Carrier Relaxation of ZnCdSe/ZnSe Quantum Wells

2008 Jpn. J. Appl. Phys. 47 7056

(http://iopscience.iop.org/1347-4065/47/9R/7056)

Japanese Journal of Applied Physics
Vol. 47, No. 9, 2008, pp. 7056–7059

Carrier Relaxation of ZnCdSe/ZnSe Quantum Wells

Der-Jun JANG*, Meng-En LEE[1], Yung-Hsien CHUNG, Chu-Shou YANG[2], and Wu-Ching CHOU[2]

Department of Physics and Center of Nanoscience, National Sun Yat-sen University, Kaohsiung, Taiwan 80441, R.O.C.
[1]*Department of Physics, National Kaoshiung Normal University, Kaohsiung, Taiwan 80264, R.O.C.*
[2]*Department of Electrophysics, National Chiao-Tung University, Hsinchu, 30056 Taiwan, R.O.C.*

(Received December 7, 2007; accepted May 10, 2008; published online September 12, 2008)

The energy relaxations of $Zn_{0.91}Cd_{0.09}Se$ multiquantum wells and epilayer structures were studied with an ultrafast time-resolved photoluminescence apparatus. The increase in the rise time of photoluminescence with decreasing photon energy and the redshift of the peak energy of time-resolved photoluminescence spectra with the delay time are attributed to the band-filling effect and energy relaxation of hot carriers. The derived carrier temperature decreases rapidly within the first 10 ps after photoexcitation and at a much slower rate thereafter. The fast carrier cooling can be explained by the longitudinal optical (LO) phonon emissions by carriers through the Fröhlich interaction. The obtained effective scattering times of carrier and LO phonons are comparable to the theoretical prediction of 20 fs for multi-quantum well (MQW) of 20 nm well thickness and the epilayer. The slow carrier capture process may account for the long effective scattering time of 35 fs for the MQWs of 5 nm well thickness. [DOI: 10.1143/JJAP.47.7056]

KEYWORDS: ZnCdSe, quantum well, carrier capture, hot carrier, energy relaxation, optical phonon

1. Introduction

The nonequilibrium properties of photoexcited carriers in semiconductors have been intensively studied for their adverse impact on the performances of optoelectric devices.[1)] The understanding of the mechanism and magnitude of the photoexcited carrier relaxation is not only of physical interest but also important for the development of fast-speed optoelectronic devices and spintronics. Recently, spin detectors composed of ZnCdSe/ZnSe quantum wells doped with the transition-metal element Mn have been studied, and it is concluded that the hot carrier relaxation may have an important effect on the efficiency of spin injection across a semiconductor heterointerface.[2,3)] Meanwhile, the quantum well[4)] and quantum dot structures[5,6)] of the II–VI semiconductor that employs ZnSe as barriers and ZnCdSe as active layers have been investigated owing to their wide applications. However, the carrier relaxations of ZnCdSe/ZnSe heterostructures and quantum wells have not been studied. Among III–V semiconductor materials, such as GaAs, the energy relaxation investigation was complicated by the fact that the excess energy of hot carriers was often larger than the Γ–L valley energy splitting (~300 meV). However, in II–VI semiconductor materials, such as ZnSe and CdSe, the energy difference between the bottoms of the Γ and L valleys is larger than 1 eV.[7,8)] In the previous study, we found that hot carriers release most of the excessive energy within 10 ps and the effective scattering time between carriers and longitudinal optical (LO) phonon is 40 fs in ZnCdSe epilayers. In this paper, we study the carrier relaxation in $Zn_{0.91}Cd_{0.09}Se/ZnSe$ multi-quantum wells (MQWs) and epilayer. We found that, for the MQW of 20 nm well thickness and the epilayer, the effective carrier–LO-phonon scattering times (τ_0) are comparable to the theoretical prediction. The effective scattering time of the MQWs of 5 nm well thickness, however, is longer than the theoretical prediction. We found that carrier capture process may have an effect on the effective scattering time. The information obtained in this work, such as effective scattering times of all samples, will facilitate the design and development of high-speed and high-performance devices with quantum well and dot structures[9)] of ZnCdSe/ZnSe doped with transition-metal elements.

