# Green's-function formalism of band-to-band Auger recombination in semiconductors. Correlation effect

Masumi Takeshima

Semiconductor Laboratory, Matsushita Electronics Corporation, Takatsuki, Osaka, Japan (Received 18 March 1982)

A general theory of band-to-band Auger recombination in semiconductors is developed on the basis of the Green's-function formalism. Starting with the density operator we obtain the excess-carrier lifetime in terms of the four-particle Green's function. This allows us to incorporate electron correlation effect as well as phonon, impurity, and some other scattering effects into the theory in a coherent way. It is shown that the correlation effect, which has never been considered in the earlier theories, can be important under some conditions for the electron-hole-hole collision process. In contrast the effect is negligible for the electron-hole collision process without any restrictions.

## I. INTRODUCTION

Since the publication of a work by Beatie and Landsberg<sup>1</sup> the Auger recombination (AR) has been considered as an efficient nonradiative process in semiconductours, especially at heavy-doping levels. A number of investigations<sup>2</sup> have been made on the basis of the pure collision Auger recombination (PCAR) and have led to fruitful results on narrow direct band-gap materials. However, with PCAR it was insufficient to explain the recombination rate in wide band-gap materials.<sup>3-7</sup> To give a better explanation, the phonon-assisted Auger recombination<sup>4,7-10</sup> (PHAR) and the impurity-assisted Auger recombination<sup>11</sup> (IAR) have been proposed. Recently, successful results have been obtained for Si and GaAs on the basis of PHAR (Ref. 12) and of IAR (Ref. 13). The unified treatment of PHAR and IAR has been given<sup>13</sup> in the theory of impurity- and phonon-assisted Auger recombination (IPHAR).

In the earlier theory of PHAR a divergence difficulty was inevitable, owing to the use of the second-order perturbation theory. Recently, the present author has given a theory based on the Green's-function formalism,<sup>14</sup> where no divergence difficulty appears. It is quite likely that phonon and impurity scattering plays an important role in real materials as in the case of transport phenomena and free-carrier absorption. Consequently, it is general and convenient to give the AR lifetime in terms of the Green's function, into which any scattering effects can in principle be incorporated. Although the Green's-function formalism has been given by the present author in the theories of IAR and PHAR, there are some defects, given as follows. The former theory starts from the statistical average of the scattering amplitude, although actually the statistical average of the absolute square of the amplitude is necessary, so that the result is of limited use. On the other hand, the latter theory relates the AR lifetime to the imaginary part of the self-energy so that the damping indifferent to the excess-carrier lifetime is involved.

In this paper rigorous treatment of the AR lifetime is given starting from the density-operator formalism. The lifetime is given in terms of the four-particle Green's function. This is finally approximated as a product of four one-particle Green's functions, neglecting the electroncorrelation effect. The validity of the neglect is discussed.

#### **II. FORMULATION**

In this section the excess-carrier lifetime of AR is expressed in terms of the Green's function, into which any scattering effects can be incorporated. First we define our model by writing down the Hamiltonian as

$$H = H_e + H_{ph} + H_{e-ph} + H_{e-i} + H_{e-e} .$$
 (2.1)

Here  $H_e$ ,  $H_{ph}$ ,  $H_{e-ph}$ ,  $H_{e-i}$ , and  $H_{e-e}$  are the Hamiltonians for the band electrons, phonons, electron-phonon interaction, electron-impurity interaction, and electron-electron Coulomb interaction, respectively. The explicit forms of the Hamiltonians are

$$H_e = \sum_{l \,\vec{k} \,\sigma} E_l(\vec{k}) a_{l \,\vec{k} \,\delta}^{\dagger} a_{l \,\vec{k} \,\sigma} , \qquad (2.2)$$

and

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$$H_{e-e} = \frac{1}{2V} \sum_{\substack{l_1 l_2 l_3 l_4\\\vec{k} \ \vec{k} \ \vec{q} \ \vec{\sigma} \ \sigma'}} \mathscr{V}(\vec{q}) \langle l_1 \vec{k} + \vec{q} \ | \ l_4 \vec{k} \rangle \langle l_2 \vec{k} \ ' - \vec{q} \ | \ l_3 \vec{k}' \rangle^a l_1 \vec{k} + \vec{q} \ \sigma^a l_2 \vec{k} \ ' - \vec{q} \ \sigma'^a l_3 \vec{k} \ \sigma'^a l_4 \vec{k} \ \sigma$$
(2.3)

We have not referred to  $H_{\rm ph}$ ,  $H_{e\text{-ph}}$ , and  $H_{e\text{-}i}$ , whose explicit forms are of no interest in this paper. In Eqs. (2.2) and (2.3)  $a_{l\,\vec{k}\,\sigma}^{\dagger}$ ,  $a_{l\,\vec{k}\,\sigma}$ , and  $E_{l}(\vec{k})$  are the creation operator, annihilation operator, and electron energy, respectively, for the band index l, wave vector  $\vec{k}$ , and spin  $\sigma$ . With V as the crystal volume  $\mathscr{V}(\vec{q})/V$  is the Fourier component of the unscreened Coulomb potential, i.e.,

$$\mathscr{V}(\vec{q}) = \frac{4\pi e^2}{q^2} .$$

 $\langle l_i \vec{k}_i | l_j \vec{k}_j \rangle$  is the overlap integral between the modulating parts of the Bloch functions  $| l_i \vec{k}_i \rangle$  and  $| l_j \vec{k}_j \rangle$ , which are normalized over the whole crystal. It is more convenient to rewrite  $H_{e-e}$  as

$$H_{e-e} = \frac{1}{4} \sum_{\substack{1234\\1234}} \Gamma_0(1234) a_1^{\dagger} a_2^{\dagger} a_3 a_4 , \qquad (2.5)$$

where  $i \ (=1,2,3,4)$  stands for the abbreviation  $i = l_i \vec{k}_i \sigma_i$  and

$$\Gamma_{0}(1\,2\,3\,4) = \frac{1}{V} [\mathscr{V}(\vec{k}_{1} - \vec{k}_{4})\langle l_{1}\vec{k}_{1} | l_{4}\vec{k}_{4}\rangle\langle l_{2}\vec{k}_{2} | l_{3}\vec{k}_{3}\rangle\Delta(\sigma_{1} - \sigma_{2}) - \mathscr{V}(\vec{k}_{1} - \vec{k}_{3})\langle l_{1}\vec{k}_{1} | l_{3}\vec{k}_{3}\rangle\langle l_{1}\vec{k}_{2} | l_{4}\vec{k}_{4}\rangle\Delta(\sigma_{1} - \sigma_{3})] \times\Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4})\Delta(\sigma_{1} + \sigma_{2} - \sigma_{3} - \sigma_{4}).$$
(2.6)

Here  $\Delta(x)=1$  for x=0 and  $\Delta(x)=0$  otherwise with x as a scalar or a vector.

Now we consider systems of the conductionband (CB) electrons and the valence-band (VB) electrons. The number operator  $\hat{N}_c$  for the CB system is defined as

$$\hat{N}_C = \sum_{\substack{i \\ l_i \in CB}} a_i^{\dagger} a_i .$$
(2.7)

With the use of the density operator  $\rho(t)$  the number  $N_C(t)$  of the CB electrons is given by

$$N_C(t) = \operatorname{Tr}[\rho(t)\hat{N}_C] . \qquad (2.8)$$

Using the well-known relation<sup>15</sup> of  $\partial \rho(t)/\partial t$  to the Hamiltonian and the nature of the trace we obtain

$$\frac{d}{dt}N_C(t) = \frac{1}{i\hbar} \operatorname{Tr}\{\rho(t)[\hat{N}_C, H]\}, \qquad (2.9)$$

where [,] is the commutator. Let us neglect the interband scattering for  $H_{e-ph}$  and  $H_{e-i}$  and consider the intraband effect only. The  $\hat{N}_C$  is commutable with all the terms in Eq. (2.1) except  $H_{e-e}$ . We obtain

$$[\hat{N}_{C},H] = [\hat{N}_{C},H_{e-e}]$$

$$= \frac{1}{4} \sum_{1234} \Gamma_{0}(1234) \Delta_{1234} a_{1}^{\dagger} a_{2}^{\dagger} a_{3} a_{4}$$

$$\equiv H_{I}^{c}, \qquad (2.10)$$

where

$$\Delta_{1234} = \Delta(l_1 - l_C) + \Delta(l_2 - l_C) - \Delta(l_3 - l_C) - \Delta(l_4 - l_C)$$
(2.11)

with  $l_C$  denoting the CB. The last equality in Eq. (2.10) is a definition. It is to be noted that  $H_I^c$  differs from  $H_{e-e}$  by the factor  $\Delta_{1234}$ . Equation (2.9) is rewritten as

$$\frac{d}{dt}N_C(t) = \frac{1}{i\hbar} \operatorname{Tr}[\rho(t)H_I^c] . \qquad (2.12)$$

