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Ambipolar diffusion of high-density electrons and holes in Ge, Si, and GaAs: Many-body effects

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Many-body effects are incorporated into a theory for the density dependence of the electron-hole ambipolar diffusion coefficient in semiconductors. Self-energy shifts of the free-carrier band edges lead to a band-gap gradient in the presence of a carrier-density gradient and therefore a diffusion coefficient which is less than that obtained from the independent-particle Boltzmann transport theory. The diffusion coefficient decreases with increasing carrier density until carrier degeneracy becomes important, after which the coefficient increases with density as in the independent-particle theory. The difference between the two theories is most apparent for high-effective-mass semiconductors and low carrier temperatures. Results are calculated for Ge, Si, and GaAs for common lattice and carrier temperatures of 100 and 300 K, with silicon showing the largest influence from many-body effects.

I. INTRODUCTION

High-density nonequilibrium electrons and holes can be produced in semiconductors through interaction with pulsed laser, ion, and electron beams. The dynamics of the carriers so produced has been the subject of numerous experimental and theoretical investigations during the past decade.[1–5] Apart from the fundamental research reasons, much of the motivation for these studies derives from the development of new semiconductor devices and semiconductor processing techniques such as laser annealing.[6–10] If the semiconductor plasmas are initially produced in a shallow layer it becomes important in many cases to take into account the spatial and temporal evolution of the carrier distribution through diffusion. In this paper we theoretically investigate diffusion of high-density electrons and holes and consider the influence of many-body effects on the ambipolar diffusion coefficient. In particular we derive expressions for the density dependence of the ambipolar diffusivity in semiconductors and apply the formalism to calculating values in Ge, Si, and GaAs.

The simplest approach to carrier diffusion is to assume that the electrons and holes are each characterized by their own density and temperature-independent diffusion coefficients, with the actual diffusion of both carriers determined by a constant ambipolar diffusion coefficient. However, for sufficiently high densities such that carrier degeneracy is important, the carrier kinetic energy increases with density. One would therefore expect that the ambipolar diffusion coefficient would differ from its low-density value. If the carriers can be considered to move independently (i.e., neglecting many-body effects), the Boltzmann transport theory in the relaxation-time approximation (hereafter referred to as IPBTT) predicts a rapid increase in the diffusion coefficient beyond the onset of carrier degeneracy,[11] which for many semiconductors occurs at a density of approximately 10^{19} cm^{-3} at room temperature.

In general, data on the high-density diffusivity in semiconductors is scarce with only a few isolated results reported, mostly in germanium. Using picosecond ellipsometric techniques, Auston and Shank[12] have reported that for a surface carrier density of 1.7×10^{20} cm^{-3} the ambipolar diffusivity in germanium at 300 K is 4 times its low-density value, in reasonable agreement with IPBTT. Jamison *et al.*,[13] however, have reported that the diffusivity does not differ substantially from its low-density value in the density range $10^{19}-10^{20}$ cm^{-3}, whereas Moss *et al.*,[14] using transient-grating techniques, report that for a surface carrier density of 5×10^{19} cm^{-3} the diffusivity is actually smaller than the low-density diffusivity for lattice temperatures of 135 and 295 K. These apparent inconsistencies can occur for at least two reasons. Because the carrier lifetime at these high densities can be subnanosecond,[15] recombination can compete with diffusion in causing density reduction, rendering quantitative separation of the two processes difficult in many experiments, with the exception, perhaps, of transient-grating experiments. Secondly, the diffusivity of high-density photogenerated carriers can be influenced by thermal effects since it is well known that the carrier diffusivity increases with carrier temperatures but decreases with lattice temperature.[16]

Notwithstanding the difficulties in procuring accurate data on the high-density diffusivity, there is reason to suspect that the values obtained would not be in agreement with IPBTT. At high carrier densities the independent-particle band picture of semiconductors will break down as carrier exchange and correlation effects become important. Many-body effects have long been recongnized, primarily through band-gap narrowing, to influence optical absorption,[17] luminescence,[18] and semiconductor laser operation.[19] Their possible influence on transport properties could occur through alteration of the carrier scattering time, effective-mass renormalization, and band-gap narrowing. Meyer and Glicksman[20] have reported that increased carrier-carrier scattering at high densities has a strong effect on the carrier drift mobility, i.e., the response of carriers to an applied electric field. However, Meyer, in a recent theoretical paper,[21] has concluded that, at least in first order, the ambipolar diffusivity would not be similarly affected. Since in ambipolar flow, in the absence of an applied field, both types of carriers travel in the same direction, the increase in carrier-carrier scattering with increased density does not occur. Similarly, it has been demonstrated[22] that effective-mass renormalization is not significant, except, perhaps for carrier densities in excess of 10^{21} cm^{-3}. Alteration of the band structure at high densities due to the self-energy shifts of the dressed carriers, or quasiparticles, can therefore be viewed, to a good approximation, as rigid shifts of the independent-particle bands. These shifts can certainly influence the diffusivity since, if a carrier-density gradient is present, a band-gap gradient will also be present as well, and each carrier type will experience a macroscopic force towards the high-density region. Many-body effects, primarily through band-gap narrowing, will therefore lead to a diffusivity which is smaller than that given by IPBTT. Wautelet[23] originally pointed out the importance of these effects for silicon. However, he has only included the exchange contribution to the self-energy and has adopted an inconsistent statistical mechanical description of the carriers, which leads to unphysical values of the diffusion coefficient. We will include correlation energy contributions to the self-energy, adopt a more conventional description of the carrier-distribution functions, and present results for the ambipolar diffusivity for Si, Ge, and GaAs for densities up to 10^{20} cm^{-3}.