2. Experiment and Samples

The $Zn_{0.91}Cd_{0.09}Se/ZnSe$ MQW structures investigated are composed 20 periods of 20 nm barrier thickness, grown on a ZnSe buffer layer of 500 nm thickness by molecular beam epitaxy on undoped GaAs(001) substrates. Two MQWs of 5 and 20 nm well thicknesses and a thick epitaxy layer of ZnCdSe were studied and are designated as samples A, B, and C, respectively. For the MQW structures, both electrons and holes are confined in ZnCdSe QWs. The details of the samples are given elsewhere.[10)] The time-resolved photoluminescence (TRPL) was measured by all optical photoluminescence (PL) up-conversion spectroscopy.[11)] The samples were illuminated by laser pulses of 3.0 eV energy, which is frequency-doubled from the fundamental of a Kerr-lens mode-locked Ti:sapphire laser with a pulsewidth of 150 fs and a repetition rate of 76 MHz. Details of the experimental configuration have been described elsewhere.[12)] Considering the absorption coefficients,[13)] the reflectance from the surface of the sample (<5%), and the Gaussian profile of the laser beam, the density of the photoexcited carriers was estimated to be 1×10^{17} cm^{-3}.

3. Results and Discussion

The inset of Fig. 1 shows the time-integrated PL of these samples. The blue shift of PL peak energy due to quantum confinement is evident for ZnCdSe MQW. The PL peak energy of sample B of thickness 20 nm is close to that of the epilayer and this may be due to the large effective masses of electrons and holes in this structure.[14)] The PL profile of sample A is similar to that studied by Netti *et al.*[15)] We have obtained the TRPL of all the three samples at 40 K and for clarity only that of sample B is shown in Fig. 1. The TRPL at photon energy far above the band edge exhibits a very fast rise and decay within 2 ps. As the photon energy decreases to the band gap energy, owing to the increasing overlap of wave functions of electrons and holes, the PL shows an increase not only in the rise and decay times but also in the intensity. The fast decays of PL at high energies and the

*E-mail address: djjang@mail.nsysu.edu.tw

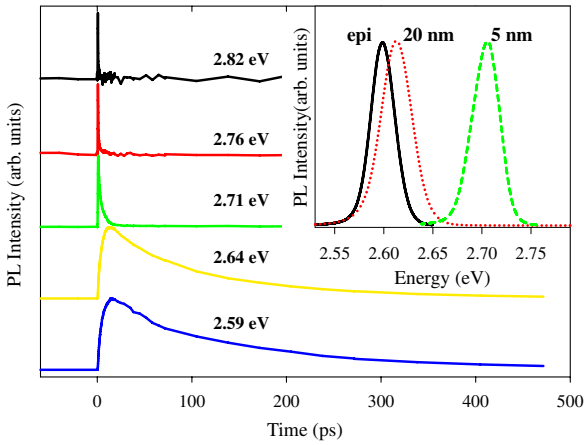

Fig. 1. (Color online) Normalized TRPL of sample B at 40 K. The inset shows the time-integrated PLs of sample A (dashed line), sample B (dotted line), and sample C (solid line) at 40 K.

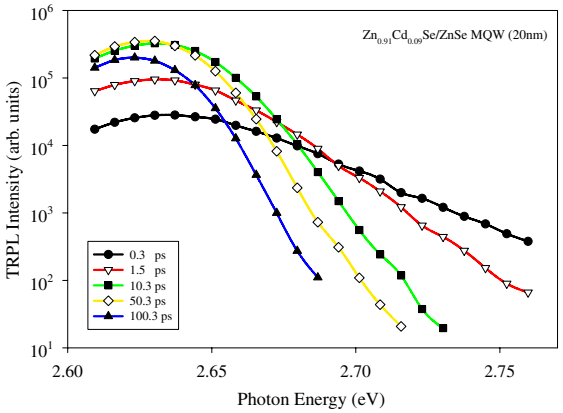

Fig. 2. (Color online) TRPL spectra of sample B at different delay times. The slopes of the high energy portions were used to determine the carrier temperatures. See text for details. The lines are for visual guidance.

slow rises of PL at energies close to the band gap energy indicate that carriers relax to the lower-energy states in a very short time. From Fig. 1, we can estimate that carriers build up at the band gap at a time scale of approximately 10 ps. This rapid carrier accumulation at the band edge suggests that carriers interact with LO phonons during the relaxation. With the excitation energy of 3.0 eV, the initial excessive energies of the photoexcited carriers vary in different samples and are at least larger than 300 meV, which is much larger than the LO phonon energy of 29.7 meV.[10] Therefore, the emission of LO phonons by hot carriers to release excessive energy is a probable and the most effective process. After the first 10 ps of fast cooling, the carriers continue to release energy by other types of carrier–phonon interactions and by carrier recombination. While the PL lifetimes at the energy close to the band edge are close for both MQW structures (samples A and B) and are comparable to the results reported in other studies,[12,16,17] the PL lifetime of sample C is smaller than those of samples A and B.