Let (1234) designate the bands to which  $a_1^{\dagger}, a_2^{\dagger}$ ,  $a_3$ , and  $a_4$  refer, by replacing 1, 2, 3, and 4 with C for the CB or V for the VB. From Eq. (2.10) we see that  $\hat{N}_C$  is commutable with those terms of  $H_{e-e}$  that have creation and annihilation operators in pairs belonging to the same bands. A sum of these terms and that of the residual terms are denoted as  $H_{e-e}^{p}$  and  $H_{I} + H_{I}^{r}$ , respectively: The relevant terms are shown in Table I. Since  $H_I + H_I^r$  is not commutable with  $\hat{N}_C$ , it is  $H_I + H_I^r$ that causes time evolution of the number of the CB electrons. We see that  $H_I$  and  $H_I^r$  represent one-electron and two-electron interband transitions, respectively. The former transition is the Auger process in question, while the latter is inconsistent with the energy conservation requirement. In view of this we write  $H = H_0 + H_I$ , where

H <sup>p</sup> <sub>e-e</sub>	(a)	(CVCV) (CVVC) (VCCV) (VCVC)
	(b)	(CCCC) (VVVV)
$H_I$	(c)	(CVVV) (VCVV) (VVCV) (VVVC)
	(d)	(VCCC) (CVCC) (CCVC) (CCCV)
$H_I'$	(e)	(CCVV) (VVCC)

TABLE I. Possible terms of  $H_{e-e}(=H_{e-e}^p+H_I+H_I^r)$ .

# $H_0 = H_e + H_{ph} + H_{e-ph} + H_{e-i} + H_{e-e}^p + H_I^r$ .

Now we consider  $H_0$  and  $H_I$  to be the unperturbed Hamiltonian and the perturbation, respectively, for the time-dependent operator. For an arbitrary operator A in the Schrödinger representation we define an operator  $\tilde{A}(t)$ ,

$$\widetilde{A}(t) = \exp\left[\frac{i}{\hbar}H_0t\right]A\exp\left[-\frac{i}{\hbar}H_0t\right].$$
 (2.13)

Then Eq. (2.12) is rewritten as

$$\frac{d}{dt}N_{C}(t) = \frac{1}{i\hbar} \operatorname{Tr}[\widetilde{\rho}(t)\widetilde{H}_{I}^{c}(t)] . \qquad (2.14)$$

where especially  $\tilde{\rho}(t)$  is defined as

$$\widetilde{\rho}(t) = \exp\left(\frac{i}{\hbar}H_0 t\right) \rho(t) \exp\left(-\frac{i}{\hbar}H_0 t\right). \quad (2.15)$$

Now we consider that  $H_I$  is adiabatically switched on at the time  $t = t_0$  ( $t_0 \rightarrow -\infty$ ). The following assumptions are made for an excited semiconductor at  $t = t_0$ . The CB electrons and the VB electrons belong to different systems, each of which is in a thermal equilibrium within itself and with the phonon system. Then the CB and VB systems can be described by quasi-Fermi-levels  $\mu_C$  and  $\mu_V$ , respectively, and by one temperature. Under these assumptions we give the boundary condition  $\rho(t_0) = \rho_0$  with

$$\rho_0 = \exp\left[\frac{\Omega - \overline{H}_0}{T}\right], \qquad (2.16)$$

where

$$\overline{H}_0 = H_0 - \mu_C \widehat{N}_C - \mu_V \widehat{N}_V , \qquad (2.17)$$

where  $\hat{N}_V$  is the number operator for the VB system, T the thermal energy for the electron systems as well as the phonon system, and  $\Omega$  is a constant defined through  $\text{Tr}(\rho_0)=1$ . Assuming  $H_I$  to be a small perturbation, we expand  $\tilde{\rho}(t)$  in power series of  $\tilde{H}_I(t)$  as

$$\widetilde{\rho}(t) = \rho_0 + \sum_{n=1}^{\infty} \widetilde{\rho}_n(t) . \qquad (2.18)$$

where  $\tilde{\rho}_n(t)$  is the term of the *n*th order in  $\tilde{H}_I(t)$ . Hereafter we write  $\operatorname{Tr}(\rho_0 \cdots)$  as  $\langle \rangle$ . Evidently we find  $\langle \tilde{H}_I^c(t) \rangle = 0$ . Retaining only the term of the lowest order in  $\tilde{H}_I(t)$  that does not vanish, we obtain

$$\frac{d}{dt}N_C(t) = \frac{1}{(i\not\!\!\!R)^2} \int_{t_0}^t dt_1 \langle [\widetilde{H}_I^c(t), \widetilde{H}_I(t_1)] \rangle .$$
(2.19)

The equation is rewritten using Eqs. (2.5), (2.10), and the definition of  $H_I$  as

$$\frac{d}{dt}N_{C}(t) = \frac{1}{16i\hbar^{2}} \sum_{\substack{1234\\5678}} {}^{\prime}\Gamma_{0}(1234)\Gamma_{0}(5678)\Delta_{1234} \int_{t_{0}}^{t} dt_{1}K_{4}(1234,5678;t-t_{1})\exp(-\delta|t_{1}|) .$$
(2.20)

Here  $\delta \rightarrow 0+$  and

$$K_4(1234,5678;t) = \frac{1}{i} \zeta(t) \langle [\tilde{C}_1(t), \tilde{C}_2(0)] \rangle , \qquad (2.21)$$

where  $C_1 = a_1^{\dagger} a_2^{\dagger} a_3 a_4$ ,  $C_2 = a_5^{\dagger} a_6^{\dagger} a_7 a_8$ , and  $\zeta(t)$  is the step function defined as  $\zeta(t) = 0$  for t < 0 and  $\zeta(t) = 1$  for  $t \ge 0$ . In Eq. (2.20) the factor  $\exp(-\delta |t_1|)$  is introduced to take into account the adiabatic switching and  $\Sigma'$  means restricting the summation over (5678) to cases (c) and (d) of Table I. Noting that the summation over (1234) is also restricted to these cases, we find

$$\widetilde{C}_{1}(t) = \exp\left[\frac{i}{\hbar}\mu_{\Delta}t\right] \exp\left[\frac{i}{\hbar}\overline{H}_{0}t\right] C_{1} \exp\left[-\frac{i}{\hbar}\overline{H}_{0}t\right], \qquad (2.22)$$

where  $\mu_{\Delta} = (\mu_C - \mu_V) \Delta_{1234}$ . Then Eq. (2.21) is rewritten as

$$K_4(1234,5678;t) = \exp\left[\frac{i}{\hbar}\mu_{\Delta}t\right] G_4^R(1234,5678;t) , \qquad (2.23)$$

where

$$G_4^R(1234,5678;t) = \frac{1}{i}\zeta(t) \left\langle \left[ \exp\left[\frac{i}{\hbar}\overline{H}_0 t\right] C_1 \exp\left[-\frac{i}{\hbar}\overline{H}_0 t\right], C_2 \right] \right\rangle.$$
(2.24)

Thus our subject is reduced to calculating  $G_4^R(t)$ :  $G_4^R(t)$  is the abbreviation of  $G_4^R(1234,5678;t)$ .

Let us consider  $\overline{H}_0$  in Eq. (2.24) to be the total Hamiltonian, where  $H_e + H_{\rm ph} - \mu_C \hat{N}_C - \mu_V \hat{N}_V$  and  $H_{e-\rm ph} + H_{e-\rm i} + H_{e-\rm i}^P + H_I^r$ , which constitute  $\overline{H}_0$ , are the unperturbed Hamiltonian and the perturbation, respectively. Then  $G_4^R(t)$  can be treated, noting Eq. (2.16), as the usual four-particle retarded Green's function, whose analytical properties are well known.<sup>16</sup> Defining the Fourier component  $G_4^R(\omega)$  as

$$G_4^R(\omega) = \frac{1}{\hbar} \int_{-\infty}^{\infty} dt \ G_4^R(t) \exp\left[\frac{i}{\hbar}\omega t\right]$$
(2.25)

and using the Lehmann representation, we obtain

$$G_4^R(t) = \frac{i}{\pi} \int_{-\infty}^{\infty} d\omega \operatorname{Im} G_4^R(\omega) \exp\left[-\frac{i}{\hbar} \omega t\right].$$
(2.26)

Using Eqs. (2.20), (2.23), and (2.26), we obtain after performing the integration over  $t_1$  under  $t \to \infty$ ,

$$\frac{d}{dt}N_{C}(t) = \frac{1}{8\hbar} \sum_{\substack{1234\\5678}}' \Gamma_{0}(1234)\Gamma_{0}(5678)\Delta_{1234} \text{Im}G_{4}^{R}(\mu_{\Delta}) .$$
(2.27)

[Notice  $\text{Im}G_4^R(\mu_{\Delta}) = \text{Im}G_4^R(\omega)$  with  $\omega = \mu_{\Delta}$ .] It is to be noted that in Eq. (2.20) the factor  $\exp(-\delta |t_1|)$  in the range  $t_1 > 0$  is taken only for convenience. In fact, we can also give the factor to be unity for t > 0. The  $dN_C(t)/dt$  becomes a constant given by Eq. (2.27) for t positive and sufficiently large. Thus the Auger lifetime  $\tau$  is defined through  $dN_C(t)/dt = -Vn_C/\tau$  for such t, where  $n_C$  is the excess concentration of the CB electrons.