It should be noted that band-gap narrowing and band-gap gradients can be induced by lattice heating as well, and to a greater or lesser degree will be present during and after carrier creation. This is also partially due to a many-body effect because of the increased carrier-phonon interaction at high temperature. The effect of thermal gradients on carrier diffusivity has been treated recently by Van Vechten and Wautelet,[24] Wautelet,[23] and van Driel *et al.*[25] In this paper we will not explicitly consider thermal effects although we will briefly compare the relative magnitudes of the thermal and carrier-density-induced influences on the diffusivity under various circumstances.

The remainder of this paper is outlined as follows. In Sec. II we discuss the framework for a statistical mechanical description of high-density electron-hole distribution. Section III presents a summary of the many-body corrections to the independent-particle band structure as calculated using the single plasmon pole approximation for the dielectric function. The ambipolar diffusivity of the carriers is derived in Sec. IV from the Boltzmann transport equation in the relaxation-time approximation with appropriate corrections for many-body effects. The theory is applied to calculate the density dependence of the diffusivity for Ge, Si, and GaAs in Sec. V for common lattice and carrier temperatures of 300 and 100 K. Our results are compared with IPBTT and the recent theory proposed by Wautelet.

II. NONEQUILIBRIUM CARRIER DISTRIBUTIONS

The transport properties of carriers in semiconductors depend on the details of the carrier-energy-distribution function. A considerable effort has been put forward in recent years to obtain this function for high-density nonequilibrium carriers which are produced by pulsed excitation. There are obviously many different parameters which are necessary to describe the state of a semiconductor plasma, and in general, they reach their equilibrium values on different time scales. For example, numerous authors[1,2,25,26] have indicated that the typical momentum relaxation time is 10^{-14} sec, the thermalization time (time required to establish a carrier temperature) is approximately the same time, the energy relaxation time (time required for the carriers and the phonons to reach a common temperature) is a few picoseconds, and the electron-hole pair recombination time can be as short as a few picoseconds and as long as seconds. Clearly the nature of the distribution function depends on the time scale that one wishes to characterize the plasma. On a time scale of picoseconds or longer, however, one should be able to describe the carriers by Fermi-Dirac distribution functions with their own temperature and chemical potentials which vary with time. This quasiequilibrium approach to nonequilibrium plasmas has been used since the introduction of quasi-Fermi-levels by Shockley[27] and its validity rests on the restriction that one not make observations on the plasma over a time scale which is long compared to the appropriate parameter's relaxation time. Because of rapid Coulomb scattering events, the electrons and holes will be described by the same temperature. However, the chemical potentials used to characterize the two distribution functions will be different from each other and their common equilibrium value. This latter condition is simply related to the fact that, on a time scale which is short compared to the carrier lifetime, the electrons (in the conduction band) and the holes (in the valence band) do not have access to each other's states. Entropy considerations therefore dictate that they have separate chemical potentials.

With the above general comments in mind the electron-distribution function is given by

$$f_e(E)=\frac{1}{e^{(1/k_B T_e)(E-F_e)}+1}\ , \tag{1}$$

where F_e is the chemical potential and T_e is the electron temperature. Because the present work considers a spatially inhomogeneous band gap, the energy E and chemical potential are referenced to a spatially invariant energy level such as the vacuum level and not the band-edge energy E_c. If one assumes a parabolic conduction-band structure, the electron density is given by

$$N_e=2\left[\frac{2\pi m_{\rm de}^{*}k_B T_e}{h^2}\right]^{3/2}\mathscr{F}_{1/2}(\eta_e)\ , \tag{2}$$

where $m_{\rm de}^{*}$ is the density of states effective mass, $\eta_e\ [=(F_e-E_c)/k_B T_e]$ is the reduced chemical potential, and $\mathscr{F}_r$ is the Fermi-Dirac integral of order r.[11] The corresponding hole-distribution function f_h and density N_h can be obtained similarly using the valence-band-edge energy E_v, the hole chemical potential F_h, density of states effective mass $m_{\rm dh}^{*}$, and reduced chemical potential, η_h $[=(E_v-F_h)/k_B T_e]$. For these nonequilibrium carrier distributions, the electron and hole densities are related by the fact that they are equal and not by the law of mass action which applies only to equilibrium situations. The quantities F_e, F_h, and T_e are determined by a knowledge of the total energy and density of the plasma. The carrier temperature will differ from the lattice temperature only during and within picoseconds of the termination of the excitation pulse and only in the region of excitation-pulse absorption. The two chemical potentials will obviously approach the common equilibrium value on the time scale of the carrier lifetime which for high densities is usually determined by Auger recombination.

Although this view of nonequilibrium carrier distributions is widely accepted, it is not universally accepted. Van Vechten and Wautelet[24,26] and Yoffa[4] have offered a different description which employs only a carrier temperature to characterize the electron and hole distributions. Both carrier chemical potentials are taken to be equal to the common, equilibrium chemical potential, which has a value near the center of the band gap. Using Eq. (2) with silicon as an example, this implies that as the carrier density increases from 10^{12} to 10^{21} cm^{-3}, the carrier temperature would be required to increase from 300 to 3000 K, independent on the details on any external excitation process. For this single chemical potential description to be valid, carrier recombination processes must be exactly balanced by generation processes for all densities at different locations within sample. For example, Auger recombination would be balanced by impact ionization. For densities as high as 10^{21} cm^{-3} the Auger lifetime is approximately 10^{-11} sec (Refs. 2 and 15) in a typical semiconductor and it increases with decreasing density as N^{-2}.[28] Auger recombination can severely limit the density, but is unlikely to induce substantial heating of the carriers since energy relaxation occurs at a faster rate. It is also argued that screening of the carrier-phonon interaction can reduce the rate of energy transfer from the carriers to the lattice so as to maintain the carrier temperature at a high value. However, it has recently been pointed out by several authors[29] that even at densities as high as 10^{21} cm^{-3} the carrier-phonon interaction is reduced by less than an order of magnitude. In what follows, therefore, we will adopt the more conventional approach to nonequilibrium carriers in our derivation of the influence on many-body effects on the ambipolar diffusivity.