To study the energy relaxation of carriers in these structures, the TRPL spectra at different delay times were measured and only that of sample B is shown in Fig. 2. Right after photoexcitaiton, the TRPL intensity is weak and spans broadly in spectrum. The TRPL spectra reveal an increase in intensity and redshift with delay time. The above observation and the increase in PL rise time shown in Fig. 1 demonstrate that the band-filling effect occurred during the carrier relaxation. The exponential behavior at high-energy tails indicates that carriers are thermalized in a very short time, and the carrier temperature T_c at a specific time delay can be derived from the slope of the TRPL spectra, assuming the Boltzmann approximation, using the equation[18]

$$I_{PL}(E) \propto \alpha(E)E^2 \exp\left(-\frac{E-E_g}{k_B T_c}\right).$$

Here, $I_{PL}(E)$ is the PL intensity at energy E, $\alpha(E)$ the absorption coefficient, E_g the band gap energy, and k_B the Boltzmann constant. The time-dependent carrier temperatures of all the three samples are shown in Fig. 3. The carrier distribution is clearly thermalized within 0.5 ps of excitation and this time scale is comparable to that shown in another report.[19] The carrier temperature decreases rapidly

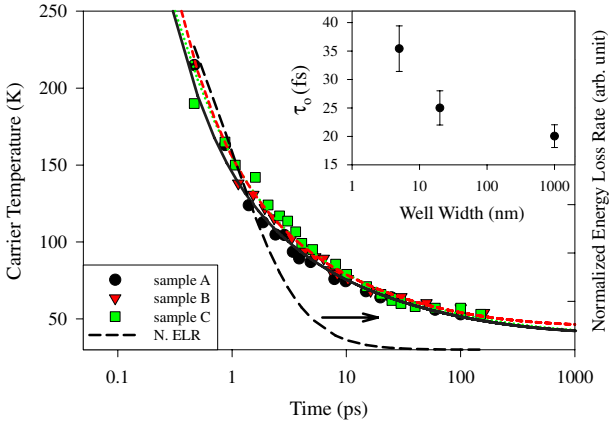

Fig. 3. (Color online) Carrier cooling curves for samples A, B, and C. The dashed, dotted, and solid lines are the fitting curves for samples A, B, and C, respectively. The scale of the normalized energy loss rate (long dashed line) is on the right. The inset shows the effective scattering times as a function of well width.

within the first 10 ps and then continues to decrease with moderate rates. This behavior can be clearly justified from the time-dependent energy loss rate (ELR) determined from the experiment data shown in Fig. 3. The ELR decreases rapidly within 10 ps and becomes insignificant after 10 ps. The carrier temperature decreases to 50 K, which is close to the lattice temperature of 40 K, for all the three samples at the time delay of 170 ps. Beyond that, T_c cannot be derived owing to the weak intensity of TRPL. The fast decay of carrier temperature is consistent with the discussion on the rise time of PL at the energy close to the band edge and can be attributed to the carrier–LO-phonon interaction to release excessive energy.

To understand the mechanism of the energy relaxation of the carrier, we fit the obtained time-dependent carrier temperature, assuming that the dominant mechanism is the carrier–LO-phonon interaction, with[20]

$$P(t) = \frac{\hbar\omega_{LO}}{\tau_0}\frac{\exp(-x_c)-\exp(-x_L)}{1-\exp(-x_L)} \times \frac{[(x_c/2)^{1/2}\exp(x_c/2)K_0(x_c/2)]}{\sqrt{\pi/2}}.$$

