Boltzmann-equation approach to nonlinear acoustoelectric interactions in piezoelectric semiconductors

Gautam Johri* and Harold N. Spector Department of Physics, Illinois Institute of Technology, Chicago, Illinois 60616 (Received 6 November 1974)

Ultrasonic second-harmonic generation in piezoelectric semiconductors is investigated using the classical Boltzmann-equation approach with the ansatz of a constant relaxation time. The flux at the second-harmonic frequency is calculated in terms of the flux in the fundamental and the linear and nonlinear conductivity tensors. It is found that when $ql \ll 1$, our results for second-harmonic generation reduce to those of Conwell and Ganguly, while when $ql \gg 1$ they reduce to those of Wu and Spector. The existence of a separate regime when $ql \gg 1$ and $\omega \tau \ll 1$ as found by Nakamura is shown not to occur.

I. INTRODUCTION

When large-amplitude acoustic flux propagates in a piezoelectric semiconductor, the nonlinearities in the acoustoelectric interaction between the phonons and the conduction electrons give rise to many frequency-mixing effects. One such effect of particular interest is second-harmonic generation, which was first observed in photoconducting CdS by Tell.¹ The physical situation is that the self-consistent electric field produced by the interaction of the ultrasonic wave and the conduction electrons contains harmonics of the fundamental piezoelectric field. Second-harmonic generation has been calculated by Spector² and Conwell and Ganguly³ using a phenomenological theory which is valid for low-mobility semiconductors where the electron mean free path is much smaller than the phonon wavelength $(ql \ll 1)$ and by Wu and Spector⁴ using the quantum Liouville-theorem approach which is valid for high-mobility semiconductors $(ql \gg 1)$ in the limit $\omega \tau \gg 1$. Recently, Nakamura⁵ has discussed three wave-mixing effects using the classical Boltzmann-equation approach. One can extract the flux distribution in the second harmonic from his calculations, though he does not calculate it directly. However, there are certain discrepancies in his calculations, and his results do not go over to those of previous calculations $^{2-4}$ in the appropriate limits. Therefore we present our calculations in this paper also following the classical Boltzmann-equation approach.

In Sec. II we present the theory of second-harmonic generation due to the interaction between the ultrasound and the conduction electrons, following the same approach as in Ref. 3. The secondharmonic flux generated is calculated in terms of linear and nonlinear conductivity tensors. In Sec. III we present the calculation of the conductivity tensors. This is done using the classical Boltzmann equation. We can use the Boltzmann distribution for the equilibrium distribution of the conduction electrons since in most materials where acoustic amplification is observed, the carrier density is low enough for the electrons to obey classical statistics. This approach is valid both in low-mobility semiconductors like CdS ($ql \ll 1$) and in high-mobility semiconductors like GaAs and *n*-InSb ($ql \gg 1$). This is of interest since amplification of large ultrasonic flux has been observed in GaAs, ^{6,7} and *n*-InSb. ⁸ Finally, in Sec. IV we present the discussion of our results and comparison with previous works, ²⁻⁵

II. SECOND-HARMONIC GENERATION IN PIEZOELECTRIC SEMICONDUCTORS

In nonlinear media, the electronic current density contains terms which are nonlinear in the electric field. The observation of second- and higherorder harmonics of the fundamental piezoelectric field can be investigated by using the nonlinear terms in the electronic current density as source terms in Maxwell's equations. In Sec. III we shall show that one can write the current density in the form

$$J_i = \sigma_{ij} E_j + \Lambda_{ijk} E_j E_k \quad . \tag{2.1}$$

In a piezoelectric semiconductor, the equation of motion

$$0 \frac{\partial^2 \xi_i}{\partial t^2} = \frac{\partial T_{ij}}{\partial x_i}$$
(2.2)

must be supplemented by the piezoelectric equation of state

$$T_{i\,i} = C_{i\,ikl} S_{kl} - \beta_{i\,ik} E_k , \qquad (2.3)$$

$$D_i = \epsilon E_i + 4\pi \beta_{ijk} S_{jk} , \qquad (2.4)$$

where

$$S_{ij} = \frac{1}{2} \left(\frac{\partial \xi_i}{\partial x_j} + \frac{\partial \xi_j}{\partial x_i} \right)$$
(2.5)

is the strain tensor, T_{ij} is the stress tensor, C_{ijkl} are the elastic constants, β_{ijk} are the piezoelectric

12

constants, \vec{E} is the electric field, \vec{D} is the electric displacement, and ϵ is the static dielectric constant of the medium. Using Eqs. (2.1)-(2.5) together with Maxwell's equations, one can solve for the electric fields induced by the ultrasound.

