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Theory of recombination time in electron-hole drops in indirect-band-gap semiconductors

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We start with a Hamiltonian containing electron-hole Coulomb, electron (hole)-phonon, and electron-hole photon interactions to derive effectively renormalized interactions appropriate to recombination in the indirect-band-gap semiconductors with the use of canonical transformations. It is found that a phonon-induced Auger process and a radiative process involving electron-hole-phonon-photon interaction leads to relaxation in the indirect-band-gap semiconductors. The electron-hole pair self-energy for these processes is calculated. The imaginary part of the self-energy which leads to the relaxation time is deduced for these processes for optical as well as acoustic phonons. The theory is applied to the electron-hole drop recombination relaxation in Ge. The predicted relaxation times for the optical phonons which dominate in Ge for both the radiative process and the phonon-induced Auger process are in reasonable agreement with the experimental values.

I. INTRODUCTION

Extensive studies of the electron-hole drop (EHD) luminescence have been carried out in the last decade.¹⁻³ However, there has been no complete theory of the relaxation times and mechanisms of recombination. In indirect-band-gap semiconductors, the electrons and holes are separated in the momentum space by a wave vector too large to be supplied by the photon. Therefore, the *e*-*h* recombination can occur only through the participation of a phonon which provides the correct momentum. The experimentally measured relaxation rate is a sum of two major contributions,

$$1/\tau_0 = 1/\tau_r + 1/\tau_A \quad . \tag{1.1}$$

Although the nonradiative Auger lifetime τ_A has been a subject of several preliminary theoretical⁴⁻¹¹ and experimental¹²⁻¹⁵ studies, there are no microscopic calculations of the radiative relaxation time τ_r . The experimental measurements of τ_0 indicate that a theory of the radiative relaxation time is much needed.

A phenomenological description of the radiative decay rate has been given by Rice¹ according to which,

$$\frac{1}{\tau_r} = \frac{2\pi}{\hbar} \sum_{\substack{k_e k_h jj' \\ k_e k_h jj'}} |M(k_e j, k_h j')|^2 f(\epsilon_e) f(\epsilon_h) \\ \times \delta[\epsilon_e(k_e, j) + \epsilon_h(k_h, j') \\ + E_g - h \nu - \hbar \omega_p(k_e - k_h + K_{jj'})] ,$$
(1.2)

where the energies and momenta are measured from

the band extrema; the label j includes spin and orbital quantum numbers, $f(\epsilon_e)$ and $f(\epsilon_h)$ are the Fermi functions, $M(k_ej,k_hj')$ is the transition amplitude for an electron of wave number k_e and hole of wave number k_h to recombine yielding a phonon of a given band (LA, TO, etc.) and polarization and a photon of wave number $\lambda \approx 0$, $\epsilon_e(k_ej)$ and $\epsilon_h(k_hj')$ are the energies of an electron and a hole of wave k_e and k_h , respectively, $h\nu$ is the energy of the emitted photon, and $\hbar\omega_p$ is the energy of the emitted phonon.

The usefulness of the transition amplitude $M(k_e,j,k_h,j')$ has been realized in a number of studies,¹⁶⁻²² but this coefficient has not been calculated to date. Hammond and others¹⁶⁻²¹ have suggested a procedure to fit the line shape of the EHD luminescence. However, Rostworowski and Bergersen²² have noted that the momentum dependence of the argument of the energy δ function has not been properly taken into account. They have suggested²² an approximate procedure to include this wave-vector dependence. Under these circumstances it appears to us that the process of the recombination through a phonon and a photon is not well understood. We therefore develop a microscopic theory of the recombination mechanisms in electron-hole drops.

In this paper we calculate both the radiative and nonradiative Auger relaxation times from the first principles. We formulate the Hamiltonian in Sec. II by performing a canonical transformation on the electron-photon-hole, electron-phonon, hole-phonon, and the electron-hole Coulomb interactions. We interpret the various terms arising in the transformed interaction and distinguish between the scattering and the recombination mechanisms. In Sec. III, the contribution of the recombination terms to the selfenergy of the e-h pair is calculated for all the per-

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tinent terms. Section IV contains a calculation of the phonon-assisted Auger and the radiative relaxation times, which are the dominant mechanisms of recombination in the indirect-band-gap materials, as there has been considerable interest in such systems. In Sec. V we discuss the enhancement of the relaxation rates by the correlations in the liquid state and in Sec. VI we obtain numerical estimates for the relaxation times in Ge in reasonable agreement with the experimental measurements.

II. HAMILTONIAN

We start with the unperturbed Hamiltonian

$$\mathcal{K}_{0} = \sum_{k} \epsilon_{k}^{e} c_{k}^{\dagger} c_{k} + \sum_{k} \epsilon_{k}^{h} b_{k}^{\dagger} b_{k} + \sum_{p} \hbar \omega_{p} \beta_{p}^{\dagger} \beta_{p} + \sum_{\lambda} \hbar \Omega_{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\lambda}$$

$$(2.1)$$

For electrons the creation and annihilation operators are c_k^+, c_k , respectively, for holes b_k^+, b_k , the phonons β_p^+, β_p , and the photons $\alpha_{\lambda}^+, \alpha_{\lambda}$. The electrons and holes being in the conduction and the valence bands, respectively, the single-particle energies are of the form

$$\epsilon_k^h = \hbar^2 k^2 / 2m_h, \quad \epsilon_k^e = E_e + \hbar^2 k^2 / 2m_e \quad , \tag{2.2}$$

where E_g is the gap energy in the condensed state. This energy requires the knowledge of the modification of the Fermi energies in the liquid state as compared with the free excitonic state. The interaction of the electrons and holes with the phonons is of the form

$$\mathcal{K}_{ehp} = i \sum_{kp} D_p (c^{\dagger}_{k+p} c_k \beta_p + b^{\dagger}_{k+p} b_k \beta_p) - \text{H.c.} , \qquad (2.3)$$

where H.c. stands for the Hermitian conjugate of the

previous terms and the coupling constant D_p depends on whether the acoustic or the optical phonons are being considered as we shall see later. The interaction of photons is given by

$$\Im C_{ehr} = i \sum_{k\lambda} G_{\lambda} c_{k+\lambda}^{\dagger} b_{-k}^{\dagger} \alpha_{\lambda} - \text{H.c.} , \qquad (2.4)$$

where the matrix element is given by Hanamura²³

$$G_{\lambda} = g_{\lambda} V^{-1/2}, \quad g_{\lambda} = (\hbar e/\mu c) (\hbar \Omega_{\lambda}/\kappa)^{1/2} \quad , \quad (2.5)$$

where μ is the reduced mass of the *e*-*h* pair. The Coulomb interaction between the electrons and the holes is of the form^{24, 25}

$$3C_{c} = \sum_{kk'q} \left[V_{q} \left(c_{k+q}^{\dagger} c_{k'-q}^{\dagger} c_{k'} c_{k} + b_{k+q}^{\dagger} b_{k'-q}^{\dagger} b_{k'} b_{k} \right) - 2 V_{q}' c_{k+q}^{\dagger} b_{k'-q}^{\dagger} b_{k'} c_{k} \right] .$$
(2.6)

