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Recombination of correlated electron-hole pairs in two-dimensional semiconductors

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The Coulomb interaction of electrons and holes in a two-dimensional semiconductor is shown to affect radiative and nonradiative recombination processes strongly. Our detailed calculations based on a many-body theory give the Coulomb correlation factors for a wide range of carrier densities and temperatures. For radiative recombination, a continuous transition between an excitonlike and a free-carrier-like behavior is revealed, which strongly influences the temperature dependence of the radiative lifetime. Auger recombination is strongly enhanced and its kinetic behavior is modified to look like classical radiative recombination up to fairly high temperatures.

Recombination processes of electrons and holes in semiconductors are an important subject both from the basic physics point of view and for their implications on devices.[1] Radiative recombination processes give a direct measure of wave-function overlap, that is, of the band structure. Nonradiative processes, on the other hand, depend on the strength of the electron-phonon and electron-electron interactions.[2,3] With many devices based on two-dimensional semiconductor structures emerging, a fundamental understanding of recombination processes in such structures is urgently required.

Recombination of electrons and holes is commonly treated on the basis of a free-carrier picture, i.e., the electrons and holes are assumed to be noninteracting particles.[1–3] The limitations of this approximation become obvious when excitons are taken into consideration: The formation of bound electron-hole pairs is due to their often neglected Coulomb interaction. The behavior of the two extremes, excitons and free electrons and holes, is quite simple to see for their radiative recombination. The really existing intermediate case is not straightforward to assess, particularly for nonradiative recombination processes such as Auger recombination.

For conventional three-dimensional semiconductor systems, the impact of Coulomb correlation of electrons and holes on recombination processes has already been studied in some detail. To our knowledge, its importance was recognized for the first time by Schlangenotto, Maeder, and Gerlach[4] for radiative recombination in silicon, although their approach was limited to low carrier densities. Auger recombination in electron-hole droplets at low temperature was later found to be enhanced by electron-hole correlations.[5–8] More recently, radiative recombination,[9] band-to-band Auger recombination,[10] and nonradiative recombination via deep impurities[11] in bulk semiconductors in general and at arbitrary temperatures and carrier densities have been shown to be strongly enhanced by electron-hole correlations.

Two-dimensional semiconductor structures exhibit a strongly increased exciton binding energy compared to bulk semiconductors. In the ideal two-dimensional case, this increase amounts to a factor of 4.[12] It is commonly argued that due to the stronger binding of the excitons, the radiative recombination of electrons and holes is excitonic up to room temperature.[13] On the other hand, it is obvious that at high carrier densities where many-body effects like screening of the Coulomb interaction come into play, the electron-hole system must approach a free-carrier-like behavior. From an analysis of the kinetics of radiative recombination in quantum wells based on a simple rate-equation approach Ridley[14] found quite complicated kinetics and temperature dependencies of the radiative recombination. Recent experimental results on radiative recombination in $In_xGa_{1-x}As/InP$ quantum-well structures[15] have been successfully explained on this basis.

In this paper we present a consistent picture of radiative and nonradiative recombination processes in two-dimensional semiconductors, valid for all carrier densities and temperatures. It includes the correlation of electrons and holes and electrons and electrons, respectively, by their attractive or repulsive Coulomb interaction in calculating the recombination rates. The enhancement of the recombination rate is evaluated quantitatively from a quantum statistical theory of interacting pair states including free-carrier screening and phase-space blocking. The results provide new insight into the dynamics of carrier recombination in two-dimensional (2D) semiconductors and suggest a reanalysis of experimental data.