III. MANY-BODY EFFECTS AND THE CARRIER BAND STRUCTURE

Over the past twenty-five years there have been several experimental and theoretical papers concerning the influence of many-body effects on the single-particle band structure in both intrinsic[18,30] and extrinsic[17,19,22,31] semiconductors. Because of the complexity of the problem it is perhaps not surprising that different approaches have led to different results.[32] In addition, for extrinsic semiconductors it is often difficult to separate carrier-induced effects from impurity-induced effects such as lattice distortion. For intrinsic situations the theoretical treatment, at least, is complicated by the presence of carriers in both the conduction and valence bands. Much of the interest in this case has centered around the electron-hole droplets which can exist in several semiconductors near liquid-helium temperatures.

It is not the purpose here to attempt to develop another theory of many-body calculations in semiconductors. Rather we will use the approach of Lundqvist[33] which has seen much success, certainly in intrinsic semiconductors, in both electron-hole droplet physics[34] and the gain and absorption spectrum of high-density plasma in direct-gap semiconductors.[35] In the Lundqvist model the inverse dielectric constant is approximated by a single plasmon pole and the results are essentially equivalent to those of the random-phase approximation. For a quasiparticle of momentum $\vec{k}$ and energy ω in the ith band, the self-energy associated with exchange and correlation Coulomb energy corrections is given by

$$\Sigma^i(\vec{k},\omega)=\frac{4\pi e^2 i}{(2\pi)^4\epsilon_0}\int\frac{d\omega' d\vec{k}'}{k'^2}\epsilon^{-1}(\vec{k}',\omega')\times G^i(\vec{k}+\vec{k}',\omega+\omega')\,, \tag{3}$$

where $\epsilon^i(\vec{k},\omega)$ is the wave vector and energy-dependent dielectric constant of the electron-hole plasma, ϵ_0 is the static dielectric constant, and $G^i(\vec{k},\omega)$ is the single quasiparticle propagator. In the single plasmon pole approximation the dielectric constant is given by

$$\epsilon^{-1}(\vec{k},\omega)=1+\frac{\omega_p^2}{\omega^2-\omega_1^2(\vec{k})}\,, \tag{4}$$

where ω_p is the zero wave vector plasmon resonances frequency which depends on the electron-hole pair density N and the electron and hole optical masses m^*_{oe} and m^*_{oh} through

$$\omega_p^2=\frac{4\pi n e^2}{\epsilon_0}\left[\frac{1}{m^*_{\text{oe}}}+\frac{1}{m^*_{\text{oh}}}\right]. \tag{5}$$

The electron optical mass is related to the transverse m_t^* and the longitudinal m_l^* effective masses by $m^{*-1}_{\text{oe}}=\frac{1}{3}(2/m_t^*+1/m_l^*)$ while the hole optical mass is given in terms of the light (m_{lh}) and heavy (m_{hh}) hole effective masses by $m^{*-1}_{\text{oh}}=\frac{1}{2}(1/m_{\text{hh}}+1/m_{\text{lh}})$. The wave-vector dependence of the plasmon frequency ω_1 is given to a good approximation[33] by

$$\omega_1^2(\vec{k})=\omega_p^2+ak^2+bk^4\,, \tag{6}$$

where the various parameters are defined by $a=\omega_p^2/k_s^2$ and $b=\hbar^2/4(m^{*-1}_{\text{de}}+m^{-1}_{\text{dh}})$ with k_s being the screening length. We have neglected damping of the plasmon resonance. In what follows we use the zeroth-order independent-particle propagator given by

$$G_0^i(\vec{k},\omega)=\frac{f_{\vec{k}}}{\omega-\omega_{\vec{k}}-i\eta}+\frac{1-f_{\vec{k}}}{\omega-\omega_{\vec{k}}+i\eta}\,, \tag{7}$$

with $f_{\vec{k}}$ being the appropriate carrier-distribution function, η a small frequency, and $\hbar\omega_k$ the energy of a carrier in a single-particle state of wave vector $\vec{k}$. The energy integration in Eq. (3) gives

$$\Sigma^i(\vec{k},\omega)=\frac{-e^2}{2\pi^2\epsilon_0}\left\{\int_{\vec{k}'}\frac{d\vec{k}'}{k'^2}\left[f_{\vec{k}+\vec{k}'}+\omega_p^2\left[\frac{f_{\vec{k}+\vec{k}'}}{(\omega_{\vec{k}+\vec{k}'}-\omega)^2-\omega_1^2(\vec{k}')}-\frac{1}{2\omega_1(\vec{k}')[\omega-\omega_1(\vec{k}')-\omega_{\vec{k}+\vec{k}'}]}\right]\right]\right\}. \tag{8}$$

For electrons the integration over k is over the various N_c valleys of a multivalley semiconductor. We do not consider the effects of nonparabolicity of the energy bands but rather consider a constant density of states effective mass which for ellipsoidal constant-energy surfaces is given by $(m_t^2 m_l)^{1/3}$ for electrons. For most semiconductors the hole constant-energy surfaces are sufficiently close to spherical so that the longitudinal and transverse effective masses are about the same. For holes $m_{\mathrm{dh}}=(m_{\mathrm{hh}}^{3/2}+m_{\mathrm{lh}}^{3/2})^{2/3}$, the effect of the anisotropy of the band structure itself on the self-energy corrections for particles of arbitrary $\vec{\mathrm{k}}$ is small,[36] yielding a correction no larger than 15% for $m_t/m_e \simeq 20$, as for electrons in germanium. This correction will be ignored below.

