Radiative recombination of excitons in disk-shaped InAs/InP quantum dots

doi: 10.1103/PhysRevB.76.205317

Radiative recombination of excitons in disk-shaped InAs/InP quantum dots

Shinichi Tomimoto,[1] Atsushi Kurokawa,[1] Yoshiki Sakuma,[2] Tatsuya Usuki,[3,*] and Yasuaki Masumoto[1]
[1]*Institute of Physics and Center for Tsukuba Advanced Research Alliance (TARA), University of Tsukuba, Tsukuba 305-8571, Japan*
[2]*National Institute for Materials Science (NIMS), Tsukuba 305-0044, Japan*
[3]*Fujitsu Laboratories Ltd., Atsugi 243-0197, Japan*
(Received 18 June 2007; revised manuscript received 4 October 2007; published 16 November 2007)

Recombination dynamics of excitons confined in disk-shaped InAs/InP quantum dots is studied by time-resolved photoluminescence measurements. By comparing the result with that in a homologous ultrathin quantum well, it is revealed that the lateral confinement of excitons suppresses the thermal variation of the radiative recombination lifetime. The oscillator strength of the radiative transition is reduced with the decrease of the disk height, which is attributed to the increasing tunneling into the InP barrier of the exciton wave function.

DOI: 10.1103/PhysRevB.76.205317 PACS number(s): 78.67.Hc, 78.55.Cr, 78.47.+p

Radiative recombination of excitons confined in semiconductor nanostructures has a possibility of being controlled by a variety of physical mechanisms and has attracted much interest in association with the recent sophistication of nanostructure fabrication techniques. One of the main interests has been the dependence of the radiative recombination lifetimes on the size, shape, and dimensionality of the confinement space. In quantum wells (QWs), where the motion of both electrons and holes is restricted to quasi-two-dimensional (2D) planes, it is known that the lifetime becomes shorter with the decrease of the well width as a result of squeezing the exciton internal motion.[1] In quantum dots (QDs), which are considered to be quasi-zero-dimensional (0D) systems where the motion of electrons and holes is three dimensionally restricted, the lifetime is supposed to become shorter as the dot size increases, because the magnitude of the oscillator strength of the radiative recombination is enhanced with increase of the so-called "exciton coherence volume."[2] It is commonly believed that the dependence of the lifetime on the confinement dimensionality manifests itself most clearly in the thermal variation of the lifetime.[3] In QWs (2D), the lifetime generally shows a linear increase with temperature,[1] while such thermal effect is expected to be suppressed in QDs (0D).

In this work, we have experimentally investigated the recombination lifetimes of excitons confined in ultrathin disk-shaped InAs/InP QDs. By comparing the result with that in a homologous InAs/InP QW, the effect of lateral confinement of excitons on the lifetime is revealed. Moreover, the lifetime has been determined as a function of the discrete disk height. It shows reduced oscillator strength of the radiative recombination in thinner disks.

The samples are ultrathin InAs QDs fabricated on InP (001) substrates by a particular process called "double capping."[4] In this process, the Stranski-Krastanow (SK) dots of InAs were first normally grown on a 200-nm-thick InP buffer layer by metal organic chemical vapor deposition. Then, an ultrathin (nominally 1 nm) InP cap layer was deposited. As the average height of the dome-shaped SK dots is 8.2 nm, this first cap layer does not cover the large portion of the dots on the upper side. This portion was removed during the subsequent exposure to the phosphine (PH_3) gas flow by As/P exchange reaction. Consequently, the height of the InAs dots was reduced to several monolayers, while their lateral size remained as large as 35–50 nm. The height-to-base ratio is roughly 1:30 in the case of 5 ML (monolayer) disk height. These disk-shaped QDs were finally capped with a 120-nm-thick InP layer. The areal density of the QDs is about 2×10^{10} cm^{-2}. For comparison, an ultrathin InAs/InP single QW sample was also fabricated (the well width is 2 ML).[5] The thickness of the InP cap layer of this sample is 100 nm.