Thus the Hamiltonain of our problem is given by

$$\mathcal{K} = \mathcal{K}_0 + \mathcal{K}_{ehp} + \mathcal{K}_{ehr} + \mathcal{K}_c \quad . \tag{2.7}$$

We seek a transformed Hamiltonian which displays the desired interactions explicitly through the canonical transformation on Eq. (2.7) such that

$$\tilde{\mathcal{K}} = e^{-S} \mathcal{K} e^{S} = \mathcal{K}_{0} + \mathcal{K}_{ehp} + \mathcal{K}_{ehr} + \mathcal{K}_{c} + [\mathcal{K}_{0}, S] + [\mathcal{K}_{ehp} + \mathcal{K}_{ehr} + \mathcal{K}_{c}, S] + \frac{1}{2} [\mathcal{K}_{0}, [\mathcal{K}_{0}, S]] + \cdots$$
(2.8)

and find S by setting

$$\Im \mathcal{C}_{c} + \Im \mathcal{C}_{ehv} + \Im \mathcal{C}_{ehr} + [\Im \mathcal{C}_{0}, S] = 0$$

so that $\mathcal{K}_{ehp} + \mathcal{K}_{ehr} + \mathcal{K}_c$ are eliminated through

$$S = i \sum_{kp} D_p [(\epsilon_k^e - \epsilon_{k+p}^e + \hbar \omega_p)^{-1} c_{p+k}^{\dagger} c_k \beta_p + (\epsilon_k^h - \epsilon_{k+p}^h + \hbar \omega_p)^{-1} b_{k+p}^{\dagger} b_k \beta_p] + i \sum_{k\lambda} G_{\lambda} (\hbar \Omega_{\lambda} - \epsilon_{k+\lambda}^e - \epsilon_{-k}^h)^{-1} c_{k+\lambda}^{\dagger} b_{-k}^{\dagger} \alpha_{\lambda} + \sum_{kk'q} \{ V_q [(\epsilon_k^e + \epsilon_{k'}^e - \epsilon_{k'-q}^e - \epsilon_{k+q}^e)^{-1} c_{k+q}^{\dagger} c_{k'-q}^{\dagger} c_{k'} c_k + (\epsilon_k^h + \epsilon_{k'}^h - \epsilon_{k+q}^h - \epsilon_{k'-q}^h)^{-1} b_{k+q}^{\dagger} b_{k'-q}^{\dagger} b_{k'} b_k] - 2 V_q' (\epsilon_{k'}^h - \epsilon_{k'-q}^h + \epsilon_{k}^e - \epsilon_{k+q}^e)^{-1} c_{k+q}^{\dagger} b_{k'-q}^{\dagger} b_{k'-q} b_{k'} c_k \} + \text{H.c.}$$

$$(2.9)$$

The transformed Hamiltonian now occurs in the form

$$\tilde{\mathfrak{K}} = e^{-S} \mathfrak{K} e^{S} = \mathfrak{K}_{0} + \frac{1}{2} [\mathfrak{K}_{ehp} + \mathfrak{K}_{ehr} + \mathfrak{K}_{c}, S] , \qquad (2.10)$$

in which the terms can be grouped as follows:

$$\mathfrak{K} = \mathfrak{K}_{0} + \mathfrak{K}_{ehs} + \mathfrak{K}_{ps} + \mathfrak{K}_{A} + \mathfrak{K}_{pA} + \mathfrak{K}_{pps} + \mathfrak{K}_{r} + \mathfrak{K}_{rp} + \mathfrak{K}_{rrs} + \mathfrak{K}_{d} , \qquad (2.11)$$

where

$$\mathcal{K}_{0} = \sum_{k} \epsilon_{k}^{e'} c_{k}^{\dagger} c_{k} + \sum_{k} \epsilon_{k}^{h'} b_{k}^{\dagger} b_{k} + \sum_{p} \hbar \omega_{p} \beta_{p}^{\dagger} \beta_{p} + \sum_{\lambda} \hbar \Omega_{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\lambda} \quad , \qquad (2.12)$$

in which the single-particle energies are slightly renormalized as

$$\epsilon_k^{e'} = \epsilon_k^e - \sum_p D_p^2 (\epsilon_k^e - \epsilon_{k+p}^e + \hbar \omega_p)^{-1} - \sum_{\lambda} G_{\lambda}^2 (\epsilon_{-k}^h + \epsilon_{k+\lambda}^e - \hbar \Omega_{\lambda})^{-1} , \qquad (2.13)$$

$$\epsilon_k^{h'} = \epsilon_k^h - \sum_p D_p^2 (\epsilon_k^h - \epsilon_{k+p}^h + \hbar \omega_p)^{-1} - \sum_{\lambda} G_{\lambda}^2 (\epsilon_{-k}^h + \epsilon_{k+\lambda}^e - \hbar \Omega_{\lambda})^{-1} \quad .$$
(2.14)

In these expressions the second term determines the change in the single-particle energy due to the electronphonon interaction whereas the third term is the change due to the electromagnetic wave. This change in the electron wavelength due to the interaction with photons is called the Kapitza-Dirac effect²⁶ which is believed to be small. These energy shifts need not be considered further. The various terms in the transformed Hamiltonian are as follows:

$$\mathfrak{K}_{ehs} = -\sum_{kk'p} D_p^2 [(\epsilon_k^e - \epsilon_{k+p}^e + \hbar\omega_p)^{-1} c_k^{\dagger} c_{k'+p}^{\dagger} c_{k'} c_{k+p} + (\epsilon_k^h - \epsilon_{k+p}^h + \hbar\omega_p)^{-1} b_k^{\dagger} b_{k'+p}^{\dagger} b_{k'} b_{k+p}] + \mathrm{H.c.}$$
(2.15)

describes the scattering of electrons or holes within the same band. This gives rise to processes which do not contribute to recombination in low orders. The interaction can be either attractive or repulsive depending on the sign of the energy denominator. Thus Eq. (2.15) is of interest for the problem of electron-electron and hole-hole pairing mediated by phonons. This effect has been well studied in the theories of superconductivity and on such possibilities in the EHD but is not of interest for the present problem. The terms