The integrations in Eq. (8) were carried out for both Maxwell-Boltzmann (MB) and totally degenerate ($T_e=0$) Fermi-Dirac (FD) statistics. For the MB case the self-energy correction to the conduction-band edge is given by

$$\Sigma_{\text{band edge}}^{\text{MB}}=\frac{-4\pi N e^2}{k_{\text{th}}^2\epsilon_0}+\frac{4\pi^{3/2}e^2 k_{\text{DH}}N}{k_{\text{th}}^3\epsilon_0}e^{(k_{\text{TF}}/k_{\text{th}})^2}\operatorname{erfc}\left[\frac{k_{\text{DH}}}{k_{\text{th}}}\right]-\frac{2e^2}{\pi\epsilon_0}\left[\frac{m_{\text{ed}}^*\omega_p}{2\hbar}\right]^{1/2}\int_0^\infty\frac{ds}{\Omega(\Omega+s)^2}\,, \tag{9}$$

where

$$k_{\text{th}}=\left[\frac{2\pi m_{\text{de}}^* k_B T_e}{h^2}\right]^{1/2}$$

is the thermal wave vector,

$$k_{\text{DH}}^2=32\pi e^2 N/\epsilon_0 k_B T_e$$

is the Debye-Hückel screening length,

$$\Omega=(1+S_0 S^2+S^4)\,,$$

and

$$S_0=\frac{2m_{\text{de}}^*\omega_p}{\hbar k_{\text{TF}}^2}\,.$$

The corresponding expression for the FD case is

$$\Sigma_{\text{band edge}}^{\text{FD}}=-\frac{2e^2k_F}{\pi\epsilon_0}+\frac{2e^2}{\pi\epsilon_0}k_{\text{TF}}\tan^{-1}\left[\frac{k_F}{k_{\text{TF}}}\right]-\frac{2e^2}{\pi\epsilon_0}\left[\frac{m_{\text{de}}^*\omega_p}{2\hbar}\right]^{1/2}\int_0^\infty\frac{ds}{\Omega(\Omega+S^2)}\,, \tag{10}$$

where $k_F=(3\pi^2 N/N_c)^{1/3}$ is the Fermi wave vector and

$$k_{\text{TF}}^2=12e^2/\epsilon_0\hbar^2\pi(\pi/3)^{2/3}N^{1/3}(m_{\text{de}}^*N_c^{2/3}+m_{\text{dh}}^*)$$

is the Thomas-Fermi screening length. Similar expressions exist for the valence band edge. In each case the first term is associated with the exchange energy, while the last two terms are associated with correlation energies.

In the intermediate degeneracy regime where neither the FD nor MB results are strictly applicable, one can smoothly interpolate between the two expressions since the two functions agree to within about 20% for all densities. This also indicates that self-energy shifts of the two bands are not extremely sensitive to carrier temperature. For all densities the shift in the conduction-band edge is negative while that of the valence-band edge is positive, leading to a reduction of the band gap with increasing density.

IV. THE AMBIPOLAR DIFFUSIVITY

To calculate the diffusivity we will use the IPBTT with suitable modifications for the self-energy of the carriers. As mentioned in the Introduction, for densities less than 10^{21} cm^{-3} one can consider the quasiparticle effective mass to be density independent to a good approximation. The Boltzmann equation in the relaxation-time approximation can be written as

$$\frac{f_{\vec{\mathrm{k}}}(\vec{\mathrm{r}})-f_{0_{\vec{\mathrm{k}}}}(\vec{\mathrm{r}})}{\tau_{\vec{\mathrm{k}}}}=\vec{\mathrm{V}}\cdot\vec{\nabla}_{\vec{\mathrm{r}}}f_{0_{\vec{\mathrm{k}}}}(\vec{\mathrm{r}})+\frac{1}{h}\vec{\mathrm{F}}\cdot\vec{\nabla}_{\vec{\mathrm{k}}}f_{0_{\vec{\mathrm{k}}}}(\vec{\mathrm{r}})\,, \tag{11}$$

where $f_{\vec{\mathrm{k}}}$ and $f_{0_{\vec{\mathrm{k}}}}$ are the instantaneous and quasiequilibrium carrier-distribution functions, respectively, which are dependent on position $\vec{\mathrm{r}}$ and wave vector $\vec{\mathrm{k}}$. $\tau_{\vec{\mathrm{k}}}$ is the carrier momentum relaxation time which depend on $\vec{\mathrm{k}}$ through the particle kinetic energy, $\tau_{\vec{\mathrm{k}}}\propto(E_k-E_{c,v})^p$, and $\vec{\mathrm{F}}$ is a macroscopic force on the carriers. The macroscopic force $\vec{\mathrm{F}}$ can be represented as

$$\vec{\mathrm{F}}\equiv\hbar\frac{d\vec{\mathrm{k}}}{dt}=-\vec{\nabla}_{\vec{\mathrm{r}}}E_{\vec{\mathrm{k}}}-e\vec{\mathrm{E}}\,, \tag{12}$$

where we have considered it to result from an internal force, related to the gradient of a carrier energy and a macroscopic electric field $\vec{\mathrm{E}}$. The internal force is a natural consequence of any $\vec{\mathrm{r}}$ dependence of N and therefore $E_{c,v}$. The solution of the linearized Boltzmann equation is therefore

$$\begin{aligned} f_{\vec{k}}(\vec{r}) &= f_{0\vec{k}}(\vec{r}) - \frac{\tau_{\vec{k}}}{k_B T_e} f_{0\vec{k}} \vec{V} \cdot \left[\frac{E_{\vec{k}} - F_e}{T_e} \vec{\nabla}_{\vec{r}} T_e + \vec{F} + \vec{\nabla}_{\vec{r}} F_e \right] \\ &= f_{0\vec{k}}(\vec{r}) - \frac{\tau_{\vec{k}}}{k_B T_e} f_{0\vec{k}} \vec{V} \cdot \left[\frac{E_{\vec{k}} - F_e}{T_e} \vec{\nabla}_{\vec{r}} T_e - e\vec{E} + \vec{\nabla}_{\vec{r}} (F_e - E_{\vec{k}}) \right], \end{aligned} \tag{13}$$

