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Density dependence of electron-hole plasma lifetime in semiconductor quantum wells

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We report on an investigation of the electron-hole (*e-h*) plasma decay time τ_{tot} versus *e-h* pair density in semiconductor quantum wells. We determine the density reached in a steady-state photoluminescence experiment from the optical spectra and compare it with the *e-h* pair generation rate. We find that in our samples only radiative *e-h* recombination is important, and that nonradiative processes and plasma expansion have negligible effects on τ_{tot}. At plasma densities larger than 5×10^{12} cm^{-2}, we observe a strong nonlinear reduction of the *e-h* capture rate into the quantum wells.

In recent years, the physics of highly excited semiconductor quantum wells (QW) has been the subject of intense investigations.[1] Their nonlinear optical properties are among the most interesting features; the accumulation in real space of a large number of elementary excitations (excitons, biexcitons, free *e-h* pairs, etc.) is often the reason for such nonlinearities. In undoped samples, at high optical or electrical excitations, the *e-h* pairs form a dense neutral plasma. The carrier density $n(\mathbf{r},t)$ in space and time is determined by the microscopic processes of *e-h* pair recombination, trapping, spatial diffusion, drift, and under certain circumstances even ballistic particle motion. In bulk GaAs, nonradiative recombinations (Auger processes) have been found to compete with the radiative ones at high densities,[2] while in confined systems very little is known. However, neither in bulk nor in QW has a precise answer for the length over which the plasma effectively expands been found yet. On one hand, observations of plasma expansions with carrier speeds up to nearly sonic velocities have been reported.[3] On the other hand, models in which big parts of the *e-h* cloud are drifting with velocities (v_d) as high as 10^6–10^7 cm/s, i.e., of the same order or even higher than the Fermi velocity (v_F), have been proposed.[4,5] From different experiments made with different materials and under different conditions, sometimes contradicting conclusions have been drawn.

The different processes influencing the *e-h* pair density reached in a particular experiment have their characteristic dependence on density. Thus, investigating the decay time of the *e-h* plasma as a function of its density provides a quantitative understanding of the relative importance of each contribution. In a steady-state experiment the generation rate of photoexcited *e-h* pairs equates the losses due to spatial drift and to radiative and nonradiative recombinations. Since the carrier density and the relative rate of *e-h* pair generation can be determined quite accurately, measuring steady-state *e-h* pair density versus generation rate is a simple and powerful tool to investigate the microscopic mechanisms controlling plasma density. On the contrary, information of transient measurements (i.e., luminescence and/or Raman scattering intensity versus time) is rather entangled due to the dependence on time and space of recombination processes and plasma expansion.[6]

Let us mention that the nonlinear optical properties of strongly excited semiconductors are of crucial importance for the performance of optoelectronic devices. In particular, the presence of a dense *e-h* plasma strongly modifies the optical properties of a semiconductor near the band-gap energy. This has been investigated by several groups in order to determine the optimum operating conditions of semiconductor lasers.[7] It is thus important to be able to check in a simple way which are the processes influencing the properties of an *e-h* plasma.

In this paper we report on an experimental investigation of *e-h* pair decay time versus density, in *e-h* plasmas confined in one direction by a semiconductor QW. We show that steady-state measurements can effectively be employed to investigate plasma recombination times, which in turn strongly influence the optical nonlinearities. The most striking result of our work is that radiative recombination alone limits the photogenerated carrier density, only once the electron and hole states with the highest energies in the QW begin to be occupied, the Pauli exclusion principle reduces the electron and hole trapping rates into the wells. We show that neither Auger processes nor drift motions affect the *e-h* pair density. We present a simple way to calculate the radiative plasma lifetime, which fully accounts for the experimental data.