For the calculation of  $G_4^R(t)$  it is convenient to use the four-particle Green's function

$$\mathcal{G}_{4}(\tau) = - \left\langle T_{\tau} \left[ \widetilde{a}_{1}^{\dagger}(\tau) \widetilde{a}_{2}^{\dagger}(\tau) \widetilde{a}_{3}(\tau) \widetilde{a}_{4}(\tau) \widetilde{a}_{5}^{\dagger}(0) \widetilde{a}_{6}^{\dagger}(0) \widetilde{a}_{7}(0) \widetilde{a}_{8}(0) \right] \right\rangle , \qquad (2.28)$$

where  $T_{\tau}$  is the Wick chronological operator,

$$\widetilde{a}_{i}^{\dagger}(\tau) = \exp(\tau \overline{H}_{0}) a_{i}^{\dagger} \exp(-\tau \overline{H}_{0}) ,$$
  
$$a_{i}(\tau) = \exp(\tau \overline{H}_{0}) a_{i} \exp(-\tau \overline{H}_{0}) .$$

Considering the Fourier component

$$\varphi_4(i\omega_n) = \int_0^{1/\tau} \tau d\tau \exp(i\omega_n T) \varphi_4(\tau) . \qquad (2.29)$$

with  $\omega_n = 2\pi nT$ ,  $G_4^R(\omega)$  is obtained from

$$G_4^R(\omega) = \varphi_4(\omega + i\delta) , \qquad (2.30)$$

with  $\delta \rightarrow 0+$ . In the following  $\varphi_4(i\omega_n)$  is discussed on the basis of the conventional diagram method.<sup>16</sup>

First we discuss the screening of interactions. It is pointed out that the terms of  $H_{e-e}^p + H_I^r$  of  $H_0$ shown in (a), (b), and (e) of Table I give the freecarrier (both electron and hole) screening and the host-lattice screening, respectively. However, the

interference between both screenings, which is represented by  $H_I$ , is not contained in  $H_0$ . In other words,  $H_0$  does not contain  $H_{e-e}$  in a complete form so that the screening involved in Eq. (2.19) is different from that given in Ref. 14. The latter form is correct, considering that the screening and the transition have been treated on an equal footing. In this paper the correct form should be obtained from Eq. (2.14) using the all-order terms of  $\tilde{\rho}(t)$ . This is not an easy task. As an alternative approximation we incorporate the contributions of all the terms of  $\tilde{\rho}(t)$  into the unperturbed Hamiltonian by defining the new Hamiltonian. This Hamiltonian is

$$H = H'_0 + H_I \exp(-\delta |t|),$$

where  $H'_0 = H_0 + H_I$ , which is the same as that given by Eq. (2.1). Thus  $H'_0$  contains  $H_{e-e}$  in a

complete form. Considering  $H'_0$  and  $H_I \exp(-\delta |t|)$  to be the unperturbed Hamiltonian and the perturbation, respectively, the discussion below Eq. (2.13) is repeated. The restriction here is that  $dN_C(t)/dt$  should be given up to the firstorder term of  $\tilde{\rho}(t)$  as in Eq. (2.19). If higher-order terms were taken into account, we would evaluate the screening effect to excess. Using the new Hamiltonian we obtain the screened interactions of electron-electron, electron-impurity, and electronphonon, as has been discussed in Ref. 14. The discussion on this subject is not repeated here. We neglect the electron-electron interaction caused by phonon emission followed by reabsorption and consider only the screened Coulomb interaction between electrons. Thus we replace the unscreened interaction  $\Gamma_0$  appearing in Eqs. (2.5), (2.10), (2.20), and (2.27) with the screened interaction  $\Gamma$ , which is obtained by replacing  $\mathscr{V}(q)$  in  $\Gamma_0$  [Eq. (2.6)] with

$$U(\vec{q}) = \frac{4\pi e^2}{\epsilon_0} \frac{1}{q^2 + \lambda^2} .$$
 (2.31)

The screening on  $H_I$  and  $H_I^c$  is an assumption, which will be justified by taking the higher-order terms of  $\tilde{\rho}(t)$  into account. Here  $\epsilon_0$  and  $\lambda$  are the static dielectric constant of the host lattice and the inverse screening length due to both free electrons and free holes, respectively. Practically,  $\lambda$  is given in the Thomas-Fermi approximation. From the discussion given just below Eq. (2.27) we obtain

$$\frac{1}{\tau} = -\frac{1}{n_C V} \frac{1}{8\hbar} \sum_{\substack{1234\\5678}}^{\prime\prime} \Gamma(1234) \Gamma(5678) \times \Delta_{1234} \text{Im} G_4^R(\mu_{\Delta}) .$$
(2.32)

Here  $\sum''$  means not only the restricted summation over (5678) but also the restriction that  $G_4^R(\mu_{\Delta})$  is obtained from  $\varphi_4(i\omega_n)$ , for which all the diagrams containing any electron-hole bubbles<sup>14</sup> constructed of internal lines are excluded.

In practical calculation of  $\mathcal{G}_4(i\omega_n)$  we start from  $\mathcal{G}_4(\tau)$ , which is given as a sum of the free part and the bound part. The free part is expressed as four equivalent diagrams shown in Fig. 1. As for the bound part we consider two-particle interactions as an approximation. One example is shown in Fig. 2: In addition to this there are five similar diagrams. In all these diagrams we have replaced all free-one-particle Green's functions with complete ones (seen below). It is to be noted that we have omitted those diagrams which do not represent the Auger process as shown by examples of Fig. 3. Taking into account equivalence of some diagrams, we obtain effectively

$$\begin{aligned}
\varphi_{4}(\tau) &= -4\varphi(7,1\tau)\varphi(8,2\tau)\varphi(3\tau,5)\varphi(4\tau,6) - 4\varphi(7,1\tau)\varphi(3\tau,5)\varphi_{2}^{b}(4\tau,8;2\tau+\delta,6\delta) \\
&+ 4\varphi(7,2\tau)\varphi(3\tau,5)\varphi_{2}^{b}(4\tau,8;1\tau+\delta,6\delta) + 4\varphi(7,1\tau)\varphi(4\tau,5)\varphi_{2}^{b}(3\tau,8;2\tau+\delta,6\delta) \\
&- 4\varphi(7,2\tau)\varphi(4\tau,5)\varphi_{2}^{b}(3\tau,8;1\tau+\delta,6\delta) + 4\varphi(7,1\tau)\varphi(8,2\tau)\varphi_{2}^{b}(3\tau+\delta,4\tau;5\delta,6) \\
&+ 4\varphi(3\tau,5)\varphi(4\tau,6)\varphi_{2}^{b}(7\delta,8;1\tau+\delta,2\tau) .
\end{aligned}$$
(2.33)

Here  $\varphi(i,j\tau)$  is the one-particle temperature Green's function defined as

$$\varphi(i,j\tau) = - \langle T_{\tau}(\widetilde{a}_{i}(0)\widetilde{a}_{j}^{\dagger}(\tau)) \rangle \equiv \varphi(i,j;-\tau) .$$

We also define

$$\varphi(i\tau,j) = -\langle T_{\tau}(\widetilde{a}_i(\tau)\widetilde{a}_j(0))\rangle \equiv \varphi(i,j;\tau) ,$$

and



FIG. 1. Free part of the four-particle Green's function, which represents AR.

$$\mathscr{G}_{2}(i\tau,j;k\tau+\delta,l\delta) = \langle T_{\tau}[\widetilde{a}_{i}(\tau)\widetilde{a}_{j}(0)\widetilde{a}_{k}^{\dagger}(\tau+\delta)\widetilde{a}_{l}^{\dagger}(\delta)] \rangle$$

with  $\delta \rightarrow 0+$ , is the two-particle temperature Green's function and  $\varphi_2^b$  represents the bound part of it. The first term and the residual terms of Eq. (2.33) are the free part and the bound part, respectively, of  $\varphi_4(\tau)$ .

To calculate Eq. (2.33) it is convenient to give



FIG. 2. An example of the bound part of the fourparticle Green's function, which represents AR.