for the electrons, with a similar expression for the holes. The macroscopic electric current density for either type of carrier can be found by integration of the velocity distribution function so that

$$\vec{J}_{e,h}(\vec{r}) = \mp e \int_{\vec{k}} \vec{V} f_{\vec{k}}(\vec{r}) d\vec{k} , \tag{14}$$

where the + sign corresponds to holes and the − sign to electrons. It is usually the case in intrinsic materials that the momentum relaxation time is determined by electron-phonon interactions so that $p = -\frac{1}{2}$.[37] Meyer[21] has shown, in particular, that carrier-carrier scattering is small in the case of ambipolar carrier flow. If we ignore temperature gradients, Eqs. (2), (11), and (13) can be combined to give

$$\begin{aligned} \vec{J}_e &= ek_B T_e \mu_e^0 \frac{\mathcal{F}_0(\eta_e)}{\mathcal{F}_{-1/2}(\eta_e)} \vec{\nabla} N_e \\ &\quad + eN_e \mu_e^0 \frac{\mathcal{F}_0(\eta_e)}{\mathcal{F}_{1/2}(\eta_e)} \left[\frac{\vec{\nabla} E_c}{e} + \vec{E} \right], \\ \vec{J}_h &= -ek_B T_e \mu_h^0 \frac{\mathcal{F}_0(\eta_h)}{\mathcal{F}_{-1/2}(\eta_h)} \vec{\nabla} N_h \\ &\quad + eN_h \mu_h^0 \frac{\mathcal{F}_0(\eta_h)}{\mathcal{F}_{1/2}(\eta_h)} \left[\frac{\vec{\nabla} E_v}{e} + \vec{E} \right], \end{aligned} \tag{15}$$

where $\mu_{e,h}^0 = e\langle \tau_{\vec{k}} \rangle / m_{e,h}^*$ is the low-density (MB) carrier mobility. Note that the only difference between the expression for the current density presented here and that of the IPBTT is the appearance of the band-edge gradient term. For either type of carrier its effect, as expected, is similar to that of an electric field with one important difference. The gradients of the two band edges have opposite signs so that the electrons and holes would be driven in the same direction, the direction that would tend to minimize their energy. The general carrier mobilities $\mu_{e,h}$ are related to the corresponding diffusivities by the generalized Einstein relation,

$$\begin{aligned} \mu_{e,h} &= \frac{eD_{e,h}^0}{k_B T} \frac{\mathcal{F}_0(\eta_{e,h})}{\mathcal{F}_{1/2}(\eta_{e,h})} \\ &= \mu_{e,h}^0 \frac{\mathcal{F}_0(\eta_{e,h})}{\mathcal{F}_{1/2}(\eta_{e,h})} , \end{aligned} \tag{16}$$

where $D_{e,h}^0$ is the MB diffusion coefficient. The charge currents are then given by

$$\begin{aligned} \vec{J}_{e,h} &= \pm eD_{e,h}^0 \frac{\mathcal{F}_0(\eta_{e,h})}{\mathcal{F}_{-1/2}(\eta_{e,h})} \left[1 - \frac{N_{e,h}}{k_B T_e} \frac{\mathcal{F}_{-1/2}(\eta_{e,h})}{\mathcal{F}_{1/2}(\eta_{e,h})} \left| \frac{\partial E_{c,v}}{\partial N_{e,h}} \right| \right] \vec{\nabla} N_{e,h} + e\mu_{e,h}^0 N_{e,h} \frac{\mathcal{F}_0(\eta_{e,h})}{\mathcal{F}_{1/2}(\eta_{e,h})} \vec{E} \\ &= \sigma_{e,h} \left[\frac{\partial}{\partial N_{e,h}} (F_{e,h} - E_{c,v}) + \frac{\partial E_{c,v}}{\partial N_{e,h}} \right] \vec{\nabla} N_{e,h} + e\mu_{e,h}^0 N_{e,h} \frac{\mathcal{F}_0(\eta_{e,h})}{\mathcal{F}_{1/2}(\eta_{e,h})} \vec{E} \\ &= D_{e,h} \vec{\nabla} N_{e,h} + \sigma_{e,h} \vec{E} , \end{aligned} \tag{17}$$

where $D_{e,h}$ is the actual diffusivity and $\sigma_{e,h}$ is the carrier conductivity. Even if no applied field exists a field can develop as a result of charge separation. This field can therefore not be set to zero since it is ultimately responsible for both carriers diffusing with the ambipolar diffusivity. In the absence of band-edge gradients and electric fields, the diffusivity, which is the coefficient of $\vec{\nabla} N_{e,h}$, increases with increasing density according to the ratio of the two Fermi-Dirac functions. For nondegenerate carrier statistics this ratio is unity and the diffusivity is density independent. However, if the carriers are degenerate, because the carrier kinetic energy in this case increases with density, the diffusivity will increase. The effect of the band-edge gradients will be a reduction of the diffusivity for both types of carriers at all densities. But since the many-body effects will be small at low densities while the ratio $\mathcal{F}_{-1/2}(\eta_{e,h})/\mathcal{F}_{1/2}(\eta_{e,h})$ (which is proportaional to $\eta_{e,h}^{-1}$ at high densities) will be small at high densities, the diffusivities will not differ appreciably from that of the IPBTT for these two limits. It is also clearly seen that the many-body corrections to the diffusivity appear through shifts of the band-edge energy only. If, however, the effective mass were also renormalized and energy dependent (as would occur for densities in excess of 10^{21} cm^{-3}), the many-body effects would reflect all the carrier energies.