Transverse electric fields induced by the ultrasound in piezoelectric materials are weaker by a factor of $(v_s/c)^2$ than the longitudinal electric fields.⁴ Using Gauss's law we can obtain the longitudinal electric fields associated with the fundamental and second harmonic. They are

$$E_{1z} = \frac{-(4\pi/\epsilon)\beta_{zjk}S_{jk1}}{1 - (4\pi/i\omega\epsilon)\sigma_{zz}(\omega)}, \qquad (2.6)$$

$$E_{2z} = \frac{-(4\pi/\epsilon)\beta_{zjk}S_{jk2}}{1 - (2\pi/i\omega\epsilon)\sigma_{zz}(2\omega)} - \frac{(2\pi i/\omega\epsilon)\Lambda_{zzz}(\omega)(4\pi\beta_{zjk}/\epsilon)^2 S_{jk1}^2}{[1 - (2\pi/i\omega\epsilon)\sigma_{zz}(2\omega)][1 - (4\pi/i\omega\epsilon)\sigma_{zz}(\omega)]^2},$$

$$(2.7)$$

where S_{jk1} and S_{jk2} are the strains associated with the fundamental and second harmonic, respectively. Using the piezoelectric fields in the equations of motion of the lattice (2, 2)-(2, 5), we find that the sound wave amplitudes for the fundamental and the second harmonic obey the following equations:

$$\rho \frac{\partial^2 \xi_1}{\partial t^2} = \left(C + \frac{4\pi \beta^2 / \epsilon}{1 - (4\pi/i\omega\epsilon)\sigma_{zz}(\omega)}\right) \frac{\partial^2 \xi_1}{\partial z^2} , \qquad (2.8)$$

$$\rho \frac{\partial^2 \xi_2}{\partial t^2} = \left(C + \frac{4\pi\beta^2/\epsilon}{1 - (2\pi/i\omega\epsilon)\sigma_{zz}(2\omega)}\right) \frac{\partial^2 \xi_2}{\partial z^2} + \left(\frac{(i/\omega)(4\pi\beta/\epsilon)^3\Lambda_{zzz}(\omega)}{[1 - (2\pi/i\omega\epsilon)\sigma_{zz}(2\omega)][1 - (4\pi/i\omega\epsilon)\sigma_{zz}(\omega)]^2}\right) \frac{\partial \xi_1}{\partial z} \frac{\partial^2 \xi_1}{\partial z^2} , \qquad (2.9)$$

where β and *C* are the appropriate components of the the piezoelectric tensor and the elastic constants.

Following the same procedure as in Refs. 2 and 3, we can write the sound-wave amplitude in the form

$$\xi_i(z, t) = u_i(z) \exp((q_i z - \omega_i t))$$
 (2.10)

Because of the smallness of the electromechanical coupling coefficient $4\pi\beta^2/\epsilon C \ll 1$, the amplitude u_i will change very little over the distance of a wavelength. Therefore, neglecting terms involving $\partial u_i/\partial z$ compared to $q_i u_i$, we obtain the following set of first-order differential equations for u_1 and u_2 :

$$\frac{\partial u_1}{\partial z} = -\alpha_1 u_1 , \qquad (2.11)$$

$$\frac{\partial u_2}{\partial z} = -\alpha_2 u_2 + C_{111} u_1^2 \exp((2q_1 - q_2)z) . \qquad (2.12)$$