The experiment has been performed on a high-quality quantum structure consisting of six 122-Å-wide wells of GaAs, separated by 180-Å-wide barriers of $\mathrm{Ga_{0.77}Al_{0.23}As}$ alloy. Between this structure and the GaAs substrate there is a 1-μm-thick layer of $\mathrm{Ga_{0.77}Al_{0.23}As}$ alloy. The GaAs substrate was chemically etched away, in order to avoid superposition of the luminescence of the QW and that of the substrate. The *e-h* pairs have been excited either directly inside the wells by a pulsed infrared dye laser ($h\nu_{\rm ir} = 1.75$ eV, $\tau_{\rm ir} = 75$ ns), or with an energy larger than the gap of the barriers by a green frequency doubled Nd:YAG (yttrium aluminum garnet) laser ($h\nu_G = 2.33$ eV, $\tau_G = 50$ ns). The diameter (d) of the laser spot on the sample was 70 and 100 μm for the infrared and green pump, respectively. The temporal width of the laser pulse was sufficiently long to ensure that a steady state has been established. Usual pin-hole and boxcar techniques[8] have been used in order to make sure that only the spatially and temporally homogeneous part of the plasma is investigated. Since the energy loss rate of the carriers to the lattice increases with temperature, a relatively high lattice temperature ($T_L = 155$ K) was intentionally chosen to have a small difference between carrier temperature and T_L.

The luminescence spectra show (see Fig. 1) an important subband filling. The second electron and hole subbands start to be occupied at an excitation intensity of about 600 W/cm^2 absorbed *per* well. At the highest pumping rates also the third subbands are partially occupied. Despite this large population inversion, no evidence of stimulated emission is present in the spectra. Having a small number of wells and a relatively high temperature, the probability of optical amplification remains low. The light emission of the thick $\mathrm{Ga_{0.77}Al_{0.23}As}$ layer and/or the barriers is very weak at the lowest excitation intensities; it becomes progressively more important at the highest pump rates. The luminescence spectra excited by the red laser light show the same main features as those visible in Fig. 1, except that there is no spontaneous emission from the $\mathrm{Ga_{0.77}Al_{0.23}As}$ layers.

The rate equation for the e-h pair density n is

$$\begin{aligned}\frac{\partial n(\mathbf{r},t)}{\partial t} &= G(\mathbf{r},t) - \frac{n(\mathbf{r},t)}{\tau_r(\mathbf{r},t)} - \sum_i \frac{n(\mathbf{r},t)}{\tau_i} \\ &\quad - \nabla(n(\mathbf{r},t)\mathbf{v}(\mathbf{r},t)) \\ &\equiv G(\mathbf{r},t) - \frac{n(\mathbf{r},t)}{\tau_{\rm tot}(n,\mathbf{r},t)}, \end{aligned} \qquad (1)$$

where G is the e-h pair generation rate, $1/\tau_i$ are the nonradiative recombination probabilities, $\mathbf{v}$ is the mean carrier velocity, and in our definition the total decay rate $n/\tau_{\rm tot}$ accounts for both e-h pair recombination and spatial carrier migration. Under stationary conditions,

$$n(\mathbf{r},t) \simeq G(\mathbf{r},t)\tau_{\rm tot}(\mathbf{r},t). \qquad (2)$$

Thus by measuring G and n independently, for a given time t and a given spatial position $\mathbf{r}$, the density dependence of $\tau_{\rm tot}$ can directly be determined.

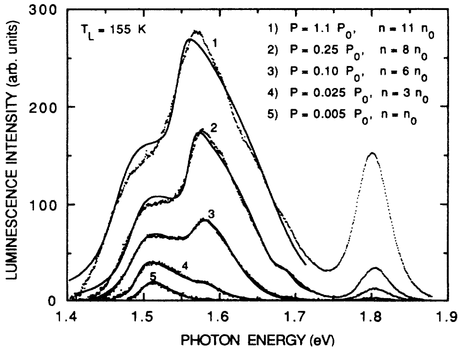

FIG. 1. Spontaneous photoluminescence spectra (dots) at different excitation intensities. Continuous curves represent fits to the experimental spectra. P is the absorbed excitation intensity per well, given in units of $P_0 = 40$ kW/cm^2. n is the e-h pair density, as obtained by the fits, $n_0 = 10^{12}\,\mathrm{cm}^{-2}$. The electron and lattice temperatures, $T_{e\text{-}h}$ and T_L, have been found to be practically equal, except for spectrum 1 where $T_{e\text{-}h} = 200$ K.

