

Time-dependent theory of hot electrons using the discrete Boltzmann equation

G. S. Canright and G. D. Mahan

*Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
and Department of Physics, The University of Tennessee, Knoxville, Tennessee 37996*

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The discrete Boltzmann equation may be applied to the time evolution of the electron distribution for spatially homogeneous systems in which optical-phonon scattering is dominant. We describe the time-dependent solution for both pulsed and steady excitation (e.g., via photoexcitation). The results for the latter case may be compared with experimental hot-electron luminescence spectra under steady photoexcitation. The agreement is generally good, but sensitive in detail to the method used to calculate recombination rates.

I. INTRODUCTION

Excitation of semiconductors by light of energy $\hbar\omega_{\text{ex}} > E_g$, where E_g is the band gap of the material, results in a nonequilibrium population of photoexcited carriers, which then relax towards equilibrium via various scattering mechanisms.^{1,2} The study of such systems under photoexcitation is of considerable interest for the information which may be gained concerning the scattering mechanisms and dynamics of nonequilibrium carriers in semiconductors. In polar semiconductors, under certain, commonly met conditions, the scattering of electrons in the conduction band is dominated by polar optical-phonon scattering. These conditions are that ϵ_c (the electron energy relative to the bottom of the band) is greater than $\hbar\omega_{\text{OP}}$, the phonon energy (assumed constant), but less than that energy which is sufficient for intervalley transitions; and that carrier densities, either from doping or from photoexcitation, are low, so that carrier-carrier scattering is not important.

The influence of optical-phonon scattering appears in various forms, all of which involve periodic oscillations in spectra: oscillatory photoconductivity,³⁻⁵ oscillations in photoluminescence intensity versus energy of photoexcitation,^{5,6} and oscillations in luminescence intensity versus luminescence energy.⁷⁻¹⁴ The last-mentioned phenomenon is observed only for low carrier densities.⁸⁻¹¹ If the carrier density is too large, then carrier-carrier scattering, in which energy exchange occurs in arbitrary amounts, tends to wash out the discrete line structure of photoluminescence spectra; in the high-density limit, the spectra may be analyzed as arising from a Maxwellian distribution at a temperature above the lattice temperature.^{2,5} The various scattering processes which affect the hot-carrier distribution, and their interpretation in terms of observed photoluminescence spectra, have been discussed in several review articles.^{1,2,15}

The present calculation applies to the low-carrier-density limit. The system most commonly studied is lightly doped *p*-type gallium arsenide.⁹⁻¹⁴ Steady photoexcitation yields a photoluminescence spectrum consisting of a series of broadened lines, beginning below the ex-

citation energy $\hbar\omega_{\text{ex}}$, and spaced by the optical-phonon energy $\hbar\omega_{\text{OP}}$. These lines have been shown¹³ to arise from recombination of electrons in the conduction band with neutral acceptors, i.e., from the (e, A^0) transition. Since the acceptor level is essentially *k* independent, one obtains almost direct information on the hot-electron distribution in the steady state; that is

$$I(\epsilon) \propto n(\epsilon)W_{cA}(\epsilon), \quad (1)$$

where I is the luminescence intensity, n the electron distribution, and W_{cA} the transition rate from conduction band to acceptor level. Hence one must assume a model for the (e, A^0) transition, based on a model for the acceptor wave function, in order to obtain $n(\epsilon)$ from $I(\epsilon)$. The commonly employed model^{1,6,9,16} uses hydrogenic wave functions for the acceptor level and incorporates an effective acceptor mass m_A .^{17,18} Within the limitations of a model for W_{cA} , then, one obtains information on the electron distribution from luminescence spectra.

In this paper we show that an analytical solution to the Boltzmann equation due to Mahan¹⁹ may be applied to the dynamics of photoexcited carriers in semiconductors. Mahan's solution applies to systems with no time dependence, and one-dimensional spatial dependence, in which optical-phonon scattering is dominant. Since the optical-phonon energy is treated as a constant, only discrete energy levels need be considered; hence the term "discrete Boltzmann equation" (DBE). In the following we show that the DBE applies equally well to systems which are isotropic (no spatial dependence), but time dependent, as long as optical phonons remain the dominant scattering mechanism. Using the DBE we will obtain the time-dependent electron distribution for both pulsed and steady photoexcitation.