Here, α_1 and α_2 are the absorption (amplification) coefficients for the fundamental and the second harmonic

$$\alpha_{i} = \frac{2\pi\beta^{2}}{\epsilon C} q_{i} \operatorname{Im}\left(\frac{1}{1 - (4\pi/i\omega_{i}\epsilon)\sigma_{zz}(\omega_{i})}\right), \quad (2.13)$$

and the wavevectors for the fundamental and second harmonic are determined by the dispersion relation

$$\omega_{i} = q_{i} v_{s} \left[1 + \operatorname{Re} \left(\frac{2\pi \beta^{2} / \epsilon C}{1 - (4\pi / i\omega_{i} \epsilon) \sigma_{zz}(\omega_{i})} \right) \right] \cdot \quad (2.14)$$

The coupling coefficient determining how the fundamental drives the second harmonic is

$$C_{111} = \frac{(4\pi\beta/\epsilon)^3 (i/\omega C) (q^2/4) \Lambda_{zzz}(\omega)}{[1 - (2\pi/i\omega\epsilon)\sigma_{zz}(2\omega)][1 - (4\pi/i\omega\epsilon)\sigma_{zz}(\omega)]^2},$$
(2.15)

where $q = \omega/v_s$ and v_s is the sound velocity in the absence of the piezoelectric coupling.

The solution of Eqs. (2.11) and (2.12) are²

$$u_1(z) = u_1(0)e^{-\alpha_1 z}$$
, (2.16)

$$u_{2}(z) = \frac{C_{111}u_{1}^{2}(0)}{i(2q_{1}-q_{2})-(2\alpha_{1}-\alpha_{2})} \left(e^{[i(2q_{1}-q_{2})-2\alpha_{1}]z}-e^{-\alpha_{2}z}\right)$$
(2.17)

The acoustic flux accompanying a wave of frequency ω_i is

$$P_{i} = \frac{1}{2} \rho \omega_{i}^{2} v_{s} |\xi_{i}|^{2} .$$
 (2.18)

Therefore, the ratio of the acoustic flux in the second harmonic to the initial flux in the fundamental is

$$P_2/P_1^2(0) = A(e^{-2\alpha_2 z} + e^{-4\alpha_1 z})$$

$$-2e^{-(2\alpha_1+\alpha_2)z}\cos(2q_1-q_2)z), \quad (2.19)$$

where

$$A = \frac{8}{(\rho v_s)^3} \left(\frac{2\pi\beta}{\epsilon}\right)^2 \times \left| \frac{\Lambda_{zgz}(\omega)}{\left[1 - (4\pi/i\omega\epsilon)\sigma_{zz}(\omega)\right] \left[2\sigma_{zz}(\omega) - \sigma_{zz}(2\omega)\right]} \right|^2 .$$
(2.20)

Therefore, to study the acoustic flux in the second harmonic, we have to calculate the linear and nonlinear conductivity tensors σ_{zz} and Λ_{zzz} .

III. CALCULATION OF CONDUCTIVITY TENSORS

The electron current density in a piezoelectric semiconductor in the presence of the ultrasound is given by

$$\dot{\mathbf{j}} = -e \int d\vec{\mathbf{v}} \, \vec{\mathbf{v}} f(\vec{\mathbf{v}}) , \qquad (3.1)$$

where $f(\vec{\mathbf{v}})$ is the electron distribution function in the semiconductor in the presence of the ultrasound. The Boltzmann equation which determines $f(\vec{\mathbf{v}})$ is

$$\frac{\partial f}{\partial t} + \vec{\mathbf{v}} \cdot \frac{\partial f}{\partial \vec{\mathbf{r}}} - \frac{e}{m} \sum_{n} \left(\vec{\mathbf{E}}_{n} + \frac{\vec{\mathbf{v}}}{c} \times \vec{\mathbf{B}}_{n} \right) \frac{\partial f}{\partial \vec{\mathbf{v}}} = -\frac{f - f_{s}}{\tau} ,$$
(3.2)

where \vec{E}_n and \vec{B}_n are the electric and magnetic fields induced by the acoustic wave, τ is the electron relaxation time, and f_s is the distribution to which the electrons relax in presence of the wave. This distribution is⁹

$$f_{s}(\vec{v}) = f_{0}\left(\vec{v} - \frac{d\vec{\xi}}{dt}, n_{0} + n_{1} + n_{2}\right)$$
$$\approx f_{0}(\vec{v}) - \left(\frac{d\vec{\xi}}{dt}\right)\left(\frac{df_{0}}{d\vec{v}}\right) + n_{1}\frac{df_{0}}{dn_{0}} + n_{2}\frac{df_{0}}{dn_{0}} \quad . \quad (3.3)$$

