Enhancement of Band-to-Band Auger Recombination by Electron-Hole Correlations

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A huge enhancement of band-to-band Auger recombination in semiconductors by electron-hole correlations is found from a quantitative calculation. In contrast to earlier attempts, our calculation of the Coulomb correlation factors is valid for all temperatures and carrier densities, including the high-temperature and low-density case. Our results nicely explain recent carrier-lifetime measurements on silicon and show that Coulomb-enhanced Auger recombination poses an intrinsic upper limit to the carrier lifetime even at room temperature, a factor of 30 lower than previously assumed.

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Nonradiative 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] The study of nonradiative recombination processes reveals important information about electron-electron and electron-phonon interactions.[2,3] Device performance is limited by the carrier lifetime which can be increased by the avoidance of defects, but which is ultimately limited by intrinsic processes such as band-to-band Auger recombination.[4]

In nonradiative band-to-band Auger recombination, an electron recombines with a hole by transferring the excess energy to another electron or hole, which is highly excited into its respective band.[5] For silicon, it has been shown only recently,[6] using extensive numerical calculations, that the electron-electron-hole (*eeh*) Auger process is a direct process whereas the electron-hole-hole (*ehh*) process is likely to be phonon assisted.[7-9] Such Auger processes are traditionally described using the picture of noninteracting free particles.[3,5-8,10,11] For carrier densities higher than about 10^{18} cm^{-3} the experimental results reported up to now are in good agreement with these assumptions. For *n*-type silicon, where the *eeh* process dominates, the recent theoretical results for the direct Auger process[6] agree even quantitatively with the data of Dziewior and Schmid.[12]

At lower carrier density, however, the experimental carrier lifetimes[12-17] in silicon are significantly lower than expected from the Auger coefficients determined at high density,[12] even for ultrapure material. Interestingly, this is found for doped material[12-14,17] as well as for highly excited samples.[15,16] At least for doped samples, this is often interpreted as being due to contributions of defect recombination,[12,14] or even measurement errors. Other authors simply deduce a higher Auger coefficient for the low-density regime[13,15,16] without giving a physical explanation. To explain our own data, we have tentatively proposed an excitonic Auger process.[17]

In fact, electron-hole correlations have already been considered for Auger recombination in electron-hole droplets (EHD) at very low temperature.[18-21] The increased Auger coefficient in the EHD compared to high temperature,[19] as well as the variation of the EHD lifetime with uniaxial stress,[18,20,21] has been interpreted in this way. Takeshima[22,23] has included electron-hole interactions in a calculation of Auger transition rates, but did not consider excitonic bound states, which are very important at low carrier density. Furthermore, electron-hole correlation has been shown to be important for radiative recombination in silicon[24] and for nonradiative recombinations via deep impurities in silicon.[25,26]

In this Letter, we present a consistent picture of nonradiative band-to-band Auger recombination valid for all carrier densities and temperatures. It is based on the inclusion of the Coulomb attraction and repulsion, respectively, of electrons and holes and of electrons and electrons in calculating the Auger rate. The enhancement of the recombination rate is evaluated quantitatively by calculating the Coulomb correlation factors from a quantum statistical theory of interacting pair states including free-carrier screening and phase-space blocking. The results explain the experimental data for the whole range of carrier densities and temperatures.

From a kinetic point of view, the band-to-band Auger recombination rate R^0 for noninteracting particles is given by[3] (for simplicity, let us consider only the *eeh* process, where c_n is the Auger coefficient)

$$R^0_{\text{Auger}} = c_n n^2 p \,, \tag{1}$$

reflecting the fact that two electrons and one hole are involved in the transition. This expression is exact in the limit of Boltzmann statistics[3] but has been shown to be approximately true for Fermi statistics as well.[8] For noninteracting particles, electrons and holes are distributed uniformly in space. However, since electrons and holes interact via their Coulomb interaction, the electron density in the vicinity of a hole is increased while it is decreased in the vicinity of another electron. In other words, the particles are no longer uniformly distributed in space. Since the Auger recombination rate strongly depends on the particle density, this nonuniform distribu-

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tion has to be included by writing[18]

$$R_{\text{Auger}}=g_{eeh}R^0_{\text{Auger}}=g_{eeh}c_n n^2 p \simeq g^2_{eh}g_{ee}c_n n^2 p\,, \qquad (2)$$

where g_{eeh} is the Coulomb enhancement factor, giving the enhancement of the probability of finding two electrons and one hole at the same position due to their Coulomb interaction. As an approximation,[18] it is factorized into the two-particle correlation factors g_{eh} and g_{ee}.

Previous theoretical work on electron-hole and electron-electron correlation factors has been somewhat limited. Vashishta, Battacharyya, and Singwi[26] have calculated g_{eh} and g_{ee} for the case of the electron-hole liquid, i.e., in the high-density and low-temperature limit, by means of many-particle plasma theories. Schmidt and Röpke[27] have calculated the electron-hole correlation function using a Green's-function approach, but have limited themselves to a crude model potential.

