Time-resolved kinetics of *e-h* plasma in GaAsP under intense picosecond laser pulse excitation

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Time-resolved photoluminescence kinetics of $GaAs_{1-x}P_x$ ($x = 0.38$) were measured by a streak camera system in order to determine the radiative and nonradiative recombination rates. The photoluminescence decay profile was found to be intensity dependent. When excitation power fluence increased above 6×10^8 W/cm^2, the decay profile of emission deviated from exponential form. This is attributed to bimolecular and Auger processes. The bimolecular and Auger rates were determined to be $B_R = 9\times10^{-10}$ cm^3/s and $C_{NR} = 3\times10^{-29}$ cm^6/s by fitting the time-resolved photoluminescence decay profiles to the solution of the rate equation which describes the dynamical behavior of the photogenerated carriers.

Information about carrier recombination rates is essential in the design and implementation of semiconductor lasers, light-emitting diodes, and electro-optical switches.[1] Over the past three decades, a great deal of effort has been devoted to the understanding of radiative and nonradiative processes in semiconductors.[2] The reliability of photonic devices has to be tested under extreme operational conditions, and any deviation from linearity may limit their applications. Most of the problems encountered regarding nonlinearity have been attributed to loss mechanisms associated with the nonradiative Auger process. Therefore, it is important to determine the radiative and nonradiative recombination rates in semiconductors under high excitation power. There is a variety of direct and indirect techniques to measure or estimate recombination rates in semiconductors under current injection or laser pulse excitation. Conventional techniques such as turn on delay, phase shift, and photon counting are limited to the nanosecond time scale.[3,4] In most of the experiments to date, semiconductors were excited under quasistationary conditions where the exciting laser pulse duration was longer or comparable to the carrier lifetime. Since direct band-gap semiconductors have inherently short radiative lifetimes, radiative and nonradiative processes have to be studied by picosecond techniques such as pump and probe,[5] optical Kerr gate,[6] up-conversion gate,[7] and streak camera.[8]

In this letter, we have determined nonradiative and radiative rates by analysis of the time-resolved photoluminescence kinetics of $GaAs_{1-x}P_x$ ($x = 0.38$) under high power picosecond laser pulse excitation at room temperature using a streak camera system.

The experimental setup used in this research has been described in detail elsewhere.[9] A 527-nm pulse of 8-ps duration was used to excite the sample of $GaAs_{1-x}P_x$ ($x = 0.38$) on the front surface. The sample was 30 μm thick, n type, Te doped ($\sim2\times10^{17}$ cm^{-3}), and grown by vapor phase epitaxy on a GaAs substrate. The composition of the sample was calculated from the calibration of band gap versus composition.[10] The band gap of the sample was determined to be 1.892 eV at room temperature from the direct transition relationship $E_g = h\nu_p + E_d - 1/2\,kT$, where $h\nu_p$ is the energy of the peak in the low power steady state photoluminescence spectra, E_d is the donor ionization energy (~5 meV), and the $1/2\,kT$ term accounts for the direct transition of the carrier. The photoluminescence was collected by a combination of lenses and imaged into a 30-μm slit of a Hamamatsu streak camera. The output was detected by a temporal analyzer and computer for data analysis. The time resolution of the detection system was approximately 12 ps. The excitation area was measured to be approximately 8×10^{-3} cm^2 and excitation power fluence was varied by placing neutral density filters in the path of the laser pulse.

The time-resolved photoluminescence profiles of $GaAs_{1-x}P_x$ ($x = 0.38$) at room temperature for different excitation power fluences are displayed in Fig. 1. The solid curves are experimental data, the dashed lines are the theoretical fit assuming a simple exponential decay, and the circles are the theoretical fit to the model which includes the bimolecular and Auger rates. The salient features of the experimental curves shown in Fig. 1 are the following. The rise time of photoluminescence did not change appreciably with excitation power fluence and was between 10 to 15 ps. The decay time of the emission decreased from 207 to 44 ps when excitation power fluence increased from 3×10^8 to 3×10^9 W/cm^2. When excitation power fluence was below 6×10^8 W/cm^2 the decay profile was exponential [see Fig. 1(a) and 1(b)]. As the excitation power fluence increased above 6×10^8 W/cm^2 the decay profile of the photoluminescence deviated from an exponential form. This was most apparent at the highest excitation power fluence of 2.8×10^9 W/cm^2 [see Fig. 1(d)]. An effective decay time $\tau_{eff}(n)$, defined as the time in which photoluminescence intensity dropped to $1/e$ of its maximum value, was determined from the experimental time-resolved profiles. The $\tau_{eff}(n)$'s are plotted versus the excitation power fluence in Fig. 2. Each experimental point is the average of three to four shots. The scattering of data points around a given excitation power fluence was due to changes in the duration of the laser pulse and could not be controlled. Figure 3 shows the maximum intensity of the time-resolved photoluminescence profiles as a function of excitation power fluence. The instantaneous intensity of photoluminescence shown in Fig. 3 increased with excitation power fluence as $I_F \propto P_L^{(1\pm0.05)}$ when excitation power fluence was below 1.5×10^9 W/cm^2. When excitation power fluence increased beyond 1.5×10^9 W/cm^2 there was some deviation from linearity. The observed sublinearity of photo-

