Local recombination mechanisms in type-II GaAs/AlAs superlattices: The role of temperature-dependent transport processes

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We study type-II GaAs/AlAs short-period superlattices by spatially resolved photoluminescence spectroscopy (μ-PL). We observe two groups of spectrally narrow luminescence lines. The origin of one group is localized type-II states while the other group is related to deeply localized type-I states. The latter result from a local crossover of the band alignment from type-II to type-I induced by a variation of the well width of 2 bilayers. The use of nonresonant excitation and the variation of the lattice temperature give a direct access to the transport processes preceding the recombination. We can distinguish between two cases. Among the localized type-II states, phonon-assisted relaxation of the excitons is identified by a redshift of the envelope of the μ-PL lines. For the deeply localized type-I states, the luminescence intensity increases exponentially with rising temperature. We model this behavior by electron tunneling into the luminescing states.
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I. INTRODUCTION

The spectra of spatially integrated photoluminescence (PL) of type-II GaAs/AlAs short-period superlattices (SPS) are well understood (see, e.g., Refs. 1–3 and references therein). The underlying radiative processes are due to Γ-X mixing (pseudodirect recombination) for the zero-phonon recombination. For the phonon replica, the photon and one or more phonons are emitted simultaneously. The phonon is necessary to pick up the zone-boundary **k** vector of the initial excitonic state during the recombination. For SPS with AlAs thicknesses comparable to those used in our study, there are two phonon sidebands. One is due to emission of AlAs transverse optical (TO) and longitudinal acoustic (LA) phonons that can be observed separately in samples with very small inhomogeneous broadening.[2,3] The other one is caused by emission of AlAs longitudinal optical (LO) phonons.

Spatially resolved PL on a length scale of $1\,\mu$m (μ-PL) reveals local optical properties that can differ significantly from the above-described averaged ones. The area of the detected spot is typically decreased by four or more orders of magnitude with respect to the spatially integrated method. Averaging over spatial inhomogenities of the sample is thereby significantly reduced. But even within the area of $1\ \mu\text{m}^2$, one finds a large number of radiative states contributing to the μ-PL signal. To gain access to individual recombination centers, spectral resolution is added to the technique. Since different local environments give rise to different transition energies, local properties of the sample even below the diffraction limit can be distinguished. They show up in μ-PL as spectrally narrow lines with a linewidth determined only by the homogenous broadening. μ-PL has been applied during the past years to study, e.g., exciton localization in width fluctuations of quantum wells,[4,5] which have been addressed as ''naturally formed quantum dots''. Even single natural quantum dots can be investigated,[6–9] where biexcitons,[6] magnetic properties of excitons and biexcitons,[7] and homogenous linewidths and the phonon bottleneck[8] have been some of the topics of interest. The latter methods use typically resonant excitation.

In order to study relaxation processes among the localized states, we investigate the temperature dependence of spectrally resolved μ-PL under off-resonant excitation. The investigated type-II GaAs/AlAs SPS are chosen such that they are close to the crossover layer thickness combination to type-I band alignment. From the temperature dependence of the μ-PL, we are able to distinguish between two distinctly different sets of localized states in these samples. For one, we observe type-II excitons that are localized at potential fluctuations due to interface roughness. Energy relaxation between these localized states is possible due to phonon assisted excitonic transport. On the other hand, we also find deeply localized type-I states. They stem from a local crossover of the band alignment. These efficient recombination centers are populated via phonon-assisted tunneling of the *electron*. They are isolated in the sense that further *excitonic* transport is not possible during the radiative lifetime.

The paper is organized as follows: we first give some details about the samples and the measurement setup (Sec. II). In Sec. III, we describe the observations of the temperature-dependent μ-PL, which will allow us to separate the spectrally narrow PL lines into two groups. Sections IV and V clarify the origin of these groups as localized type-II and isolated type-I states. In Sec. V we further identify an electron-transfer mechanism into the type-I states. We summarize the most important results in Sec. VI.

II. SAMPLES AND EXPERIMENTAL DETAILS

We investigated a series of four GaAs/AlAs superlattices grown by molecular beam epitaxy. The labels and the properties of the samples are summarized in Table I. Details about the growth and the interface properties of the samples are described in Ref. 10. The AlAs-on-GaAs interface is intermixed while the GaAs-on-AlAs one is abrupt, which is indicated in the upper left of Fig. 3. The quality of the samples is extremely high: we observed confined optical

TABLE I. Properties and labels for the samples investigated in this article. For samples *A* and *B*, a variation of the well width by 2 bilayers results in a change of the band alignment from type-II to type-I. This is confirmed by the band alignment of sample *D*. For sample *C*, such a crossover is not possible.