FIG. 3. Examples of the free part and the bound part of the four-particle Green's function, which do not represent AR.

the one-particle temperature Green's function in terms of the retarded one-particle Green's function  $G^{R}(i,j;\omega)$ , which is defined as

$$G^{R}(i,j;\omega) = \frac{1}{\hbar} \int_{-\infty}^{\infty} dt \ G^{R}(i,j;t) \exp\left[\frac{i}{\hbar}\omega t\right]$$
(2.34)

with

Use of the Lehmann representation under 
$$\tau > 0$$
  
leads to

$$\varphi(i\tau,j) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega [1 - \theta(\omega)] \mathrm{Im} G^{R}(i,j;\omega) \exp(-\omega\tau)$$
(2.36)

 $=\frac{1}{i}\zeta(t)\left\langle \left[\exp\left[\frac{i}{\hbar}\overline{H}_{0}t\right]a_{i}\exp\left[-\frac{i}{\hbar}\overline{H}_{0}t\right],a_{i}^{\dagger}\right]\right\rangle .$ 

and

 $G^{R}(i,j;t)$ 

$$\varphi(i,j\tau) = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \,\theta(\omega) \mathrm{Im} G^{R}(i,j;\omega) \exp(\omega\tau) , \qquad (2.37)$$

where

$$\theta(\omega) = \frac{1}{\exp\left(\frac{\omega}{T}\right) + 1}$$
(2.38)

is the Fermi-Dirac distribution. As for  $\varphi_2^b$  we have a general expression,<sup>16</sup>

$$= -\int_{0}^{1/T} d\tau_{1'} \int_{0}^{1/T} d\tau_{2'} \int_{0}^{1/T} d\tau_{3'} \int_{0}^{1/T} d\tau_{4'} \sum_{1'2'3'4'} \varphi(1\tau_{1}, 1'\tau_{1'}) \varphi(2\tau_{2}, 2'\tau_{2'}) \\ \times \widetilde{\Gamma}(1'\tau_{1'}, 2'\tau_{2'}, 3'\tau_{3'}, 4'\tau_{4'}) \varphi(3'\tau_{3'}, 3\tau_{3}) \varphi(4'\tau_{4'}, 4\tau_{4}) ,$$
(2.39)

where  $\tilde{\Gamma}$  represents the vertex parts. For the calculation of  $\varphi_2^b$  we neglect vertex corrections giving

$$\Gamma(1'\tau_{1'}, 2'\tau_{2'}, 3'\tau_{3'}, 4'\tau_{4'}) = \Gamma(1'2'3'4')\delta(\tau_{1'} - \tau_{4'})\delta(\tau_{1'} - \tau_{2'})\delta(\tau_{3'} - \tau_{4'}) .$$
(2.40)

Neglecting the interband scattering, the summation over (1'2'3'4') is on the wave vectors and the spins. To further facilitate the calculation we consider the intraband and spin-conserving processes for the phonon scattering and for the impurity scattering. Then we have

$$G^{R}(i,j;\omega) = G^{R}(l_{i}\vec{k}_{i},l_{i}\vec{k}_{j};\omega)\Delta(l_{i}-l_{j})\Delta(\sigma_{i}-\sigma_{j}) .$$

The wave vector is not conserved in the presence of randomly distributed impurities. However, it has been shown<sup>11</sup> that in the limit of  $V \rightarrow \infty$  the Green's function is equal to its ensemble average over all the impurity sites. Then the wave vector conservation is restored giving  $G^R(l\vec{k}_i, l\vec{k}_j; \omega) = G^R(l\vec{k}_i, \omega)\Delta(\vec{k}_i - \vec{k}_j)$ , where  $G^R(l\vec{k}_i, \omega)$  is the retarded Green's function obtained for the average impurity field in the above sense. Therefore we obtain

$$G^{R}(i,j;\omega) = G^{R}(l_{i}\vec{k}_{i},\omega)\Delta(l_{i}-l_{j})\Delta(\vec{k}_{i}-\vec{k}_{j})\Delta(\sigma_{i}-\sigma_{j}) .$$
(2.41)

From the definitions (2.17) and (2.35) it is found that the energy in  $G^{R}(l_{i}\vec{k}_{i},\omega)$  should be measured from the quasi-Fermi-level relevant to the band  $l_{i}$ . With the use of Eqs. (2.29), (2.30), (2.32), (2.33), and (2.36)-(2.41), we obtain the formula for the AR lifetime. We see that for (1234) of  $H_{I}^{c}$  (CVVV), (VCVV), (VVCV), and (VVVC) are one equivalent quartet in contribution and (VCCC), (CVCC), (CCVC), and (CCCV) are the other. The former and the latter are designated as (VVVC) process and (VCCC) process, respectively. For each process we obtain

(2.35)

$$\frac{1}{\tau} = \frac{2}{n_C V \hbar \pi^3} \left[ \exp\left[\frac{\mu_C - \mu_V}{T}\right] - 1 \right]$$

$$\times \sum_{\vec{k}_1 \vec{k}_2 \vec{k}_3 \vec{k}_4} \int d\omega_1 \int d\omega_2 \int d\omega_3 \int d\omega_4 \delta(\omega_1 + \omega_2 - \omega_3 - \omega_4 - \mu_C + \mu_V)$$

$$\times \theta_1 \theta_2 (1 - \theta_3) (1 - \theta_4) \prod_{i=1}^4 \operatorname{Im} G^R(i) \left[ S_0 + \sum_{n=1}^6 S_n \right], \qquad (2.42)$$

where

$$S_{0} = \sum_{\text{spin}} \Gamma(1234)\Gamma(3412) , \qquad (2.43)$$

$$S_{1} = \frac{1}{\pi^{2}} \sum_{\substack{\vec{k}_{5} \vec{k}_{6} \\ \text{spin}}} [\Gamma(1234)\Gamma(4526)\Gamma(3615) + \text{c.c.}]$$

$$\times \int d\omega_5 \int d\omega_6 \frac{1}{\omega_2 + \omega_6 - \omega_4 - \omega_5} (\theta_5 - \theta_6) \operatorname{Im} G^R(5) \operatorname{Im} G^R(6) , \qquad (2.44)$$

$$S_{2} = \frac{1}{\pi^{2}} \sum_{\substack{\vec{k}_{5} \neq \vec{k}_{6} \\ \text{spin}}} [\Gamma(1234)\Gamma(4516)\Gamma(3625) + \text{c.c.}] \times \int d\omega_{5} \int d\omega_{6} \frac{1}{\omega_{1} + \omega_{6} - \omega_{4} - \omega_{5}} (\theta_{5} - \theta_{6}) \text{Im} G^{R}(5) \text{Im} G^{R}(6) , \qquad (2.45)$$

$$S_{3} = \frac{1}{\pi^{2}} \sum_{\substack{\mathbf{k}_{5} \mathbf{k}_{6} \\ \text{spin}}} [\Gamma(1234)\Gamma(3516)\Gamma(4625) + \text{c.c.}] \times \int d\omega_{5} \int d\omega_{6} \frac{1}{\omega_{2} + \omega_{6} - \omega_{3} - \omega_{5}} (\theta_{5} - \theta_{6}) \text{Im} G^{R}(5) \text{Im} G^{R}(6) , \qquad (2.46)$$

$$S_{4} = \frac{1}{\pi^{2}} \sum_{\substack{\vec{k}_{5} \mid \vec{k}_{6} \\ \text{spin}}} [\Gamma(1234)\Gamma(3526)\Gamma(1234) + \text{c.c.}] \times \int d\omega_{5} \int d\omega_{6} \frac{1}{\omega_{1} + \omega_{6} - \omega_{3} - \omega_{5}} (\theta_{5} - \theta_{6})\text{Im}G^{R}(5)\text{Im}G^{R}(6) , \qquad (2.47)$$

$$S_{5} = -\frac{1}{\pi^{2}} \sum_{\substack{\vec{k}_{5} \mid \vec{k}_{6} \\ spin}} [\Gamma(1234)\Gamma(3456)\Gamma(5612) + c.c.] \times \int d\omega_{5} \int d\omega_{6} \frac{1}{\omega_{3} + \omega_{4} - \omega_{5} - \omega_{6}} (1 - \theta_{5} - \theta_{6}) \text{Im} G^{R}(5) \text{Im} G^{R}(6) , \qquad (2.48)$$

$$S_{6} = -\frac{1}{\pi^{2}} \sum_{\substack{\vec{k}_{5} \mid \vec{k}_{6} \\ \text{spin}}} [\Gamma(1234)\Gamma(3456)\Gamma(5612) + \text{c.c.}] \times \int d\omega_{5} \int d\omega_{6} \frac{1}{\omega_{1} + \omega_{2} - \omega_{5} - \omega_{6}} (1 - \theta_{5} - \theta_{6})\text{Im}G^{R}(5)\text{Im}G^{R}(6) .$$
(2.49)

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In the above equations we have used the abbreviations  $\theta_i = \theta(\omega_i)$  and  $G^R(i) = G^R(l_i \vec{k}_i, \omega_i)$  and c.c. means the complex conjugate. In Eqs. (2.44) – (2.49), 5 and 6 represent the intermediate states to which the intraband carrier-carrier scattering occurs from pairs (2,4), (1,4), (2,3), (1,3), (3,4), and (1,2) for  $S_1$ ,  $S_2$ ,  $S_3$ ,  $S_4$ ,  $S_5$ , and  $S_6$ , respectively. The factor

$$\left| \exp\left[\frac{\mu_C - \mu_V}{T}\right] - 1 \right| \theta_1 \theta_2 (1 - \theta_3) (1 - \theta_4)$$

can be rewritten as

$$(1-\theta_1)(1-\theta_2)\theta_3\theta_4 - \theta_1\theta_2(1-\theta_3)(1-\theta_4) .$$
(2.50)

This is the statistical factor correctly characterizing occupancy and vacancy of states for the transition  $(3,4) \rightarrow (1,2)$  and the reverse processes.