In this work, plasma density and temperature have been determined through a line-shape fit of luminescence spectra.[9] The spontaneous emission rate $r_s(h\nu)$ (Ref. 10) has been calculated using an approximation already used in Ref. 11 and discussed in Refs. 12 and 13,

$$\begin{aligned} r_s(h\nu) &\equiv \sum_{i,j} r_{ij}(h\nu) \\ &= \left(\frac{2\nu n_r}{c}\right)^2 \frac{2\pi e^2}{n_r c m^2 \nu} \\ &\quad \times \sum_{i,j} \int d^2\mathbf{k}\, |M_{ij}(\mathbf{k})|^2 A_{ij}(\mathbf{k},h\nu) \\ &\qquad \times f_e(E_i(\mathbf{k}))\, f_h(E_j(\mathbf{k})), \end{aligned} \qquad (3)$$

where $\sum_{i,j}$ runs over the optical transitions between the electron subbands i and the hole subbands j, n_r and m are the refractive index and the free electron mass, $M_{ij}(\mathbf{k})$ are the (momentum conserving) transition matrix elements, $\mathbf{k}$ is the two-dimensional (2D) wave vector of the carriers in the well plane, $E_i(\mathbf{k})$ and $E_j(\mathbf{k})$ are electron and hole energies, f_e and f_h are the electron and hole Fermi distribution functions, and $A_{ij}(\mathbf{k},h\nu)$ are the spectral functions of the e-h pair states describing collision broadening. Our model includes further a realistic description of the subband dispersion.[12] The excitonic enhancement[10] is negligible at the present plasma temperature ($T_{e\text{-}h} \sim 155$ K) and therefore it has been omitted. We find that the e-h plasma has nearly the same temperature as the lattice (within an uncertainty of about 10%), at all excitation wavelengths and all but the highest pump rates. This greatly simplifies our investigation of the e-h pair recombination rate versus carrier density.

As shown in Fig. 1, the plasma density n does not scale linearly with excitation intensity; as the pump rate is increased 100 times n rises only by a factor of 10. The exact dependence of the total lifetime $\tau_{\rm tot}$ versus plasma density is shown in Fig. 2. $\tau_{\rm tot}$ has been determined using Eq. (2) (steady state). To determine $\tau_{\rm tot}$ we have to know the generation rate $G = \alpha I/h\nu$, where α is the absorption coefficient and I is the excitation intensity. It is difficult to determine the absolute value of I; we know it within roughly a factor of 2. The relative excitation intensities however, as obtained by the variation of the exciting laser light using calibrated attenuators, are known with an accuracy of a few percent. Thus the density dependence of $\tau_{\rm tot}$ can be investigated with precision. For the infrared excitation, the fraction of the absorbed light per well, α, has been determined by transmission measurements. For an initial determination of $\tau_{\rm tot}$ (squares in Fig. 2),

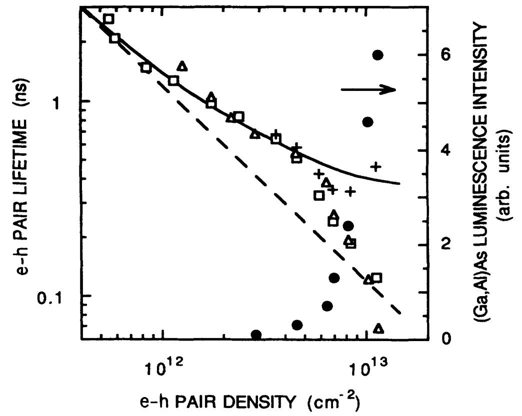

FIG. 2. Total and radiative lifetimes vs *e-h* pair density n. Squares and triangles are the measured total lifetimes τ_{tot} for plasmas excited by the infrared (ir) and green laser, respectively. To obtain the points denoted by pluses, the saturation of the absorption of the ir laser light has been accounted for. The full curve is the radiative lifetime $\tau_r(n)$ calculated using Eqs. (3) and (4); and the dashed curve is the low-density approximation, $\tau_r \simeq BT/n$, where a proportionality constant $B = 7.74$ s/(K cm^2) has been used. The black circles are the peak luminescence intensities $r_s(h\nu)$ emitted by the $Ga_{0.77}Al_{0.23}As$ barriers, plotted vs the *e-h* pair density in the wells.