Here $f_0(\vec{\mathbf{v}})$ is the equilibrium distribution of the electrons, $\vec{\xi}$ the amplitude of the acoustic wave, and the second and third terms on the right-hand side of (3.3) arise from the collision drag effect and the fact that scattering is local^{9,10} and therefore does not change the electron density. The second term can be neglected in semiconductors where the electron-phonon coupling is either via the deformation potential or piezoelectric coupling.¹¹ The transverse electric fields induced can be neglected, since they are smaller by a factor $(v_s/c)^2$ than the longitudinal fields so that $\vec{B}_n = (c\vec{q}_n/\omega_n) \times \vec{E}_n = 0$. The solution of Eq. (3.2) can be written

$$f(\vec{\mathbf{v}}) = f_0(\vec{\mathbf{v}}) + \sum_n g_n(v) e^{i(\vec{\mathbf{d}}_n \circ \vec{\mathbf{r}} - \omega_n t)}, \qquad (3.4)$$

where $g_n(\vec{\mathbf{v}})$ represents the part of the distribution function induced by the acoustic wave of frequency ω_n and wave vector $\vec{\mathbf{q}}_n$.

The part of the distribution function which is associated with the fundamental obeys the equation

$$\frac{\partial g_1}{\partial t} + \vec{\mathbf{v}} \cdot \frac{\partial g_1}{\partial \vec{\mathbf{r}}} + \frac{g_1}{\tau} = \frac{e}{m} \vec{\mathbf{E}}_1 \cdot \frac{\partial f_0}{\partial \vec{\mathbf{v}}} + \frac{n_1}{\tau} \frac{df_0}{dn_0} , \qquad (3.5)$$

and the part of the distribution function associated with the second harmonic obeys the equation

$$\frac{\partial g_2}{\partial t} + \vec{\mathbf{v}} \cdot \frac{\partial g_2}{\partial \vec{\mathbf{r}}} + \frac{g_2}{\tau} = \frac{e}{m} \vec{\mathbf{E}}_2 \cdot \frac{df_0}{d\vec{\mathbf{v}}} + \frac{n_2}{\tau} \frac{df_0}{dn_0} + \frac{e}{m} \vec{\mathbf{E}}_1 \cdot \frac{\partial g_1}{\partial \vec{\mathbf{v}}}.$$
(3.6)

We choose our coordinate system so that the wavevector of the acoustic wave lies along the z axis, and we also treat the electrons as obeying Boltzmann statistics in the absence of the acoustic wave. This is justified since in most materials of interest the electron density is low enough for them to obey classical statistics. Therefore,

$$f_0(\vec{\mathbf{v}}) = n_0 \left(\frac{m}{2\pi k_B T}\right)^{3/2} \exp\left(-\frac{mv^2}{2k_B T}\right).$$
 (3.7)

Then the solution of (3.5) can be written

$$g_{1}(\vec{v}) = \left(-\frac{e\tau}{k_{B}T} E_{1z}v_{z} + \frac{n_{1}}{n_{0}}\right) \frac{f_{0}(\vec{v})}{1 + i(q_{z}v_{z} - \omega)\tau} .$$
(3.8)