The electron and hole currents are, of course, not independent but are linked by the electric field which develops during charge separation as the carriers attempt to diffuse at different rates. The electric field is related to the carrier densities by Poisson's equation,

$$\vec{\nabla}\cdot\vec{\mathrm{E}}=\frac{e}{\epsilon_0}(N_h-N_e)\ . \tag{18}$$

Combining Eqs. (17) and (18) gives for the ambipolar particle current in terms of the electron-hole pair density, $N=N_{e,h}$,

$$\vec{\mathrm{J}}=\left[\frac{\sigma_e D_h}{\sigma_e+\sigma_h}+\frac{\sigma_h D_e}{\sigma_e+\sigma_h}\right]\vec{\nabla}N\ , \tag{19}$$

with the coefficients of $\vec{\nabla}N$ defining the ambipolar diffusivity including many-body corrections.

The evolution of the carrier density can be determined from Eq. (19) and the equation of continuity. For example, if the carriers are generated by the absorption of a light pulse of photon energy $\hbar\omega$ and intensity $I(t)$ by a semiconductor with reflectivity R and absorption coefficients α, the equation of continuity is given by

$$\frac{\partial N(\vec{\mathrm{r}},t)}{\partial t}+\vec{\nabla}\cdot\vec{\mathrm{J}}=\frac{\alpha(1-R)I(t)e^{-\alpha x}}{\hbar\omega}-\gamma N^3\ , \tag{20}$$

where we have assumed that recombination occurs by the Auger process with coefficient γ. Combining Eqs. (19) and (20), one obtained the temporal and spatial evolution of the electron-hole density from

$$\frac{\partial N}{\partial t}=\vec{\nabla}\cdot\left|\left[\frac{\sigma_e D_e+\sigma_h D_e}{\sigma_e+\sigma_h}\right]\nabla\vec{\mathrm{N}}\right| +\frac{\alpha I(t)(1-R)e^{-\alpha x}}{h\omega}-\gamma N^3\ . \tag{21}$$

This completes the general formalism. At this point it is worth noting three limitations of the theory besides those already mentioned. First, the linearized Boltzmann equation can only be considered valid provided the mean free path of the carriers is much less than the distance over which the density changes appreciably. Since a typical carrier mean free path is about 100 Å at room temperaure and about 1000 Å at liquid-nitrogen temperature, the linearized equation should be valid except for highly inhomogeneous carrier densities. Second, the expressions for the many-body shift of the band edge were based upon the plasmon pole approximation for the inverse dielectric coefficient of a bulk homogeneous electron-hole plasma. The presence of a surface and carrier-density inhomogeneities might be expected to lead to modification of our results. It is generally agreed, however, that the local-density approximation offers accurate quantitative descriptions for inhomogeneous plasmas on distance scales of much less than a micron. The influence of surface effects is still an extremely active area of research and controversy. Third, vertex corrections should be considered in the calculation of the carrier self-energies as has been done for electrons in metals by Lundqvist[33] with a simplified theory offered by Overhauser.[38] Recently Berggren and Sernelius[39] have indicated that the corrections are significant for semiconductors, at least for extrinsically doped materials. For ambipolar plasmas, however, Overhauser[40] has shown that there are four vertex functions as opposed to just one for single component plasmas. Although particular expressions for these functions are not available to date, in general vertex corrections reduce the one-carrier correlation energy by no more than 50%. For the density range of interest here ($N\gtrsim10^{17}$ cm^{-3}) one might expect therefore that the more exact treatment of the carrier self-energies will lead to reductions of the diffusivities by ~10–20% over those reported here. This margin is within the spirit of other approximations made here. These modifications will be considered elsewhere.

V. AMBIPOLAR DIFFUSIVITY FOR Ge, Si, AND GaAs

Using the theory of the preceding section we have calculated the density dependence of the ambipolar diffusivity for Ge, GaAs, and Si at common lattice-carrier temperatures of 100 and 300 K. The various parameters used in the calculations are given in Table I with most of the values taken from Neuberger.[41] Figures 1–3 illustrate the calculated results for the three materials. Also indi-

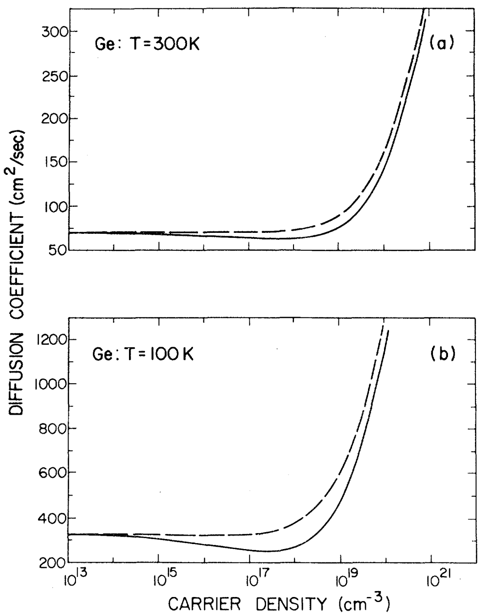

FIG. 1. Density dependence of the ambipolar diffusivity in Ge at 100 and 300 K according to the Boltzmann transport theory on the independent-particle approximation (— — —) and with the inclusion of many-body effects (——).

cated are the results predicted by IPBTT. In Fig. 3 for Si we also show the density dependence of the ambipolar diffusivity as predicted by the many-body theory of Wautelet.[23] In comparing results of the three theories we note that the IPBTT shows the diffusivity to be density independent up to the point of carrier degeneracy, after which it increases monotonically. Our results show that the diffusivity decreases as a function of density as many-body effects become more important. After the onset of carrier degeneracy the diffusivity increases for the same reasons as in the IPBTT and the two curves actually come together again because the carrier mobility decreases with increasing density, as indicated by Eq. (16). The difference between these two theories is dependent on carrier temperature, with the many-body effects becoming more important at low temperatures. This is not, per se, related to the temperature dependence of the many-body effects, which we have indicated to be small, but rather due to the electron (hole) carrier chemical potential decreasing (increasing) with temperature for constant carrier density. From Eq. (17) one therefore sees that the many-body effects become relatively more important at the lower temperatures.