We have determined the recombination lifetimes of confined excitons by time-resolved photoluminescence (PL) measurements based on the frequency up-conversion technique.[6] The light source is a femtosecond mode-locked Ti:sapphire laser operating at the wavelength of 894 nm (1.386 eV) with the pulse repetition rate of 82 MHz. The output is separated into two beams, and they are used for the excitation of the samples mounted in a cryostat and for the sum-frequency generation at a gating nonlinear optical crystal ($LiIO_3$, thickness of 1 mm). The sum-frequency light is spectrally resolved with a single-grating monochromator and detected by a photomultiplier tube with the photon-counting method. The overall time resolution of the system is about 0.5 ps. All the experiments have been done with the excitation density $I_0\sim6.2$ μJ/cm^2 except for the excitation-density-dependence measurement. We have also measured the time-integrated PL spectra of the samples on the same excitation condition using a single-grating monochromator equipped with an InGaAs photodiode array detector.

Figure 1 shows the PL spectra of the QD and QW samples. The PL of the QW is characterized by a single band peaked at 1.16 eV [Fig. 1(a)]. As the temperature rises, this peak slightly shifts toward lower energy, and the intensity decreases steeply up to $T=100$ K due to the thermally activated nonradiative decay of excitons. The intensity, however, begins to increase around 100 K. This can be understood as follows. The excitation photon energy (1.386 eV) in our experiment is lower than the energy gap of the barrier material, InP at low temperatures (about 1.42 eV around 10 K).[7,8] Thus, we can consider that the optical excitation is selective to the InAs well at the low temperatures. However, as the temperature rises, the InP band gap decreases, which leads to the absorption of the excitation photons in the barrier. Then,

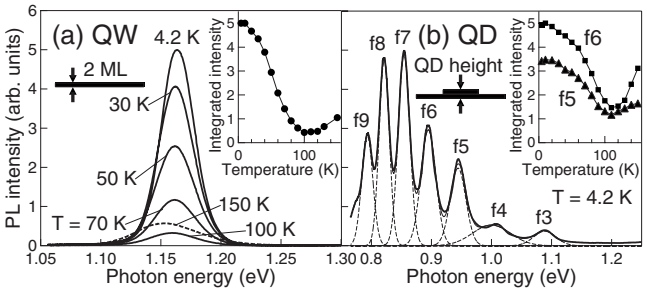

FIG. 1. PL spectra of the samples of (a) the single InAs/InP QW and (b) the disk-shaped QDs. Schematic illustrations of the cross-sectional view of the InAs layer are drawn in each panel. In (a), spectra at several different temperatures are shown. The integrated intensity in arbitrary units is plotted as a function of temperature in the inset. In (b), the thick line shows a spectrum obtained at $T=4.2$ K. It can be divided into seven Gaussian components which are shown as broken lines. The sum of these components is also drawn as a thin solid line in the panel (0.78–1.12 eV), but it is almost completely overlapping with the experimental data (thick line). Then, the temperature dependence of the integrated intensity of the f5 and f6 bands is obtained as shown in the inset.

a large number of photocarriers are created in the barrier, and they can drift and flow into the InAs well. It will enhance the PL intensity as we have observed it above 100 K. Looking at this consideration from another side, we can safely say that the excitation is certainly selective to the InAs layer below 70 K where the PL intensity is monotonously decreasing. Therefore, as will be seen afterward, our analysis of the time-resolved PL is concentrated on the temperature range below 70 K. This is to avoid the influence of the drifting carriers on our exciton lifetime analysis.