Using (3.8), the solution for (3.6) can be written

$$g_{2}(\vec{\nabla}) = \left(-\frac{e\tau}{k_{B}T} E_{2z}v_{z} + \frac{n_{2}}{n_{0}}\right) \frac{f_{0}(\vec{\nabla})}{1 + 2i(q_{z}v_{z} - \omega)\tau} + \frac{e\tau}{m} \left[-\frac{(e\tau/k_{B}T)E_{1z}^{2}}{1 + i(q_{z}v_{z} - \omega)\tau} \times \left(1 - \frac{mv_{z}^{2}}{k_{B}T} - \frac{iq_{z}v_{z}\tau}{1 + i(q_{z}v_{z} - \omega)\tau}\right) + \frac{(n_{1}/n_{0})E_{1z}}{1 + i(q_{z}v_{z} - \omega)\tau} \left(-\frac{mv_{z}}{k_{B}T} - \frac{iq_{z}\tau}{1 + i(q_{z}v_{z} - \omega)\tau}\right) \right] \times \frac{f_{0}(\vec{\nabla})}{1 + 2i(q_{z}v_{z} - \omega)\tau} .$$
(3.9)

Here we have used the approximate dispersion relation $qv_s = \omega$. Substituting from (3.8) and (3.9) into (3.1), we can write the ac current densities $j_1 \propto \exp i(\mathbf{\vec{q}} \cdot \mathbf{\vec{r}} - \omega t)$ and $j_2 \propto \exp 2i(\mathbf{\vec{q}} \cdot \mathbf{\vec{r}} - \omega t)$ as

$$j_{1z} = \sigma_{zz}(\omega) E_{1z} - R_z(\omega) n_1 e v_s \tag{3.10}$$

and

$$j_{2z} = \sigma_{zz}(2\omega)E_{2z} - R_{z}(2\omega)n_{2}ev_{s} + \tau_{zzz}(\omega)E_{1z}^{2} - S_{zz}(\omega)n_{1}ev_{s}E_{1z} , \qquad (3.11)$$

where

$$\sigma_{zz}(\omega) = (2\sigma_0/v_0^2) (1/\pi^{1/2}v_0) I_2 , \qquad (3.12)$$

$$R_{z}(\omega) = (1/\pi^{1/2} v_{s} v_{0}) I_{1} , \qquad (3.13)$$

$$\tau_{zzz}(\omega) = \frac{2\sigma_0\mu}{\pi^{1/2}v_0^3} \left(J_1 - \frac{2}{v_0^2} J_3 - iq\tau J_2' \right) , \qquad (3.14)$$

$$S_{zz}(\omega) = -\frac{\mu}{v_s} \frac{1}{v_0 \pi^{1/2}} \left(\frac{2}{v_0^2} J_2 + iq\tau J_1'\right), \qquad (3.15)$$

where

$$I_n = \int_{-\infty}^{\infty} \frac{dx \, x^n \, e^{-dx^2}}{a + ibx} \, , \qquad (3.16)$$

$$J_n = \int_{-\infty}^{\infty} \frac{dx \, x^n \, e^{-dx^2}}{(a + ibx)(c + 2ibx)},$$
 (3.17)

$$J'_{n} = -(d/da) J_{n} , \qquad (3.18)$$

with $a=1-i\omega\tau$, $c=1-2i\omega\tau$, $b=iq\tau$ and $d=1/v_0^2$. The integrals (3.16) and (3.17) can easily be done using the integral representation for the function¹²

$$w(z) = e^{-z^2} \operatorname{erfc} - iz = \frac{i}{\pi} \int_{-\infty}^{\infty} \frac{e^{-t^2} dt}{z - t} \quad . \tag{3.19}$$

Therefore, we get for the conductivity tensors

$$\sigma_{zz}(\omega) = -\frac{2\sigma_0 \pi^{1/2}}{ql} \left[\frac{(1-i\omega\tau)^2}{(ql)^2} w \left(-\frac{1-i\omega\tau}{iql} \right) -\frac{1-i\omega\tau}{\pi^{1/2}ql} \right] , \qquad (3.20)$$

$$R_{z}(\omega) = \frac{v_{0}}{v_{s}iql} \left[1 - \frac{1 - i\omega\tau}{iql} i\pi^{1/2} w \left(-\frac{1 - i\omega\tau}{iql} \right) \right],$$
(3.21)

