

ized by Seitz are responsible for the significant reduction of Mössbauer effect. While a detailed computer model such as that employed by Vinyard and collaborators to trace the history of an energetic ion in a lattice would be much more satisfactory, no such calculation is available.

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## New Approach to the Solution of the Time-Dependent Boltzmann Transport Equation\*

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We present a new method of solving analytically the time-dependent Boltzmann transport equation. Two examples are given to demonstrate the use of this method to predict ac and dc conductivities in nondegenerate single-valley semiconductors.

The time-dependent Boltzmann transport equation has been solved analytically on the basis of certain approximations,<sup>1</sup> or solved numerically using computer techniques.<sup>2</sup> The purpose of this Letter is to present a new approach to the exact solution of this equation by means of an iterative perturbation technique for single-valley semiconductors.

The time-dependent Boltzmann transport equation for a single-valley semiconductor which is spatially homogeneous and free of traps can be written as

$$\partial f / \partial t = (eF \partial / \partial p_F + Q)f, \quad (1)$$

where  $f$  is the carrier distribution function,  $e$  is the electronic charge,  $F$  is the applied electric field,  $p_F$  is the momentum of the carriers in the direction of the electric field, and  $Q$  is the scattering operator such that  $Qf$  is the rate of change of  $f$  due to the scattering of carriers. When  $F=0$ , Eq. (1) has the steady-state solution  $f=f_0$ , where  $f_0$  takes the form of a Maxwell-Boltzmann function for nondegenerate semiconductors and a Fermi-Dirac function for degenerate semiconductors. Taking  $f_0$  as the zero-order solution and using an iterative perturbation technique similar to that used in the quantum field theory,<sup>3</sup> we can find the solution of Eq. (1) to the  $n$ th order as

$$f = f_0 + \int_0^t dt_1 \left[ eF(t_1) \frac{\partial}{\partial p_F} + Q \right] f_0 + \int_0^t dt_1 \int_0^{t_1} dt_2 \left[ eF(t_1) \frac{\partial}{\partial p_F} + Q \right] \left[ eF(t_2) \frac{\partial}{\partial p_F} + Q \right] f_0 + \dots \\ + \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{n-1}} dt_n \left[ eF(t_1) \frac{\partial}{\partial p_F} + Q \right] \dots \left[ eF(t_n) \frac{\partial}{\partial p_F} + Q \right] f_0. \quad (2)$$

The exact solution of Eq. (1) is then given by

$$f(t) = \lim_{n \rightarrow \infty} f_n(t). \quad (3)$$

If  $Qf$  is linear in  $f$ , which is the case for nondegenerate semiconductors, the term in the first order of  $F$ , denoted by  $g_1(t)$ , is

$$g_1(t) = \lim_{n \rightarrow \infty} \left[ \int_0^t dt_1 e^{F(t_1)} \frac{\partial f_0}{\partial p_F} + \int_0^t dt_1 \int_0^{t_1} dt_2 Q e^{F(t_2)} \frac{\partial f_0}{\partial p_F} + \dots + \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{n-1}} dt_n Q^{n-1} e^{F(t_n)} \frac{\partial f_0}{\partial p_F} + \dots \right]. \quad (4)$$

In this equation  $\int_0^t dt_1 e^{F(t_1)} (\partial f_0 / \partial p_F)$  is the first-order deviation from  $f_0$  produced by the external applied field, and the other terms describe the effect of repeated scattering of this deviation. Thus,  $g_1(t)$  satisfies the following equation:

$$\partial g_1(t) / \partial t = Qg_1(t) + e^{F(t)} \partial f_0 / \partial p_F. \quad (5)$$

This equation expresses the balance of the rate of change of  $f$  to the first order in  $F$ . In general, it can be shown that the deviation from  $f_0$  in the  $n$ th order of  $F$ , denoted by  $g_n(t)$ , is governed by the following rate equation:

$$\frac{\partial g_n(t)}{\partial t} = Qg_n(t) + \frac{e^{F(t)}}{n} \frac{\partial g_{n-1}}{\partial p_F}. \quad (6)$$

Thus  $g_1(t)$ ,  $g_2(t)$ ,  $\dots$  can be solved successively. The exact expression for  $f(t)$  in the case of nondegenerate semiconductors is then

$$f(t) = f_0 + \sum_{n=1}^{\infty} g_n(t). \quad (7)$$

It can be seen that if  $F(t)$  oscillates with frequency  $\omega$ ,  $g_1(t)$  contains components of frequency  $\omega$ ,  $g_2(t)$  contains components of frequencies 0 and  $2\omega$ ,  $g_3$  contains components of frequencies  $\omega$  and  $3\omega$ , and so on; so that  $f(t)$  contains components of  $\omega$  and all the higher harmonics of  $\omega$ .