We have calculated the electron-hole and electron-electron correlation factors using a more realistic interaction potential and taking into account details of the host band structure. By definition,[28] the two-particle correlation factors are calculated from the thermal average of $|\psi(0)|^2$ of the pair states

$$g_{ab}=\frac{\langle|\psi_{ab}(0)|^2\rangle}{n_a n_b}=\frac{\langle|\psi_{ab}(0)|^2\rangle_{\text{corr}}+n^0_a n^0_b}{n_a n_b}\,, \qquad (3)$$

where a,b stands for the particle species, n_a,n_b for the total density of particles of species a,b, and n^0_a,n^0_b for the free-particle densities.

The total particle densities n_a,n_b are calculated following the quantum statistical theory of Stolz and Zimmermann,[29] based on a Green's-function approach, where they are decomposed into contributions of correlated particles n_a^{corr} and free particles n_a^0,

$$n_a=n_a^0+n_a^{\text{corr}}\,, \qquad (4)$$

with

$$n_a^{\text{corr}}=\frac{2}{V_0}\sum_{q,b}\left[\sum_\alpha g(E_{\alpha ab}(q))\{2-\delta_{ab}e^{i\phi}\}-\sum_k g(\epsilon_a+\epsilon_b)\{2-\delta_{ab}\delta_{k+q,-k}\}\right],$$

where $E_{\alpha ab}(q)$ are the pair energy eigenvalues, ϵ_a and ϵ_b are the single-particle energies, q is the center-of-mass momentum, and g is the Bose distribution function.

The thermal average of $|\psi(0)|^2$ is calculated analogously using

$$\langle|\psi(0)|^2\rangle_{\text{corr}}=\frac{2}{V_0}\sum_{q,b}\left[\sum_\alpha g(E_{\alpha ab}(q))\{2-\delta_{ab}e^{-i\phi}\}|\psi^\alpha_{ab}(0)|^2-\sum_k g(\epsilon_a+\epsilon_b)\{2-\delta_{ab}\delta_{k+q,-k}\}|\psi^0_{ab}(0)|^2\right]. \qquad (5)$$

The energies and wave functions of the pair states are calculated numerically from the eigenvalue problem[29]

$$\{\epsilon_a(k+q)+\epsilon_b(-k)-E_{\alpha ab}(q)\}\psi^\alpha_{ab}(k,q)+s_a s_b\sum_{k'}V_s(k-k')\{1-f_a(k'+q)-f_b(k')\}\psi^\alpha_{ab}(k',q)=0\,, \qquad (6)$$

where s_a,s_b are the signs of the Coulomb potentials, V_s is the screened Coulomb potential, and f_a,f_b are the Fermi distribution functions for species a,b. For the screened Coulomb potential, we use static Thomas-Fermi screening[29] by free carriers only. Additional contributions to the screening by bound states and by scattering states are of second order[29] and will be neglected here.

The eigenvalue problem was solved numerically using a procedure described previously,[25] and the convergence of the sums in Eqs. (4) and (5) was checked carefully. The material parameters entering the calculations are the effective masses (optical and density-of-states masses)[30] and the excitonic Rydberg R_{ex} and Bohr radius a_0,[31] all of which are temperature dependent.[32]

Our calculations give the pair correlation factors as functions of carrier density and temperature. Figure 1 shows the results for the case of electron-hole pairs and electron-electron pairs in an electron gas (n-type silicon) at a temperature of 300 K. Taking into account the temperature dependence of the effective masses, this temperature corresponds to $1.4R_{\text{ex}}$. It is instructive to analyze this result in some detail. The electron-hole correlation factor can be split up into contributions of bound states (i.e., excitons) and of scattering states. The bound-state contribution vanishes at the metal-insulator transition[33] (Mott density) since there are no more bound states at higher density. This discontinuity is exactly compensated by the scattering part, so that the total correlation factor remains continuous. The physical reason is that the bound states do not really disappear but rather merge into the continuum. The electron-electron correlation factor of course has only a scattering part which is completely continuous with only a slight bump when the electron gas becomes degenerate.

Towards low carrier density, both correlation factors approach a constant value, determined by thermal equilibrium among the pair states, corresponding to the unscreened limit. It is interesting to note that even for such a high temperature, the electron-hole correlation factor in silicon is dominated by bound excitonic states.

Using the results discussed above, we are now able to calculate the Auger enhancement factor g_{eeh} using Eq. (2). On the other hand, in order to allow for a comparison with experimental results, we have evaluated g_{eeh} from our experimental data obtained from carrier-lifetime measurements on doped silicon,[17] by assuming that all deviations from classical Auger recombination according to (1) are due to the Coulomb enhancement. This corresponds to inverting Eq. (2), obtaining

$$g^{\text{expt}}_{eeh}=R^{\text{expt.}}_{\text{Auger}}/c_n n^2 p\,, \qquad (7)$$