$$\tau_{zzz}(\omega) = \frac{2\sigma_{0}\mu}{\pi^{1/2}v_{0}} \left(\frac{i\pi}{(ql)^{2}} \left[\frac{1}{2} (1 - 2i\omega\tau) w \left(\frac{1 - 2i\omega\tau}{2iql} \right) - (1 - i\omega\tau) w \left(-\frac{1 - i\omega\tau}{iql} \right) \right] + \frac{i\pi^{1/2}}{2(ql)^{3}} (3 - 4i\omega\tau) - \frac{2i\pi}{(ql)^{4}} \\ \times \left[(1 - i\omega\tau)^{3} w \left(-\frac{1 - i\omega\tau}{iql} \right) - \frac{1}{8} (1 - 2i\omega\tau)^{3} w \left(-\frac{1 - 2i\omega\tau}{2iql} \right) \right] - iql \left\{ \frac{2\pi^{1/2} (1 - i\omega\tau)^{2}}{(ql)^{4}} + \frac{\pi}{(ql)^{3}} \right\} \\ \times \left[2(1 - i\omega\tau) \left(-i\omega\tau - \frac{(1 - i\omega\tau)^{2}}{(ql)^{2}} \right) w \left(-\frac{1 - i\omega\tau}{iql} \right) - \frac{1}{2} (1 - 2i\omega\tau)^{2} w \left(-\frac{1 - 2i\omega\tau}{2iql} \right) \right] \right\} \right]$$
(3.22)

and

$$S_{zz}(\omega) = -\frac{\mu}{v_s \pi^{1/2}} \left(-\frac{\pi^{1/2}}{(ql)^2} + \frac{2\pi}{(ql)^3} \left[(1 - i\omega\tau)^2 w \left(-\frac{1 - i\omega\tau}{iql} \right) - \frac{1}{4} (1 - 2i\omega\tau)^2 w \left(-\frac{1 - 2i\omega\tau}{2iql} \right) \right] + iql \left\{ -\frac{2i\pi^{1/2}(1 - i\omega\tau)}{(ql)^3} + \frac{i\pi}{(ql)^2} \left[(1 - 2i\omega\tau) w \left(-\frac{1 - 2i\omega\tau}{2iql} \right) - \left(1 - 2i\omega\tau - \frac{2(1 - i\omega\tau)^2}{q^2 l^2} \right) w \left(-\frac{1 - i\omega\tau}{iql} \right) \right] \right\} \right\}, \quad (3.23)$$

where σ_0 is the dc conductivity, μ is the carrier mobility, and *l* is the mean free path of the electrons. Using the continuity equation we can write (3.10) and (3.11) in the form (2.1),

$$j_{1z} = \sigma'_{zz}(\omega) E_{1z}$$
, (3.24)

$$j_{2z} = \sigma'_{zz}(2\omega)E_{2z} + \Lambda_{zzz}(\omega)E_{1z}^2, \qquad (3.25)$$

where

and

$$\sigma'_{zz}(\omega) = \sigma_{zz}(\omega) / [1 - R_z(\omega)] , \qquad (3.26)$$

$$\Lambda_{zzz}(\omega) = \frac{\tau_{zzz}(\omega)}{1 - R_z(2\omega)} + \frac{S_{zz}(\omega)\sigma'_{zz}(\omega)}{1 - R_z(2\omega)}.$$
 (3.27)

In the short-wavelength limit, i.e., ql > 1, the linear and nonlinear conductivities reduce to

$$\sigma_{zz}'(\omega) = -\frac{i\omega\epsilon}{4\pi} \left(\frac{q_d}{q}\right)^2 \left(1 + i\pi^{1/2}\frac{v_s}{v_0}\right)$$
(3.28)

 $\Lambda_{zzz}(\omega) = \frac{\epsilon}{4\pi} \left(\frac{q_d}{q}\right)^2 \frac{ev_s}{mv_0^2} , \qquad (3.29)$

where $q_d = (4\pi n_0 e^2/\epsilon k_B T)^{1/2}$ is the electron Debye wave vector, $v_0 = (2k_B T/m)^{1/2}$ is the thermal velocity of the carriers, and v_s is the sound velocity in the piezoelectric semiconductor. Equations (3.28) and (3.29) are valid for either limits $\omega \tau \gg 1$ or $\omega \tau \ll 1$ as long as $ql \gg 1$.