Since the LO-phonon scattering time $\tau_{\text{OP}} \sim 100$ fs,⁹ direct time-resolved photoluminescence spectra from photoexcited carriers have not been obtained on a time scale which enables the observation of phonon-dominated transients. Carrier dynamics have been studied on nano- and picosecond time scales.^{16,20-22} Photoexcited phonon populations have been studied on the subpicosecond time scale by Kash *et al.*²³ and Collins and Yu.²⁴ Erskine,

Taylor, and Tang measured carrier lifetimes, due to phonon and intervalley scattering, on the order of tens of femtoseconds, using a pulse correlation technique.²⁵ Finally, we mention the experiment of Oudar *et al.*,²⁶ who measured subpicosecond relaxation phenomena for photoexcited carriers at energies $\epsilon_c < \hbar\omega_{OP}$.

Theoretical treatment of photoexcited carriers has been confined primarily to numerical solutions.^{1,27,28} Levinson and Levinsky have shown that, where the phonon population is small (low T , and small nonequilibrium densities),

$$n(\epsilon_n) \sim \begin{cases} \tau_{OP}(\epsilon_n), & \epsilon_n < \epsilon^* \\ 0, & \epsilon_n > \epsilon^* \end{cases}, \quad (2)$$

where ϵ_n is the discrete carrier energy equal to $\epsilon^* - n\hbar\omega_{OP}$, and ϵ^* is the energy of the photoexcited electrons.^{29,30} Equation (2) is valid only for the steady-state solution under steady illumination. It describes a rectangular distribution (if τ_{OP} is independent of ϵ), which, to the level of approximation assumed, shows agreement with experimental results,^{7,9,11} and with our calculation (see below). Our solution neglects the nonequilibrium phonons generated by the excited electrons, but incorporates scattering from thermal phonons at large T .¹⁹

The theoretical treatment of Collins and Yu²⁴ is most similar to ours. They find an analytical solution to the Boltzmann equation for a discrete electron distribution (i.e., scattering only by LO phonons), which is linearized in the distribution function. Their work differs from ours in the following respects. (1) They consider only spontaneous emission of phonons, which restricts the applicability of the result to low T . (2) Their solution is only valid for a pulsed excitation $\delta(t)$. (3) They extended their formulation to incorporate intervalley scattering and non-parabolic band structures, and solved the resulting equations numerically. (4) They also derived and solved the linearized Boltzmann equation for the phonon distribution, and compared the results with experimental Raman-scattering data.

Our method, as noted above, is not restricted to low temperatures. Further, it may be applied to excitations of arbitrary time dependence, although in the following we treat only pulsed and steady excitations.

II. PULSED EXCITATION

In the case that the distribution depends only upon time, the Boltzmann equation is

$$\frac{df}{dt} = \left[\frac{df}{dt} \right]_{\text{scatt}}. \quad (3)$$

Discretizing the above, we get¹⁹

$$\begin{aligned} \frac{d}{dt} f_l = & -\frac{f_l}{\tau_l} [N_0 + (N_0 + 1)\Theta(l-1)] \\ & + \frac{f_{l-1}}{\tau_{l-1}} [N_0\Theta(l-1)] + \frac{f_{l+1}}{\tau_{l+1}} (N_0 + 1), \end{aligned} \quad (4)$$

where $\Theta(x)$ is the step function, N_0 is the phonon occupa-

tion number equal to $[\exp(G) - 1]^{-1}$, and $G = \hbar\omega_{OP}/k_B T$.

Next we make the reasonable approximation³¹ that $\tau_l = \text{const} = \tau$; then, normalizing time so that $\tau = 1$, we get

$$\dot{\mathbf{f}} = -\underline{\mathbf{M}}\mathbf{f} \quad (5)$$

where $\underline{\mathbf{M}}$ is an infinite, tridiagonal matrix identical to that of Eq. (7) in Ref. 19. Hence one knows immediately, from Ref. 19, that

$$f_l(t) = C_0 e^{-Gt} + e^{-Gt/2} \int_0^\pi \frac{d\theta}{\pi} C(\theta) e^{-\lambda(\theta)t} \times \cos(l\theta + \phi), \quad (6)$$

where

$$\lambda(\theta) = \frac{e^G + 1 - 2e^{G/2} \cos\theta}{e^G - 1}$$

and

$$\tan\phi = \frac{e^{G/2} - \cos\theta}{\sin\theta}$$

using the spectrum of eigenvalues and eigenfunctions of $\underline{\mathbf{M}}$. The eigenvalues of $\underline{\mathbf{M}}$ consist of a discrete value $\lambda=0$, and a continuous spectrum of values $\lambda=\lambda(\theta)$. The constants C_0 and $C(\theta)$ then need to be determined by initial conditions.