Label	GaAs/AlAs thicknesses in bilayers (=2.83 Å)	Number of periods	Band alignment
A	8/8	140	type-II
B	11/10	140	type-II
C	9/14	180	type-II
D	10/5	50	type-I

phonons up to the 5th order in those samples.[10] The long luminescence decay times at low temperatures of 1.6 μs (sample *A*) and 5.5 μs (sample *C*) confirm the relatively small number of nonradiative centers.

Samples *A*, *B*, and *C* all have a type-II band alignment. This means that the conduction band minimum is in the AlAs layers at the *X* point while the valence-band maximum is in the GaAs slabs at the Γ point. As a consequence, electron and hole have only a small wave-function overlap, and the exciton ground state has a nonvanishing **k** vector. Mainly the latter feature leads to the large luminescence decay times.[11] A combination of PL and PL excitation reveals that the lowest type-I transition is about 100 meV higher in energy than the type-II bandgap.

There is a distinct difference in the properties of samples *A* and *B* with respect to sample *C*. In samples *A* and *B*, a fluctuation of the GaAs layer thickness by 2 bilayers results in a local change of the band alignment to type-I. The conduction- and valence-band extrema are then both in the GaAs at the Γ point. This assumption is confirmed by a Kronig-Penney calculation and is supported by the properties of sample *D*. The layer thicknesses of sample *D* are such that its band alignment is of type I. It is suitable for a direct comparison with the type-I recombination caused by thickness fluctuations in sample *A*. The band alignment of sample *C* cannot change through a similar thickness fluctuation.

All the spectra shown in this article have been measured on sample *A*. We made similar observations on sample *B*. The reference samples *C* and *D* allow us to identify local type-I states in the μ-PL (see Sec. V).

One of the advantages of μ-PL over spatially integrated PL comes from the fact that local environments are studied. Already one sample gives access to a valuable number of different ''samples in the sample,'' i.e., different detection spots. The needed statistics for reliable statements can thus be obtained easily.

In order to design a setup for spatially resolved spectroscopy, there are two distinct starting points. On the one hand, one can use a *local excitation* and a global detection (e.g., cathodoluminescence or most optical near-field studies). The spatial resolution of the technique is then not given by the diameter of the excitation spot but rather by the diffusion length of the excitons or carriers. On the other hand, for a global excitation combined with a *local detection* the spatial resolution is independent of diffusion lengths because always the same sample volume is probed. The spectral shape and intensity of the μ-PL however are affected by transport processes preceeding the recombination. Using the latter method, we can demonstrate the importance of phonon-assisted tunneling processes in the population dynamics of the investigated radiative states.

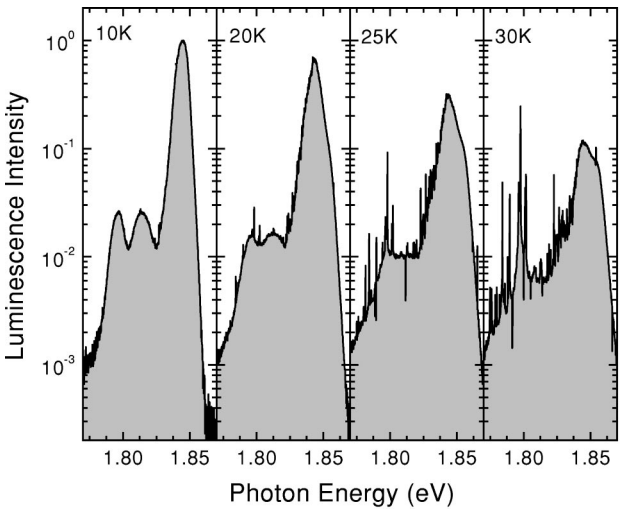

FIG. 1. μ-PL spectra of sample *A* at the same detection spot for different temperatures. All spectra are normalized with respect to the maximum of the 10 K spectrum to reveal the temperature evolution of the luminescence intensity.