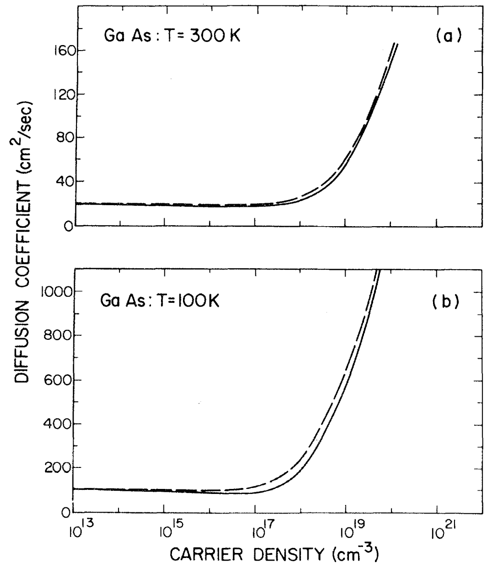

FIG. 2. Same as Fig. 1 except for GaAs.

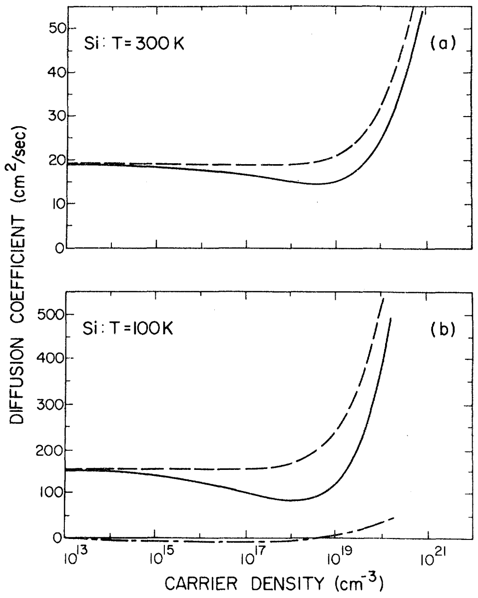

FIG. 3. Same as Fig. 1 except for Si. The -·-·- curve represents a calculation of Wautelet.

The value of the ambipolar diffusivity derived by Wautelet's theory differs substantially from that of the other two theories. The general expression for the individual carrier diffusivities given by Wautelet is of the form

$$D_{e,h}=\frac{D^0_{e,h}}{k_B T_e}(cN^{2/3}-dN^{1/3}) , \tag{22}$$

where c and d are constants and T_e is the carrier temperature corresponding to the density N in his theory. The first term in parentheses represents the average kinetic energy of the carriers and the

TABLE I. Materials parameters taken from Ref. 41. The unit of mass is the electron fundamental mass, while the diffusion coefficients are given in units of cm^2/sec.

Material	m_t	m_l	m_{lh}	m_{hh}	ϵ_0	D_e (300 K)	D_h (300 K)	D_e (100 K)	D_h (100 K)
GaAs	0.07	0.07	0.09	0.46	13.1	207	11	863	54
Ge	0.082	1.59	0.043	0.34	15.8	103	54	583	232
Si	0.19	0.98	0.16	0.59	12.0	35	13	86	690

second term is associated with the exchange-energy contribution to the carrier self-energy. As seen in Fig. 3, the predicted diffusivities are much lower than those of the other two theories. Although part of this difference is due to the inclusion of only exchange energy contributions to the many-body energy shifts, most of the difference is actually attributable to the statistical mechanical description of the carrier-distribution functions as discussed in Sec. II. At low densities the diffusivity is predicted to be negative for lattice temperatures up to at least room temperature. This is due to the assumption that the density dependence of the carrier kinetic energy is $N^{2/3}$, which is valid for degenerate distributions only. The diffusivity does not go over to the MB limit for low densities. We, too, are able to obtain negative diffusivities, although only at temperatures close to liquid-helium temperature, where the phenomenon of electron-hole droplets is well known. Figure 4 illustrates the individual electron and hole diffusion coefficients in the limit of zero temperature (complete degeneracy), although the actual values would be influenced by the presence of excitons at low temperatures. The vertical axis is taken to have arbitrary units because of the lack of knowledge of the actual momentum relaxation time in this limit. For this limit the expression for the individual diffusivities is of the form

$$D_{e,h}=\frac{\tau_{F_{e,h}}}{m^*_{d_{e,h}}}(gN^{2/3}-pN^{1/3}-qN^{1/4}+rN^{1/6}) , \qquad (23)$$

where $\tau_{F_{e,h}}$ is the momentum relaxation time at the Fermi surface. It can be seen that, apart from his

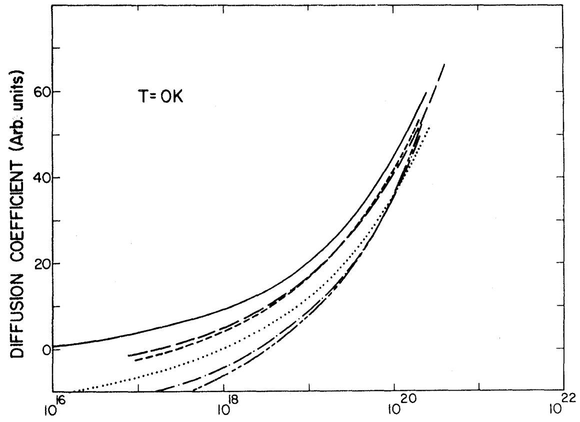

FIG. 4. Density dependence of the electron and hole diffusivities in the limit of $T=0$: Ge, D_e (---), D_h (— — —); GaAs, D_e (——), D_h (· · ·); Si, D_e (— · · —), D_h (— · —).