If we rewrite (6) as

$$\mathbf{f}(t) = C_0 \mathbf{f}_0 + \int_0^\pi \frac{d\theta}{\pi} C_\theta e^{-\lambda_\theta t} \mathbf{f}_\theta, \quad (7)$$

where \mathbf{f}_0 , \mathbf{f}_θ are eigenvectors corresponding to $\lambda=0$, $\lambda=\lambda(\theta)$, respectively, then we find that

$$\tilde{\mathbf{f}}_0 \mathbf{f}_0 = 1, \quad (8a)$$

$$\tilde{\mathbf{f}}_0 \mathbf{f}_\theta = 0, \quad (8b)$$

$$\tilde{\mathbf{f}}_\theta \mathbf{f}_0 = 0, \quad (8c)$$

$$\tilde{\mathbf{f}}_\theta \mathbf{f}_{\theta'} = \frac{\pi}{2} \delta(\theta - \theta'), \quad (8d)$$

if

$$\tilde{\mathbf{f}}_0 = (1 - e^{-G})(1 \ 1 \ 1 \ \cdots) \quad (9a)$$

and

$$(\tilde{\mathbf{f}}_\theta)_l = e^{Gl/2} \cos(l\theta + \phi), \quad (9b)$$

i.e., these are the left eigenvectors of $\underline{\mathbf{M}}$. Then, evaluating (7) at $t=0$ and multiplying on the left by $\tilde{\mathbf{f}}_\theta$, we get

$$C_\theta = 2\tilde{\mathbf{f}}_\theta \mathbf{f}(0) = 2 \sum_{l=0}^{\infty} e^{Gl/2} \cos(l\theta + \phi) f_l(0). \quad (10)$$

If we multiply by $\tilde{\mathbf{f}}_0$ we get

$$C_0 = \tilde{\mathbf{f}}_0 \mathbf{f}(0) = (1 - e^{-G}) \sum_{l=0}^{\infty} f_l(0). \quad (11)$$

Now we can imagine an initial distribution $\mathbf{f}(0)$ injected into an empty band by a short pulse of light, where the pulse time is small compared to τ . For monochromatic

light we have that

$$f_l(0) = a \delta_{ln}, \quad (12)$$

i.e., the light populates only the n th level. Clearly,

$$f_l(t) = C_0 e^{-Gl} + e^{-Gl/2} e^{Gn/2} \int_0^\pi \frac{d\theta}{\pi} 2e^{-\lambda_\theta t} \cos(l\theta + \phi) \cos(n\theta + \phi). \quad (13)$$

Equation (13) is the complete solution for the initial condition (12).

III. STEADY EXCITATION

In the case of continuous illumination, which is more typical of experiments, carriers are injected into the conduction band at a steady rate. Hence we rewrite (5) as

$$\dot{\mathbf{f}} + \underline{M} \mathbf{f} = \mathbf{k}, \quad (14a)$$

where \mathbf{k} is a constant vector representing the steady injection of carriers by photoexcitation; for monochromatic illumination \mathbf{k} has only one nonzero entry.

Equation (14a) has no steady-state solution, since we are exciting a system possessing a $\lambda=0$ eigenvalue with the frequency $\omega=0$. More physically, we have not provided a mechanism for carriers to leave the band; without one, the population grows indefinitely. Hence we must add a term representing radiative recombination; however, we will neglect other possibilities (e.g., intervalley transitions). So (14a) becomes

$$\dot{\mathbf{f}} + (\underline{M}^0 + \underline{M}') \mathbf{f} = \mathbf{k}, \quad (14b)$$

where \underline{M}^0 is the matrix of Sec. II [Eq. (7) Ref. 19] and \underline{M}' is a diagonal matrix representing recombination. We write (14b) in a form suggesting a perturbative treatment, since radiative transitions are very slow relative to phonon scattering (i.e., $M'_{ll} \ll M^0_{ll}$).