In Fig. 1(b), a PL spectrum of the disk-shaped InAs QD sample is shown. The spectrum consists of clearly split several peaks, which is consistent with the previous results.[4] As the series of the PL peaks were observed to remain unchanged over a wide range of the excitation density, they are not arising from the excited states of the QDs. It has been shown that they are reflecting the discrete distribution of the QD height in the monolayer unit.[4] This is a feature different from the InAs dots grown on an InP(311)B substrate whose heights are controlled by the same double-capping method.[9] It may be attributed to good flatness of the interfaces of our disk-shaped dots, which has been assured also from cross-sectional microscopic images.[4] The disks lie on a wetting layer of 2 ML. Here, we define the QD height as including the thickness of the wetting layer [Fig. 1(b), inset] and refer to the split PL peaks using the monolayer numbers of the QD height (f numbers). In the spectrum of Fig. 1(b), we can see seven peaks from f3 to f9. As the temperature rises, each peak shifts toward lower energy, and the intensity decreases, like the single band of the QW in Fig. 1(a). We estimated the integrated intensity of each peak by fitting the sum of seven Gaussian bands shown as broken lines in Fig. 1(b). Then, the temperature dependence of the integrated intensity of the f5 and f6 bands is obtained, as presented in the inset of the figure. It shows a decrease up to around 100 K and recovers due to the InP barrier absorption above 100 K, just as the

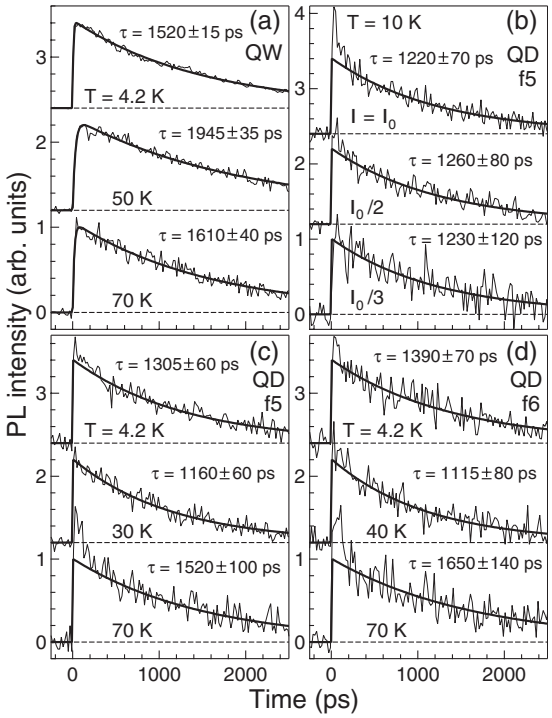

FIG. 2. Time evolution of the PL observed at the peaks (a) in the QW sample and of [(b) and (c)] the QD sample f5 and (d) f6 bands. Thick lines are the fitting results. The obtained PL lifetimes τ are shown in each panel together with the estimated error spans, which are generally less than 10% in the QD sample [(b)–(d)] and less than 3% in the QW sample (a). The data of (a), (c), and (d) are taken with the excitation density $I_0 \sim 6.2\ \mu\mathrm{J/cm}^2$ at the indicated temperatures. The data of (b) are obtained at $T=10$ K with the indicated excitation densities. (Note that the obtained τ is almost independent of the excitation density.)

QW sample does. The observed thermal quenching of the PL suggests the existence of nonradiative decay processes of excitons, which are characterized by much smaller activation energy than the excitation to the wetting layer or the InP barrier. We can notice that there is no detectable peak around the f3 energy (about 1.09 eV) in the QW spectra of Fig. 1(a). This shows that the effect of the QW width fluctuation in the sample is actually so small that the majority of excitons remain and recombine in the 2 ML layer.