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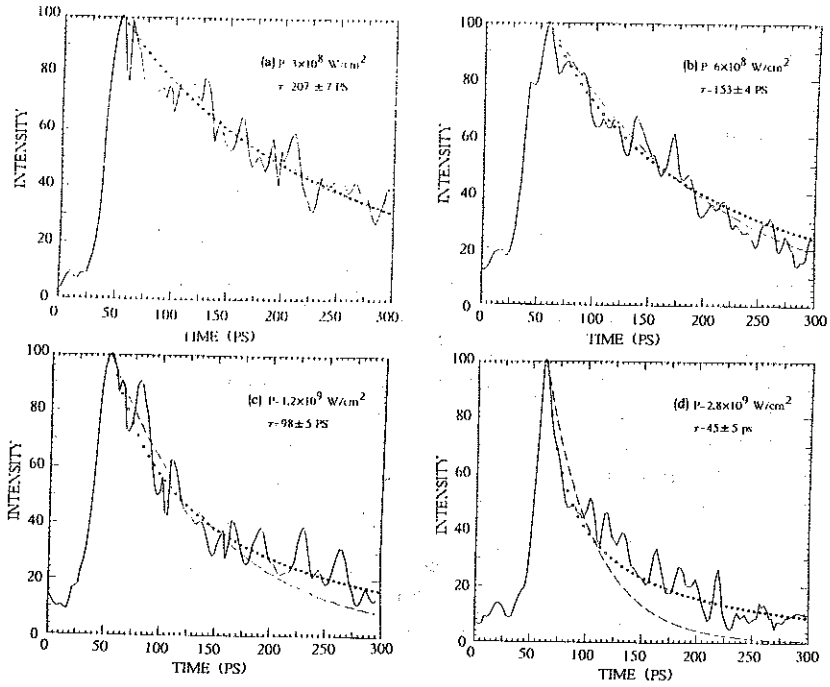

FIG. 1.(a)–(d) Time-resolved photoluminescence profiles of $GaAs_{1-x}P_x$ ($x = 0.38$) at room temperature for excitation powers of 3×10^8, 6×10^8, 1.2×10^9, and 2.8×10^9 W/cm^2, respectively. The solid lines are the experimental points, dashed lines are drawn assuming exponential model, and circles are calculated from model (2) for $A = 3.4\times10^9$/s, $B_R = 9\times10^{-10}$ cm^3/s, and $C_{NR} = 3\times10^{-29}$ cm^6/s.

luminescence intensity may be attributed to loss mechanisms at high carrier densities most likely arising from the nonradiative Auger process.

To analyze and explain the changes observed in the decay profile of the time-resolved photoluminescence of GaAsP at room temperature under different excitation power fluence, the following two models for rate equations have been developed. In model 1, the rate equation describing the time dependence of photogenerated carriers is given by

$$dn/dt = g(t) - A_{NR}n - A_R n - B_R n^2, \quad (1)$$

where $g(t)$ is the generation term which depends on the actual pulse shape of the exciting laser, A_{NR}, A_R, and B_R are nonradiative, radiative, and bimolecular recombination coefficients, respectively. The rate equation for the second model includes the Auger nonradiative rate and is given by

