

there is only one, rather than three, mutually perpendicular one-dimensional bands, the resistivity should be highly anisotropic. Magnetoresistance and Hall effect in oriented samples of V-doped  $\text{Ti}_2\text{O}_3$  were measured by Honig and collaborators.<sup>6</sup> Data from these experiments established that conduction was by holes in the low-lying  $a_{1g}$  band we have been discussing, but the huge anisotropy the one-dimensional model predicts was not observed. However, since only a very few of the data points in in Ref. 5 bear on this question, we measured the resistivity as a function of temperature for two oriented samples taken from adjacent slices of the boules used by Sjöstrand and Keesom. The data

are plotted in Fig. 3. Measurements were made by a standard four-probe technique described in Ref. 5.

Some anisotropy is observed, as is virtually inevitable in this low-symmetry system, but only about 50%; more important, note that the resistivity is *higher* in the  $c$ -axis direction than in the basal plane. The resistivity measurements are totally inconsistent with the one-dimensional interpretation of the specific-heat anomalies.

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## Collision-Induced Nonlinear Excitations

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We demonstrate that a velocity-dependent collision frequency can introduce significant nonlinearities in the theory of a homogeneous, unmagnetized plasma. By means of a practical collision model which allows us to obtain useful information on nonlinear plasma phenomena without involving the mathematical complexity usually associated with collision operators, we investigate the problems of self-focusing, parametric excitation, and third-order frequency mixing. Our results are compared with those of previous authors. It is shown that the collision-induced nonlinear excitations considered here may be more important than those due to other mechanisms.

### I. INTRODUCTION

Model collision operators with velocity-independent collision frequencies are often used to consider the effect of collisions on plasma waves. Since, in reality, particles with high and low velocities have quite distinct collision characteristics, the use of these models cannot generally be justified, except perhaps for a few cases in which the exact form of the collision frequency happens to be unimportant. It is therefore necessary to use more realistic, but often mathematically complicated, collision models, such as the full Boltzmann or Fokker-Planck operators. In considering linear problems, one may still occasionally obtain solu-

tions to these complicated kinetic equations by employing some sort of perturbation or numerical techniques. However, it would be impractical, in general, to use the latter equations in considering nonlinear problems.

In this paper, we consider collisional wave excitation in a spatially uniform plasma by using a fairly realistic, yet simple, collision model. In fact, we use the Lorentz collision operator<sup>1</sup> with the velocity-dependent collision frequency represented by the so-called Harp model.<sup>2</sup> This model, which will be discussed in Sec. II, assumes that electrons with energy less than a certain fixed value experience no collisions, while those with higher energy suffer an infinite number of colli-

sions. The model, therefore, emphasizes the difference between the collisional characteristics of high- and low-energy electrons. This property makes it particularly suitable for our purpose here, since it is precisely from the energy dependence of the collision frequency that the nonlinear effects considered in this paper originate. No such effects<sup>3</sup> will appear if the collision frequency is independent of energy. For similar reasons, previous authors<sup>2,4</sup> have employed this useful model to compare theoretical predictions with experimental observations on nonlinear plasma phenomena, such as echoes and stimulated emissions.

Using the model discussed above to describe collisions between electrons and heavy scatterers, we derive the current density and the nonlinear dielectric constant for electrostatic long-wavelength oscillations in a plasma. Application of our results to self-focusing and parametric excitation problems in semiconductors is discussed and compared with previous work on similar subjects. It is demonstrated that some previous conclusions should be reconsidered. We also investigate the problem of third-order frequency mixing in a plasma due to collisional effects.

Finally, we introduce a simple generalization of the Harp collision frequency so that the real situation can be better represented when necessary.

## II. KINETIC EQUATION

The distribution function  $F(\vec{v}, t)$  of the electrons in a spatially uniform plasma is described by the kinetic equation

$$\frac{\partial F}{\partial t} + \frac{q}{m} \vec{E} \cdot \frac{\partial F}{\partial \vec{v}} = C(F), \quad (1)$$

where  $q$  and  $m$  are, respectively, the charge and effective mass<sup>5</sup> of the electron and  $\vec{E}(t)$  is the total electric field. No external magnetic fields are present, and effects due to the energy dependence of the mass are neglected.

The collision integral  $C(F)$  will here be represented by the Lorentz model in which electrons collide elastically with "infinitely" heavy particles,<sup>1,6</sup> i. e.,

$$C(F) = -\nu F + (\nu/4\pi) \int F d\Omega, \quad (2)$$

where  $\nu(v^2)$  is the electron-neutral-particle collision frequency. The integration is over all solid angles  $\Omega$  in velocity space. It is assumed that the collision frequency only depends on the magnitude of the electron velocity. Electron-electron scattering is neglected. We use the Harp model<sup>2,4</sup> for the velocity dependence of the collision frequency, i. e.,

$$\nu(v^2) = \begin{cases} 0, & v^2 < v_0^2 \\ \infty, & v^2 > v_0^2 \end{cases} \quad (3)$$

where  $v_0$  is chosen to be in a region where the actual collision frequency increases rapidly with velocity. This model was originally proposed for gases showing a strong Ramsauer effect, such as argon, which exhibits a sharp increase of the electron-neutral-particle collision frequency within a certain electron velocity range. For other plasmas,  $v_0$  cannot be uniquely defined, since the range in which the collision frequency increases may be large. The Harp model then only gives a rough estimate of the effects of the velocity dependence of the collision frequency. Such quantitative results will, hopefully, stimulate further investigations using more realistic models.