Our μ-PL setup consists of a He flow cryostat with only one thin (approximately 1-mm thick) window. The excited spot on he sample is magnified through a microscope lens by a factor of 20 and imaged onto an exchangeable pinhole. The pinhole defines the actual spatial resolution in the detection with an optimum resolution given by the diffraction limit. The pinhole is imaged onto the entrance slit of a 0.46-m focal length single grating spectrometer with a LN_2-cooled charge-coupled device (CCD) attached. The energy resolution is approximately 100 μeV.

One has to keep in mind that the cryostat window has to be regarded as a ''cover glass.'' To obtain a sharp image of the sample, the microscope lens must be able to correct for beam aberrations by the window. Our lens can be adjusted to window thicknesses between 0 and 1.5 mm. In the measurements reported here, we used a pinhole with 60 μm diameter. The resulting detection spot size of 3 μm in diameter is about three times the diffraction limit for the regarded wavelengths. This spot size ensures a good lines-to-background ratio (Fig. 1) for reasonably short integration times.

In these measurements, we used off-resonant excitation with an unfocused red HeNe laser. The excitation intensity was a few W cm^{-2}. The laser beam struck the sample under oblique incidence to reduce stray light of, e.g., the laser plasma lines. We carefully checked that none of the lines shown in Figs. 1 and 2 are due to such artifacts.

III. TEMPERATURE DEPENDENT μ-PL

We performed temperature-dependent μ-PL measurements in the low-temperature range from below 10 to 40 K. Typical results between 10 and 30 K are shown in Fig. 1. It is important to note that these spectra are recorded at exactly the same spot on the sample. The excellent reproduction of the series of narrow lines at energies around 1.795 eV gives

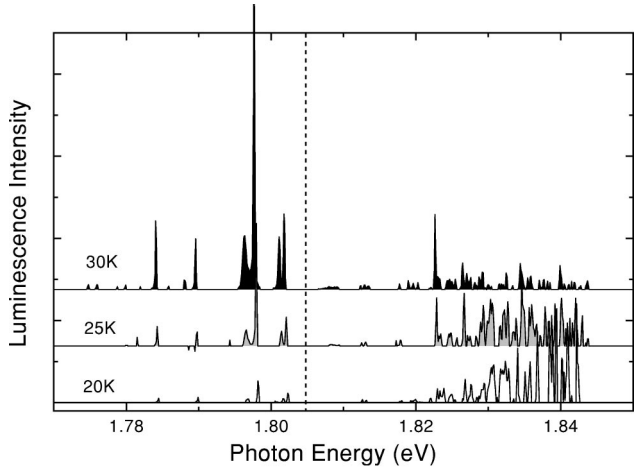

FIG. 2. The narrow luminescence lines extracted from Fig. 1 after subtraction of the smooth background. The vertical line indicates the separation of the spectra into two groups. Its position is in the spectral range of the AlAs LA/TO phonon replica.

evidence for the good spatial stability of the measurement setup.

At 10 K and below, the spectrum is almost identical to spatially integrated measurements, i.e., we observe a smooth line shape. It is composed of the zero-phonon line and the two phonon replica (Fig. 1). The specifics of the spatial resolution can be seen as soon as the temperature is raised slightly (Fig. 1, 20...30 K): as a new feature, spectrally narrow lines emerge from the spectrum. Their spectral width of $\hbar\Gamma \leqslant 100\ \mu$eV is given by the spectral resolution of the spectrometer. For this experiment, the spectral resolution is sufficient to separate the narrow PL lines. We have determined line widths of 30 μeV for the same lines in combination with a 0.75-m focal length double grating spectrometer.

We observe that some lines are situated on the low-energy side of the zero-phonon emission. In the spectral range of the AlAs LA/TO phonon replica (around 1.815 eV), the intensity of such lines is very low. On the AlAs LO phonon replica

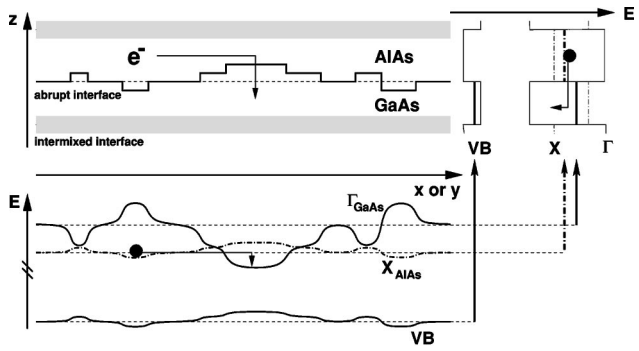

FIG. 3. A local crossover from type-II to type-I can occur due to a deviation from the ideal quantum well geometry. A locally thicker GaAs well combined with a locally thinner AlAs barrier forms a type-I center in which both, hole *and electron*, can be localized in the GaAs well. At elevated temperatures, the *X*-point electron in the AlAs can tunnel into the type-I center. The loss of energy and **k** vector during this transfer is achieved by AlAs LO phonon emission.

