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Photoluminescence (PL) dynamics of localized excitons in a strongly coupled 40 Å/40 Å $GaAs/Al_{0.2}Ga_{0.8}/As$ superlattice under an electric field along the growth direction is reported. At zero field, a delayed PL formation due to the relaxation of dark excitons into radiative states is observed. With increasing field, both the PL decay time and the PL amplitude are strongly reduced. A rate equation analysis of the measured PL transients gives evidence that the field-induced dissociation of nonradiative excitons is important. © *1995 American Institute of Physics.*

The dynamical behavior of carriers in superlattices in an electric field along the growth direction has been extensively studied due to the potential of these artificial structures for future electronic and optoelectronic device applications. Transit times are important for the saturation properties of modulators and detectors.[1] The electroabsorption properties of Stark ladder superlattices[2] have led to novel concepts for electro-optical modulators[3] and switches.[4] In the ultrafast regime, coherent oscillations of resonantly excited excitons have been observed.[5,6]

The photoluminescence (PL) of these structures is rapidly quenched at higher electric fields and therefore difficult to access. In addition, localized excitons arising from barrier- and well width fluctuations can give rise to additional luminescence lines[7] which are not associated with Stark localization. Further complications occur at small temperatures due to the delayed PL formation after the excitation, arising from a nonthermalized distribution of dark excitons.[8] In the present study, we report on the field dependence of the low-temperature photoluminescence dynamics in a Stark ladder superlattice. Our experiments and numerical simulations indicate that the PL quenching is mostly due to the dissociation of dark excitons.

The sample under study is a nominally undoped 70 period superlattice consisting of 40 Å GaAs wells and 40 Å $Al_{0.2}Ga_{0.8}As$ barriers. The whole structure is sandwiched between doped $Al_{0.3}Ga_{0.3}As$ layers, 1 μm of nominally 1×10^{18} cm^{-3} n-type (Si) on the substrate side and 0.2 μm of 5×10^{18} cm^{-3} p-type (Be) on the top side. The measurements are performed on 150 μm^2 contact windows of 200×300 μm^2 mesa diodes processed by photolithography and wet-chemical etching. The built-in voltage is estimated to be about 1.2 V, which is based on the observed invariance of the PL decay behavior at >1.2 V forward bias.

A Kronig–Penney calculation yields a conduction miniband width of 35 meV. Photocurrent spectra (not presented here) show spatially indirect absorption peaks revealing clear evidence of Stark localization, with a coherent excitonic coupling[9] across at least five SL periods at intermediate fields.

Time-resolved photoluminescence measurements are performed using a mode locked Ti:sapphire laser (≈2 ps pulse width, 1.70 eV excitation energy) for excitation and a synchroscan streak camera equipped with a 34 cm monochromator for temporally and spectrally resolved detection. The time resolution obtained in these experiments is about 15 ps (measured full a width at half maximum of the laser pulse).

Figure 1 shows the intensity dependence of the time-integrated PL spectra under flatband condition at photon densities ranging from 5×10^{8} to 5×10^{10} cm^{-2}. This corresponds to excitation densities from about 2.5×10^{6} to 2.5×10^{8} cm^{-2} excitons per SL period (assuming an absorption of 0.5% per SL period). The PL spectra consist of

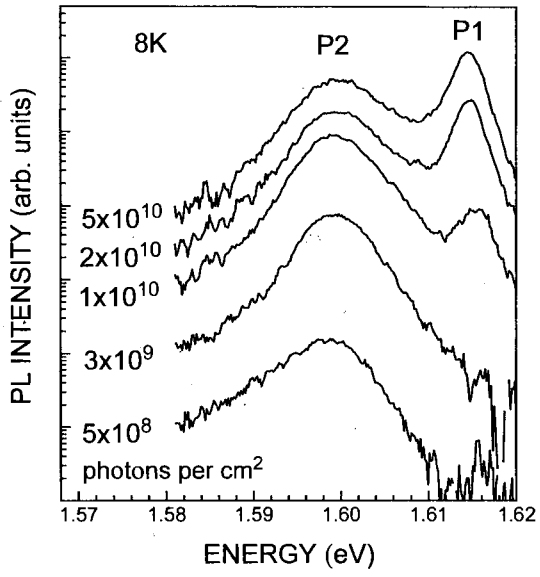

FIG. 1. Time-integrated PL spectra at different excitation intensities at 8 K. Indicated are the incident *photon densities* of the excitation pulses.