Figure 2 is the results of the time-resolved PL measurements of the QW and QD samples. The thick smooth lines represent the fitting results by a single exponential-decay function,[10] and the obtained PL lifetime τ is presented in each panel. In the QW case [Fig. 2(a)], the experimental data are fitted well throughout the entire temporal range, and the τ is plotted as a function of temperature in Fig. 3(a). As the temperature rises, it first shows an increase and reaches a maximum around 50 K [the middle panel of Fig. 2(a)]. Then, it steeply decreases. This nonmonotonous behavior with temperature represents the competition of two different processes whose rates have opposite temperature dependences. One is the radiative recombination of excitons. In general, the radiative lifetime τ_r of excitons in QWs rises linearly with temperature[1,11] because of the in-plane k conservation

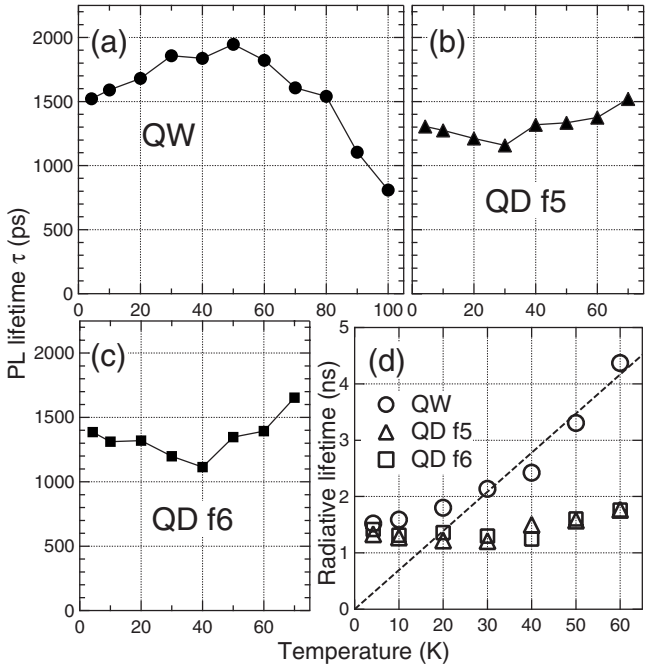

FIG. 3. Temperature dependence of the PL lifetimes τ of (a) the QW, (b) the QD f5 peak, and (c) the QD f6 peak. (d) shows the obtained radiative recombination lifetimes τ_r for the three cases. The broken line represents the linear relation with the gradient of 69.4 ps/K.

during the recombination process and the thermalization of excitons in the k space.[12] The PL lifetime shown in Fig. 3(a) becomes longer with temperature up to about 50 K, although the time-integrated intensity of the PL [Fig. 1(a), inset] considerably decreases in the same temperature range. This should be considered as a manifestation of the above temperature variation of the radiative recombination lifetime in the QW. For this reason, we assume that the PL lifetime below 50 K in this sample is determined mainly by the radiative recombination. The other process, which competes with the radiative process, is the nonradiative decay of excitons from which the thermal quenching of the PL shown in Fig. 1 arises. The nonradiative recombination lifetime τ_{nr} becomes shorter at higher temperatures. Therefore, we can say that the temperature dependence of τ in Fig. 3(a) is governed by the radiative recombination below 50 K and by the nonradiative decay at higher temperatures. From $\tau(T)$ and the PL intensity $\eta(T)$ shown in the inset of Fig. 1(a), we have obtained the radiative lifetime $\tau_r(T)$ in Fig. 3(d) (QW, open circles) by assuming that the radiative process dominates ($\tau_r \ll \tau_{nr}$) at the lowest experimental temperature (4.2 K).[13] As expected, τ_r rises linearly above 30 K. The deviation from the linear relationship below 20 K may be attributed to the localization of the excitons around impurities or at interface fluctuations.