Thus we let

$$\begin{aligned} \lambda_0 &= \lambda_0^0 + \lambda_0' = \lambda_0', \\ \lambda_\theta &= \lambda_\theta^0 + \lambda_\theta' \end{aligned} \quad (15)$$

and

$$\begin{aligned} \mathbf{f}_0 &= \mathbf{f}_0^0 + \mathbf{f}_0', \\ \mathbf{f}_\theta &= \mathbf{f}_\theta^0 + \mathbf{f}_\theta', \end{aligned} \quad (16)$$

and similarly for left eigenvectors. If we assume that (8) holds for the *perturbed* system, and the band is assumed empty at $t=0$, then it is easily shown that the solution to Eq. (14b) is

$$\begin{aligned} \mathbf{f}(t) &= (\tilde{\mathbf{f}}_0 \mathbf{k}) \left[\frac{1 - e^{-\lambda_0 t}}{\lambda_0} \right] \mathbf{f}_0 \\ &+ \int_0^\pi \frac{d\theta}{\pi} (2\tilde{\mathbf{f}}_\theta \mathbf{k}) \left[\frac{1 - e^{-\lambda_\theta t}}{\lambda_\theta} \right] \mathbf{f}_\theta \end{aligned} \quad (17)$$

which, given the perturbed eigensystem, is a complete solution.

First-order perturbation theory gives

$C_0 = a(1 - e^{-G})$. Equation (10) becomes

$$C_\theta = 2e^{Gn/2} \cos(n\theta + \phi)$$

so that

$$\lambda_0' = (1 - e^{-G}) \sum_{l=0}^{\infty} M'_{ll} e^{-Gl}, \quad (18)$$

$$\lambda_\theta' = 0. \quad (19)$$

Since¹⁹ $\lambda_\theta^0 \approx 1$, while $\lambda_0^0 \approx 10^{-6}$, we see that Eq. (17) consists of a part which evolves slowly, at the rate λ_0 , and a fast part which is basically driven by phonon scattering. Physically, the occurrence of the two very different time scales reflects the fact that the phonon scattering is very much faster (by $\sim 10^6$) than recombination.

As a first approximation, then, we set $\mathbf{f}'_\theta = \tilde{\mathbf{f}}'_\theta = 0$, i.e., we assume no perturbation to the phonon dynamics. The perturbation on \mathbf{f}_0 is found to be

$$\begin{aligned} (\mathbf{f}'_0)_l &= - \sum_{m=0}^{\infty} M'_{mm} e^{-Gm/2} e^{-Gl/2} \\ &\times \int_0^\pi \frac{d\theta}{\pi} \frac{2 \cos(l\theta + \phi) \cos(m\theta + \phi)}{\lambda_\theta^0} \end{aligned} \quad (20a)$$

while

$$(\tilde{\mathbf{f}}'_0)_l = (1 - e^{-G}) e^{Gl} (\mathbf{f}'_0)_l. \quad (20b)$$

As noted above, $\lambda_\theta^0 \approx 1$; the approximation improves with decreasing temperature.³² If we remove λ_θ^0 from (20a), the remaining integral is known analytically,¹⁹ and we get

$$\begin{aligned} (\mathbf{f}'_0)_l &\approx M'_{ll} e^{-Gl} + e^{-Gl} \sum_{m=0}^{\infty} M'_{mm} e^{-Gm} \\ &= (-M'_{ll} + \lambda_0') e^{-Gl} \end{aligned} \quad (21)$$

which is parallel to \mathbf{f}_0^0 ; hence,

$$\tilde{\mathbf{f}}_0 \approx \tilde{\mathbf{f}}_0^0 = (1 - e^{-G}) (1 \ 1 \ 1 \ \cdots). \quad (22)$$

Equations (17)–(19), (21), and (22) thus constitute an approximate solution for steady excitation.

IV. NUMERICAL RESULTS

First, we show a numerical example for the solution in Sec. II to a pulsed initial state. The pulsed solution (13) requires only the specification of $G = (\hbar\omega_0)/k_B T$, and an initial density a . Figure 1 shows a solution for $\hbar\omega_0 = .037$ eV (GaAs), $T = 300$ K, $a = 1/(1 - e^{-G})$, and n (the pulsed level) = 5. It should be emphasized that the distribution is actually a set of discrete points; the continuous curves are added to aid the eye. The distribution relaxes smoothly towards the equilibrium distribution (e^{-Gl}); however, note that it is still significantly different from equilibrium after ten scattering times.