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two lines $P1$ and $P2$ centered at 1.615 and 1.599 eV, respectively. The $P1$ line is associated with the fundamental heavy hole exciton of the SL since it corresponds essentially to the fundamental absorption edge observed in the low-field photocurrent spectra. The $P2$ line is attributed to excitons which are localized perpendicularly due to well width fluctuations[7] or to impurity-related excitons. At small densities ($<10^{10}$ photons per cm^2), the $P2$ emission shows a quadratic dependence on the excitation density, indicating the presence of a nonradiative recombination channel. This nonradiative recombination can partially arise from a finite residual electric field in the SL region. Above 1×10^{10} cm^{-2},we observe a weaker $P2$ intensity dependence, which is either due to the screening of the residual field or to saturation effects. Simultaneously, the $P1$ line appears and becomes even stronger than the $P2$ line, suggesting that saturation is actually important. It is characteristic for localized excitons that they can easily be saturated.[10] In a systematic study using similar SL structures,[7] the $P2$ transition has been attributed to perpendicular localization arising from barrier- and well width fluctuations, rather than transitions related to residual impurities.

In order to avoid additional complications associated with time-dependent field-screening effects and the observed two-line behavior, the time dependent PL measurements were performed at a small excitation density of 3×10^9 photons per cm^2. Figure 2(a) shows a series of PL transients measured at different forward bias. At 1.2 V, the PL maximum is reached after about 150 ps after excitation, indicating a rather large PL formation time. We observe several decay components, suggesting that there exist several (at least two) sublevels of the $P1$ band with different decay times. Indeed, the spectral PL position shifts by 2 meV to lower energies within the initial 500 ps, although a line splitting is not observed. With increasing electric field, not only are the PL decays faster, but the initial PL amplitude also decreases strongly. The latter observation indicates that both the dark and the radiative excitons are dissociated by the electric field. After dissociation, the carriers can in principle form new excitons with other electrons and holes contained in the SL. This exciton recycling becomes very inefficient at higher fields and is therefore neglected in the present study.[11]

In order to simulate the observed behavior numerically, we have to consider a minimum of three levels. We assume that there exists a nonradiative level (0) which is populated quasi-instantaneously. This level represents excitons which cannot decay radiatively (due to, e.g., their large momentum). The exciton formation time is neglected here. We also have to take into account two further levels (1 and 2) which contribute to the PL by radiative recombination.

A linear rate equation within this three-level scheme always generates at most three relevant time constants. Although there are many possible transition schemes giving rise to one formation and two decay times, only a few of them describe realistically the variation of the strength of the individual components on changing the dissociation time constant. Reasonable results are obtained with the scheme indicated by the inset of Fig. 2(b). The population of level 0 decays by relaxation into level 1 (time constant t_1) and by nonradiative dissociation with decay time t_2; t_3 may be interpreted as a radiative decay time and t_4 describes the relaxation into level 2 by diffusion or related processes. For simplicity, we also use the previous time constant t_3 for the decay of level 2 and assume that the PL signal is proportional to the populations n_1+n_2.