It has been clearly shown experimentally that for quantum-well structures the radiative recombination at low temperature is excitonic. From a kinetic point of view, this means that the recombination rate is simply proportional to the density of particles (excitons):

$$R^x_{\mathrm{rad}} = \frac{n_x}{\tau_x}, \tag{1}$$

where n_x is the exciton density and τ_x is their radiative lifetime. For free electrons and holes, on the other hand, the radiative rate is proportional to both densities n and p,

$$R^0_{\mathrm{rad}} = Bnp. \tag{2}$$

For the case of Auger recombination it is commonly assumed that electrons and holes can be treated as non-

interacting particles, although their Coulomb interaction mediates the Auger process.[16] The recombination rate is written as

$$R^0_{\text{Auger}} = c_p n p^2 \tag{3}$$

for an electron-hole-hole (ehh) process, with c_p being the Auger coefficient.

The correlation of electrons and holes due to their Coulomb interaction manifests itself in a modified probability density of the particles at zero interparticle separation. It is increased by the attractive Coulomb interaction for electron-hole pairs and decreased for electron-electron or hole-hole pairs due to their repulsive interaction. The correlation leads to an enhancement of the recombination rates due to a modified local carrier density,

$$R_{\text{rad}} = B g_{eh} n p, \tag{4}$$

$$R_{\text{Auger}} = c_p g_{ehh} n p^2, \tag{5}$$

expressed by the correlation factors g_{eh} and g_{ehh}.

The two-particle correlation factor actually contains the physical information about the interaction of the particles. We calculate it using the very definition of a correlation function for particles of species a, b (Ref. 17)

$$g_{ab} \equiv \frac{\langle|\psi(0)|^2\rangle}{n_a n_b} = \frac{\langle|\psi(0)|^2\rangle^{\text{corr}} + n_a^{\text{corr}} n_b^0 + n_a^0 n_b^{\text{corr}} + n_a^0 n_b^0}{n_a n_b}, \tag{6}$$

where $|\psi(0)|^2$ is averaged over all pair states.

The wave functions and densities of the interacting particles have to be calculated from a microscopic theory. This is done using a quantum statistical approach[18] already applied to correlation effects in bulk semiconductors,[10] which includes screening of the mutual Coulomb potential and Pauli blocking. For our two-dimensional system, we use the screened Coulomb potential in the form given by Stern and Howard,[19]

$$V(r) = \frac{e^2}{\epsilon} \int_0^\infty \frac{k}{k + k_s} J_0(kr) dk, \tag{7}$$

where k_s is the screening vector and J_0 is a Bessel function of order zero. The two-dimensional Schrödinger equation is transformed into Bessel space and solved using matrix techniques.[10] Numerical calculations have been carried out using the material parameters of the $In_xGa_{1-x}As/InP$ material system.[20] The numerical results were confirmed to be within a few percent of the exact result by varying the number of points in k space and the number of angular momentum states involved in the integrations. It must be noted that we use a strictly two-dimensional model system, which does not include any complications arising from the finite thickness of realized quasi-2D structures (quantum wells). This should be a reasonably good approximation for thin quantum wells with high barriers.

Our calculations give the electron-hole and hole-hole correlation factors as functions of carrier density and temperature. Results for the above-mentioned material system are given in Fig. 1 for three different temperatures. For the lowest temperature (20 K) the electron-hole correlation factor g_{eh} varies like $\frac{1}{n}$ in a good approximation for densities below 10^{-1} (in units of the exciton Bohr radius a_0). Introducing this into Eq. (4) yields $R_{\text{rad}} \propto n$, i.e., the result expected for a purely excitonic system. For densities higher than 1 the Coulomb interaction is completely screened and the correlation factor approaches unity.

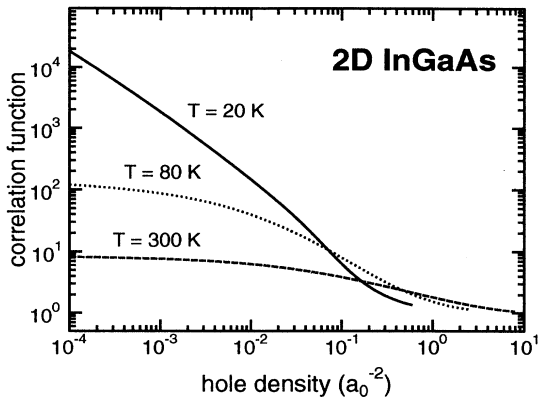

FIG. 1. Electron-hole and hole-hole correlation factors vs carrier density [in units of the exciton Bohr radius a_0 (227 Å for $In_xGa_{1-x}As/InP$)] for several different temperatures.