All the formulas for PCAR, PHAR, and IAR, which have been used, can be derived from  $S_0$  in Eq. (2.42). The residual terms represent correction for the electron correlation effect. Considering  $S_0$  alone, we give here a formula for the AR lifetime  $\tau_0$  in a more tractable form. We define a way of writing  $\Gamma(1234)=(f,g)$  which means

$$\Gamma(1234) = V^{-1}[f\Delta(\sigma_1 - \sigma_4) - g\Delta(\sigma_1 - \sigma_3)]\Delta(\vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4)\Delta(\sigma_1 + \sigma_2 - \sigma_3 - \sigma_4) .$$

From Eq. (2.6) we have

$$f = U(\vec{k}_1 - \vec{k}_4) \langle l_1 \vec{k}_1 | l_4 \vec{k}_4 \rangle \langle l_2 \vec{k}_2 | l_3 \vec{k}_3 \rangle$$
(2.51)

and

$$g = U(\vec{k}_1 - \vec{k}_3) \langle l_1 \vec{k}_1 | l_3 \vec{k}_3 \rangle \langle l_2 \vec{k}_2 | l_4 \vec{k}_4 \rangle , \qquad (2.52)$$

noting that  $\mathscr{V}$  should be replaced with U defined by Eq. (2.31). Summation over all possible spin states gives

$$S_{0} = \frac{2}{V^{2}} (|f|^{2} + |g|^{2} + |f-g|^{2}) \Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4}) .$$
(2.53)

Converting  $\sum_{\vec{k}} \cdots$  to  $[V/(2\pi)^3] \int d\vec{k} \cdots$ , we obtain<sup>17</sup>

$$\frac{1}{\tau_0} = \frac{4}{n_C \hbar \pi^3} \left[ \exp\left[\frac{\mu_C - \mu_V}{T}\right] - 1 \right]$$

 $\times \int d\omega_1 \int d\omega_2 \int d\omega_3 \int d\omega_4 \delta(\omega_1 + \omega_2 - \omega_3 - \omega_4 - \mu_C + \mu_V) \theta_1 \theta_2 (1 - \theta_3) (1 - \theta_4)$ 

$$\times \int \frac{d\vec{k}_{1}}{(2\pi)^{3}} \int \frac{d\vec{k}_{2}}{(2\pi)^{3}} \int \frac{d\vec{k}_{3}}{(2\pi)^{3}} (|f|^{2} + |g|^{2} + |f-g|^{2}) \prod_{i=1}^{4} \operatorname{Im} G^{R}(i) , \quad (2.54)$$

with  $\vec{k}_4 = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$ . This expression should be the same as that in Ref. 14 but there is a difference concerning the statistical factors contained. The factor in Ref. 14 reads

$$\theta_4[\theta_3(1-\theta_1)(1-\theta_2)+(1-\theta_3)\theta_1\theta_2]$$

for (VVVC) process. This factor is by a difference of  $(1-\theta_3)\theta_1\theta_2$  larger than the correct one given by Eq. (2.50). However, the difference is unimportant since we usually have  $\theta_3=1$  for (VVVC) process in a good approximation. The difference arises from the fact that in Ref. 14 the lifetime is calculated from the imaginary part of the self-energy, which contains the damping indifferent to the excesscarrier lifetime.

Let us now take into account the correction due to the electron correlation effect by rewriting Eq. (2.42) as

$$\frac{1}{\tau} = \frac{1}{\tau_0} \left[ 1 + \sum_{n=1}^{6} \phi_n \right], \qquad (2.55)$$

where  $\phi_n$  comes from  $S_n$ . Since accurate calculation of  $\phi_n$  is too difficult to be practical, it is desir-

able that we can use an approximation  $\tau = \tau_0$ . In this hope we estimate  $\phi_n$  in the next section on the specified processes as follows. We consider the heavy-hole band (HB), the light-hold band (LB), and the spin split-off band (SB) as the VB. For (*VVVC*) process we restrict the discussion to (*HHSC*) process, which is known to be dominant<sup>8,18</sup> under  $E_G \ge \Delta_0$ , where  $E_G$  and  $\Delta_0$  are the band-gap energy and the spin split-off energy, respectively. For (*VCCC*) process we consider (*HCCC*) process only. Both processes are shown in Fig. 4. Using the free-particle retarded Green's function  $G_0^R$  the estimation of  $S_n$  is made under an approximation

$$\operatorname{Im} G^{R}(i) = \operatorname{Im} G^{R}_{0}(i)$$
$$= -\pi \delta(\omega_{i} - \xi_{i}) \text{ for } i = 5 \text{ and } 6$$

where  $\xi_i = \xi_{l_i \vec{k}_i}$  is defined as the band energy measured from the relevant quasi-Fermi level. Another approximation is to replace  $\omega_i$  (i=1,2,3,4) in  $S_n$ with  $\xi_i$ . The above approximations are adopted in view of our main interest, which is in electron correlation effect on the lifetime: The effect of the level broadening on the correction  $S_n$  may be subsidiary. As for the bands we assume spherical energy surfaces, which are characterized by the effective masses  $m_C$ ,  $m_H$ , and  $m_S$  for CB, HB, and SB, respectively. The overlap integrals are estimated using the relations<sup>19,20</sup>

$$\langle l_{\nu}\vec{\mathbf{k}}_{i} | l_{H}\vec{\mathbf{k}}_{j} \rangle = \vec{\mathbf{w}}_{\nu H} \cdot (\vec{\mathbf{k}}_{i} - \vec{\mathbf{k}}_{j})$$
(2.56)

and

$$|\vec{\mathbf{w}}_{\nu H} \cdot \vec{\mathbf{k}}| = w_{\nu} k \tag{2.57}$$

with v = C or S. Here  $l_C$ ,  $l_S$ , and  $l_H$  designate CB, SB, and HB, respectively.  $\vec{w}_{vH}$  and  $w_v$  are con-



FIG. 4. AR in (VVVC) process (a) and (VCCC) process (b).

stants. We take  $\langle l\vec{k}_i | l\vec{k}_j \rangle = 1$  for the intraband overlap integral. The overlap integrals, which make the calculation quite tedious, are given in more simplified forms using the above relations.

#### **III. CORRECTION TERMS**

### A. (VVVC) process

We discuss (HHSC) process in direct-gap materials as shown in Fig. 4(a). It is found that  $S_1$  and  $S_2$  come from the interaction between the CB electrons (4) and the HB holes (1 and 2) with  $\phi_1 = \phi_2$  while  $S_3$  and  $S_4$  from the interaction between the SB electrons (3) and the HB holes (1 and 2) with  $\phi_3 = \phi_4$ . Thus interactions are between carriers in different bands. For  $S_1$  we define  $\Gamma(4526) = (f_1,g_1)$  and  $\Gamma(3615) = (f_2,g_2)$ . We consider only the intraband scattering. This corresponds to taking  $\langle l_5 \vec{k}_5 | l_6 \vec{k}_6 \rangle = 0$  so that we have  $g_1 = 0$ . Using the approximations given at the end of the last section we obtain

$$S_{1} = \frac{2}{V^{2}} \int \frac{d\vec{k}_{5}}{(2\pi)^{3}} [(f-g)(f_{1}-g_{1})(f_{2}-g_{2})-gf_{1}f_{2}+c.c.] \\ \times \frac{1}{\xi_{2}+\xi_{6}-\xi_{4}-\xi_{5}} [\theta(\xi_{5})-\theta(\xi_{6})]\Delta(\vec{k}_{1}+\vec{k}_{2}-\vec{k}_{3}-\vec{k}_{4})$$
(3.1)

under  $\vec{k}_6 = \vec{k}_5 + \vec{k}_1 - \vec{k}_3$ . As for  $S_3$  we make new definitions  $\Gamma(3516) = (f_1, g_1)$  and  $\Gamma(4625) = (f_2, g_2)$  and obtain  $g_1 = 0$ . Then the expression for  $S_3$  is formally given from Eq. (3.1) under replacement  $\xi_4 \rightarrow \xi_3$  and  $\vec{k}_6 = \vec{k}_5 + \vec{k}_2 - \vec{k}_3$ .