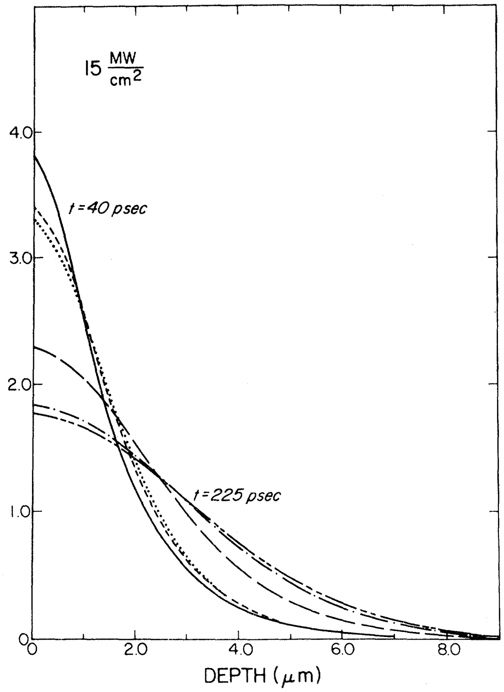

FIG. 5. Carrier-density profiles in Si at 40 and 225 psec after the beginning of a 15-MW/cm^2, 25-psec, 0.53-μm-square pulse for the independent-particle theory (· · · , — -- —), the many-body theory (——, — — —), and a constant, low-density diffusivity (---, — · —).

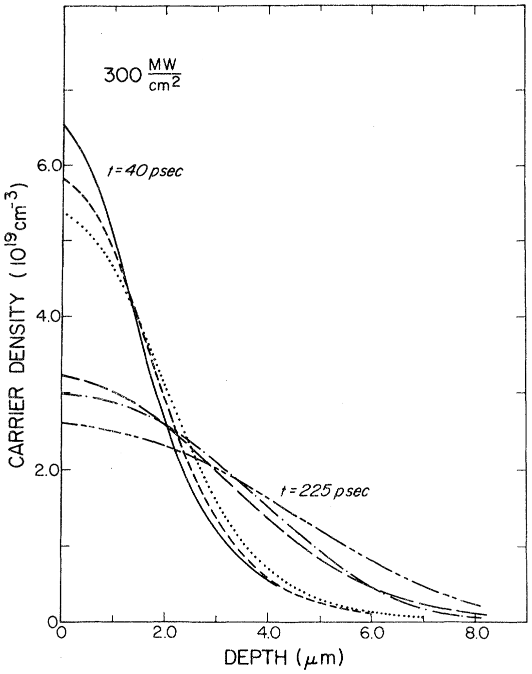

FIG. 6. Carrier-density profiles in Si at 40 and 225 psec after the beginning of a 300-MW/cm^2, 25-psec, 0.53-μm-square pulse for the independent-particle theory (· · · , — - - - —), the many-body theory (---, — · —), and a constant, low-density diffusivity (——, — — —).

T_e dependence, Wautelet's results are equivalent to ours, at least in form, only in the $T_e = 0$ limit.

From our theory one can also easily understand the role of the carrier effective mass in yielding the different results in the various materials, especially when carrier degeneracy becomes important. Semiconductors with larger carrier effective masses demonstrate a larger deviation from IPBTT, as shown in Figs. 1–4. But, just as for the temperature dependence discussed above, the effective-mass dependence results primarily from the effective-mass dependence of the carrier chemical potentials. For constant density the change of the carrier chemical potentials with increasing effective mass is larger than that of the many-body shift of the band edge so that, from Eq. (7), the many-body effects become relatively more important in influencing the value of the diffusivities as the carrier mass increases.

To compare our results with those of IPBTT in an actual diffusion problem we have calculated the evolution of the carrier density in silicon at 100 K after excitation with 0.53-μm, 25-psec pulses. By way of illustration we also include results derived from a density-independent diffusivity which has the MB value. The absorption,[42] reflection,[41] and Auger[43] coefficients were taken to be 8×10^3 cm^{-1}, 0.50, and 4×10^{-31} cm^6/sec, respectively. Figure 5 shows the carrier-density profiles at 40 and 225 psec after the beginning of a 15-MW/cm^2 pulse. The effect of the density-dependent diffusion coefficients in increasing carrier diffusion away from the surface is clear. It can be seen that the surface density at a particular time varies approximately inversely as the square root of the diffusivity and the differences between the models are more apparent at the lower densities, reflecting the larger differences between the diffusivities, as expected. More dramatic differences, of course, would appear at lower temperatures and correspondingly lower densities.

Before attempting to verify the influence of many-body carrier effects presented here one should be aware of two experimental difficulties. First, since diffusion occurs in the presence of density gradients, a given experiment will not be able to obtain the diffusion coefficient for a particular density. Second, as was discussed in the Introduction, any technique which will allow high-density carriers to be created in semiconductors will always give rise to some source of inhomgeneous lattice heating. The temperature gradient will give rise to an enhanced band-gap gradient since the lattice temperature will most likely be highest where the density is highest: Our calculations[25] have indicated that if the lattice temperature spatial profile has a depth on the order of a micrometer, a surface temperature which is only about 50 K higher than the bulk temperature can cause a reduction in the diffusivity comparable to the largest many-body-induced effects discussed here. At the lower densities, however, one might hope to avoid heating effects.

In conclusion, we have demonstrated the importance of many-body effects in the density dependence of the ambipolar diffusivity of Ge, Si, and GaAs. Because of the usual complications in performing any many-body calculation, we consider our results to be accurate to within 20%. However, we have shown how the theory is influenced by the effective masses of the carriers and the carrier and lattice temperatures. The lattice-temperature dependence essentially only influences the carrier momentum relaxation time, while the carrier temperature can influence both the relaxation time and the kinetic energy of the carriers. The theory presented here is seen to differ dramatically from that of Wautelet, primarily because of the different statistical mechanical description of the nonequilibrium carrier distributions.

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