Here, $P(t)$ is the mean power lost per carrier, τ_0 the effective carrier optical-phonon scattering time, K_0 the modified Bessel Function of order zero, and $x_{c(L)} = E_{op}/k_B T_{c(L)}$, with $T_{c(L)}$ being the carrier (lattice) temperature. The carrier cooling curves were fitted with the initial temperature T_0 of the carriers and τ_0 with

$$T_c = T_0 - \frac{1}{3k_B C}\int_0^t P(t)\,dt.$$

A scaling factor C is included to take into account the reduction of the carrier ELR that may be caused by the hot phonon effect or other effects. While the time-dependent carrier temperatures of these samples are comparable, the difference in the band gap energies that result in different initial carrier temperatures may affect the fitted effective scattering times. The best fits give the effective scattering times τ_0's of 35, 25, and 20 fs for samples A, B, and C, respectively, as shown in the inset of Fig. 3. The result is consistent with the reported τ_0's for ZnSe and CdSe.[21,22] Both τ_0's of samples B and C are close to the theoretical predictions for ZnSe and CdSe,[23] and this result concludes that the hot phonon effect is insignificant in both samples at a carrier density of $1 \times 10^{17}\,cm^{-3}$. The hot phonon effect has been reported for carrier densities above $1 \times 10^{18}\,cm^{-3}$ in III–V and II–VI semiconductor materials.[24]

The slightly long τ_0 of sample A is irrelevant to the hot phonon effect or plasma screening effect because of the low carrier density. It may be associated with the relatively larger excessive energy than those for samples B and C, and also with carrier relaxation at barriers. As shown in Fig. 4, for all these samples, the PLs at the barrier energy of 2.82 eV rise steeply to maxima as fast as 450 fs and decay very rapidly with different decay times. This implies that the photogenerated carriers are created and decay to the band edges of barriers at the same time scale and this observation is helpful for analyzing the carrier cooling under the same conditions. While the PL at the barrier energy decays comparably for samples B and C, one can see that the PL of sample C decays apparently at a much slower rate. It has been reported that the carrier relaxation at the barrier energy is a function of the well thickness of MQW structures.[25] The longer PL decay of sample C indicates that it may take a longer time for carriers generated in the barrier region to be captured by quantum wells and thus prolong the energy relaxation. Therefore, a longer τ_0 is to be expected. The dependence of τ_0 on the well thickness is contingent on the energy difference of the ground state of the MQW and that of its barrier. As in the case for carrier capture time,[25] an oscillation of τ_0 is likely to occur as the well width changes. The minor increase in τ_0 of sample B compared with that of sample C is also attributed to the well width and the smaller excessive energy of carriers in sample B. The scattering time is representative of the complicated convolution of all electrons and holes energy relaxation and the carrier capture.

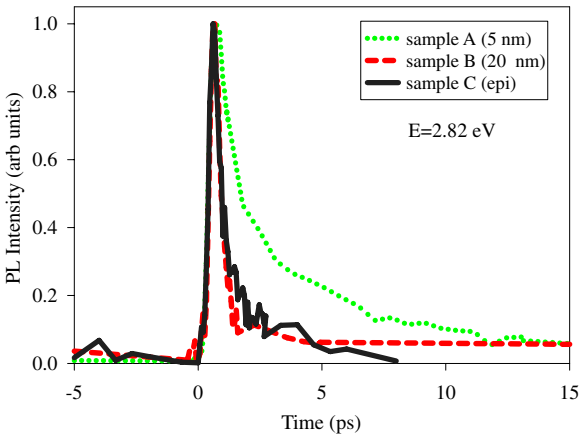

Fig. 4. (Color online) TRPLs at photon energy of 2.82 eV of sample A (dotted line), sample B (dashed line), and sample C (solid line).

4. Conclusions

In summary, the energy relaxations of $Zn_{0.91}Cd_{0.09}Se$ of MQWs and epilayer structures were studied with an ultrafast time-resolved photoluminescence apparatus. The increase in the rise time of PL with decreasing photon energy and the redshift of the peak energy of time-resolved PL spectra with the time delay demonstrate the band-filling effect and the energy relaxation of carriers by interacting with the LO phonon. The carrier temperatures at different delay times were determined from the time-resolved PL spectra. The carrier temperature decreases rapidly within the first 10 ps after photoexcitation and with much slower rate thereafter. The fast carrier cooling can be explained by the LO-phonon emissions by carriers through the Fröhlich interaction. The obtained effective scattering times of carrier and LO phonons are comparable to the theoretical prediction of 20 fs for samples B and C. The hot phonon effect cannot account for the slightly long of the effective scattering time for sample A. Instead, from the observation of the TRPL at the barrier energy, it can be attributed to the slow carrier capture process to quantum wells in sample A.