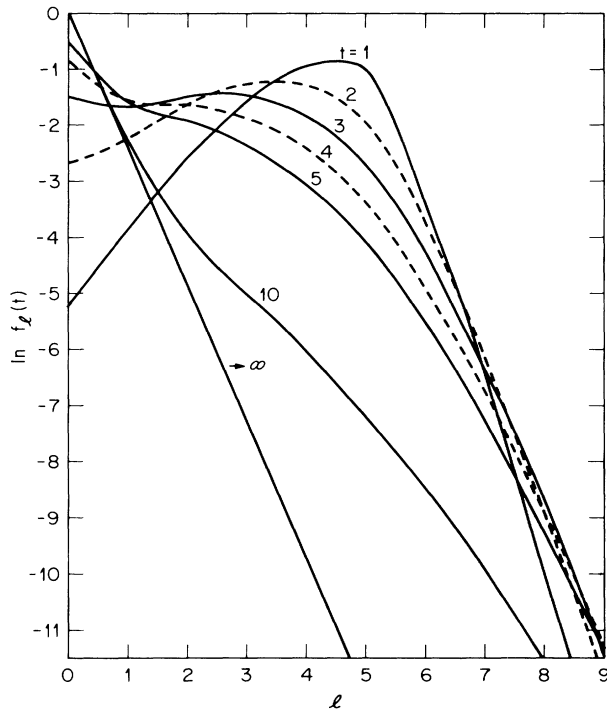


FIG. 1. Decay of electron distribution from an initial spike at $l=5$, $t=0$; $\hbar\omega_0=0.037$ eV, $T=300$ K. The distribution is discrete in energy; the smooth curves are a visual aid.

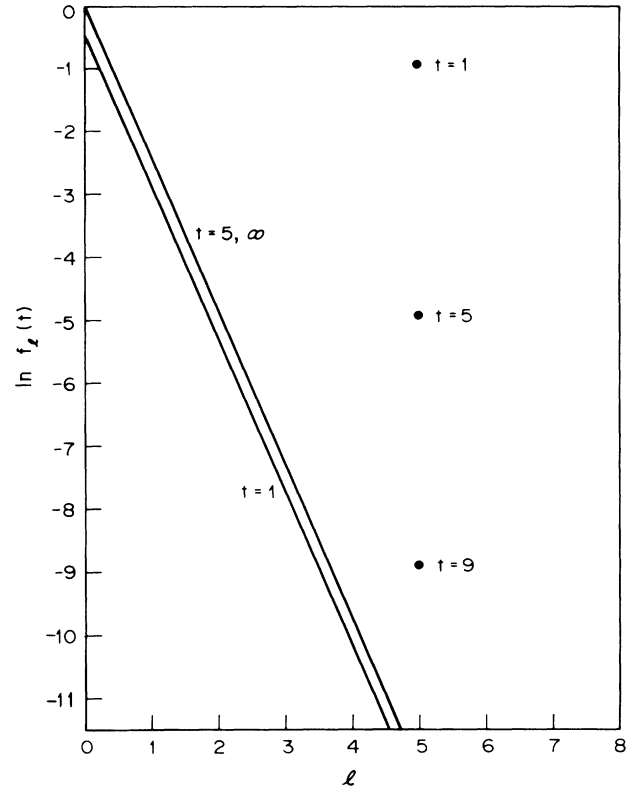


FIG. 2. The same condition as in Fig. 1, in the relaxation-time approximation.

It should be noted that this solution, normalized to an energy-independent scattering time τ_{OP} , is nevertheless *not* equivalent to the relaxation-time approximation.²⁸ Figure 2 shows the relaxation-time solution to the DBE, for the same initial condition; it consists of a decaying spike and a growing Maxwell-Boltzmann distribution. In the relaxation time approximation interactions between energy levels are not modeled, unlike the present formalism; the result is a more rapid, but less physical, approach to equilibrium.