Figures 2(b)–2(d) show the results in the QD sample. Here, the fitting lines have been obtained with masking the experimental data from 0 to 500 ps.[10] This is because an additional fast component within 200 ps often appears in this sample. We consider that this component is related to some sort of many-body states like excitonic molecules because it is more likely to appear at a higher excitation density, as seen in Fig. 2(b).[14] When that temporal range is masked, we can obtain an almost constant value of τ regardless of the excitation density. Thus, the measurement of the data in Figs. 2(c) and 2(d), the temperature dependences of the f5 and f6 bands, respectively, was performed under rather high excitation density I_0 for the experimental easiness. The PL lifetimes $\tau(T)$ of the f5 and f6 bands are summarized in Figs. 3(b) and 3(c). From them and the PL intensity $\eta(T)$ [Fig. 1(b), inset], the radiative lifetimes $\tau_r(T)$ of Fig. 3(d) (triangles and squares for f5 and f6, respectively) have been obtained in the same way as the QW case. Here, we assume again that the low-temperature PL lifetime is determined mainly by the radiative recombination. As the thermal quenching of the PL intensity in the QD sample [Fig. 1(b), inset] is rather slower than that in the QW sample [Fig. 1(a)], we consider that the effect of the nonradiative decay is even smaller in the QDs than that in the QW. We can notice that the temperature dependence of τ_r in the QDs is much smaller than that in the QW. This should be attributed to the effect of the lateral confinement of excitons in the QDs. With the removal of the translational invariance, the condition of the in-plane k conservation in the recombination process will be alleviated; moreover, the discrete distribution of energy levels in QDs will suppress the population change with temperature among them. These effects make the temperature variation of τ_r smaller in comparison with the QW case. We can see, however, that the τ_r show a slow increase above 50 K for both f5 and f6. This can be explained by the thermal population of higher exciton states having no or lower oscillator strength. A calculation by Gotoh *et al.* has shown that it can elongate the average lifetime of excitons above 50 K in a thin box-shaped QD with the lateral width of 30 nm, although the assumed QD is rather thicker (10 nm) than ours (about 1.5 and 1.8 nm for the f5 and f6 bands, respectively).[15]

In the discussion described above, the transfer of excitons between QDs has been neglected. We consider that this is appropriate as long as the sample temperature does not exceed 100 K. For the transfer to be possible, excitons (or carriers) localized in QDs have to be thermally excited into the wetting layer (or the InP barrier) first. It is, however, improbable when the temperature is below 100 K because the energy separation between the exciton states in the wetting layer (the thickness is 2 ML) and even in the thinnest QD (3 ML) corresponds to the temperature of about 1300 K (k_BT =75 meV). (This value is estimated from the difference of the PL peak energies of the QW sample [1.163 eV at T =4.2 K, Fig. 1(a)] and the f3 band [1.088 eV, Fig. 1(b)].) Therefore, it is not necessary to take account of the exciton transfer process in the lifetime analysis limited below 70 K.

We mention next the QD height dependence of the radiative recombination. By varying the photon energy of the time-resolved PL measurement, we can determine the lifetimes of excitons confined in QDs with different heights, which are specified in the monolayer unit [Fig. 1(b)]. Figure 4 shows the time evolution of the PL observed at the f3 and f6 peaks and the dependence of the PL lifetime τ on the peak

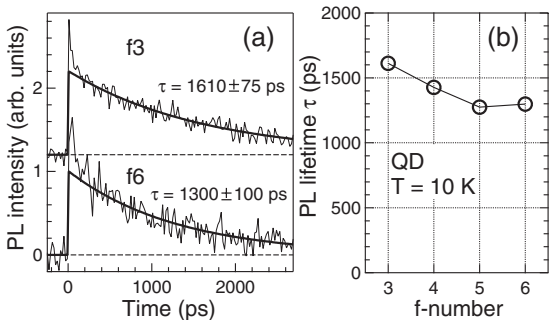

FIG. 4. QD height dependence of the time-resolved PL. (a) PL time evolution observed at the f3 and f6 peaks ($T=10$ K). (b) Peak f-number dependence of the PL lifetime $\tau(\simeq\tau_r)$.