Acknowledgment

This research was supported by the National Science Council of the Republic of China, Grant No. NSC 96-2112-M-110-012.

1) P. Battacharya, J. Singh, H. Yoon, X. Zhang, A. G. Aitken, and Y. Lam: IEEE J. Quantum Electron. **32** (1996) 1620.
2) W. M. Chen, I. A. Buyanova, K. Kayanuma, Z. H. Chen, A. Murayama, Y. Oka, A. A. Toropov, S. V. Sorokin, S. V. Ivanov, and P. S. Kop'ev: Appl. Phys. Lett. **85** (2004) 5260.
3) O. Maksimov, S. P. Guo, L. Zeng, M. C. Tamargo, F. C. Peiris, and J. K. Furdyna: J. Appl. Phys. **89** (2001) 2202.
4) E. Vanelle, B. Zhang, T. Yasuda, W. Wang, Y. Segawa, T. Itoh, D. Brinkmann, and G. Fishman: Jpn. J. Appl. Phys. **37** (1998) 1474.
5) R. Cingolani, R. Rinaldi, L. Calcagnile, P. Prete, P. Sciacovelli, L. Tapfer, L. Vanzetti, G. Mula, F. Bassani, L. Sorba, and A. Franciosi: Phys. Rev. B **49** (1994) 16769.
6) Y.-D. Kim, S. L. Cooper, M. V. Klein, and B. T. Jonker: Appl. Phys. Lett. **62** (1993) 2387.
7) J. R. Chelikowsky and M. L. Cohen: Phys. Rev. B **14** (1974) 556.
8) D. J. Dougherty, S. B. Gleischer, E. L. Warlick, J. L. House, G. S. Petrich, L. A. Kolodziejski, and E. P. Ippen: Appl. Phys. Lett. **71** (1997) 3144.
9) N. Matsumura, E. Tai, T. Kimura, T. Saito, M. Ohira, and J. Saraie: Jpn. J. Appl. Phys. **39** (2000) 1104.
10) C. S. Yang, Y. P. Hsieh, M. C. Kuo, P. Y. Tseng. Z. W. Yeh, K. C. Chiu, J. L. Shen, A. H. M. Chu, W. C. Chou, and W. H. Lan: Mater. Chem. Phys. **78** (2003) 602.
11) J. Shah: IEEE J. Quantum Electron. **24** (1988) 276.
12) D.-J. Jang, C. S. Yang, W. C. Chou, K. T. Kuo, and M. S. Lee: Jpn. J.

Appl. Phys. **42** (2003) 7370.
13) S. Ninomiya and S. Adachi: J. Appl. Phys. **78** (1995) 4681.
14) H. K. Ng, Y. A. Leem, R. Knobel, I. P. Somrchkova, A. A. Sirenko, and N. Samarth: Appl. Phys. Lett. **75** (1999) 3662.
15) M. C. Netti, C. Gadaleta, N. Del Fatti, F. Vallee, and R. Tommasi: Phys. Rev. B **61** (2000) 15637.
16) Y. Masumoto and S. Shionoya: Phys. Rev. B **30** (1984) 1076.
17) H. Wang, K. S. Wong, B. A. Foreman, Z. Y. Yang, and G. K. L. Wong: J. Appl. Phys. **83** (1998) 4773.
18) S. A. Lyon: J. Lumin. **35** (1986) 121.
19) H. Wang, I. K. Sou, G. K. L. Wong, and K. S. Wong: Superlattices Microstruct. **29** (2001) 301.
20) J. Shah: in *The Physics of the Two-Dimensional Electron Gas*, ed. J. T. Devreese and F. M. Peeter (Plenum, New York, 1986) p. 183.
21) M. Mehendale, S. Sivananthan, and W. Andreas Schroeder: Appl. Phys. Lett. **71** (1997) 1089.
22) F. Sasaki, T. Mishina, Y. Masumoto, B. Fluegel, K. Meissner, and N. Peyghambarian: Semicond. Sci. Technol. **7** (1992) B160.
23) E. M. Conwell: in *Solid State Physics*, ed. F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1967) Suppl. Vol. 9.
24) S. S. Prabhu, A. S. Vengurlekar, S. K. Roy, and J. Shah: Phys. Rev. B **51** (1995) 14233.
25) P. W. M. Blom, C. Smit, J. E. M. Haverkort, and J. H. Wolter: Phys. Rev. B **47** (1993) 2072.