Substituting from Eqs. (3.28) and (3.29) into (2.20), we find the flux in the second harmonic is

$$A = \frac{2}{9\rho v_s^3} \left(\frac{8\pi\beta}{\epsilon}\right)^2 \frac{e^2}{(mv_0^2)^2} \frac{q^2}{(q^2+q_d^2)^2} .$$
 (3.30)

The acoustic flux in the second harmonic has a maximum when $q = q_d$, i.e., when the phonon wave-vector is equal to the electron Debye wave vector. The magnitude of the flux at this maximum is

$$A = (2/9\rho v_s^3)\pi\beta^2/\epsilon n_0 k_B T$$
 (3.31)

and is inversely proportional to both the electron

12

3219

density and the absolute temperature. These results agree with those of Wu and Spector⁴ using a Liouville-equation approach and neglecting the effects of collisions. This indicates that this approach is valid as long as $ql \gg 1$. In the long-wavelength limit, i.e., $ql \ll 1$, we can use the asymptotic form of the error functions to obtain the limiting forms of the conductivity tensors. For large X, ¹³

$$\operatorname{erfc} X = \frac{\exp - X^2}{\pi^{1/2} X} \left(1 - \frac{1}{2X^2} \right).$$
 (3.32)

Then we have

$$\sigma_{zz}'(\omega) = \frac{\epsilon \omega_c}{4\pi (1 + i\omega/\omega_D)}, \qquad (3.33)$$

$$\Lambda_{zzz}(\omega) = \frac{-\epsilon\omega_c \,\mu}{4\pi_s(1+i\omega/\omega_D)(1+2i\omega/\omega_D)},\qquad(3.34)$$

where $\omega_c = 4\pi n_0 e \,\mu/\epsilon$ is the dielectric relaxation frequency, and $\omega_D = v_s^2/D$, where *D* is the diffusion coefficient. The flux in the second harmonic can be obtained using (3.33) and (3.34) in (2.20):

$$A = \frac{8(2\pi\mu\beta/\epsilon v_s)^2 \left[1 + (\omega/\omega_D)^2\right]}{\rho v_s^3 [1 + 9(\omega/\omega_D)^2] \left[1 + (\omega_c/\omega + \omega/\omega_D)^2\right]} .$$
 (3.35)

Therefore, we see that the frequency dependence of the second harmonic will depend on the relative values of ω_c and ω_D . In low-resistivity materials $\omega_c \gg \omega_D$, and (3.35) has a maximum at $\omega = (\omega_c \omega_D)^{1/2}$, or a wave vector $q = q_d$. The magnitude of the second-harmonic flux at this maximum is

$$A = (1/9\rho v_s^3) \pi \beta^2 / \epsilon n_0 k_B T$$
 (3.36)

and has the same form as the second-harmonic flux at the maximum $q = q_d$ for a high-mobility semiconductor, given by (3.31). However, the magnitude of second-harmonic generation will be higher for low-mobility semiconductors ($ql \ll 1$), because the limit $ql \gg 1$ will occur for the frequency of peak generation at higher electron densities. In highresistivity materials we have $\omega_c \ll \omega_D$, and (3.27) predicts a plateau region for $\omega_c < \omega < \omega_D$. The maximum second-harmonic flux in this region is

$$A = (8/\rho v_s^3) (2\pi \,\mu\beta/\epsilon v_s)^2 \,. \tag{3.37}$$

This predicts a smaller second-harmonic flux than Eq. (3.31) for materials of the same electron density and at the same temperature. The results derived here also agree with those obtained by Conwell and Ganguly³ using a phenomelogical approach. When $ql \approx 1$, the exact expression for the

linear and nonlinear conductivities have to be evaluated numerically.