f number. The measurement has been done at $T=10$ K where it is very likely that the radiative process governs the PL dynamics ($\tau\simeq\tau_r$). As seen in Fig. 4(b), the height dependence of τ is not very large; it slightly increases (about 1.2 times) while the QD height becomes half (from 6 to 3 ML). As the radiative recombination rate has the relationship $1/\tau_r\propto f\omega^2$ with the oscillator strength f and the optical transition frequency ω,[16] this result means that the f at f3 is only about 54% of that at f6. [The ω at f3 is about 22% higher than that at f6, as seen in Fig. 1(b).] This height dependence of f cannot be ascribed to the change of the exciton lateral extent with the QD height because the lateral size of the QDs scarcely depends on the height.[4]

Although the f may seem roughly in proportion with the QD height (or volume), it is not appropriate to attribute this dependence to the variation of the exciton coherence volume. In general, the concept of the exciton coherence volume is used to explain radiative lifetimes only when the confinement dimensions are sufficiently large in comparison with the exciton Bohr radius.[2] If this is not the case, it is more common to consider the electron-hole relative motion to explain the radiative lifetime evolution with the change of confinement. The Bohr radius of the three-dimensional exciton in InAs is about 37 nm, and it is much larger than the disk heights. It is even larger than half the lateral size of the QDs. Therefore, it is more appropriate to explain the disk-height dependence of f in terms of the electron-hole wave function overlap than the exciton coherence volume.

The height dependence of f is significantly different from the intuitive expectation that the increased overlap of the electron and hole wave functions in lower QDs will enhance the exciton recombination. Indeed, it has been commonly observed in QWs that the radiative lifetime has an almost linear dependence on the well width.[1,17] The data in Fig. 4(b), however, can be understood if the tunneling of the wave functions into the InP barrier is taken into consideration. It can occur in ultrathin structures. Recently, Maes *et al.* have experimentally determined the exciton extent in thin ribbon-shaped InAs/InP quantum wires and reported that, when the height of the ribbon is 4 ML (about 1.2 nm), the exciton extent along the direction is as large as 5.8 nm.[18] This large tunneling of the wave function is caused by the high confinement energy which is comparable to the band offset of the InAs/InP heterostructure and tends to be increased as the InAs layer height decreases. In addition, the tunneling is expected to be far larger for electrons than for holes because their effective mass is much smaller. Therefore, the overlap of the electron and hole wave functions decreases as the InAs layer becomes thinner. Cebulla *et al.* observed in thin $In_xGa_{1-x}As/InP$ QWs that the exciton lifetime slightly increases as the well width decreases from about 4 nm by the above-mentioned mechanism.[19] In Fig. 4(b), we have varied the height of the disk-shaped QD from about 1.8 nm (6 ML) to 0.9 nm (3 ML). Thus, the observed decrease of the oscillator strength in lower QDs should be attributed to the increasing tunneling of the exciton wave function into the barrier layer.

The plot of Fig. 4(b) suggests that the PL lifetime of the "f2" band in the QD sample would be longer than that of the QW sample, if we could have 2-ML-thick QDs. This may hint that the lateral confinement size of the QDs is smaller than that of localized excitons in the QW at low temperature because the radiative recombination lifetime is expected to become longer as the lateral confinement size decreases.[16,20,21]

In summary, we have investigated the recombination lifetimes of excitons confined in disk-shaped InAs/InP quantum dots by time-resolved PL measurements. By comparing the temperature dependence with that in a homologous quantum well, it is revealed that the temperature variation of the radiative recombination lifetime is suppressed due to the lateral confinement of excitons. The oscillator strength of the radiative transition appreciably decreases as the disk height becomes smaller. It is attributed to the increasing tunneling of the exciton wave function into the barrier layer.

This work was supported by the Grant-in-Aid for the Scientific Research (Grant No. 18204028) and for Young Scientists (B) (Grants No. 17740185 and No. 19740174) from the MEXT of Japan.

*Present address: Institute for Nano Quantum Information Electronics, the University of Tokyo, Meguroku, Tokyo 153-8505, Japan.

[1] J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, and R. J. Elliott, Phys. Rev. Lett. **59**, 2337 (1987).

[2] Y. Kayanuma, Phys. Rev. B **38**, 9797 (1988).