α has been taken to be independent of density. For the green excitation, we supposed that the main part of the photoexcited *e-h* pairs falls down into the wells. Since I is known only within a factor of 2, and in order to be able to easily compare experimental with theoretical lifetimes, the former have been multiplied by a constant factor equal to 0.8.

From the experimental results shown in Fig. 2, we establish that the total *e-h* plasma lifetime is nearly independent of the excitation wavelength. An increase of the *e-h* pair density by somewhat more than a factor of 10 causes a reduction of τ_{tot} by about one order of magnitude. At a concentration of about 5×10^{12} cm^{-2} an evident change of slope in the experimental τ_{tot} versus n curve is present. Since temperature variations are negligibly small, all the observed changes in the slope of the $\tau_{\text{tot}}(n)$ are to be ascribed to subband filling.

The mean radiative *e-h* pair lifetime τ_r is given by

$$\frac{1}{\tau_r} \equiv \frac{1}{n} \int_0^{+\infty} r_s(h\nu)\, dh\nu = \frac{1}{n} \sum_{i,j} \int r_{ij}(h\nu) dh\nu, \qquad (4)$$

i.e., the integrated spontaneous emission rate divided by the *e-h* pair density.[10] It is worth noting that the radiative lifetime depends only weakly on the well thickness, through the optical matrix elements $M_{ij}(\mathbf{k})$ and the subband structure.

In the limit of highly degenerate *e-h* plasmas, electron and hole states with energies below the respective chemical potentials are occupied with a probability of $\simeq$ 1. Thus, in this limit and combining Eqs. (3) and (4), $r_s(h\nu)$ is found to be almost independent of energy and of the number of occupied subbands, because of the nearly constant and equal 2D density of states of each subband, because of the selection rules permitting practically only transitions between conduction- and valence-band states having the same wave vector and the same subband index, and because the matrix elements M_{ii} are nearly independent of i. In a QW the large valence subband interactions and the ensuing k dependence of the optical matrix element $M_{ij}(\mathbf{k})$ could suggest that this conclusion is not correct. However, summing over all the allowed and "forbidden" transitions contributing to the valence-to-conduction oscillator strength, a staircaselike optical density with flat plateaus is obtained, i.e., as if intersubband coupling were absent.[14] Therefore, we expect that at high carrier densities and low temperatures the radiative lifetime τ_r is practically independent of density. At lower densities, the electrons are still degenerate while the holes are not, due to the large hole to electron mass ratio. If the condition $1 < \mu_e/k_BT < m_h/m_e$ is satisfied, a simple analysis shows that $\tau_r \propto T/n$. $\tau_r(n)$, as obtained by using Eqs. (3) and (4), is plotted in Fig. 2. The calculated dependence of τ_r on density is completely smooth; there are no changes in the slope caused by the onsets of occupation in the second and third subbands. This is not surprising, since the recombination probability is almost independent on the subband index. The low-density approximation $\tau_r \propto T/n$ is also shown in Fig. 2; the deviation from the exact calculations becomes more and more pronounced as the density increases and the full degeneracy is approached. In this limit, the calculated saturation lifetime of about 0.4 ns agrees quite well with a recently reported value.[15]