For  $S_6$ , which comes from the HB hole-hole (1 and 2) interaction, we define  $\Gamma(3456) = (f_3, g_3)$  and  $\Gamma(5612) = (f_4, g_4)$ . We obtain

$$S_{6} = -\frac{2}{V^{2}} \int \frac{d\vec{k}_{5}}{(2\pi)^{3}} [(f-g)(f_{1}-g_{1})(f_{2}-g_{2}) - f(f_{3}g_{4}+g_{3}f_{4}) + \text{c.c.}] \\ \times \frac{1}{\xi_{1}+\xi_{2}-\xi_{5}-\xi_{6}} [1-\theta(\xi_{5})-\theta(\xi_{6})]\Delta(\vec{k}_{1}+\vec{k}_{2}-\vec{k}_{3}-\vec{k}_{4}), \qquad (3.2)$$

under  $\vec{k}_6 = \vec{k}_1 + \vec{k}_2 - \vec{k}_5$ . As for  $S_5$ , which comes from the interaction between the CB electrons (4) and the SB electrons (3), we obtain the expression from  $S_6$  under replacment  $\xi_1 + \xi_2 \rightarrow \xi_3 + \xi_4$  and  $\vec{k}_6 = \vec{k}_3 + \vec{k}_4 - \vec{k}_5$ . Definitions of  $(f_3, g_3)$  and  $(f_4, g_4)$  are the same as above.

## B. (VCCC) process

We discuss (HCCC) process in direct-gap materials as shown in Fig. 4(b). It is found that  $S_1$  and  $S_3$  come from the interaction between the energetic (2) and the nonenergetic (3 and 4) electrons with  $\phi_1 = \phi_3$  while  $S_2$  and  $S_4$  from the interaction between the nonenergetic CB electrons (3 and 4) and the HB holes (1) with  $\phi_2 = \phi_4$ .  $S_5$  comes from the nonenergetic electron-electron (3 and 4) interaction.  $S_6$  comes from the interaction between the HB holes (1) and the energetic CB electrons (2). The definitions of  $(f_1,g_1)$ ,  $(f_2,g_2)$ ,  $(f_3,g_3)$ , and  $(f_4,g_4)$ are the same as those given in A. We consider only small-energy scattering and neglect largeenergy scattering. This corresponds to taking  $\langle l_5 \mathbf{k}_5 | l_6 \mathbf{k}_6 \rangle = 0$  so that we have  $g_1 = 0$ . Thus the expression for  $S_n$  is the same as that given in A.

# C. The correction effect on PCAR in (*VVVC*) process for $E_G \gg \Delta_0$

The PCAR lifetime is obtained by a special choice of  $G^{R}(i) = G_{0}^{R}(i)$  for i = 1, 2, 3, and 4. The

threshold energy for the (*VVVC*) process is given as  $\eta(E_G - \Delta_0)$ , where  $E_G$ ,  $\Delta_0$ , and  $\eta$  are the band-gap energy, the spin split-off energy, and a constant of the order of 0.1, respectively. We consider the case of  $\eta(E_G - \Delta_0) \gg T$ . Then some approximations can be made to evaluate the overlap integrals considering the threshold condition. This condition is given<sup>1</sup> as  $\vec{k}_1 = \vec{k}_2 = (b/2)\vec{k}_3$  and  $\vec{k}_4 = (b-1)\vec{k}_3$ , where  $b = (1 + \mu_H/2)^{-1}$  with  $\mu_H = m_C/m_H$ . For direct gap materials we have  $\mu_H \ll 1$  so that we have approximately b=1 or

$$\vec{k}_1 = \vec{k}_2 = \frac{1}{2}\vec{k}_3, \quad \vec{k}_4 = 0,$$
  
 $k_3 = [2m_S(E_G - \Delta_0)/\hbar^2]^{1/2}$ 
(3.3)

as the threshold condition. Since the overlap integrals in  $S_n$  are found to be slowly varying functions of  $\vec{k}_i$  (i=1,2,3,4) as compared with the statistical factor in Eq. (2.42), they are taken out of the integral in  $S_n$  and evaluated for the threshold condition (3.3).

For  $S_1$  we have  $\vec{k}_5 \sim \vec{k}_2$   $(l_5 = l_H)$  and  $\vec{k}_6 \sim \vec{k}_4$  $(l_6 = l_C)$  so that we give  $\vec{k}_5 = \vec{k}_2$  and  $\vec{k}_6 = \vec{k}_4$ . Taking into account the condition (3.3), we find f = g. Noting  $f_1 = U(\vec{k}_5 - \vec{k}_2)$  and

$$\xi_{2} + \xi_{6} - \xi_{4} - \xi_{5} = \frac{\hbar^{2}}{2m_{C}} (k_{4}^{2} - k_{6}^{2}) + \frac{\hbar^{2}}{2m_{H}} (k_{2}^{2} - k_{5}^{2})$$

we obtain

$$S_{1} = -\frac{8}{V^{2}\vec{n}^{2}} \frac{m_{H}m_{C}}{m_{H}+m_{C}} |f|^{2} \int \frac{d\vec{k}_{5}}{(2\pi)^{3}} U(\vec{k}_{5}-\vec{k}_{2}) \frac{1}{|\vec{k}_{5}-\vec{k}_{2}|^{2}+(\vec{k}_{5}-\vec{k}_{2})\cdot\vec{k}_{24}} \Theta_{1}\Delta(\vec{k}_{1}+\vec{k}_{2}-\vec{k}_{3}-\vec{k}_{4}),$$
(3.4)

where  $\Theta_1 = \theta(\xi_5) - \theta(\xi_6)$  and  $\vec{k}_{24} = (m_C \vec{k}_2 + m_H \vec{k}_4)/(m_H + m_C)$ . Considering extreme cases of nondegeneracy or degeneracy for CB and VB,  $\Theta_1$  is a constant. Then we obtain

$$S_{1} = -\Theta_{1} |f|^{2} \frac{m_{H}}{m_{H} + m_{C}} \frac{8}{V^{2} \pi a_{C} k_{24}} F\left[\frac{\lambda}{k_{24}}\right] \Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4}) , \qquad (3.5)$$

where  $a_C = \hbar^2 \epsilon_0 / (m_C e^2)$  corresponds to the Bohr radius and F(y) is defined as

$$F(y) = \int_0^\infty dx \frac{x}{x^2 + y^2} \ln \left| \frac{x + 1}{x - 1} \right| \,. \tag{3.6}$$

In a similar way we obtain

$$S_{3} = -\Theta_{3} |f|^{2} \frac{m_{H}}{m_{H} - m_{S}} \frac{8}{V^{2} \pi a_{S} k_{23}} F\left[\frac{\lambda}{k_{23}}\right] \Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4}) .$$
(3.7)

Here  $\Theta_3 = \theta(\xi_5) - \theta(\xi_6)$ , where  $l_5 = l_H$  and  $l_6 = l_C$ , is taken to be a constant as above. We define  $a_S = \hbar^2 \epsilon_0 / (m_S e^2)$  and  $\vec{k}_{23} = (m_C \vec{k}_2 + m_H \vec{k}_3) / (m_H + m_C)$ .

As for  $S_6$  we take  $\vec{k}_5 = \vec{k}_1$  and  $\vec{k}_6 = \vec{k}_2$  as an approximation. Thus the overlap integrals are evaluated for the condition (3.3), giving f = g and  $f_3 = g_3$ . Noting  $f_4 = U(\vec{k}_5 - \vec{k}_2)$  and  $g_4 = U(\vec{k}_6 - \vec{k}_2)$  we obtain

$$S_6 = \Theta_6 |f|^2 \frac{16}{V^2 \pi a_H k_{21}} F\left[\frac{\lambda}{k_{21}}\right] \Delta(\vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4)$$

where  $\Theta_6 = 1 - \theta(\xi_5) - \theta(\xi_6)$  is taken to be a constans,  $\vec{k}_{21} = \vec{k}_2 - \vec{k}_1$ , and  $a_H = \hbar^2 \epsilon_0 / (m_H e^2)$ . We neglect  $S_5$ , which is small as compared to  $S_6$ .