[3] C. Klingshirn, *Semiconductor Optics*, 2nd ed. (Springer, Berlin, 2005), p. 629.

[4] Y. Sakuma, M. Takeguchi, K. Takemoto, S. Hirose, T. Usuki, and N. Yokoyama, J. Vac. Sci. Technol. B **23**, 1741 (2005); Y. Sakuma, K. Takemoto, S. Hirose, T. Usuki, and N. Yokoyama, Physica E (Amsterdam) **26**, 81 (2005).

[5] The thin InAs layer of 2 ML is regarded as a wetting layer in the SK dot growth in nature. For fabrication of the QW sample, the deposition of InAs was ceased just before the three-dimensional

dot growth begins. (Nominal amount of the supplied InAs corresponds to 2.4 ML.)

[6] J. Shah, IEEE J. Quantum Electron. **QE-24**, 276 (1988).

[7] W. J. Turner, W. E. Reese, and G. D. Pettit, Phys. Rev. **136**, A1467 (1964).

[8] L. Pavesi, F. Piazza, A. Rudra, J. F. Carlin, and M. Ilegems, Phys. Rev. B **44**, 9052 (1991).

[9] C. Paranthoen, N. Bertru, O. Dehaese, A. Le Corre, S. Loualiche, B. Lambert, and G. Patriarche, Appl. Phys. Lett. **78**, 1751 (2001).

[10] The fitting function has a formula of $I(t)=I_0[1-\exp(-t/\tau_{\mathrm{rise}})]\exp(-t/\tau)$. The τ_{rise} represents the rise time of the PL, which has values of 10–60 ps in the QW case [Fig. 2(a)]. This is assumed to be much shorter than the experimental data point separation (20 ps) in the QD case [Figs. 2(b)–2(d)].

[11] J. Martinez-Pastor, A. Vinattieri, L. Carraresi, M. Colocci, Ph. Roussignol, and G. Weimann, Phys. Rev. B **47**, 10456 (1993).

[12] L. C. Andreani, F. Tassone, and F. Bassani, Solid State Commun. **77**, 641 (1991).

[13] The PL intensity η is proportional to $\tau_r^{-1}/(\tau_r^{-1}+\tau_{nr}^{-1})$. From this and the relation $\tau^{-1}=\tau_r^{-1}+\tau_{nr}^{-1}$, we can find $\tau_r(T)\propto\tau(T)/\eta(T)$.

[14] We have not noticed systematic changes of the fast component with temperature until now, which may be partly due to the signal to noise ratio of our data (Fig. 2) which is not very good.

[15] H. Gotoh, H. Ando, and T. Takagahara, J. Appl. Phys. **81**, 1785 (1997); H. Gotoh, H. Ando, T. Takagahara, H. Kamada, A. Chavez-Pirson, and J. Temmyo, Jpn. J. Appl. Phys., Part 1 **36**, 4204 (1997).

[16] J. Hours, P. Senellart, E. Peter, A. Cavanna, and J. Bloch, Phys. Rev. B **71**, 161306(R) (2005).

[17] R. Höger, E. O. Göbel, J. Kuhl, and K. Ploog, in *Proceedings of the Seventeenth International Conference on the Physics of Semiconductors, San Francisco, 1984*, edited by D. J. Chadi and W. A. Harrison (Springer, New York, 1985), p. 575.

[18] J. Maes, M. Hayne, Y. Sidor, B. Partoens, F. M. Peeters, Y. González, L. González, D. Fuster, J. M. García, and V. V. Moshchalkov, Phys. Rev. B **70**, 155311 (2004).

[19] U. Cebulla, G. Bacher, A. Forchel, G. Mayer, and W. T. Tsang, Phys. Rev. B **39**, 6257 (1989).

[20] D. S. Citrin, Phys. Rev. B **47**, 3832 (1993).

[21] M. Sugawara, Phys. Rev. B **51**, 10743 (1995).