$$\mathcal{K}_{ps} = i \sum_{kk'qp} 2D_p V_q \{ [(\epsilon_{k'}^{e} + \epsilon_{k}^{e} - \epsilon_{k+q}^{e} - \epsilon_{k'-q}^{e})^{-1} - (\epsilon_{k'}^{e} + \epsilon_{k+p}^{e} - \epsilon_{k'-q}^{e} - \epsilon_{k+q+p}^{e})^{-1} + (\epsilon_{k}^{e} - \epsilon_{k+p}^{e} + \hbar\omega_{p})^{-1} - (\epsilon_{k+q}^{e} - \epsilon_{k+q+p}^{e} + \hbar\omega_{p})^{-1}] c_{k+q+p}^{\dagger} c_{k'-q}^{\dagger} c_{k'} c_{k} \beta_{p} + [(\epsilon_{k}^{h} + \epsilon_{k'}^{h} - \epsilon_{k'-q}^{h} - \epsilon_{k+q}^{h})^{-1} - (\epsilon_{k+p}^{h} + \epsilon_{k'}^{h} - \epsilon_{k'-q}^{h} - \epsilon_{k+q+p}^{h})^{-1} + (\epsilon_{k}^{h} - \epsilon_{k+p}^{h} + \hbar\omega_{p})^{-1} - (\epsilon_{k+q}^{h} - \epsilon_{k+q+p}^{h} + \hbar\omega_{p})^{-1}] b_{k+q+p}^{\dagger} b_{k'-q}^{\dagger} b_{k'} b_{k} \beta_{p} \} + \text{H.c.}$$
(2.16)

and

$$\mathcal{K}_{pps} = -\sum_{kpp'} D_p D_{p'} \{ [(\epsilon_{k-p}^e - \epsilon_k^e + \hbar \omega_p)^{-1} - (\epsilon_{k+p'-p}^e - \epsilon_{k+p'}^e + \hbar \omega_p)^{-1}] c_{k+p'-p}^{\dagger} c_k \beta_p^{\dagger} \beta_{p'} + [(\epsilon_{k-p}^h - \epsilon_k^h + \hbar \omega_p)^{-1} - (\epsilon_{k+p'-p}^h - \epsilon_{k+p'}^h + \hbar \omega_p)^{-1}] b_{k-p+p'}^{\dagger} b_k \beta_p^{\dagger} \beta_{p'} \} + \text{H.c.}$$
(2.17)

give the intraband scattering of electrons or holes accompanied by the scattering of one [Eq. (2.16)] or two [Eq. (2.17)] phonons. The scattering process accompanied by the emission of one photon and the absorption of another photon is given by

$$\mathfrak{K}_{rrs} = \sum_{k\lambda\lambda'} G_{\lambda} G_{\lambda'} [(\hbar \Omega_{\lambda} - \epsilon^{h}_{-k} - \epsilon^{e}_{k+\lambda})^{-1} c^{\dagger}_{k+\lambda'} c_{k+\lambda} \alpha_{\lambda'} \alpha^{\dagger}_{\lambda} + (\hbar \Omega_{\lambda} - \epsilon^{h}_{\lambda-k} - \epsilon^{e}_{k})^{-1} b^{\dagger}_{\lambda'-k} b_{\lambda-k} \alpha_{\lambda'} \alpha^{\dagger}_{\lambda}] + \mathrm{H.c.}$$
(2.18)

Since these terms describe scattering within the same band, these do not contribute to the recombination relaxation times and may hence forth be ignored for the present problem.

The terms which describe relaxation are

$$\mathcal{K}_{d} = i \sum_{kq\lambda} 2G_{\lambda} V_{q}' [(\epsilon_{k+q+\lambda}^{e} - \epsilon_{\lambda-k}^{e} + \epsilon_{k'+q}^{h} - \epsilon_{k'}^{h})^{-1} - (\hbar \Omega_{\lambda} - \epsilon_{k}^{h} - \epsilon_{k+\lambda}^{e})^{-1}] c_{k+\lambda}^{\dagger} b_{-k}^{\dagger} \alpha_{\lambda} - \text{H.c.}$$
(2.19)

This interaction gives recombination in *direct*-band-gap semiconductors only. In indirect-band-gap semiconductors, which we are considering, this term has no contribution because of momentum limitations on the Fermi surfaces in the condensed state. In fact these processes give rise to fast relaxation in the direct-band-gap semiconductors. It is for this reason that relaxation in the direct-band-gap semiconductors is several orders of magnitude faster than in the indirect-band-gap materials. This explains as to why the relaxation times in the direct-band-gap materials are of the order of nsec whereas in the indirect-band-gap materials these are of the order of μ sec. The next term is given by

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$$\mathfrak{K}_{A} = -\sum_{kk'p} D_{p}^{2} F_{A1}(k,k',p) c_{k}^{\dagger} b_{k'+p}^{\dagger} b_{k'} c_{k+p} - \sum_{kk'\lambda} G_{\lambda}^{2} F_{A2}(k,k',\lambda) c_{k'+\lambda}^{\dagger} b_{-k'}^{\dagger} c_{k+\lambda} b_{-k} + \text{H.c.} , \qquad (2.20a)$$

with

$$F_{A1}(k,k',p) = (\epsilon_{k}^{e} - \epsilon_{k+p}^{e} + \hbar\omega_{p})^{-1} + (\epsilon_{k'}^{h} - \epsilon_{k'+p}^{h} + \hbar\omega_{p})^{-1} , \quad F_{A2}(k,k',\lambda) = (\hbar\Omega_{\lambda} - \epsilon_{-k}^{h} - \epsilon_{k+\lambda}^{e})^{-1} , \quad (2.20b)$$

which are the phononless Auger recombination terms. These terms again are of importance in the direct-bandgap semiconductors but because of limitations on the Fermi wave vectors in the indirect-band-gap materials, these cannot give rise to a relaxation process with simultaneous conservation of momentum. This is not quite so when the temperature is raised up. In the indirect-band-gap systems, the phonon-induced Auger process with the simultaneous emission or absorption of one phonon dominates over the phononless process at low temperatures only. At room temperature both the processes can be of comparative magnitude. Next we see that

$$\Im C_{pA} = i \sum_{kk'qp} 2D_p V_q' F_{pA}(k,k',q,p) c_{k+q+p}^{\dagger} b_{k'-q}^{\dagger} b_{k'} c_k \beta_p + \text{H.c.} , \qquad (2.21a)$$

with

$$F_{p,4}(k,k',q,p) = (\epsilon_{k'}^{h} - \epsilon_{k'-q}^{h} + \epsilon_{k+q}^{e} - \epsilon_{k+q+p}^{e})^{-1} + (\epsilon_{k'}^{h} - \epsilon_{k'-q}^{h} + \epsilon_{k}^{e} - \epsilon_{k+q}^{e})^{-1} + (\epsilon_{k}^{e} - \epsilon_{k+q+p}^{e} + \epsilon_{k'-q}^{h})^{-1} + (\epsilon_{k}^{e} + \epsilon_{k'}^{h} - \epsilon_{k'-q-p}^{h} - \epsilon_{k+q+p}^{e})^{-1} + (\epsilon_{k+q}^{e} - \epsilon_{k+q+p}^{e} + \hbar\omega_{p})^{-1} - (\epsilon_{k}^{e} - \epsilon_{k+p}^{e} + \hbar\omega_{p})^{-1} + (\epsilon_{k'-q-p}^{h} - \epsilon_{k'-q}^{h} + \hbar\omega_{p})^{-1} - (\epsilon_{k'}^{h} - \epsilon_{k'-p}^{h} + \hbar\omega_{p})^{-1}$$

$$(2.21b)$$

describes the phonon-assisted Auger recombination. This is the dominant mechanism in the indirect-band-gap materials since the phonon can provide the required momentum. The phononless radiative term is given by