Inserting  $S_n$ 's thus obtained into Eq. (2.42) we determine  $\phi_n$ . To facilitate the calculation we give a simulation function for F(y) as

$$F(y) = \frac{\pi^2/2}{1 + (\pi/2)y} , \qquad (3.9)$$

which agrees with the original function in the limits of  $y \rightarrow 0$  and  $y \rightarrow \infty$  and within an error of 10% in all the range of y. As for  $\phi_1$  we make an approximation

$$\vec{k}_{24} = \vec{k}_1 + \vec{k}_2 - b \vec{k}_3$$
,

which is valid under  $m_H \gg m_C$ . As for  $S_3$  an approximation is

$$\vec{k}_{23} = (m_H + m_S/2)\vec{k}_3/(m_H - m_S)$$
,

which is obtained for the condition (3.3). Let us define a function

$$H(y) = \int_0^1 dx \frac{x^2}{x+y} (1-x^2)^{1/2} , \qquad (3.10)$$

which is simulated by

$$H(y) = \frac{\pi/16}{y + (3\pi/16)} . \tag{3.11}$$

Using the function we calculate  $\phi_n$  on the basis of the method similar to that used<sup>20</sup> in the analysis of PCAR, assuming nondegenerate statistics. We approximately obtain the following under  $m_H \gg m_C$ and  $m_H \gg m_S$ :

$$\phi_1 = \phi_2 = -\frac{2}{a_C \left[\lambda + \frac{3}{8} \left[\frac{5ab}{2\beta_S}\right]^{1/2}\right]} \Theta_1,$$
(3.12)

$$\phi_3 = \phi_4 = -\frac{\pi}{a_S \left[k_S + \frac{\pi}{2}\lambda\right]} \Theta_3 , \qquad (3.13)$$

$$\phi_6 = \frac{4}{a_H \left[\lambda + \frac{3}{8} \left[\frac{5a}{\mu_H \beta_S}\right]^{1/2}\right]} \Theta_6 , \qquad (3.14)$$

where

$$\begin{split} k_{S} = & [2m_{S}(E_{G} - \Delta_{0})/\hbar^{2}]^{1/2} , \\ a = & \mu_{S} - 1 + b , \ \mu_{S} = & m_{C}/m_{S} \end{split}$$

and

$$\beta_S = \hbar^2 / (2m_S T)$$

We have  $\Theta_1 = 1$ ,  $\Theta_3 = 0$ , and  $\Theta_6 = -1$ , which is obtained for nondegenerate statistics. We see that the electron-hole interaction ( $\phi_1$  and  $\phi_2$ ) and the hole-hole interaction ( $\phi_6$ ) reduce the AR rate. The latter effect is especially predominant due to  $m_H \gg m_C$ . Using the data of the band parameters for GaAs, we find  $\sum \phi_n = -2$  to -5 for the carrier concentrations below  $10^{18}$  cm<sup>-3</sup> and for the temperatures between 77 and 500 K. Therefore the conventional calculation of AR lifetime neglecting the correction effect is not valid. Since the effect is too large, more rigorous treatment of the electron correlation is required.

In the case when degenerate statistics is assumed for VB, we cannot give simple formulas for  $\phi_n$ 's. Roughly speaking, the hole-hole interaction effect is predominant giving  $\phi_6 \sim (a_H \lambda)^{-1}$  (note  $\Theta_6 = 1$ ). This shows an enhancement of the AR rate in contrast with the case of nondegenerate statistics. We find that the electron correlation effect is too large for the present treatment to be justified for the carrier concentrations up to about  $10^{19}$  cm<sup>-3</sup>.

# D. The correction effect on IPHAR in (*VVVC*) process for $E_G \gg \Delta_0$

The threshold energy for IPHAR is reduced to zero in the presence of the impurity and/or phonon scattering. The reduction is caused by violation of the wave vector conservation. As an approximation we consider the scattering effect only for HB, for which the effective mass is far larger than those for CB and SB. Then the threshold condition is given<sup>8</sup> as  $\vec{k}_1 = \vec{k}_4 = 0$  and  $\vec{k}_2 = \vec{k}_3$  or alternatively  $\vec{k}_2 = \vec{k}_4 = 0$  and  $\vec{k}_1 = \vec{k}_3$ , where  $k_3$  is just given in Eq. (3.3). Under the condition the overlap integrals are evaluated as in C. We use the same definition of  $\Theta_n$  as in C, considering extreme cases of nondegeneracy or degeneracy again.

For  $S_1$  we find f=0,  $g_1=0$ , and  $gf_1g_2 = U(\vec{k}_5 - \vec{k}_2) |g|^2$ . Retaining a dominant term we obtain

(3.8)

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$$S_{1} = \Theta_{1} \frac{4m_{C}}{V^{2} \hbar^{2}} |g|^{2} \int \frac{d\vec{k}}{(2\pi)^{3}} U(\vec{k}) \left[ \frac{1}{k^{2} + \mu_{H} |\vec{k} + \vec{k}_{3}|^{2}} + \frac{1}{(1 + \mu_{H})k^{2} + 2\mu_{H} \vec{k} \cdot \vec{k}_{3}} \right] \Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4}).$$
(3.15)

Usually we have  $k_3 \gg \lambda$  so that we approximately obtain

$$\phi_1 = \Theta_1 \left[ \left[ \mu_H / (1 + \mu_H) \right]^{1/2} + \frac{2}{\pi} \right] \frac{1}{a_H k_S} .$$
(3.16)

 $\phi_3$  is negligible as compared to  $\phi_1$ . As for  $S_6$  we obtain

$$S_{6} = -\Theta_{6} \frac{4m_{H}}{V^{2} \hbar^{2}} \int \frac{d\vec{k}}{(2\pi)^{3}} \left[ U(\vec{k}) [U(\vec{k}) - 2U(\vec{k} - \vec{k}_{3})] \frac{1}{k^{2} + |\vec{k} - \vec{k}_{3}|^{2}} + U(\vec{k} + \vec{k}_{3}) [U(\vec{k} + \vec{k}_{3}) - 2U(\vec{k})] \frac{1}{k^{2} + |\vec{k} + \vec{k}_{3}|^{2}} \right] \Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4}) \times (g\vec{w}_{CH} \cdot \vec{k} \vec{w}_{SH} \cdot \vec{k} + \text{c.c.}) .$$

$$(3.17)$$

It is to be noted that here the overlap integrals containing  $\vec{k}_5$  and  $\vec{k}_6$  cannot be evaluated for the threshold condition as in C. If this were done, the integrals would be zero. The last factor in Eq. (3.17) appears owing to this situation. Using the relation

 $|g\vec{\mathbf{w}}_{CH}\cdot\vec{\mathbf{k}}\vec{\mathbf{w}}_{SH}\cdot\vec{\mathbf{k}}+\text{c.c.}|<2|g||\vec{\mathbf{w}}_{CH}\cdot\vec{\mathbf{k}}||\vec{\mathbf{w}}_{SH}\cdot\vec{\mathbf{k}}|=2|g|w_{C}w_{S}k^{2},$ 

the upper limit of  $S_6$  is found. We obtain

$$|\phi_6| < \frac{10}{\pi} \frac{1}{a_H k_S} . \tag{3.18}$$

 $\phi_5$  is negligible as compared to  $\phi_6$ . For GaAs we find  $|\phi_1| = 0.15$  and  $|\phi_6| < 0.55$ . Thus  $|\sum \phi_n|$  is at most 0.85. This may be somewhat an overestimation. In view of this the neglect of the correction terms is not so bad an approximation. This situation is in remarkable contrast with that for PCAR. The difference is ascribed to that between the threshold conditions for PCAR and for IPHAR: For the former the correction is mainly due to the interaction between holes having nearly the same wave vectors, while for the latter the interaction is predominantly between holes having very different wave vectors.

## E. The correction effect for (VCCC) process

For (VCCC) process the threshold condition is nearly the same for PCAR and for IPHAR. For the former the threshold condition is given<sup>1</sup> as  $\vec{k}_3 = \vec{k}_4 = (\mu_H/2)ck_2$  and  $\vec{k}_1 = \vec{k}_2$ , where  $c = (\mu_H + \frac{1}{2})^{-1}$ . Noting  $\mu_H \ll 1$  and  $k_2 = (2m_C E_G/\hbar^2)^{1/2}$  we can approximately give the condition as  $\vec{k}_3 = \vec{k}_4 = 0$  and  $\vec{k}_1 = \vec{k}_2$ . This is also the threshold condition for IPHAR as far as the effect of impurity and phonon scattering is considered only for HB. Thus the correction  $S_n$  is evaluated as equal for both PCAR and IPHAR. We take  $\langle l\vec{k}_i | l\vec{k}_j \rangle = 1$  only for the intraband overlap integrals where  $|\vec{k}_i - \vec{k}_j|$  is small.

As for  $S_2$  we have f = g and  $g_1 = 0$  under the threshold condition. We approximately obtain

$$S_{2} = -\Theta_{2} \frac{4m_{C}}{V^{2} \hbar^{2}} \int \frac{d\vec{k}_{6}}{(2\pi)^{3}} U(\vec{k}_{6}) U(\vec{k}_{6} + \vec{k}_{2}) \frac{1}{k_{6}^{2} + \mu_{H} |\vec{k}_{6} + \vec{k}_{2}|} \times [g\vec{w}_{CH} \cdot (\vec{k}_{6} + \vec{k}_{2}) + \text{c.c.}] \Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4}), \qquad (3.19)$$

where  $\Theta_2 = \theta(\xi_5) - \theta(\xi_6)$  is taken to be a constant with  $l_5 = l_H$  and  $l_6 = l_C$ . Using  $|g\vec{w}_{CH}\cdot\vec{k} + c.c| \le 2|g|w_Ck$ , we obtain

$$|\phi_2| < \frac{1}{a_C k_C} , \qquad (3.20)$$

where  $k_C = (2m_C E_G / \hbar^2)^{1/2}$ . For GaAs we find  $\phi_2 < 0.06$ .  $\phi_1$  is comparable to  $\phi_2$ . As for  $\phi_5$  we obtain in a similar way

$$|\phi_5| < \frac{4}{\pi} \frac{1}{a_C k_C}$$
 (3.21)

For GaAs we find  $\phi_5 \leq 0.08$ .  $\phi_6$  is negligible as compared to  $\phi_5$  since the interaction is between energetic carriers. Thus we obtain  $|\sum \phi_n| < 0.2$ . This may be somewhat an overestimation. In view of this the neglect of the correction terms is a good approximation for both PCAR and IPHAR.