(around 1.795 eV), the lines grow absolutely with rising temperature and can even become stronger in peak intensity than the zero-phonon emission. This means that the latter lines cannot be due to phonon replica. It is an indication for a new physical mechanism. In addition, we note that the global luminescence intensity drops with rising temperature.

Figure 2 shows the sharp lines extracted from the spectra shown in Fig. 1 by subtracting the remaining smooth ''background'' line shape. The background still resembles the spatially integrated measurements very much. A first look at this figure suggests to divide the sharp lines into two different groups indicated here by the vertical line. Qualitatively, the spectral weight shifts red for the energetically higher lines with rising temperature, and their summed intensity drops. This is different for the group of lines in the spectral range of the AlAs LO phonon replica (left of the vertical line): its spectral weight does not change significantly, but even more important is that the individual intensities *increase* with rising temperature.

We will now carefully investigate both groups separately. We will show that the lines on the low-energy side of the zero-phonon line stem from localized type-II states. Excitonic transport between the states is possible and results in the red shift of the spectral weight. The energetically lower lines originate from localized type-I states. Those type-I states result from a local fluctuation of the quantum well thickness. Once these states are populated, no further excitonic transport is possible during the exciton lifetime. From the temperature dependence of the lines' intensity we will deduce the population mechanism to be electron tunneling. First, we discuss the type-II states since they can be expected in this kind of SPS system.

IV. LOCALIZED TYPE-II STATES

We want to study phonon-assisted relaxation within the localized states on the low-energy side of the zero-phonon line. In spatially integrated PL, this phenomenon manifests itself as a redshift of the PL maximum with rising temperature (see, e.g., Ref. 12). Compared to the shift of the band gap, this redshift is by far larger.

For the lines in Fig. 2, we have to define a quantity that corresponds to the properties of the PL maximum. We have chosen here the spectral weight $SW(a,b)$ which is defined by

$$SW(a,b) := \frac{\int_a^b \hbar\omega\, I_{\text{lines}}\, d(\hbar\omega)}{\int_a^b I_{\text{lines}}\, d(\hbar\omega)}, \tag{1}$$

where I means the intensity of the indexed quantity, i.e., I_{lines} is the intensity of the sharp lines. $SW(1.805\ \text{eV},\infty)$ shifts red by 5 meV between 20 and 30 K, which is clearly more than the expected shift of the band gap.[13] This behavior reflects that at higher temperatures excitons transfer more efficiently to localized states that are lower in energy.

The nature of the localized states in this energy range can be deduced from the temperature evolution of the PL intensity. It was already noted that both, the global luminescence intensity and the integrated intensity of the lines regarded

here, drop with rising temperature. It is thus reasonable to compare the two temperature dependencies. The important quantity is the fraction

$$F_{upper} := \frac{\int_{1.805\,eV}^{\infty} I_{\text{lines}}\, d(\hbar\omega)}{\int_{0}^{\infty} I_{\text{background}}\, d(\hbar\omega)}, \tag{2}$$

which is found to be almost constant. This result suggests that the origin for both, the ''background'' of the smooth zero-phonon line/phonon replica and the energetically higher lines, is the same. The narrow lines stem from localized type-II states.

The fact that the lines under investigation occur predominantly on the low-energy side of the zero-phonon band raises one question: why do these lines only occur on the low-energy side of the emission band and not, e.g., also on the high-energy side? We can compare our result to observations in type-I GaAs/AlAs single quantum wells[4] and theoretical considerations[14] in the same system. In those cases, also most of the lines occur on the low energy tail of the luminescence spectrum. The high-energy side and the maximum are more or less smooth. The underlying reason according to Ref. 14 is that at the luminescence maximum, there is a large number of states with rather small matrix elements. This results in a smooth line shape. At energies on the low-energy side of the PL band, the number of states is smaller. They exhibit large matrix elements, though. Together, this results in relatively intense separable lines. Thus, our result compares well with previous observations in the same material system, but here with a different band alignment.