To our knowledge, experimental resolution of the carrier distribution on this time scale ($\tau_{OP} \sim 100$ fs) has not been achieved. We may compare our results with those obtained by Lugli and Ferry,²⁷ who have solved the same problem using a Monte Carlo technique, both with and without $e-e$ scattering; their results without $e-e$ scattering are qualitatively similar to ours. Ulbrich has solved a similar problem numerically, except that the initial spike is at low energy (5 meV) and scattering is by acoustic phonons.¹⁶

The second example we solve is the solution for steady illumination (17). It requires a knowledge of the matrix elements M'_{ll} , i.e., the recombination rates. We choose to model transitions from the conduction band to an acceptor impurity level, as this phenomenon is typical of experiments.⁹⁻¹⁴ For these transition rates we used expressions from Dumke,¹⁷ normalized to $\tau_{OP}=10^{-13}$ s.

In Figs. 3 and 4 we show time-dependent solutions for an initially unoccupied band, into which carriers are injected at a steady rate at the level $l=8$ ($\epsilon=8\hbar\omega_0$), for

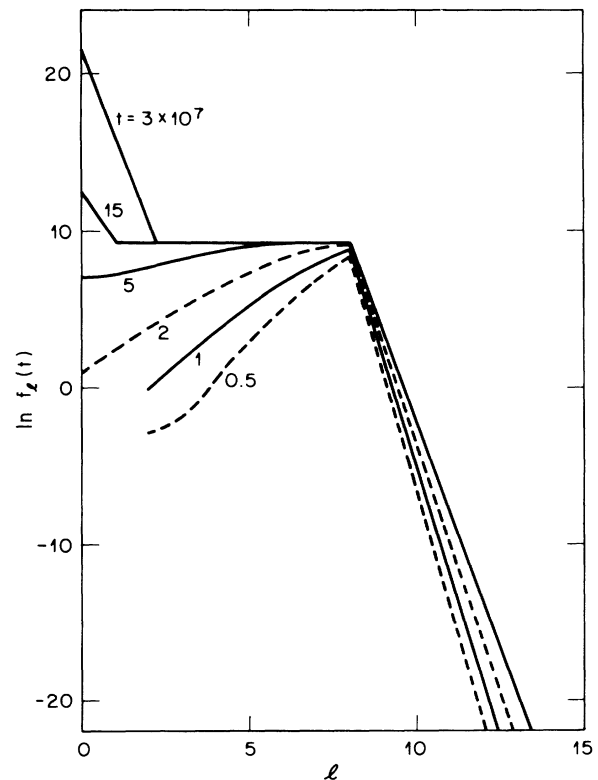
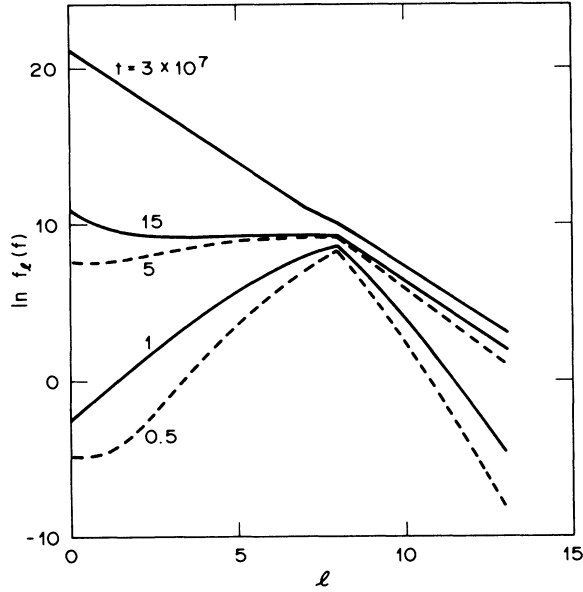


FIG. 3. Electron distribution vs time, for steady injection at $l=8$; GaAs, $T=77$ K.

FIG. 4. Same as Fig. 3 except $T = 300$ K.

GaAs, at $T = 77$ and 300 K, respectively. The distribution achieves a broad plateau, at either temperature, after $\sim 10\tau_{OP}$. However at room temperature the plateau is masked by a slow-growing quasithermal distribution which appears after much longer times ($\sim 10^7\tau_{OP}$). At 77 K this same distribution appears but falls more rapidly with energy, so that the plateau persists; note the resemblance to the rectangular distribution mentioned in Sec. I.

