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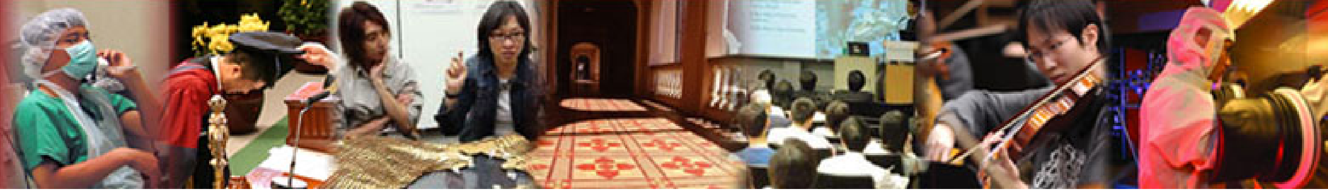

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Interpretation of anomalous temperature dependence of anti-Stokes photoluminescence at $GaInP_2/GaAs$ interface

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In this letter, we report on temperature-dependent anti-Stokes photoluminescence (ASPL) at an interface between partially ordered $GaInP_2$ epilayer and GaAs substrate. It is found that the intensity of the ASPL depends strongly on temperature accompanying with a clear blueshift in energy. A localized-state luminescence model was employed to quantitatively interpret temperature dependence of the ASPL. Excellent agreement between the theory and experiment was obtained. Radiative recombination mechanism of the up-converted carriers was discussed. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1691496]

Anti-Stokes photoluminescence (ASPL) or up-conversion photoluminescence already observed in some different semiconductor heterostructure systems is an interesting nonlinear optical phenomenon to which people have paid intensive attention in the past decade.[1–10] Among them, the lattice-matched $GaInP_2/GaAs$ is a typical one. The ASPL at the interface region between the partially ordered $GaInP_2$ epilayer and GaAs substrate was first observed by Driessen[4] and was later confirmed by other groups.[5,7,10] Most of the studies focus on the energy gain mechanism of the ASPL at the $GaInP_2/GaAs$ interface; that is, how the carriers gain energy from the thermal source to become up-converted. Two types of energy gain mechanisms—the cold Auger[4,6] and the two-step two photon absorption[5,7,8,10] were proposed. At the moment, the energy gain mechanism is still under debate, due mainly to the highly complex interface situation. On the other hand, another aspect of the ASPL—the recombination mechanism of the already up-converted carriers—has been paid little attention to, partially because it is naturally viewed as being the same as the normal PL process. In this letter, we focus on the observed anomalous temperature dependence of the ASPL at the $GaInP_2/GaAs$ interface. We attempt to quantitatively interpret the experimental data using a model that accounts for luminescence from the localized state ensemble.

The $GaInP_2$ film used in this study was grown on GaAs (001) substrate by metalorganic vapor phase epitaxy. The substrate temperature during epitaxy growth was 700 °C while the V/III gas-flow ratio was maintained at 200. The thickness of the film is 1 μm. The variable-temperature PL setup employed in the experiment was described elsewhere.[11] For the temperature-dependent ASPL measurements, the excitation light was the 647 nm line of a coherent argon-krypton mixed gas laser. In order to test the dependence of the ASPL signal of the sample on the energy (or wavelength) of incident photons, a Spectra-Physics femtosecond Ti/sapphire laser was employed to excite the sample which was cooled at 12 K in a closed-cycle cryostat. The ASPL signal was dispersed with a Jobin–Yven H250 monochromator and detected with a GaAs photomultiplier. The ASPL signal is always noticeable, although the incident photon energy is varied from 1.675 to 1.442 eV. The result seems to not support the cold Auger process being a main energy gain mechanism, as suggested in Refs. 4 and 6. As mentioned earlier, the unusual temperature dependence of the ASPL is our main concern in the present work.

Figure 1 depicts the variable-temperature ASPL spectra of the sample excited by the 647 nm laser line. It can be seen that the intensity of the ASPL peak depends strongly on temperature. At about 50 K, the ASPL becomes almost completely quenched. Moreover, the thermal quenching of the ASPL line is accompanied with a blueshift of the peak position for about 4 meV. The temperature-induced blueshift of the luminescence peak is an anomalous behavior observed in many different material systems,[12,13] and is believed to be closely related to the localized states participating in the radiative recombination. Besides the peak position, the line shape of the ASPL peak also exhibits unusual variation with increasing temperature, as shown in Fig. 1. The decrease of

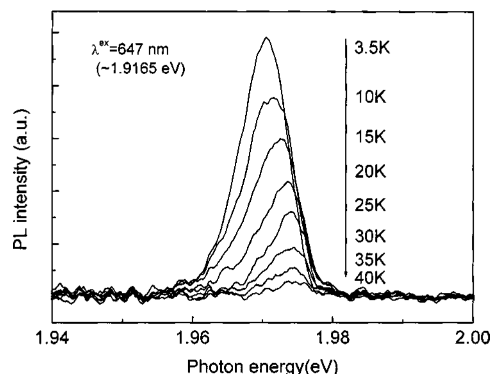

FIG. 1. ASPL spectra recorded at different temperatures when the sample was excited by the 647 nm line of a coherent argon-krypton mixed gas laser.