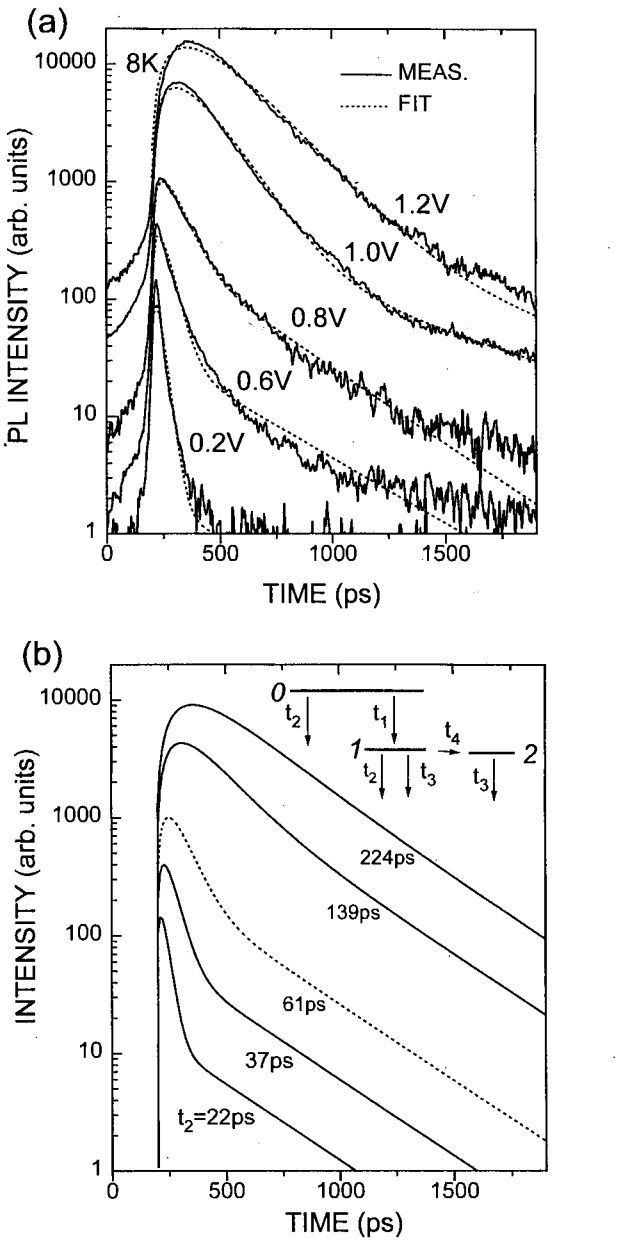

FIG. 2. (a) Transient PL of the $P2$ band at 3×10^9 photons per cm^2, 8 K and different forward bias (full lines) and numerical fits (dashed) using one formation and two decay times. (b) Numerical simulation of PL transients using the rate equation shown schematically in the inset. The dashed line is the theoretical curve from (a) for 0.8 V (t_1=833 ps, t_2=61 ps, t_3=653 ps, t_4=335 ps). The solid lines are obtained by changing only t_2.

The simulations in Fig. 2(a) have been obtained with a fixed reference time and prefactor under variation of t_1-t_4. We note that there is a certain tradeoff between t_2 and the initial PL amplitude. In Fig. 2(b), we have used the fitting parameters obtained for 0.8 V and varied only t_2, resulting in a reasonable description of the field dependent PL intensities and lifetimes over the whole voltage range. The correct

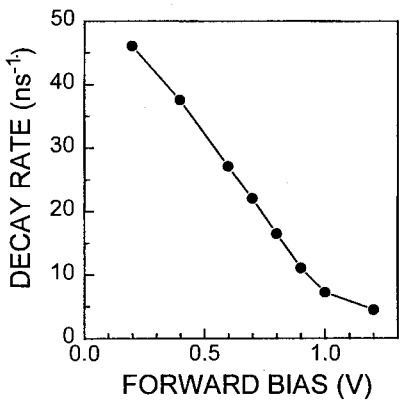

FIG. 3. Decay rate $1/t_2$ due to the field dissociation process of the photoexcited excitons vs applied forward bias.

description of the field dependent PL maximum value is a direct consequence of the nonradiative dissociation of the excitons in level 0 and is also reproduced when omitting level 2. The slow decay constant associated with level 2 apparently depends only weakly on the applied bias. We point out that a strongly different behavior is observed when the dissociation channel (t_2) of level 0 is omitted.

We have also tried a few different three-level schemes in order to optimize the simulations. In each case we obtained approximately (within 10%–15%) the same values of t_2. The observed dissociation rates are therefore summarized in Fig. 3. Except for large forward bias where the PL starts to decay mainly radiatively, we observe an almost linear relationship between these decay rates and the electric field. In the reverse bias regime, the PL becomes quenched very efficiently. It therefore, becomes difficult to distinguish the observed signal from residual stray light of the exciting laser.

In conclusion, we have studied the PL dynamics from localized excitons in a strongly coupled superlattice under an electric field, at small excitation density and at low temperature. Field dissociation of dark excitons contributes significantly to the observed rapid PL quenching.

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[11] Exciton recycling can be described phenomenologically by a transition rate from level 1 into level 0.