For higher temperatures, the slope of g_{eh} is smaller and is excitonlike only in a limited range of carrier densities. For very low carrier densities, we find a constant correlation factor which is considerably larger than unity. This is due to the fact that the probability of forming excitonlike states is proportional to np at low density. In the limit of high carrier density, the correlation factor again approaches unity. Consequently, radiative recombination behaves like expected classically [Eq. (2)] for very low and for very high carrier density from a kinetic point of view, but with a significantly enhanced rate at low carrier densities. In the intermediate density range there are deviations due to electron-hole correlation up to fairly high temperatures.

The temperature dependence of the electron-hole correlation factors is illustrated in Fig. 2 from a different

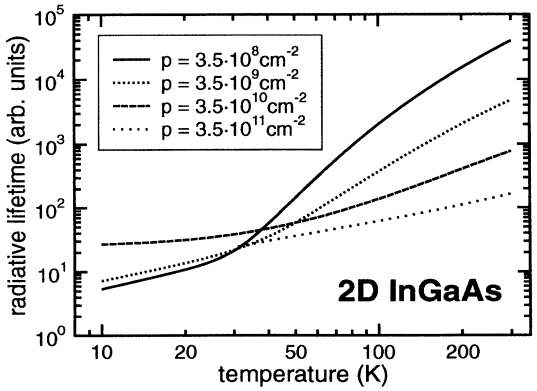

FIG. 2. Temperature dependence of the radiative lifetime in 2D $In_xGa_{1-x}As$ for a range of background carrier densities.

point of view. There we have plotted the radiative lifetime in 2D-$In_xGa_{1-x}As$ as a function of temperature for several values of the background hole density. For noninteracting electrons and holes, or for a purely excitonic system we would expect $\tau \propto T$, due to the steplike density of states in two dimensions. In fact, such a behavior is found in Fig. 2 for low temperatures, where excitons are only weakly ionized. At higher temperature, excitonic bound states get ionized and the radiative lifetime increases more rapidly, depending on carrier density. It is important to note that the slope of $\tau(T)$ can be much larger than unity. This behavior has only recently been observed experimentally.[15]

For Auger recombination, which is a three-particle process, we have to deal with a three-particle correlation function instead of the simple two-particle correlation. Chou and Wong have proposed to approximate the three-particle correlation function using the two-particle correlation functions according to[5]

$$g_{ehh} \simeq g_{eh}^2 g_{hh}. \tag{8}$$

However, there is a fundamental difficulty associated with this approach. Auger processes are a consequence of the electron-electron interaction, in our case the interaction between the two holes. Auger transition probabilities are usually calculated from Fermi's "golden rule" treating the electron-electron interaction as the perturbation. On the other hand, the naive approach corresponding to Eq. (8) implicitly assumes that the initial state of the transition corresponds to an eigenstate of the electron-electron interaction, which is in contradiction to using the same interaction as a perturbation.

We propose to overcome this difficulty by not implicitly including the interaction between the like particles (here, the holes) in the correlation function. In order to do this, we express the probability to find one electron and two holes at the origin as

$$w \propto \langle |\psi_{eh}(0)|^2 |\psi_{hx}(0)|^2 \rangle, \tag{9}$$

with $\psi_{eh}(0)$ being the wave function of an electron-hole pair and $\psi_{hx}(0)$ being the wave function of the second hole interacting with the electron quite like in a H^- ion. In principle one has to take into account all bound and all scattering states of the particles. In order to make it computationally feasible, we approximate the sum of products in Eq. (9) by the product of sums and write

$$g_{ehh} \simeq g_{eh} g_{hx}, \tag{10}$$

where g_{eh} is the electron-hole correlation function and g_{hx} describes the correlation of a hole with a correlated electron-hole pair ("exciton"). We note that this is exact if one particular state (e.g., a $1s$ state) dominates either of the correlation functions.