The calculated radiative lifetimes agree well with the experimentally determined ones, at least for $n \leq 5 \times 10^{12}$ cm^{-2}. As far as the excitation with the infrared laser is concerned, once the third subbands are occupied the absorption of the laser light decreases because the occupation factor $[1 - f_e(E_e) - f_h(h\nu_{\text{ir}} - E_e)]$ becomes smaller than 1. This effect has been taken into account in calculating the generation rate G (plus symbols in Fig. 2). As for the *e-h* pairs excited by the green laser, after their creation they are captured into the QW by interaction with optical phonons and/or with the already thermalized particles in the wells. The Pauli exclusion principle diminishes the capture rate as the plasma density increases, due to the increasing occupation probability of the electron (hole) states with energies $E(k) \geq E_{\text{barrier}} - h\nu_{\text{LO}}$ ($h\nu_{\text{LO}}$ is the longitudinal-optical phonon energy $\simeq$ 36 meV). Subband structure calculations show that for the electrons these energies roughly coincide with the energies of the third subband.[16,17] A reduction of the *e-h* capture rate into the QW is therefore expected once the third subbands start to be occupied. In Fig. 1 the luminescence intensity $r_s(h\nu)$ of the bulk $Ga_{0.77}Al_{0.23}As$ layers is also visible. A strong nonlinear increase of this luminescence intensity is observed for $n \geq 5 \times 10^{12}$ cm^{-2}. At these densities the luminescence spectra of the QW show that the third subbands indeed are occupied, indicating that a saturation of the carrier capture rate into the QW occurs, which in turn implies an apparent decrease of τ_{tot}. Thus, we have to conclude that radiative *e-h* pair recombination alone accounts for the measured total plasma lifetime.

In bulk GaAs and GaAs-AlAs alloys, the probability for nonradiative Auger recombination, $1/\tau_{\rm nr}$, is large enough to influence the total *e-h* recombination time at high plasma densities.[2] We expect $1/\tau_{\rm nr} \propto n^m$, with $m \geq 2$. The density dependence of the lifetime $\tau_{\rm tot}$ we observed rules out that Auger processes play a significant role in QW. As far as we know, no detailed calculations exist in literature for 2D plasmas. In bulk GaAs Takeshima[18] calculated Auger recombination rates for *e-h* pair densities corresponding to the highest we reached in our experiment ($\sim 10^{19}/{\rm cm}^3$), he found $\tau_{\rm nr} \sim 10^{-8}$ s, i.e., one to two orders of magnitude longer than the lifetimes measured in the present work.

The diffusive motion of the particles leads to expansions (l_d) of the *e-h* pair cloud in the 10-μm range.[19] In our experiment with the infrared pump, the diameter d of the excitation spot is about 70 μm; from this point of view we expect that particle diffusion will have a small influence on our experimental results. On the other hand, however, d is comparable with the distance over which a particle having Fermi velocity v_F can travel during its collision time. As the *e-h* plasma density n increases from 0.6 to 11×10^{12} cm^{-2}, the electron Fermi velocity $v_F \propto n^{0.5}$ increases roughly from 2 to 9×10^7 cm/s. Assuming a mean electron velocity of $2v_F/3$, that half the electrons move away from the excited region of the QW, and that plasma expansion occurs on a 100 ps time scale as suggested by Tsen and Morkoc,[5] we find that l_d increases from about 7 μm at the lowest plasma density to 30 μm at the highest density. This means that under the mentioned assumptions, *e-h* plasma expansion lowers the carrier density by a factor which is increasing from 1.4 to 3.4, as the carrier density rises from its lowest to its highest value. Since the carrier temperature is not influenced by how the carriers are excited, this effect does not depend on the wavelength of excitation. Looking at the plus symbols in Fig. 2 which were obtained using the infrared laser, such a reduction with respect to theory is not observable. Concluding this paragraph, we observe that the noise on our experimental data could hide an expansion of the plasma volume by about 20%, corresponding to an upper limit of the expansion speed of $0.1v_F$.

In conclusion, we determined the total *e-h* plasma lifetime as a function of its density. The analysis of our experimental data shows that (a) the radiative decay is the predominant mechanism of *e-h* recombination in a plasma confined in a QW, i.e., $\tau_{\rm tot} \simeq \tau_r$, (b) the experimentally observed transition from a partially degenerate plasma ($\tau_r \propto T/n$) to a fully degenerate one ($\tau_r \simeq$ const) is reproduced quantitatively by theory, (c) the *e-h* plasma does not expand with a drift velocity comparable to the Fermi velocity, (d) once the energetically highest states of the QW start to be occupied, the rate of particle capture from the $\rm Ga_{0.77}Al_{0.23}As$ layers into the wells is strongly reduced.

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