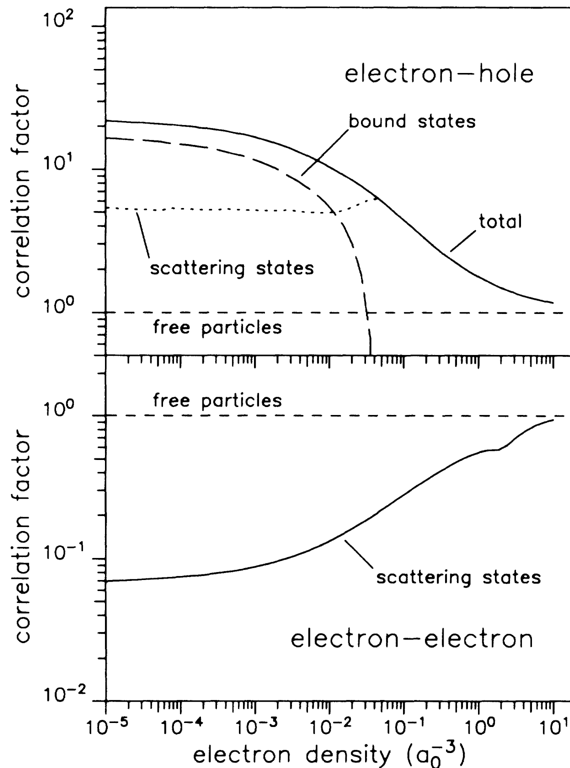

FIG. 1. Calculated electron-hole and electron-electron correlation factors for *n*-type silicon at $T=300$ K. The electron density is in units of the exciton Bohr radius.

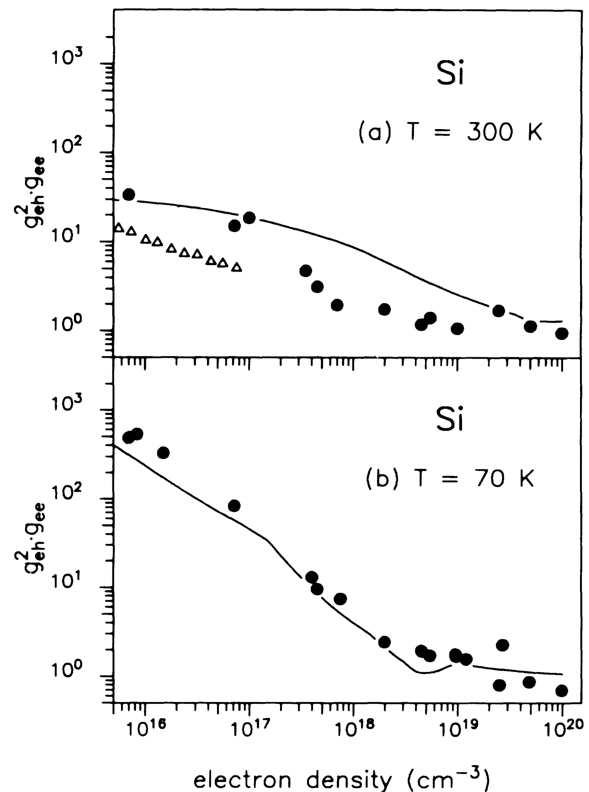

FIG. 2. Enhancement of Auger recombination in silicon at (a) $T=300$ K and (b) $T=70$ K. The experimental values are derived from Ref. 17 (circles) and from Ref. 15 (triangles). The theoretical curves (solid lines) have been calculated as described in the text.

where for c_n the values determined by Dziewior and Schmid[12] at high carrier densities have been used. We prefer this procedure over a comparison of the Auger lifetimes because small details are more clearly seen in this way.

The results are shown in Fig. 2(a) for $T=300$ K and in Fig. 2(b) for $T=70$ K. At room temperature (300 K) the theory is in qualitative agreement with experimental data derived from Ref. 17 (circles). The theory gives the correct result in the high- and low-density limits, but seems to be shifted towards high density compared to the experimental values. Qualitatively, the experimental results of Yablonovitch and Gmitter[15] (triangles) are also consistent with our theory although these are not strictly comparable because of the high excitation conditions used in those measurements. Under high excitation, the electron and hole densities are equal, leading to stronger screening and to a difference in the total particle densities.

At low temperature (70 K), where the Coulomb correlations are much stronger, the agreement between our theory and the experimental data[17] is almost quantitative. To appreciate this, one has to keep in mind that the calculations were performed without any adjustable parameter, using only well known material parameters. The deviations observed at room temperature might be caused by a change in the exciton Bohr radius, not reflected in the known temperature dependence of the density-of-states masses. Possibly, this is caused by the broadening of the excitonic states due to electron-phonon coupling which is particularly important at high temperature.

Our results clearly demonstrate that Auger recombination in silicon is strongly enhanced by electron-hole correlations at low carrier density. Even at room temperature the enhancement factor is about 30, increasing exponentially towards lower temperature. As a consequence, the intrinsic upper limit of the carrier lifetime is much lower than previously assumed. Our theory provides a consistent picture of nonradiative recombination in pure silicon, reconciling several recent experimental results with the idea of Auger recombination. The Coulomb enhancement is particularly large in silicon, because of its large exciton binding energy, but may be appreciable in other materials as well.

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