$$dn/dt = g(t) - A_{NR}n - A_R n - B_R n^2 - C_{NR}n^3, \quad (2)$$

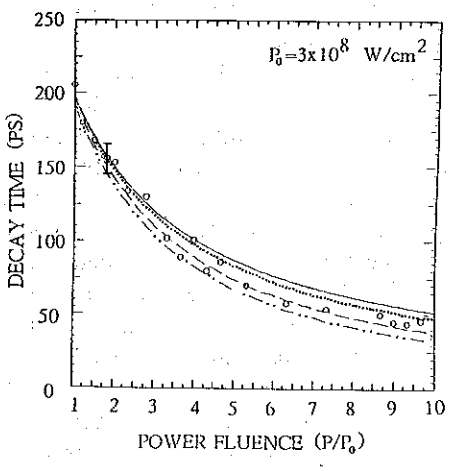

FIG. 2. Plot of $\tau_{eff}(n)$ vs the excitation power fluence. The circles are the experimental points, solid line is calculated from model (1) for $A = 3.4\times10^9$/s, $B_R = 9\times10^{-10}$ cm^3/s, and the dots, dashs, dot-dashs curves are calculated from model (2) for $C_{NR} = 1,3,5\times10^{-29}$ cm^6/s, respectively.

where C_{NR} is the nonradiative Auger rate. At low excitation power fluence where the density of photogenerated carrier is low and $(A_R n + A_{NR} n + B_R n^2) \gg C_{NR} n^3$, the two models will approach each other in the calculation of the decay time. The absence of any term representing the population of excitons is justified by the fact that at high carrier densities ($n > 5\times10^{17}$ cm^{-3}) the exciton state becomes unstable due to the screening of Coloumb interaction.[11] The Mott transition in $GaAs_{1-x}P_x$ ($x = 0.38$) at room temperature from the exciton state to the plasma state occurs at a carrier density of 3×10^{17} cm^{-3}. The photogenerated[12] carrier densities here are in the range of 3×10^{18}–3×10^{19} cm^{-3}.

Since we are concerned with the changes in the decay profile of the time-resolved photoluminescence, we set $g(t) = 0$ for $t > t_r$, where t_r is the rise time of the time-resolved profiles. The solution of model (1) becomes[13]

$$n(t) = \frac{A}{B_R}\frac{n(0)}{[A/B_R + n(0)]\exp(At) - n(0)} \quad \text{for } t > t_r, \quad (3)$$

where $A = A_R + A_{NR}$. The time for which the carrier density drops to $1/e$ of its initial value $\tau_{eff}(n)$ for the model (1) is given by

$$\tau_{eff}(n) = \frac{1}{A}\ln\frac{2.718 + B_R n(0)/A}{1 + B_R n(0)/A}. \quad (4)$$

In order to determine A and B_R the experimental data of Fig. 2 were fitted to Eq. (4) for excitation power fluence below 8×10^8 W/cm^2. The best fit was obtained for $A = 3.4\times10^9$ s^{-1} and $B_R = 9\times10^{-10}$ cm^3/s. Modesti *et al.* estimated the bimolecular recombination rate in $GaAs_{1-x}P_x$ ($x = 0.38$) to be 3.6×10^{-8} cm^3/s in terms of hydrodynamic expansion model.[14] This rate is 40 times larger than the rate we have determined. A simple calculation assuming only bimolecular recombination yields an expression for lifetime $\tau = 1/B_R$. If we use the $B_R = 3.6\times10^{-8}$ cm^3/s and the lifetime of 98 ps measured by a streak camera at $P = 1.2\times10^9$ W/cm^2 we see that the carrier density

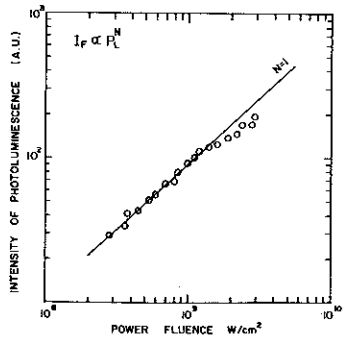

FIG. 3. Peak intensity of time-resolved photoluminescence profile vs the excitation power fluence.

should be around $3\times10^{17}\,\mathrm{cm}^{-3}$. This value is about 40 times smaller than the estimated carrier density of $1.3\times10^{19}\,\mathrm{cm}^{-3}$ at $P=1.2\times10^{9}\,\mathrm{W/cm}^2$, consequently Modesti's estimate of B_R does not agree with our results. Also the value of $B_R=9\times10^{-10}\,\mathrm{cm}^3/\mathrm{s}$ determined by us is within an order of magnitude of the values reported for GaAlAs[15] and GaInAsP.[16]