To summarize this section, the narrow lines on the low-energy tail of the zero-phonon line stem from localized type-II excitons. Within these localized states, phonon-assisted relaxation of localized excitons is observed.

V. ISOLATED TYPE-I STATES

Before one can try to understand the temperature dependence of the lines around 1.795 eV, one needs to know the origin of those lines. We already pointed out that although these lines are energetically in the spectral range of a phonon replica, they cannot be due to phonon-assisted recombination. We will first describe a model for deeply localized states which explains the origin of the narrow PL lines. Then we will discuss a transport model for the filling mechanism of these states, which can reproduce the temperature evolution of the intensity of the lines.

In Fig. 3 (upper left), a schematic cross section of a quantum well is shown. A variation of the GaAs well width also affects the thickness of the AlAs layer, but here in the opposite direction. This behavior is also reflected in the change of the quantization energies (Fig. 3, lower left). Here, a change in layer thickness of 1 bilayer does not affect the band alignment. But if the GaAs layer becomes thicker by 2 bilayers, the electronic state in the GaAs has a lower energy than the global conduction-band minimum in an ideal structure. A local crossover from type-II to type-I band alignment has taken place.

This proposed scenario is supported by a comparison of the four samples. The respective layer thicknesses of samples *A* and *D* fit almost perfectly to the situation described above. Sample *A* has got a type-II band alignment, but a local crossover as the one sketched in Fig. 3 is possible. The transition energy in sample *D*, which definitely has a type-I band alignment, compares well with the spectral position of the narrow lines in sample *A*. In addition to this experimental evidence, we carried out Kronig-Penney calculations. These calculations predict that a local band alignment crossover is possible for samples *A* and *B*. This explains why we have similar observations for samples *A* and *B*. On the other hand, this kind of crossover is not possible for sample *C*. Indeed, we do not observe the group of intense narrow lines for sample *C* in the μ-PL experiment. This fact is also an indication that the discussed lines in sample *A* and *B* are not due to impurity related transitions.[15]

When a fluctuation induced crossover from type-II to type-I takes place, the local conduction-band minimum is in the GaAs layer. The result is an additional lateral confinement of the electron. At the same spatial position, there is a local maximum in the GaAs valence band. Consequently, we find a deeply localized center for an exciton with quasi-zero-dimensional character.

The presence of such local type-I centers can explain that there are no sharp features on the zero-phonon emission an order of magnitude higher in intensity. The type-I states get filled by an electron transfer across the interface under emission of one AlAs LO phonon. The subsequent radiative decay is direct, and the electron and hole wave functions have a large spatial overlap. The optical matrix element is therefore larger by orders of magnitude compared to the one for the indirect type-II transitions.[11] Consistently, these sharp lines have a rather high intensity compared to the type-II luminescence (Fig. 1). As a second feature, this model explains why we only observe so few lines: all the relevant states are deep in the tail of the type-I density of states (approximately 150 meV below the type-I band edge).

The temperature dependence of the narrow lines in this energy range exhibits a completely different behavior compared to the one discussed in Sec. IV. First, their intensity rises exponentially compared to the background

$$F_{lower} := \frac{\int_{0}^{1.805\,eV} I_{\text{lines}}\, d(\hbar\omega)}{\int_{0}^{\infty} I_{\text{background}}\, d(\hbar\omega)} \propto \exp\left(\frac{T}{T_0}\right) \tag{3}$$

over *two orders of magnitude* (Fig. 4) in the temperature range under investigation.[16] The value of T_0 varies only slightly for different detection spots. Second, the lines can be traced *individually* over the temperature range. They shift very slightly by some 500 μeV, which is due to the usual shift of the band gap with temperature.[13,17]

The exponential rise of the intensity with temperature is qualitatively different from an activation law. For an activation law, the intensity would rise proportional to $\exp(T_0/T)$. But, a behavior depending on $\exp(T/T_0)$ is deduced from our data. Such a temperature law has already been reported for nonradiative recombination in amorphous semiconductors.[18,19] The fraction of the radiative and nonra-