Suppose  $F(t)$  is given by

$$F(t) = F_0 \exp(i\omega t), \quad (8)$$

where  $F_0$  is the peak amplitude of  $F(t)$ .  $Qf$  can be written as

$$Qf = (1/\tau - H)f, \quad (9)$$

where  $\tau$  is a scalar quantity depending on the direction of  $\vec{p}$  in general, so that  $(1/\tau)f$  is the rate of change of  $f$  due to the carriers being scattered out of state  $\vec{p}$ , and  $H$  is an operator such that  $Hf$  is the rate of change of  $f$  due to the carriers being scattered into state  $\vec{p}$ . Thus, using an iterative technique,<sup>4</sup>  $g_1(t)$  can be written as

$$g_1(t) = -e^{F_0} \exp(i\omega t) \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} \left[ 1 + H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} + H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} + \dots \right] \frac{\partial f_0}{\partial p_F}. \quad (10)$$

In the following are given two examples to show the application of our approach for predicting some experimental phenomena in nondegenerate semiconductors.

In most ac conductivity experiments the applied electric field is a superposition of a dc bias field  $F_d$  and a comparatively much smaller ac component  $F_0' \exp(i\omega t)$ . For this case the deviation of  $f$  from  $f_0$  in the first order of  $F_0'$  is given by

$$\Delta f = \sum_{n=1}^{\infty} \left\{ -\frac{e\tau(1-i\omega\tau)}{1+\omega^2\tau^2} \left[ 1 + H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} + H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} + \dots \right] \frac{\partial}{\partial p_F} \right\}^n \times \frac{f_0}{(n-1)!} F_d^{n-1} F_0' \exp(i\omega t). \quad (11)$$

The series

$$\frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} \left[ 1 + H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} + \dots \right] \frac{\partial f_0}{\partial p_F}$$

can be regarded as equivalent to  $[\tau^*(1-i\omega\tau^*) / (1+\omega^2\tau^{*2})] \partial f_0 / \partial p_F$ , in which  $\tau^*$  is an effective scattering

time taking into account all successive scattering events

$$H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2}, H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2}, \dots$$

For most cases,  $\tau^*$  can be calculated without resorting to numerical techniques.<sup>4</sup> Thus, the term independent of  $F_d$  in  $\Delta f$  leads to the expression for the ac conductivity similar to that derived by Herring<sup>5</sup> for the low dc bias field. For higher dc bias fields the terms of higher powers in  $F_d$  have to be included in  $\Delta f$  for the determination of the ac conductivity. On the basis of the time-reversal symmetry, the carrier energy is symmetric in  $\vec{p}$ ,

$$E(\vec{p}) = E(-\vec{p}). \quad (12)$$

Therefore, the terms in  $\Delta f$  are symmetric in  $\vec{p}$  when  $n$  is even and those are antisymmetric in  $\vec{p}$  when  $n$  is odd. The terms with odd  $n$  contribute to the average carrier momentum. For such terms, the limit of the series

$$\frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} \left[ 1 + H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} + \dots \right] h_o(\vec{p})$$

is determined by the degree of anisotropy of the scattering, and this limit is likely to be of the same order of magnitude as  $[\tau(1-i\omega\tau)/(1+\omega^2\tau^2)]h_o(\vec{p})$  even in the most anisotropic cases,<sup>4</sup> where  $h_o(\vec{p})$  is an odd function of  $\vec{p}$ . The terms with even  $n$  contribute to the average carrier energy. For such terms, each term in the series

$$\frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} \left[ 1 + H \frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} + \dots \right] h_e(\vec{p})$$

is approximately  $(1 - \langle \hbar\omega \rangle / \langle E \rangle)$  times the previous term, and the limit of the series is then approximately

$$\frac{\tau(1-i\omega\tau)}{1+\omega^2\tau^2} h_e(\vec{p}) \frac{\langle E \rangle}{\langle \hbar\omega \rangle},$$

where  $h_e(\vec{p})$  is an even function of  $\vec{p}$ ,  $\langle \hbar\omega \rangle$  is a typical amount of energy exchange during scattering, and  $\langle E \rangle$  is the average carrier energy. For nearly elastic scattering  $\langle E \rangle / \langle \hbar\omega \rangle$  is much larger than unity. Thus, it is reasonable to introduce momentum and energy relaxation times in the phenomenological approach to the ac conductivity at high dc bias fields, and to assume that the energy relaxation time is much larger than the momentum relaxation time for nearly elastic scattering.<sup>6</sup> Gibson, Granville, and Paige<sup>6</sup> have predicted that a negative  $\partial\tau/\partial E$  will decrease the magnitude of the imaginary part of the ac conductivity. This is obvious from Eq. (11).