$$\mathbf{3C}_{r} = i \sum_{kk'q\lambda} 2G_{\lambda} V_{q}' [F_{r1}(k,k',q,\lambda) c_{k+q+\lambda}^{\dagger} c_{k'-q}^{\dagger} c_{k'} b_{-k}^{\dagger} \alpha_{\lambda} + F_{r2}(k,k',q,\lambda) b_{-k}^{\dagger} b_{k'-q}^{\dagger} b_{k'} c_{k+q+\lambda} \alpha_{\lambda}] + \text{H.c.} , \qquad (2.22a)$$

with

$$F_{r1}(k,k',q,\lambda) = (\epsilon_{k+q+\lambda}^{e} + \epsilon_{k'-q}^{e} - \epsilon_{k+\lambda}^{e} - \epsilon_{k'}^{e})^{-1} - (\epsilon_{k'}^{e} - \epsilon_{k'-q}^{e} - \epsilon_{k}^{h} + \epsilon_{q-k}^{h})^{-1} + (\hbar \Omega_{\lambda} - \epsilon_{-k}^{h} - \epsilon_{k+\lambda}^{e})^{-1} - (\hbar \Omega_{\lambda} - \epsilon_{q-k}^{h} - \epsilon_{k+q+\lambda}^{e})^{-1} ,$$

$$F_{r2}(k,k',q,\lambda) = (\epsilon_{-k}^{h} - \epsilon_{q-k}^{h} + \epsilon_{k'+q}^{h} - \epsilon_{k'}^{h})^{-1} - (\epsilon_{k+\lambda}^{e} - \epsilon_{k+q+\lambda}^{e} + \epsilon_{k'-q}^{h})^{-1} + (\hbar \Omega_{\lambda} - \epsilon_{-k}^{h} - \epsilon_{k+\lambda}^{e})^{-1} - (\hbar \Omega_{\lambda} - \epsilon_{q-k}^{h} - \epsilon_{k+q+\lambda}^{e})^{-1} .$$

$$(2.22b)$$

In this process an electron is created and annihilated to provide the momentum for the recombination of an electron and a hole with the emission of a photon. However, in the electron-hole liquid the Fermi wave vectors are three orders of magnitude smaller than in metals. Therefore in EHD the expression (2.22) cannot meet the requirements of the momentum δ function. Next we write,

$$\mathfrak{R}_{pr} = -\sum_{kp\lambda} D_p G_{\lambda} [F_{pr1}(k,p,\lambda) c^{\dagger}_{k+p+\lambda} b^{\dagger}_{-k} \alpha_{\lambda} \beta_p - F_{pr2}(k,p,\lambda) c^{\dagger}_{k+\lambda-p} b^{\dagger}_{-k} \alpha_{\lambda} \beta_p^{\dagger}] + \mathrm{H.c.} , \qquad (2.23a)$$

where

$$F_{pr1}(k,p,\lambda) = (\hbar \Omega_{\lambda} - \epsilon^{h}_{-k} - \epsilon^{e}_{k+\lambda})^{-1} + (\hbar \Omega_{\lambda} - \epsilon^{h}_{p-k} - \epsilon^{e}_{k+p+\lambda})^{-1} - (\epsilon^{e}_{k+\lambda} - \epsilon^{e}_{k+p+\lambda} + \hbar \omega_{p})^{-1} - (\epsilon^{h}_{p-k} - \epsilon^{h}_{-k} + \hbar \omega_{p})^{-1}$$

$$(2.23b)$$

$$F_{pr2}(k,p,\lambda) = (\hbar \Omega_{\lambda} - \epsilon_{k}^{h} - \epsilon_{k+\lambda}^{e}) - (\hbar \Omega_{\lambda} - \epsilon_{p-k}^{h} - \epsilon_{k+p+\lambda}^{e})^{-1} - (\epsilon_{k+\lambda-p}^{e} - \epsilon_{k+\lambda}^{e} + \hbar \omega_{p})^{-1} - (\epsilon_{-k}^{h} - \epsilon_{p-k}^{h} - \epsilon_{p-k}^{h} + \hbar \omega_{p})^{-1} - (\epsilon_{-k}^{h} - \epsilon_{p-k}^{h} + \hbar \omega_{p})^{-1} - (\epsilon_{-k}^{h} - \epsilon_{p-k}^{h} - \epsilon_{$$

In this process an electron and a hole recombine with emission of a photon and emission or absorption of a phonon. This gives rise to τ_r of Eq. (1.1) in indirect-band-gap materials. Finally there are terms in the transformed Hamiltonian which contain products of six or more quasiparticle operators, the contribution of these higher-order terms to the physical properties of the EHD is small. The interactions (2.21) and (2.23) are the only important recombination mechanisms in the indirect-band-gap materials at low temperatures. At this time the effect of these terms on the ground state^{25, 27–31} is not obvious which has been studied either with the Coulomb interactions alone or with the electron-phonon interaction in polar materials. The electron-phonon interaction in nonpolar materials such as Ge has not been considered.³¹ We will consider the effect of radiative interactions on the ground state in a later paper. It may be noted that thermal nonradiative scattering as well as radiative mechanisms have been recognized by one of the present authors³² as experimentally observable mechanisms of magnon relaxation in magnetic materials. The present problem of nonradiative Auger and radiative relaxation is thus some what analogous to that of the magnon relaxation.

III. RECOMBINATION SELF-ENERGY

There are only four types of processes which contribute to recombination, the phononless Auger, one-phonon Auger, phononless radiative, and the one-phonon radiative. The phononless processes dominate in the direct-band-gap materials. The selfenergy of an electron-hole pair due to the recombination is now calculated for these four processes using the standard rules. The contribution to the pair selfenergy due to the Auger recombination is obtained from Eq. (2.20) as

$$\Sigma_{A}(k) = \sum_{k'p} D_{p}^{4} F_{A1}^{2} \left(\epsilon_{k'+p}^{h} + \epsilon_{k}^{e} - \epsilon_{k+p}^{e} - \epsilon_{k'}^{h} \right)^{-1} \eta_{1}(k,k',p) + \sum_{k'\lambda} G_{\lambda}^{4} F_{A2}^{2} \left(\epsilon_{k'+\lambda}^{e} + \epsilon_{-k'}^{h} - \epsilon_{k+\lambda}^{e} - \epsilon_{-k}^{h} \right)^{-1} \eta_{2}(k,k',\lambda) + \tilde{\Sigma}_{A}(k)$$
(3.1)

where

$$\eta_1(k,k',p) = f_{k'}^h f_{k+p}^e (1 - f_{k'+p}^h) + (1 - f_{k'}^h) (1 - f_{k+p}^e) f_{k'+p}^h , \qquad (3.2)$$

$$\eta_2(k,k',\lambda) = f^h_{-k-\lambda} (1 - f^e_{k'+\lambda}) (1 - f^h_{k'}) + (1 - f^h_{-k-\lambda}) f^e_{k'+\lambda} f^h_{k'}$$
(3.3)