The approximations used in the perturbation solution are considered acceptable if the results converge to the steady-state distribution. The latter may be obtained by solving (14b) subject to $\dot{\mathbf{f}}=0$, i.e.,

$$\underline{M}\mathbf{f} = (\underline{M}^0 + \underline{M}')\mathbf{f} = \mathbf{k}. \quad (23)$$

Equation (23) is soluble by matrix inversion, as the addition of \underline{M}' makes the matrix nonsingular. For temperatures below ~ 50 K, we find that it is not a good approximation to use the unperturbed \mathbf{f}_θ and $\tilde{\mathbf{f}}_\theta$ eigenvectors in the time-dependent solution [Eq. (17)]. We have analytical expressions for these vectors, to first order in \underline{M}' , but have not implemented them numerically.

Steady-state results are easily available at any temperature from (23). Also, it is possible to drop the assumption that τ_{OP} is independent of energy, in the steady-state case. Figure 5 shows the steady-state distribution for injection at $l = 8$, for $T = 30$ K. Results are plotted for three cases:

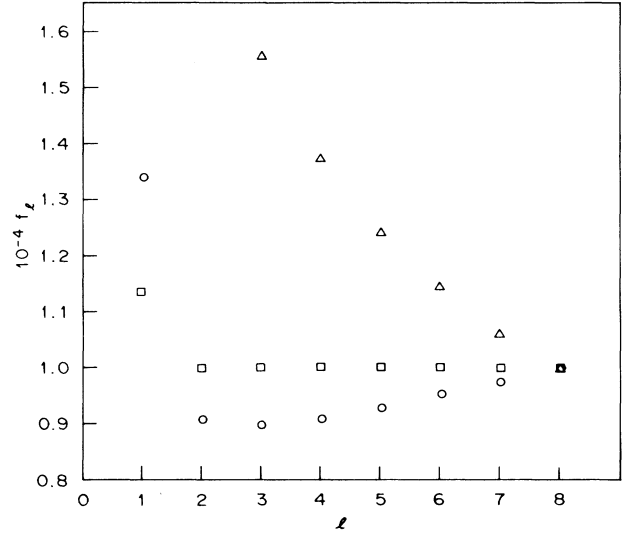
- (i) $\tau_{OP} = \text{constant}$.
- (ii) $l_{MFP} = \text{const} = v(\epsilon)\tau(\epsilon)$, where l is the mean path and $v(\epsilon)$ is the group velocity.

This gives

$$\frac{1}{\tau_l(\epsilon)} = \frac{V_l(\epsilon)}{\text{const}} = \frac{\sqrt{l + \delta l}}{\text{const}}. \quad (24)$$

In the results shown in Fig. 5 we used $\delta l = \frac{1}{2}$ so that $1/\tau_0 \neq 0$, and normalized to $1/\tau_8 = 1$.

- (iii) $1/\tau_{OP} = -2 \text{Im}(\Sigma)$ or

FIG. 5. Steady-state distribution for excitation at $l = 8$, GaAs, $T = 30$ K, using various expressions for τ_{OP} (see text). \square $\tau = \text{const}$; \triangle , $l_{MFP} = \text{const}$; \circ , $1/\tau = -2 \text{Im}(\Sigma)$.

$$\frac{1}{\tau_l} = \frac{(\text{const})}{\sqrt{l+1/2}} \left[N_0 J_n \left| \frac{(l+\frac{3}{2})^{1/2} + (l+\frac{1}{2})^{1/2}}{(l+\frac{3}{2})^{1/2} - (l+\frac{1}{2})^{1/2}} \right| + (N_0 + 1) J_n \left| \frac{(l-\frac{1}{2})^{1/2} + (l+\frac{1}{2})^{1/2}}{(l+\frac{1}{2})^{1/2} - (l-\frac{1}{2})^{1/2}} \right| \right], \quad (25)$$

where $\text{Im}(\Sigma)$ is the imaginary part of the electron self-energy, obtained for single-photon scattering by Mahan,³³ and as in (ii), we used $\delta l = \frac{1}{2}$ and set $\tau_8 = 1$.