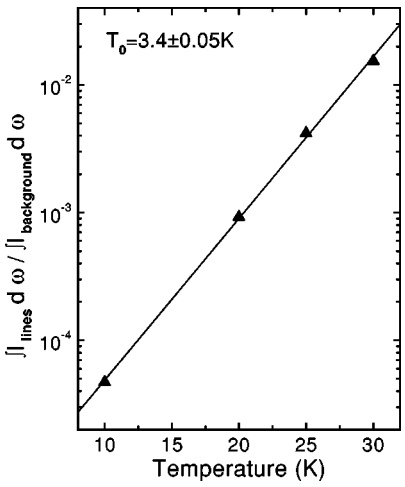

FIG. 4. The fraction F_{upper} for the spectra in Fig. 1. The solid line is an exponential fit with T_0 as a fit parameter.

diative recombination rates (p_r and p_{nr}, respectively) exhibits the same temperature dependence: $p_{nr}/p_r \propto \exp(T/T_0)$.

Street *et al.*[18] explain this law with the following model: they assume that a nonradiative process is due to tunneling of one of the carriers, e.g., the electron, away from the site of radiative recombination to a trap. The second assumption is a spatially varying band gap, i.e., an electron in a conduction-band minimum sees traps at different distances with different potential barriers. Their calculation then starts from the tunneling transition probability $p=\nu_{ph}\exp(-2\alpha R-W/kT)$ where α is the localization parameter and W the height of the tunneling barrier. Assuming that the minima are parabolic, the tunneling length is given by $R=R_0-2\gamma\sqrt{W}$. Here, γ defines the shape of the parabola and R_0 is the distance of the minima. Under the assumption of a constant α, they find after a small calculation that the rate for the most probable transition follows

$$p=\nu_{ph}\exp(-2\alpha R_0)\exp(4\alpha^2\gamma^2 k\,T)\equiv p_0\exp\left(\frac{T}{T_0}\right). \quad (4)$$

We notice that T_0 is more or less a geometrical quantity that combines the curvature of the potential and the localization parameter.

The microscopic situation in our case is similar to the one described above. The type-I states play the role of the traps, and the spatially varying band gap is here due to well width fluctuations (see Fig. 3). The main difference is that the type-I states are centers of *radiative* recombination: there is already a hole localized in the local valence-band maximum after the initial relaxation processes. After the tunneling process of the electron (Fig. 3) occurs the efficient radiative recombination which manifests itself in the intense spectrally narrow lines.

The real-space and **k**-space transfer in addition to the energy loss of the electron is mediated by phonon emission. We see in the spectra that only AlAs LO phonons contribute to this process and that the AlAs TO and LA phonons play no or only a minor role. This is in contrast to the phonon replica in the recombination where these contributions exhibit approximately equal transition probabilities. But the mechanisms are different: in the case of the phonon replica of the type-II excitonic recombination, no real-space charge transfer is required. The wave functions have enough overlap for a nonvanishing matrix element.[11] Emission of all phonons capable of conserving the **k** vector is possible and sufficient. This is different for the real filling of the type-I states since a real-space charge transfer is needed. An electric field perpendicular to the interface is needed, and the only phonon providing such a field is the LO phonon.

We finally compare the energy relaxation between the type-I and the the type-II states, respectively. In Sec. IV, we had concluded by the redshift of the spectral weight $SW(1.805\,eV,\infty)$ that phonon-assisted relaxation between the localized type-II states takes place. For the case of localized type-I states, we have no evidence for a similar behavior. The spectral weight $SW(0,1.805\,eV)$ just reflects the redshift of the individual lines. In connection with the perfect exponential rise of the intensity, we conclude that an exciton in a localized type-I states shows no further transport within its lifetime. The latter states are thus considered to be isolated.

VI. CONCLUDING REMARKS

We have demonstrated that we observe two different kinds of spectrally narrow lines in μ-PL spectroscopy of GaAs/AlAs type-II short-period superlattices. One kind is related to localized type-II excitons, which show an increasing relaxation into tail states upon raising the lattice temperature. Due to a local band-alignment crossover from type-II to type-I, also deeply localized type-I states are formed. These type-I states are filled via tunneling of the electrons across the interface. The holes are already localized in that region after the initial fast relaxation. The resulting excitons are ''trapped'' in the type-I states until they recombine radiatively.

ACKNOWLEDGMENTS

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