For the case  $\omega=0$ ,  $g_1$  in Eq. (10) becomes time independent, and from Eq. (7) the carrier distribution function can be written as

$$f = \sum_{n=0}^{\infty} \left[ -eF\tau(1+H\tau+\dots) \frac{\partial}{\partial p_F} \right]^n \frac{1}{n!} f_0. \quad (13)$$

An important consequence of Eq. (13) is that the average of a physical variable  $\varphi$ , defined by

$$\langle \varphi \rangle = \int f \varphi d^3p / \int f d^3p, \quad (14)$$

will change with the field, and this change as a function of applied field can be determined by

$$\frac{d\langle \varphi \rangle}{dF} = \frac{\int f \varphi d^3p \int f_0 d^3p - \int f_0 \varphi d^3p \int f d^3p}{(\int f d^3p)^2 F}. \quad (15)$$

Letting  $\varphi$  be the carrier velocity, we can conclude that the average drift velocity of carriers always increases with increasing field, and that there is no negative differential resistance region as long as the number of carriers remains constant and the phonon distribution function remains undisturbed. This conclusion is valid irrespective of the band structure of the semiconductors provided that the effective mass of the carriers is not negative. For the band structures with negative effective mass, Krömer<sup>7</sup> has predicted that the occurrence of negative differential resistance is possible.

The above discussion provides a qualitative justification of our new approach to the solution of the time-dependent Boltzmann transport equation.

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## Microscopic Origin for the Apparent Uncertainty Principle Governing the Anomalous Attenuation of Third Sound

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A macroscopic uncertainty principle has been invoked as an explanation for the anomalously high attenuation of third sound in certain thickness regimes of liquid-He films. The propagation criterion deduced from this idea appears to be in accord with experiment. We outline here a microscopic mechanism which gives rise to a large attenuation and which yields a propagation criterion essentially equivalent to the one proposed as a macroscopic uncertainty principle. We find that the nonlinearities inherent in the problem play the significant role in producing this attenuation.

Third sound is a surface-wave propagation in an adsorbed thin film of liquid He below the  $\lambda$  point. This phenomenon is associated with the superfluidity of liquid He. It does not occur for a nonsuperfluid. The accepted<sup>1</sup> approximate formula for the velocity of third sound exhibits this fact in its dependence on the superfluid fraction:

$$c^2 = D\sigma(D, T) \partial u(D)/\partial D, \quad (1)$$

where  $c$  represents the velocity of third sound,  $D$  is the unperturbed mean thickness of the He film,  $\partial u/\partial D$  represents the magnitude of the Van der Waals force per unit mass between the substrate wall and atoms of He at a distance  $D$  from that wall, and

$$\sigma(D, T) = (\rho_s/\rho)_{\text{film}} \quad (2)$$

represents the effective superfluid fraction in the film. Equation (1) neglects the small temperature variations which accompany the third-sound wave and which modify this equation by a few percent.

It has been found experimentally that there are regimes of film thickness at which the attenuation of third sound becomes anomalously high. "Classical" theory<sup>2-4</sup> does not account for attenuations of this magnitude. The third-sound signal

disappears in very thin films somewhat earlier than expected—in a film about 35% thicker than derived from a healing-length calculation.<sup>2, 5</sup> What is perhaps much more unexpected, however, is the thick-film anomalous regime. For relatively thick films the third-sound attenuation is found to be considerably higher than deduced from a "classical" mechanism. It has been suggested that these effects are evidence for a macroscopic quantum uncertainty principle.<sup>6</sup> This notion is embodied in the idea that third-sound waves can only propagate under the condition that

$$h/McD < 2, \quad (3)$$

where  $M$  represents the mass of one He atom. And, indeed, this condition appears to be well satisfied experimentally.<sup>2</sup> The purpose of this note is to indicate a microscopic mechanism which gives rise to this apparent uncertainty principle. In so doing we derive the counterpart of Eq. (3) from microscopic considerations.

Our calculation is restricted to the thick-film regime in that only in this limit is our hydrodynamic formalism valid. Some rather serious modifications would have to be made to carry the calculation to the very thin-film limit. Hence this presentation is to be viewed as a preliminary