IV. DISCUSSION

The results of our calculations using the Boltzmann equation to obtain the linear and nonlinear conductivities are in agreement with those of Conwell and Ganguly³ for the case $ql \ll 1$, appropriate to low-mobility semiconductors, and with those of Wu and Spector⁴ for the case $ql \gg 1$, appropriate to high-mobility semiconductors. Our results disagree with those obtained by Nakamura, ⁵ which seemed to indicate the existence of a separate regime, where $ql \gg 1$ and $\omega \tau \ll 1$. However, our results seem to indicate that there are just two regimes, at least for the case of a constant relaxation time, which depends upon whether $ql \gtrless 1$. Since the calculation of Wu and Spector neglected the effect of collisions, it was not apparent whether the results of their paper were valid when $ql \gg 1$, or whether they required the stronger condition $\omega \tau \gg 1$. The present calculation indicates that only the weaker condition $ql \gg 1$ is required for their results to be valid. Also, Nakamura's final result for the nonlinear conductivity seems to diverge in the limit $ql \gg 1$. Therefore, the present calculations resolve the apparent differences in the earlier calculations.

The calculation we have done in this paper has been in the absence of a dc electric field. However, although the presence of a dc electric field can have a drastic effect on the absorption coefficient (in fact, changing a linear loss to a linear gain), the coefficient A in Eq. (2.20), which determines the amplitude of the harmonic generated. is only a very weak function of such a drift field except at very low frequencies [compare Eq. (3.35)] of this paper to Eq. (26) of Ref. 2]. Therefore, although the presence of a dc electric field will change the rate at which the second harmonic generated will grow or decay with position, it will not greatly modify our calculation of the amplitude A. Also, even in the absence of a dc electric field, the electronic losses in high-mobility semiconductors such as InSb and GaAs are low enough because of their relatively low electromechanical coupling coefficients so that the second harmonic generated should still be detectable in samples of reasonable length. For example, in n-InSb, ¹⁴ for a frequency of 3.8 GHz and at a temperature of 77 °K, where ql is estimated to be about 15, the total absorption coefficient (electronic and lattice) in the absence of a drift field has been determined to be less than 2 cm^{-1} .

Technology, Chicago, Ill. 60616.

^{*}Submitted in partial satisfaction of the requirements for the degree of Ph. D. in physics, Illinois Institute of

¹B. Tell, Phys. Rev. 136, A772 (1964).

- ²H. N. Spector, Phys. Rev. B <u>6</u>, 2409 (1972).
- ³E. M. Conwell and A. K. Ganguly, Phys. Rev. B <u>4</u>, 2535 (1971).
- ⁴Chhi-Chong Wu and H. N. Spector, J. Appl. Phys. <u>43</u>, 2937 (1972).
- ⁵Ki-ichi Nakamura, J. Phys. Soc. Jpn. <u>32</u>, 365 (1972).
- ⁶E. Palik and R. Bray, Phys. Rev. B <u>3</u>, <u>3</u>302 (1971). ⁷D. G. Carlson, A. Segmüller, E. Mosekilde, H. Cole,
- and J. A. Armstrong, Appl. Phys. Lett. <u>18</u>, 330 (1971). ⁸K. W. Nill and A. L. McWhorter, J. Phys. Soc. Jpn.
- Suppl. <u>21</u>, 755 (1966).

- ⁹M. H. Cohen, M. J. Harrison, and W. A. Harrison, Phys. Rev. <u>117</u>, 937 (1960).
- ¹⁰T. Holstein, Phys. Rev. <u>113</u>, 479 (1959).
- ¹¹H. N. Spector, Solid State Phys. <u>19</u>, 291 (1966).
 ¹²Handbook of Mathematical Functions, edited by M. Abramowitz and I. A. Stegun, U. S. Dept. of Commerce, Natl. Bureau Stds., Appl. Math. Ser. No. 55
- (U. S. GPO, Washington, D. C., 1964), p. 297. ¹³Reference 12, p. 298.
- ¹⁴J. Gorelik, B. Fisher, B. Pratt, M. Zinman, and A. Many, J. Appl. Phys. <u>43</u>, 3614 (1972).