A rigorous calculation of g_{hx} would have to treat the interaction of the hole with all the excited states of the "exciton" separately. Instead we approximate the hole already "bound" to the electron as giving rise to an additional screening of the Coulomb potential of the electron with a screening parameter k_s^x. To calculate it we consider the case that the 2D Coulomb potential of the electron is screened by a bound hole in its lowest bound state with

$$\psi_{eh}(r) = \left(\frac{8}{\pi a^2}\right)^{1/2} e^{-2r/a}, \tag{11}$$

and a being the 3D excitonic Bohr radius. Using the procedure of Stern and Howard (see Appendix B of Ref. 19) for this case we find that the Bessel transform of the potential now reads

$$V(k) = \frac{e^2}{\epsilon} \frac{1}{k + k_s} \left[1 - \left(\frac{k_s^{x2}}{k^2 + k_s^{x2}}\right)^{3/2}\right], \tag{12}$$

where

$$k_s^x = 4/a. \tag{13}$$

Assuming that $1s$-like states dominate the *averaged* wave function it follows from a comparison of Eqs. (13) and (11) that the additional screening vector k_s^x due to the "bound" hole is

$$k_s^x = \sqrt{2\pi \langle |\psi_{eh}(0)|^2 \rangle}. \tag{14}$$

On this basis we can now calculate the hole-pair correlation function g_{hx} using the same numerical procedure as for g_{eh} and finally get the special three-particle (Auger) correlation function g_{ehh} according to Eq. (10).

Figure 3 shows a comparison of the Auger correlation functions for the 2D (quantum well) and for the 3D (bulk) case at room temperature. The most prominent feature is the huge enhancement of about a factor of 50 at low carrier densities for the 2D case. Nevertheless, this has only minor practical importance, even in the $In_xGa_{1-x}As$ material system, where the Auger coefficients are fairly large, since at such low carrier densities radiative recombination dominates over Auger recombination even when including the enhancement. More relevant is the steep slope of g_{ehh} in the 10^{17}–10^{18}-cm^{-3} density range. The maximum slope is around -1 which means that R_{Auger} varies only like n^2 instead of n^3, i.e., Auger recombi-

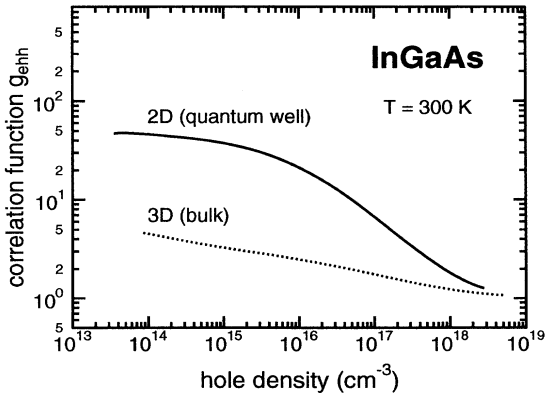

FIG. 3. Enhancement factor for electron-hole-hole (ehh) Auger processes vs carrier density in quantum well, 2D and bulk (3D) $In_xGa_{1-x}As$.

nation behaves like classical radiative recombination [cf. Eq. (2)]. This is in contrast to the 3D case, where the correlation function increases only slowly towards low carrier densities. As a consequence, the Auger coefficient in 2D systems, which is usually derived from experiments by extracting the n^3 component of the recombination rate,[21,22] may have been somewhat underestimated, particularly for thin quantum-well structures.

In conclusion, we have shown that electron-electron interactions in two-dimensional semiconductors can significantly enhance radiative as well as nonradiative recombination processes. The interplay between excitonic and free-carrier-like states changes the recombination kinetics and makes it much more difficult to analyze. For a basic understanding of recombination processes in two-dimensional semiconductors it is therefore essential to consider the many-particle nature of the recombination processes.

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