Here f_k^{h} and f_k^{h} denote the Fermi occupation-number density of electrons and holes, respectively, for example,

$$f_{\ell}^{\epsilon} = \{\exp\left[\left(\epsilon_{\ell}^{\epsilon} - \epsilon_{\ell}^{\epsilon}\right)/k_{B}T\right] + 1\}^{-1}$$
(3.4)

The term $\tilde{\Sigma}_{\mathcal{A}}(k)$ is obtained by interchanging the electrons and the holes in the preceding terms. The phononassisted Auger process (2.21) has a contribution of

$$\Sigma_{pA}(k) = -\sum_{k'qp} 4D_p^2 V_q'^2 F_{pA}^2 (\epsilon_{k+q+p}^e + \epsilon_{k'-q}^h - \epsilon_{k'}^e - \hbar \omega_p)^{-1} \varphi(k,k',q,p) + \tilde{\Sigma}_{pA}(k) , \qquad (3.5)$$

where

$$\varphi(k,k',q,p) = N_p f_{k'}^h (1 - f_{k'-q}^h) (1 - f_{k+q+p}^e) + (N_p + 1) (1 - f_{k'}^h) f_{k'-q}^h f_{k+q+p}^e$$
(3.6)

and N_p is the occupation number of the phonons. There is an additional contribution to Eq. (3.5) from a diagram which is energetically unfavorable and hence ignored. The phononless radiative contribution to the pair self-energy is calculated to be

$$\Sigma_{r}(k) = -\sum_{k'q\lambda} 4G_{\lambda}^{2} V_{q}^{\prime 2} [F_{r1}^{2}(k',k,q,\lambda) (\epsilon_{k'+q+\lambda}^{e} + \epsilon_{k-q}^{e} - \epsilon_{k}^{e} + \epsilon_{-k'}^{h} - \hbar \Omega_{\lambda})^{-1} \chi_{1}(k,k',q,\lambda) + F_{r1}^{2}(-k,k',q,\lambda) (\epsilon_{-k+q+\lambda}^{e} + \epsilon_{k'-q}^{e} - \epsilon_{k'}^{e} + \epsilon_{k}^{h} - \hbar \Omega_{\lambda})^{-1} \chi_{2}(k,k',q,\lambda)] + \tilde{\Sigma}_{r}(k) , \quad (3.7)$$

$$\chi_{1}(k,k',q,\lambda) = (1 - f_{k'+q+\lambda}^{e})(1 - f_{k-q}^{e})(1 - f_{-k}^{h})n_{\lambda} + f_{k'+q+\lambda}^{e} f_{k-q}^{e} f_{-k}^{h}(n_{\lambda}+1) ,$$

$$\chi_{2}(k,k',q,\lambda) = (1 - f_{q+\lambda-k}^{e})(1 - f_{k'-q}^{e}) f_{k'}^{e}n_{\lambda} + f_{q+\lambda-k}^{e} f_{k'-q}^{e}(1 - f_{k'}^{e})(n_{\lambda}+1) ,$$
(3.8)

and the one-phonon radiative contribution to the pair self-energy is found to be

$$\Sigma_{pr}(k) = \sum_{p\lambda} D_p^2 G_{\lambda}^2 [F_{pr1}^2 (k - p - \lambda, p, \lambda) (\epsilon_k^e + \epsilon_{p+\lambda-k}^h - \hbar \omega_p - \hbar \Omega_{\lambda})^{-1} \xi_1 + F_{pr2}^2 (p - \lambda + k, p, \lambda) (\epsilon_{\lambda-p-k}^h - \epsilon_k^e + \hbar \omega_p - \hbar \Omega_{\lambda})^{-1} \xi_2] + \tilde{\Sigma}_{pr}(k) , \qquad (3.9)$$

where

$$\xi_1 = 2n_{\lambda}N_p + f_{p+\lambda-k}^h(1+n_{\lambda}+N_p) \quad , \quad \xi_2 = 2n_{\lambda}(N_p+1) + f_{\lambda-p-k}^h(n_{\lambda}-N_p) \quad .$$
(3.10)

$$f_{\text{intra}}^2 \simeq 1 + O(q^2)$$
 ,
 $f_{\text{inter}}^2 \simeq 2\hbar^2 q^2 / (\mu E_g)$,

as discussed by Pines²⁴ and utilized by Haug¹⁰ and Lochmann,¹¹ so that the effective potential is

$$V_q = 4\pi e^2 / (V \kappa q^2) ,$$

$$V_q' = [4\pi e^2 / (V \kappa q^2)] [2\hbar^2 q^2 / (\mu E_g)]^{1/2} ,$$
(3.11)

where q is the momentum transfer along the interaction line.

IV. RELAXATION TIME IN INDIRECT-BAND-GAP SEMICONDUCTORS

The relaxation time is determined by the imaginary part of the self-energy,

$$1/\tau(k) = (2/\hbar) \operatorname{Im}\Sigma(k) \quad . \tag{4.1}$$

Since we are interested in the indirect-band-gap materials it is necessary to separate the imaginary parts only near the particular pole of interest which give the phonon-assisted Auger τ_{pA} and phonon-assisted radiative relaxation τ_{pr} . For this purpose we use Dirac's identity

$$\lim_{\epsilon \to 0} \frac{1}{x \pm i\epsilon} = \frac{P}{x} \mp i\pi\delta(x) \quad . \tag{4.2}$$

A. Phonon-assisted Auger recombination

The phonon-assisted Auger recombination time is given by

$$\frac{1}{\tau_{pA}(k)} = \frac{2}{\hbar} \operatorname{Im} \sum_{k'qp} (2D_p V'_q F_{pA})^2 (\epsilon^e_{k+q+p} + \epsilon^h_{k'-q} - \epsilon^h_{k'} - \epsilon^e_k - \hbar\omega_p)^{-1} \varphi(k,k',q,p) + \frac{1}{\tilde{\tau}_{pA}(k)} , \qquad (4.3)$$

where $\tilde{\tau}_{pA}(k)$ is obtained from the previous terms by interchanging the electrons and the holes. Separating the imaginary part of the self-energy, we obtain,

$$\frac{1}{\tau_{pA}(k)} = -\frac{2\pi}{\hbar} \sum_{k'q} \frac{4\pi V}{(2\pi)^3} \int p^2 dp \ (2D_p V'_q F_{pA})^2 \varphi(k,k',q,p) \,\delta(\epsilon^e_{k+q+p} + \epsilon^h_{k'-q} - \epsilon^h_{k'} - \epsilon^e_k - \hbar\omega_p) + \frac{1}{\tilde{\tau}_{pA}(k)}$$
(4.4)