## F. Correction effect in (VVVC) process for $E_G \leq \Delta_0$

At first we consider the case of  $E_G \simeq \Delta_0$ . Then the threshold condition for both PCAR and IPHAR is given approximately by  $\vec{k}_1 = \vec{k}_2 = \vec{k}_3 = \vec{k}_4$ . Then we may take f = g. Noting equivalence of holes 1 and 2 in contribution we obtain

$$S_{6} = -\Theta_{6} \frac{8m_{H}}{V^{2} \tilde{H}^{2}} \int \frac{d\vec{k}}{(2\pi)^{3}} U(\vec{k}) \frac{1}{k^{2} + \vec{k} \cdot \vec{k}_{12}} \times [fU(\vec{k} + \vec{k}_{2} - \vec{k}_{3})\vec{w}_{CH} \cdot (\vec{k} + \vec{k}_{2} - \vec{k}_{3})\vec{w}_{SH} \cdot (\vec{k} + \vec{k}_{2} - \vec{k}_{3}) + c.c.] \times \Delta(\vec{k}_{1} + \vec{k}_{2} - \vec{k}_{3} - \vec{k}_{4}).$$

We note that in its contribution to the integrations of Eqs. (3.22) and (2.42) the range near around  $k = \lambda$  and  $k_i = (2m_i T/\hbar^2)^{1/2}$  (i=1,2,3,4) is important, giving  $k_i \ge \lambda$ . We approximately evaluate the quantity in the square brackets of Eq. (3.22) by giving  $\vec{k} = 0$  and take it out of the integral. That quantity becomes  $2 |g|^2$ . We find that the expression for  $S_6$  is given just by Eq. (3.8). However, it is to be noted that this expression is useful for both PCAR and IPHAR in the present case. For PCAR under nondegenerate statistics  $\phi_6$  is given by Eq. (3.14). More generally we can give

$$\phi_6 = -\frac{4}{a_H \lambda + \eta_H a_H (2m_H T/\hbar^2)^{1/2}}$$
(3.23)

and

$$\phi_6 = \frac{4}{a_H \lambda + \eta_d a_H k_F} , \qquad (3.24)$$

assuming nondegenerate statistics and degenerate statistics, respectively, for VB. Here  $\eta_n$  and  $\eta_d$  are constants of the order of unity, and  $k_F$  is the magnitude of the Fermi wave vector for HB. Other  $\phi_n$ 's are negligible as compared to  $\phi_6$ , showing that the hole-hole interaction effect is predominant. From Eqs. (3.23) and (3.24) we find the correction effect to be so large that the present treatment is not valid. This results from the fact that at the threshold the interaction can be between holes having nearly the same wave vectors.

Next we consider the case of  $\Delta_0 - E_G \gg T$ . Then (*HHLC*) process is the dominant AR process. This case is analogous to that of (*VVVC*) process under  $E_G - \Delta_0 \gg T$  so that the same conclusion is reached.

#### IV. DISCUSSION AND CONCLUSION

The correction effect on AR is important especially for (VVVC) process in the following cases: PCAR under  $|E_G - \Delta_0| >> T$  and IPHAR as well as PCAR under  $E_G \simeq \Delta_0$  as far as  $\Theta_6$  is well approximated as +1 or -1. The correction is so large that the present treatment of the electron correlation is not valid. More rigorous treatment of the four-particle Green's function is necessary taking into account higher-order terms. However, in a doping range where the Fermi level is around the relevant band edge, we have  $\Theta_6 \sim 0$  so that the correction may be small. In many III-V compounds of p type this corresponds to the acceptor concentrations ranging roughly from  $10^{18}$  to  $10^{19}$  $cm^{-3}$ . In fact we find a considerable agreement between the theory<sup>13</sup> and the experiments<sup>21</sup> for ptype GaSb at 77 K for the acceptor concentrations ranging from  $4 \times 10^{18}$  to  $1 \times 10^{19}$  cm<sup>-3</sup> as follows. The Auger coefficient, which is calculated from

(3.22)

Eq. (2.54) assuming (HHSC) process of IPHAR, is  $3 \times 10^{-29}$  cm<sup>6</sup>s<sup>-1</sup> while the experimental value is  $2.5 \times 10^{-29}$  cm<sup>6</sup>s<sup>-1</sup>. Outside the above range of the acceptor concentration the discrepancy between the theory and the experiments tends to be large with increasing or decreasing concentration.

We have been considering PCAR and IPHAR as if they could be treated separately. Actually, however, Eq. (2.42) is understood to involve both PCAR and IPHAR effects. Let us consider PHAR for  $E_G - \Delta_0 \gg T$  under nondegenerate statistics, for which a calculation<sup>14</sup> based on Eq. (2.54) shows a considerably rapid increase of the AR rate with temperature. At low temperatures PHAR is dominant but PCAR becomes comparable to PHAR with increasing temperature. In view of this and of the reduction due to the large correction effect on PCAR, the AR rate may be-

- <sup>1</sup>A. R. Beatie and P. T. Landsberg, Proc. R. Soc. London, Ser. A <u>249</u>, 16 (1959).
- <sup>2</sup>For review articles see: P. T. Landsberg, Phys. Status Solidi A <u>41</u>, 457 (1970); A. Haug, in *Festkörperprobleme XII*, Advances in Solid State Physics, edited by O. Madelung (Pergamon, Braunschwieg, 1972), p. 411; R. Conradt, *ibid.*, p. 411; P. T. Landsberg and M. J. Adams, J. Lumin. <u>7</u>, 3 (1973).
- <sup>3</sup>L. Huldt, Phys. Status Solidi A <u>8</u>, 173 (1971).
- <sup>4</sup>L. Huldt, Phys. Status Solidi A <u>24</u>, 221 (1974).
- <sup>5</sup>M. Takeshima, J. Appl. Phys. <u>46</u>, 3082 (1975).
- <sup>6</sup>D. Hill and P. T. Landsberg, Proc. R. Soc. London, Ser. A <u>347</u>, 547 (1976).
- <sup>7</sup>G. Benz and R. Conradt, Phys. Rev. B <u>16</u>, 843 (1977).
- <sup>8</sup>W. Lochmann, Phys. Status Solidi A <u>40</u>, 285 (1977).
- <sup>9</sup>W. Lochmann, Phys. Status Solidi A <u>42</u>, 181 (1977).
- <sup>10</sup>W. Lochmann, Phys. Status Solidi A <u>45</u>, 423 (1978).
- <sup>11</sup>M. Takeshima, Phys. Rev. B 23, 771 (1981).
- <sup>12</sup>W. Lochmann and A. Haug, Solid State Commun. <u>35</u>, 553 (1980).
- <sup>13</sup>M. Takeshima, Phys. Rev. B <u>25</u>, (1982).
- <sup>14</sup>M. Takeshima, Phys. Rev. B <u>23</u>, 6625 (1981).

come less dependent on temperature than that based on Eq. (2.54) if the correction effect is correctly taken into account.

Though we have been considering IPHAR alone, it is evident that the discussion can be extended to some other scatterings which violate the wave vector conservation. It is concluded that AR can be analyzed on the basis of Eq. (2.54) neglecting the correction effect for (*VVVC*) process only under some restrictions and for (*VCCC*) process without any restrictions.

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- <sup>15</sup>D. ter Haar, in *Reports on Progress in Physics* (Benjamin, New York, 1969).
- <sup>16</sup>V. L. Bonch-Bruevich and S. L. Tyablikov, *The Green Function Method in Statistical Mechanics* (North-Holland, Amsterdam, 1962); A. A. Abrikosov, L. P. Gor'kov, and L. E. Dzyaloshinskii, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Englewood Cliffs, 1965); S. Doniach and E. H. Sondheimer, *Green's Function for Solid State Physicists* (Benjamin, London, 1974).
- <sup>17</sup>There are errors in Ref. 14. All the theoretical inverse lifetimes given there should be multiplied by  $\pi^{-1}$ . All the calculated Auger coefficients should also be multiplied by  $\pi^{-1}$ .
- <sup>18</sup>M. Takeshima, J. Appl. Phys. <u>43</u>, 4114 (1972).
- <sup>19</sup>E. Antončik and P. T. Landsberg, Proc. Phys. Soc. London 82, 337 (1963).
- <sup>20</sup>A. R. Beatie and G. Smith, Phys. Status Solidi <u>19</u>, 577 (1967).
- <sup>21</sup>A. N. Titkov, G. V. Benemanskaya, B. L. Gelmont, G. N. Iluridthe, and Z. N. Sokolova, J. Lumin. <u>24/25</u>, 697 (1981).