Figure 6 shows the discrete luminescence spectra obtained from the distributions of Fig. 5, using Eq. (1) and the transition rates M'_{ll} . The results resemble the spectra in Ref. 13; however, the differences among models (i)–(iii) for τ_{OP} are less obvious in these spectra than they are in Fig. 5. There are other published spectra for which the resemblance is weaker. In Ref. 12 the peaks are very broad and not well resolved, while in Ref. 9 (Fig. 1) only the first three peaks are not masked by luminescence from electrons excited from the light-hole band. Figure 3 of the same source,⁹ the luminescence spectrum of carriers injected by intervalley transitions from the L valley, is not similar to Fig. 6, being considerably flatter.

Mirlin and co-workers used Eq. (1) and models for W_{cA} to obtain the distribution $N(\epsilon)$ from their spectra.^{9,11} Using an expression for W_{cA} similar to ours, they obtained a distribution (Fig. 4, Ref. 9) very much like that of (iii) in Fig. 5; the agreement is not as good with a different model.¹¹ Nevertheless both models used by them, and the three reported here in the steady-state calculation, give essentially indistinguishable results on the coarser scale of Fig. 3, i.e., a very flat distribution above the “pileup” at the bottom of the band.

We have not done time-dependent calculations for $T < 77$ K. The steady-state results merely approximate a

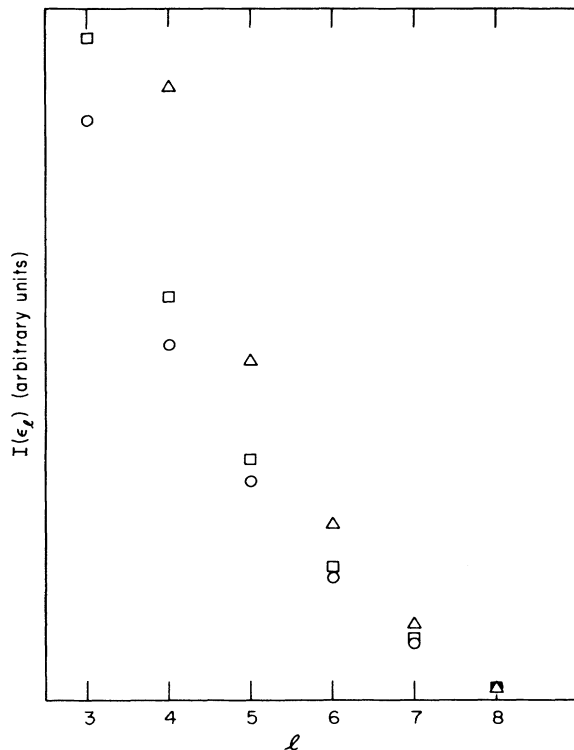


FIG. 6. Luminescence spectra for the distributions of Fig. 5, obtained from Eq. (1) and M'_{ij} (see text).

rectangular electron distribution more and more closely as $T \rightarrow 0$, showing no qualitative difference from the temperature regime in which we have time-dependent results. Further work on the low-temperature, time-dependent solution may be warranted as experimental techniques advance to yield comparable information experimentally. Similarly, more detailed comparison of steady-state results requires better modeling of the conduction-band-acceptor transition rates.

V. CONCLUSIONS

We have presented a theory for the time dependence of the nonequilibrium electron distribution, which is valid for the case of dilute carrier concentrations such that carrier-carrier scattering is unimportant. Our solution is analytical and thus avoids the computational burden of numerical solutions. It is also more general than previous analytical work in the following ways. Both the time-dependent and the steady-state solution are readily obtained in our approach, with the steady solution serving as an independent check of the time-dependent calculation. Our DBE solution is not restricted to low temperatures. We have solved both the case of steady excitation and the delta-function case; and the approach may be extended to a time-dependent excitation of arbitrary form (e.g., a broadened pulse). Hence, although it will perhaps always be necessary to resort to numerical solutions in order to incorporate all possible scattering mechanisms, our approach represents an attractive and flexible solution for systems in which optical-phonon scattering is dominant.

Such systems correspond to an important class of experimentally realizable cases which have been and remain of considerable interest, both for their pertinence to the understanding of practical devices involving hot electrons, and for the improvement in understanding of basic physical processes which they offer. Further steps toward the merging of theoretical and experimental insight will require, on the theoretical side, refinements in models for recombination rates, and on the experimental side, advances in femtosecond spectroscopy.

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