As found by Hammond *et al.*¹⁹ the temperature-dependent part of the Fermi energies is very small, so that $f_k^e \simeq f_k^h \simeq \frac{1}{2}, \varphi \simeq \frac{1}{8}(2N_p+1)$. This is the reasonable approximation if we are interested in the relaxation time at temperatures of the order of 2 K. For optical phonons in nonpolar materials such as Ge, the coupling constant of the electron-phonon interaction is given by

$$D_p = D_{0p} (\hbar \omega_L / 2c_2 V)^{1/2} , \qquad (4.5)$$

where D_{0p} is the nonpolar optical deformation potential, c_2 is the longitudinal elastic constant, and ω_L is the optical phonon frequency at the zone boundary. The zeros of the argument of the δ function in Eq. (4.4) occur for values of p given by

$$p_{1,2} = -(k+q) \pm \{(k+q)^2 - [2kq+q^2 + (m_e/m_h)(q^2 - 2k'q) - 2\hbar\omega_L m_e/\hbar^2]\}^{1/2} .$$
(4.6)

For slow electron and holes, $\epsilon_k^e \ll \hbar \omega_L$, $\epsilon_k^h \ll \hbar \omega_L$, Eq. (4.6) becomes,

$$p_{1,2} = -(k+q) \pm p_c, \quad p_c = (2m_e\omega_L/\hbar)^{1/2} \quad .$$
(4.7)

The expression (4.4) can then be written

$$\frac{1}{\tau_{pA}^{0}(k)} = -\sum_{k'q} \frac{128 \pi D_{\delta p}^{0} \omega_{L} e^{4} m_{e}}{c_{2} V \kappa^{2} \mu E_{g} q^{2}(p_{1} - p_{2})} \int p^{2} dp \ F_{pA}^{2} \phi(k', k, q, p) \left[\delta(p - p_{1}) - \delta(p - p_{2})\right] + \frac{1}{\tilde{\tau}_{pA}^{0}} \ . \tag{4.8}$$

Evaluating the δ functions we find

$$\frac{1}{\tau_{pA}^{0}(k)} = \sum_{k'q} \frac{64\pi D_{0p}^{2}\omega_{L}e^{4}m_{e}\varphi}{c_{2}V\kappa^{2}\mu E_{g}p_{c}q^{2}} [F_{pA1}^{2}(k+q-p_{c})^{2} - F_{pA2}^{2}(k+q+p_{c})^{2}] + \frac{1}{\tilde{\tau}_{pA}^{0}} , \qquad (4.9)$$

where

$$F_{pA1} = 2\left\{ \left(2\epsilon_{F}^{e}\right)^{-1} + \left[\hbar\omega_{L}\left(\frac{m_{e}}{m_{h}} - 1\right)\right]^{-1} + \left[\hbar\omega_{L}\left(\frac{m_{e}}{m_{h}} + 1\right)\right]^{-1} - \left(4\epsilon_{F}^{e}\hbar\omega_{L}\right)^{-1/2} \right\} ,$$

$$F_{pA2} = 2\left\{ \left(2\epsilon_{F}^{e}\right)^{-1} + \left[\hbar\omega_{L}\left(\frac{m_{e}}{m_{h}} - 1\right)\right]^{-1} + \left[\hbar\omega_{L}\left(\frac{m_{e}}{m_{h}} + 1\right)\right]^{-1} + \left(4\epsilon_{F}^{e}\hbar\omega_{L}\right)^{-1/2} \right\} .$$
(4.10)

The summations over k' and q in the expression (4.9) are evaluated by changing them into integration. The result of this calculation is given by,

$$1/\tau_{pA}^{0}(k) = B_{0}k_{F}^{6}\left[\left(\frac{1}{3} + \sigma + \sigma^{2} + \sigma_{c}^{2}\right)\left(F_{pA2}^{2} - F_{pA1}^{2}\right) + \sigma_{c}(1 + 2\sigma)\left(F_{pA1}^{2} + F_{pA2}^{2}\right)\right] + 1/\tilde{\tau}_{pA}^{0}(k) , \qquad (4.11)$$

where
$$\sigma = k/k_F$$
, $\sigma_c = p_c/k_F$, and
 $B_0 = \frac{2D_{0p}^2 \hbar \omega_L m_e e^4 (2N_L + 1)}{3\pi^3 \hbar c_2 \mu E_g \kappa^2 p_c}$. (4.12)

For $\sigma = 1(k = k_F)$ we have

$$1/\tau_{pA}^{0}(k_{F}) = B_{1}k_{F}^{6} + B_{2}k_{F}^{5} + B_{3}k_{F}^{4} + 1/\tilde{\tau}_{pA}^{0}(k_{F}) , \qquad (4.13)$$

with

$$B_{1} = \frac{7}{3} B_{0} (F_{pA2}^{2} - F_{pA1}^{2}) ,$$

$$B_{2} = 3 B_{0} p_{c} (F_{pA1}^{2} + F_{pA2}^{2}) ,$$

$$B_{3} = B_{0} p_{c}^{2} (F_{pA2}^{2} - F_{pA1}^{2}) .$$
(4.14)

In Ge, p_c/k_F is of the order of 3 and $B_1k_F/B_2 \simeq 0.1$, the term containing k_F^6 is smaller than that containing k_F^5 . Further $B_2k_F/B_3 \sim 1$, and $k_F = (3\pi^2 n)^{1/3}$, various terms in the relaxation rate vary as $n^{5/2}$, $n^{4/3}$, and n^2 . This result is to be compared with that of Haug who has found only the n^2 term. This difference between our calculation that of Haug arises because we have explicitly evaluated the momentum dependence in the argument of the δ function. It may be noted that the relaxation time of the phonon does vary as the square of the carrier concentration when momentum dependence is correctly treated.³³

In the case of acoustic phonons, the electronphonon coupling is given by $^{34-36}$

$$D_p = d_1 (p/V)^{1/2}, \quad d_1 = c_1 (\hbar/\rho v)^{1/2}, \quad (4.15)$$

where ρ is the mass density, v is the phonon velocity, and c_1 is the elastic energy. In this case $\omega_p = vp$, so that the δ function in Eq. (4.4) has zeros at

$$p' = -2(k + q - m_e v/\hbar), \quad p'' = 0$$

for slow electrons and holes. In this case Eq. (4.4) gives

$$1/\tau_{pA}^{a}(k) = B_{a}k_{F}^{6}[4(\frac{1}{3} + \sigma + \sigma^{2}) + 4(m_{e}v/\hbar k_{F})(2\sigma + \sigma^{2}) + 4m_{e}^{2}v^{2}/(\hbar^{2}k_{F}^{2})] + 1/\tilde{\tau}_{pA}^{a}(k) , \qquad (4.16)$$

where

$$B_a = \frac{8d_1^2 F_{p'A}^2 m_e e^4}{3\pi^3 \hbar \mu E_g \kappa^2} \quad . \tag{4.17}$$

 $F_{p'A} \simeq 1/3\epsilon_F$ which at $k = k_F$ leads to

$$1/\tau_{pA}^{a}(k_{F}) = B'k_{F}^{6} + B''k_{F}^{5} + B'''k_{F}^{4} + 1/\tilde{\tau}_{pA}^{a}(k_{F}) \quad ,$$
(4.18)

$$B' = \frac{28}{3} B_a, \quad B'' = 12 m_e v/\hbar, \quad B''' = 4 m_e^2 v^2/\hbar^2 \quad . \quad (4.19)$$

So that the dependence of the relaxation rate on the carrier concentration in the case of acoustic phonons is similar to that found for optical phonons as a result of self-cancelling effects.