The radiative and nonradiative rates at low excitation power are related to the internal efficiency of a semiconductor by the relationship

$$\eta_{\mathrm{int}}=\tau_{\mathrm{NR}}/(\tau_{\mathrm{NR}}+\tau_R), \tag{5}$$

assuming the internal efficiency is independent of the density of photogenerated carriers. The external efficiency of this sample was measured to be 2×10^{-4} by comparing the area under low power steady state photoluminescence spectra with $Ga_{0.5}In_{0.5}P$.[17] Taking into account the geometry of the emission, a value of 7×10^{-2} was estimated for the internal efficiency of this smple.[18] Note that the decay times measured by the streak camera are the photoluminescence lifetimes and the relation between photoluminescence lifetime τ, radiative lifetime τ_R, and nonradiative lifetime τ_{NR} is given by

$$1/\tau=1/\tau_R+1/\tau_{\mathrm{NR}}. \tag{6}$$

Because Eq. (5) is true only at low excitation power and we have measured the external efficiency of our sample at low carrier density ($2\times10^{17}\,\mathrm{cm}^{-3}$) we have estimated τ for $n=2\times10^{17}\,\mathrm{cm}^{-3}$. Using Eq. (4) and the values of A and B_R obtained by fitting, a value of 285 ps has been calculated for τ at low carrier density. Since the photoluminescence lifetime was calculated to be 285 ps at low carrier density, a simple calculation using Eqs. (5) and (6) yields a value of 2.5×10^{8} and $3.25\times10^{9}\,\mathrm{s}^{-1}$ for the radiative ($A_R=1/\tau_R$) and nonradiative ($A_{\mathrm{NR}}=1/\tau_{\mathrm{NR}}$) rates, respectively. So far, we have determined all the recombination rates involved in model (1), therefore $\tau_{\mathrm{eff}}(n)$ for this model has been calculated from Eq. (4) and is shown in Fig. 2 by a solid line. This curve as expected fits the experimental points at low excitation power fluence but does not fit the experimental points at higher excitation power fluence (above $1.5\times10^{9}\,\mathrm{W/cm}^2$). This is due to the fact that at high carrier densities the Auger effect cannot be ignored. Clearly model (2) is a more complete model to describe the changes observed in the decay profile of photoluminescence in $GaAs_{1-x}P_x(x=0.38)$ at room temperature, particularly at high carrier densities.

Since C_{NR} has not been determined experimentally, it was treated as the only adjustable parameter to obtain the best fit between the experimental decay time and the theoretical value calculated numerically by computer based on model (2) at different excitation power fluences. The $\tau_{\mathrm{eff}}(n)$ for model (2) versus the excitation power fluence is shown in Fig. 2 by a dashed line for $C_{\mathrm{NR}}=3\times10^{-29}\,\mathrm{cm}^6/\mathrm{s}$ and it fits the experimental data well. The Auger rate of $3\times10^{-29}\,\mathrm{cm}^6/\mathrm{s}$ is larger than the rate in GaAs (Ref. 19) by a factor of 2 and is three times smaller than the rate estimated for InGaAsP.[16] In order to determine the uncertainty in the value of C_{NR}, $\tau_{\mathrm{eff}}(n)$ from model (2) has been calculated as a function of excitation power fluence for $C_{\mathrm{NR}}=(5\pm2)\times10^{-29}\,\mathrm{cm}^6/\mathrm{s}$. These curves are shown by dots and dot-dash in Fig. 2. In further support of the second model, each decay profile was calculated using the recombination rates that we have determined. The circles in the time-resolved photoluminescence profiles of Fig. 1 are the numerical solution of model (2) for $A_R=2.5\times10^{8}/\mathrm{s}$, $A_{\mathrm{NR}}=3.25\times10^{9}/\mathrm{s}$, $B_R=9\times10^{-10}\,\mathrm{cm}^3/\mathrm{s}$, and $C_{\mathrm{NR}}=3\times10^{-29}\,\mathrm{cm}^6/\mathrm{s}$. The theoretical curves fit all the time-resolved photoluminescence profiles well at different excitation power. The significance of model (2) was apparent at the highest excitation power [see Fig. 1(d)] where there was a significant deviation from an exponential form.

In conclusion, the transient behavior of photogenerated carriers by picosecond laser pulses in $GaAs_{1-x}P_x(x=0.38)$ at room temperature can be described in terms of a recombiantion model where the nonradiative Auger term played an important role. An Auger process is included in order to explain the changes observed in the decay profile of the time-resolved photoluminescence kinetics.

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