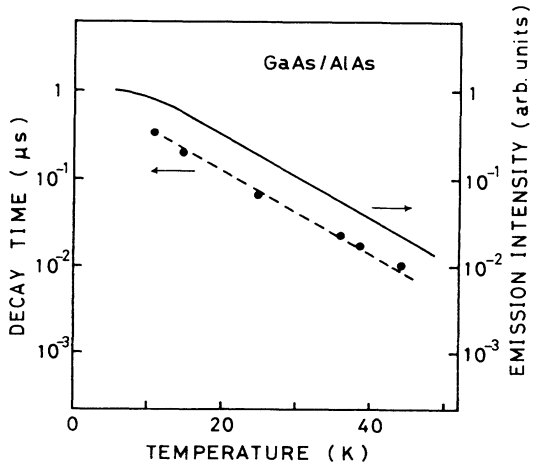

FIG. 4. Temperature dependence of the decay time and of the cw luminescence intensity of the I_{ex} line.

other hand, we suggest that it is an indirect exciton on the Λ line. Since the existing theories predict that the conduction-band minimum is at the Γ point[4–6] or on the Λ line,[3] we think our assignment is more reasonable. According to Ref. 20, however, there seems to exist a theory predicting that the unmixed X minima are the lowest. A full understanding of this state must involve a more complete investigation including, for example, the effect of uniaxial stress.

In conclusion, we have studied the optical properties of the short-period superlattice consisting of 20.4-Å GaAs and 14.7-Å AlAs layers. The superlattice behaves as an indirect-gap material. The emission at 4.2 K shows a slow and nonexponential decay. This behavior can be interpreted by the decay of indirect excitons on the Λ line made partially allowed by disorder at the GaAs/AlAs interfaces. The decay time of the excitons exhibits a e^{-T/T_0} temperature dependence, which is explained in terms of phonon-assisted tunneling of excitons to nonradiative centers.

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