B. Radiative relaxation

We now proceed to calculate the imaginary part of Eq. (3.10), the phonon-assisted radiative contribution to the recombination relaxation time. The photon wave vector is very small compared to the other momenta which enter into the problem; i.e., $\lambda = \Omega_{\lambda}/c$ ≈ 0 . In the low-temperature limit we obtain,

$$\frac{1}{\tau_{pr}(k)} = \frac{2\pi}{\hbar} \sum_{\lambda} \frac{4\pi V}{(2\pi)^3} \int p^2 dp \ D_p^2 G_{\lambda}^2 \left[F_{pr1}^2 \xi_1 \delta(\epsilon_k^e - \epsilon_{p+\lambda-k}^h - \hbar \omega_p - \hbar \Omega_{\lambda}) + F_{pr2}^2 \xi_2 \delta(\epsilon_{\lambda-p-k}^e + \epsilon_k^h + \hbar \omega_p - \hbar \Omega_{\lambda}) \right]$$

$$+ \frac{1}{\tilde{\tau}_{pr}(k)} , \qquad (4.20)$$

where $1/\tilde{\tau}_{pr}(k)$ is obtained from the previous terms by interchanging the electrons and holes. For optical pho-

nons the zeros of the argument of the δ functions are given by

$$p_{1,2} = k \pm [k^2 (1 - m_h/\mu) - (2m_h/\hbar^2) (E_g - \hbar \Omega_\lambda - \hbar \omega_L)]^{1/2} , \qquad (4.21)$$

$$p_{3,4} = -k \pm \left[k^2 (1 - m_h/\mu) - (2m_h/\hbar^2) (E_g - \hbar \Omega_\lambda + \hbar \omega_L) \right]^{1/2} , \qquad (4.22)$$

respectively. Hammond et al.¹⁹ have discussed that the lowest-energy photon which can be emitted in the recombination process is of the frequency

$$\hbar \Omega_{\lambda} = E_{g} - \hbar \omega_{g} \tag{4.23}$$

and occurs when an electron and a hole, each at its band extrema recombine. However this condition when substituted in the expression for $p_{1,2}$ in Eq. (4.21) gives complex momenta. In order to have real $p_{1,2}$ the condition (4.23) must be modified as

$$\hbar \Omega_{\lambda} = E_{g} - \hbar \omega_{g} + \hbar^{2} k^{2}/2\mu \quad . \tag{4.24}$$

The condition for the highest-energy photon as given in Ref. 19 is

$$\hbar \Omega_{\lambda} = E_g - \hbar \omega_p + \epsilon_F^e + \epsilon_F^h \quad . \tag{4.25}$$

Our condition (4.24) is consistent with Eqs. (4.23) and (4.25) qualitatively, if we require k to go from zero to k_F , but k cannot be zero since its minimum value is fixed by Eq. (4.24) itself. Using Eq. (4.24), we find that, $p_1=0$, $p_2=2k$, $p_3=-2k$, $p_4=0$. Then in the low-temperature limit, $N_L \ll 1$, Eq. (4.20) can be written

$$\frac{1}{\tau_{pr}^{0}(k)} = \frac{D_{0p}^{*} \kappa_{L} \Omega_{0} e^{2} m_{h} k}{\pi c^{2} \kappa c_{2} \mu^{2} V} \sum_{\lambda} \left[F_{pr1}^{2}(0) (n_{\lambda} + 1) + \frac{5}{2} F_{pr2}^{2}(0) n_{\lambda} \right] + \frac{1}{\tilde{\tau}_{pr}^{0}(k)} , \qquad (4.26)$$

where

$$F_{pr1}(0) = 2[(\hbar\omega_L)^{-1} - (\hbar\Omega_0 - E_g)^{-1}], \quad F_{pr2}(0) = 2[(\hbar\omega_L)^{-1} + (\hbar\Omega_0 - E_g)^{-1}] \quad .$$
(4.27)

In the case of acoustic phonons, we have $\omega = vp$ and D_p is given by Eq. (4.15). The zeros of the argument of the δ function (4.24) lead to

$$\frac{1}{\tau_{pr}^{a}(k)} = -\frac{2m_{h}}{\pi\hbar^{3}} \sum_{\lambda} \int p^{2} dp \, d_{1}^{2} G_{\lambda}^{2} \left[F_{pr1}^{2} \chi_{1}(k,p,\lambda) \delta(p-2k) + F_{pr2}^{2} \chi_{2}(k,p,\lambda) \delta(p+2k) \right] + \frac{1}{\tilde{\tau}_{pr}^{a}(k)} \quad (4.28)$$

Upon integration this result becomes

- '1

$$\frac{1}{\tau_{pr}^{a}(k)} = \frac{4d_{1}^{2} \Omega_{0} e^{2} m_{h} k^{2}}{\pi c^{2} \kappa \mu^{2} V} \sum_{\lambda} \left[(n_{\lambda} + 1) F_{pr1}^{\prime 2}(0) + \frac{5}{2} F_{pr2}^{\prime 2}(0) n_{\lambda} \right] + \frac{1}{\tilde{\tau}_{pr}^{a}(k)} , \qquad (4.29)$$

where

$$F'_{pr1} = 2[(2\hbar v k_F)^{-1} - (\hbar \Omega_0 - E_g)^{-1}],$$

$$F'_{pr2} = -2[(2\hbar v k_F)^{-1} + (\hbar \Omega_0 - E_g)^{-1}].$$
(4.30)

This completes the calculation of the recombination relaxation time owing to the dominant recombination mechanisms in the indirect-band-gap materials in a two band model. The generalization to multiple-band systems is straightforward.

V. ENHANCEMENT

In our theory the Fourier transform of the pair Green's function is of the form,

$$G(k,E) = \langle \langle b_k^{\mathsf{T}} c_k^{\mathsf{T}} | b_k c_k \rangle \rangle$$
$$= (\hbar/2\pi) [E - \epsilon_k^e - \epsilon_k^h - \Sigma(k)]^{-1} , \quad (5.1)$$

where the pair self-energy is given by $\Sigma(k)$. In the present work we are interested only in the recombination relaxation contributions to the self-energy which have been calculated in the previous sections. The pair correlation functions are related to the pair Green's function.³⁷ Extensive studies of the correlation functions $g_{ee}(r)$, $g_{eh}(r)$, and $g_{hh}(r)$ have been reported by Vashishta *et al.*^{29,38} These authors also study the enhancement factor due to correlations, $g_{eh}(0)$ for several models. It is found that the value of $g_{eh}(0)$ depends considerably on the model, the approximation scheme, and the values of the parameters chosen. In Ge, if one replaces the two-hole bands of different masses by two bands each having the same mass, $g_{eh}(0)$ is 1.8 in the Hubbard approximation and 2.2 with electron-hole multiple scattering. If one takes the actual hole masses $g_{eh}(0)$ is found to be 2.3 in absence of multiple scattering which changes to 2.8 if multiple scattering is taken into ac-