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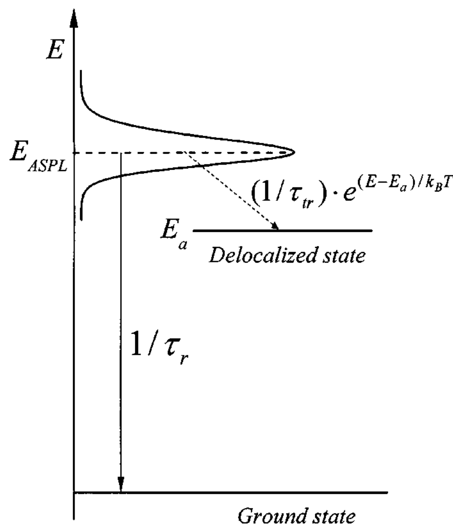

FIG. 2. Physical picture of ASPL for the localized carriers.

the emission intensity of the ASPL at the low-energy side is more rapid than that of the ASPL at the high-energy side with increasing temperature. Therefore, the ASPL peak becomes more asymmetric and narrow. This can be explained as a result of the carriers' thermal transfer from the localized states at lower energy to the localized states at higher energy. The carriers' transfer and thus redistribution within the localized states as the temperature increases will lead to the blueshift of the ASPL peak. Eventually, when the temperature is high enough, the carriers will thermally escape from the localized states at higher temperatures, and ASPL quenching then takes place. Therefore, the prevention of the carriers captured by the localized states from thermal escaping is a key element to maintaining ASPL, as concluded by Cheong *et al.*[9] From these discussions, a physical picture of temperature-dependent ASPL is suggested in Fig. 2. Recently, we have developed an analytical model of localized state luminescence based on a rate equation proposed by Xu *et al.*[14] In the model, radiative recombination, transfer, recapture, and thermal escape are taken into account. The expression for this model is derived as follows:[11]

$$n(E,T)=\rho(E)/[e^{(E-E_a)/k_\mathrm{B}T}+\tau_\mathrm{tr}/\tau_r], \tag{1}$$

where $\rho(E)$, the density distribution of localized states, is assumed to be a Gaussian-type function of $\rho(E) = \rho_0 e^{-(E-E_0)^2/2\sigma^2}$.[11,14] We have demonstrated that the function $n(E,T)$ essentially measures the "shape" of carrier distribution within the localized state ensemble. Notice that the temperature dependence of the peak position of $n(E,T)$ is due to the thermal redistribution of the carriers within the localized states. The temperature-induced gap narrowing, described by the Varshni empirical formula, should also be taken into account to interpret the temperature dependence of the luminescence peak, unless the contribution from the thermal redistribution of the carriers is dominant.[11] Therefore, a general expression describing the temperature dependence of the localized state luminescence is given by

$$E(T)=E_0-\frac{\alpha T^2}{\Theta+T}-xk_\mathrm{B}T. \tag{2}$$

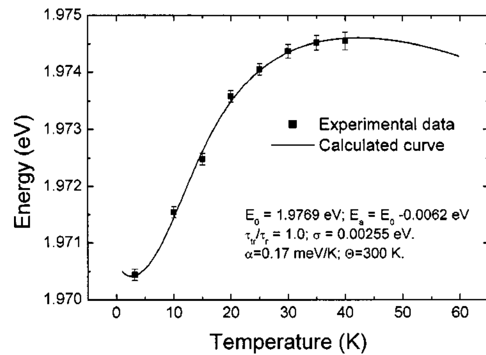

FIG. 3. Peak positions of the ASPL spectra versus temperature. Solid squares represent the experimental data while solid line is the calculated curve.

The second term on the right of Eq. (2) is the Varshni empirical formula. α and Θ are the Varshni parameter and Debye temperature of the materials, respectively. The third term on the right stems from the thermal redistribution of the localized carriers. The parameter x is obtained by numerically solving the following equation.[11]

$$xe^x=\left[\left(\frac{\sigma}{k_\mathrm{B}T}\right)^2-x\right]\left(\frac{\tau_r}{\tau_\mathrm{tr}}\right)e^{(E_0-E_a)/k_\mathrm{B}T}. \tag{3}$$

Figure 3 shows the calculated curve (solid line) according to Eq. (2) and the experimental data (solid squares). Values of the parameters adopted in the calculation are listed inside the figure. From Fig. 3, it can be seen that excellent agreement between the calculation and experiment is achieved.

Note that the energetic position E_a of the delocalized state is ~6 meV lower than the central position E_0 of the localized state distribution investigated in the present work. It is a key point of the physical model shown in Fig. 2. For usual cases, such as the self-assembled InGaAs/GaAs quantum dots[14] and cubic InGaN ternary alloy,[11] the delocalized states located on the energetic positions above the central position of the localized state distribution. The band alignment and electronic structure of $GaInP_2$/GaAs interface are rather complex and depend on the atomic ordering degree of $GaInP_2$.[15] The 6 meV shift of the delocalized state can be caused by the partially ordered $GaInP_2$ phase. Further investigations are needed to be done for identification of this delocalized state. Another interesting feature of the ASPL is that the scattering time constant τ_tr of localized carriers from the localized states to the delocalized state is equal to their radiative recombination time constant τ_r. This is quite different from the cases of InAs self-assembled quantum dots[14] and InGaN material,[11] in which τ_tr is much less than τ_r.

In conclusion, the temperature-dependent anti-Stokes PL at $GaInP_2$/GaAs interface was investigated. It was found that the localization of the up-converted carriers plays a key role in radiative recombination producing the ASPL. A localized state luminescence model was employed to interpret the temperature dependence of the peak position of the ASPL. Excellent agreement between the theory and experiment was achieved. The microscopic mechanism of the thermal quenching of ASPL at $GaInP_2$/GaAs interface was unmasked.

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