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count. If the effect of anisotropy of the bands is included in the fully self-consistent approximation $g_{eh}(0)$ is calculated³⁹ to be 2.3. The phonon scattering is not considered in these calculations while we know that in the indirect-band-gap semiconductors the phonons play an important role. In our problem,⁴⁰

$$\frac{1}{\tau} \simeq \frac{2}{\hbar} \left[1 - \frac{d}{dE} \operatorname{Re}\Sigma(k) \right]^{-1} \operatorname{Im}\Sigma(k) \quad , \qquad (5.2)$$

where $\Sigma(k)$ is the pair self-energy. In the lowestorder decoupling the effect of the interaction occurs only in the shift of the single-particle frequencies and hence in the real part of the self-energy. Accordingly Hensel *et al.* suggest that

$$1/\tau' = (1/\tau) g_{eh}(0) \quad . \tag{5.3}$$

Unfortunately, the experimental values of $g_{eh}(0)$ are as uncertain as the theoretical ones. Benoit à la Guillaume *et al.*^{41,42} have suggested on the basis of qualitative arguments that the enhancement factor may be obtained from,

$$g_{eh}(0) = \tau_r^{ex} / (\tau_r n \pi a_x^3) \quad , \tag{5.4}$$

where in Ge, using $a_x = 114$ Å, $n = 2.38 \times 10^{17}$ cm⁻³, they obtain $(n \pi a_x^3) = 1.2$. In order to find the ratio of the free-exciton radiative relaxation time to that of the radiative relaxation time in the electron-hole drop, the authors of Ref. 41 make use of the ratio of intensities of the luminescence of the free exciton to that of the EHD and the ratio of the number densities of the excitons and the *e*-*h* pairs in the EHD as

$$\tau_r^{\text{ex}}/\tau_r \simeq (N_{\text{ex}}/N_{\text{EHD}})(I_{\text{EHD}}/I_{\text{ex}}) \approx 8$$
 , (5.5)

which leads to the value of $g_{eh}(0) \sim 7$. Westervelt et al.⁴³ obtained a value of $\tau_r^{ex}/\tau_r \sim 6.5$ and hence $g_{eh}(0) \sim 5$. As pointed out by Hensel *et al.* the results of $g_{eh}(0)$ in Ge are not known with desirable accuracy. In the condensed phase the Fermi energies are three orders of magnitudes smaller than in the gas phase. Therefore the mechanisms of recombination in the EHD are not the same as in the exciton gas. The approximation $\epsilon_k \ll \hbar \omega_L$ is valid for EHD whereas reverse $\epsilon_k >> \hbar \omega_L$ is the case for free exciton gas. Therefore it is not quite correct to compare the relaxation times and intensities as in Eqs. (5.3)-(5.5) which assume the same scheme of integration in the Fermi space for both the liquid and the gas phase. In the Hartree-Fock approximation relaxation occurs only due to the electron(hole)phonon and electron-hole photon interactions. However in the indirect-band-gap semiconductors these approximations cannot lead to recombination.

VI. APPLICATION TO Ge

A. Experimental measurements

Using a variable-frequency technique, Pokrovskii and Svistunova⁴⁴ deduced the lifetime of the drop in Ge to be 20 μ sec at a temperature of 2.1 K. Benoit à la Guillaume et al.^{41,42} obtained $\tau_0 \sim 40 \ \mu \text{sec}$ by measuring the luminescence in a pulsed experiment whereas a value of $\tau_0 = (45 \pm 5) \ \mu \text{sec}$ is deduced⁴⁵ in a transparent sample at low excitations, 0.5 W/mm² and Westervelt *et al.*⁴³ obtain $\tau_0 = (41 \pm 1) \mu \text{sec}$ at high powers indicating that this value is independent of the power of excitation and hence most likely associated with nonradiative process. Recent measurements by Katyrin et al.¹³ lead to a lifetime of 30 μ sec but Leheny et al.⁴⁶ give 80 µsec for the nonradiative lifetime and suggest a radiative efficiency of about 0.5. Therefore it appears that the experimental values themselves have a large spread being of the order of 40 ± 20 µsec have an error of the order of 50%. In Sec. VIB we make an effort to predict this value from a purely theoretical viewpoint.

B. Calculations

In this section we make an effort to predict the recombination relaxation times in Ge. In this material, the intraband transition from the conduction-band minimum at point L in the Brillouin zone to the point Γ is allowed only by the longitudinal-optical phonon scattering. Accordingly we employ Eq. (4.11) to calculate the phonon-assisted Auger relaxation time. We take the band structure into account in the BRAC approximation scheme.^{1,25} Although the scheme of Combescot and Nozieres²⁷ which takes into account the shape of the bands, being more accurate, is algebraically more involved and as admitted by the authors themselves gives only a 3% correction over the BRAC model. Bhattacharyya et al.²⁹ while calculating the correlation energy also replace the anisotropic structure of the conduction bands by isotropic structure corresponding to optical masses in the self-consistent particle-hole approximation and point out that though the approach of Combescot and Nozieres is more sophisticated, the approximation scheme is quite reasonable. The parameters for Ge are well known. The optically reduced masses are $m_{0e} = 0.12$, $m_{0h} = 0.07$, and $\mu = 0.046$ in the units of electron rest mass. The remaining parameters are, $E_g = 664 \text{ meV}, D_{0p} = 25 \text{ meV}, \hbar \omega_L = 31 \text{ meV},$ $r_s = 0.6, a_x = 177 \times 10^{-8} \text{ cm}, \kappa = 15.4, c_2 = 1.5 \times 10^{12}$ dyn/cm², $k_F = (2\mu\epsilon_F/\hbar)^{1/2}$, and $\epsilon_F = 6.2 \text{ meV}$. The value deduced on the basis of Eq. (4.11) is found to be $\tau_{pA}^0 = 16 \ \mu \text{sec}$ at lowest temperatures. This value is of the correct order of magnitude, as seen by comparing with the experimental data.

We now calculate the relaxation rate due to the phonon-induced radiative process for optical phonons

on the basis of Eq. (4.26). This relaxation time depends essentially on the radiative power through the factor $\sum_{\lambda} n_{\lambda}/V$ as the value of $\sum_{\lambda} 1/V$ is negligible. The value of $\sum_{\lambda} n_{\lambda}/V$ is of the order of 10¹⁸ corresponding to a power of 75 mW. All the other quantities are assumed to have the same value as for the phonon-assisted Auger process. Then following the same scheme $\tau_{pr}(k_F) = 360 \ \mu$ sec is found. Therefore it appears that the radiative process will not lead to a dominant relaxation and the major contribution is due to the phonon-assisted Auger process in agreement with the experimental observation that the low- and high-excitation power lead to the same

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lifetime since the experiment notices only the shorter time if the other time is an order of magnitude too long. In general τ_0 of Eq. (1.1) has both the contributions. However τ_r does not significantly alter the value of τ_0 .

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