It is clear that when the Harp frequency is used in the Lorentz model electrons with speeds less than  $v_0$  behave as a collisionless gas, while those with higher speeds behave as a collision-dominated gas. It follows that high-energy electrons will always be isotropic, since they are randomly scattered rapidly in velocity space. These electrons therefore do not participate in the dynamic behavior of the plasma. In this respect, the model may also be applicable to a collisionless plasma in which high-energy electrons are lost in some manner, such as recombination, leaving the electron distribution function with a rather sharp cutoff at some velocity  $v_0$ .

## III. CURRENT DENSITY AND DIELECTRIC CONSTANT

The solution of the kinetic equation (1) is given by

$$F(\vec{v}, t) = \begin{cases} F_0[\vec{v} - (q/m) \int_{-\infty}^t \vec{E} dt], & v^2 < v_0^2 \\ G(v^2, t), & v^2 > v_0^2 \end{cases} \quad (4)$$

where

$$G(v^2, -\infty) = F_0(\vec{v}).$$

The function  $F_0(\vec{v}) = G(v^2, -\infty)$  is the distribution of the electrons in the absence of the electric field, say at  $t = -\infty$ , and is, therefore, taken to be isotropic in velocity space. When the field changes, the number of particles in each region may vary; hence, the distribution function of the high-energy electrons is also time dependent.

The current density is given by

$$\vec{j} = q \int \vec{v} F_0[\vec{v} - (q/m) \int_{-\infty}^t \vec{E} dt] d\vec{v}, \quad (5)$$

where the velocity-space integration is to include only electrons with speeds less than  $v_0$ . Electrons with higher speeds do not contribute to the current, since they remain isotropic in velocity space due to the strong scattering.

Assuming that the electric field is sufficiently small so that the perturbed velocity is much less than the thermal velocity, the integrand in Eq. (5) can be expanded as a Taylor series. Thus we have

$$j \approx 2\pi q \int_0^{v_0} \int_{-1}^1 \mu v^3 \left[ F_0 - \frac{q}{m} \left( \int^t E dt \right) \frac{\partial F_0}{\partial v_x} + \frac{1}{2} \left( \frac{q}{m} \int^t E dt \right)^2 \frac{\partial^2 F_0}{\partial v_x^2} - \frac{1}{6} \left( \frac{q}{m} \int^t E dt \right)^3 \frac{\partial^3 F_0}{\partial v_x^3} \right] dv d\mu, \quad (6)$$

where  $\mu = v_x/v$  and the  $x$  coordinate is taken to be in the direction of the electric field  $\vec{E} = E\hat{x}$  and the current density  $\vec{j} = j\hat{x}$ .

The terms proportional to even powers of the electric field vanish in the integration over  $\mu$ , so that after some straightforward manipulation one obtains

$$\vec{j} \approx (N_0 q^2/m) \int^t \vec{E} dt - (8\pi q^4/15m^3) v_0^5 F_0'' \left( \int^t \vec{E} dt \right)^3, \quad (7)$$

where

$$F_0'' = \left( \frac{\partial^2 F_0(\vec{v})}{\partial (v^2)^2} \right)_{v^2=v_0^2}$$

and

$$N_0 = -\frac{4}{3} \pi \int_0^{v_0} v^3 \frac{\partial F_0}{\partial v} dv.$$

We note that if  $v_0 = \infty$ , then  $N_0$  reduces to the electron number density and the nonlinear terms vanish. Thus, besides introducing nonlinear effects, collisions also modify the linear response of the plasma. This small linear effect is, however, of little interest here.

The current density and the electric field are related to the displacement vector  $\epsilon_0 \epsilon \vec{E}$  by the equation

$$\vec{j} + \epsilon_0 \frac{\partial \vec{E}}{\partial t} = \epsilon_0 \frac{\partial}{\partial t} (\epsilon \vec{E}), \quad (8)$$

where  $\epsilon$  is the plasma dielectric constant.

We define the nonlinear dielectric constant  $\epsilon_2$  by the relation<sup>7</sup>

$$\epsilon \approx \epsilon_1 + \epsilon_2 E_1^2, \quad (9)$$

where  $\epsilon_1 = 1 - \omega_p^2/\omega^2$  is the well-known dielectric constant when nonlinear effects are absent and  $\omega_p = (N_0 q^2/\epsilon_0 m)^{1/2}$  is the plasma frequency. The amplitude of the electric field  $\vec{E}$  is here denoted by  $\vec{E}_1$ . Combining Eqs. (7)–(9), one obtains

$$\epsilon_2 = \frac{4\pi q^4}{15m^3 \epsilon_0 \omega^4} v_0^5 F_0'', \quad (10)$$

where  $\omega$  is the wave frequency. We have considered here waves with slowly varying amplitudes.

#### IV. APPLICATIONS IN SEMICONDUCTOR-PLASMA THEORY

In this section we first apply our calculations to two problems of recent interest in laser-semiconductor interaction theory, namely, self-focusing and parametric amplification. Our results are compared with those of Tzoar and Gersten (TG),<sup>7,8</sup> who investigated similar problems. Their non-

linear mechanism is, however, originated from the nonparabolicity of the electron energy-momentum relation due to band-structure effects. We also consider the theory of third-order frequency mixing.

##### A. Self-Focusing

Two basic parameters of interest in self-focusing problems are the focal length  $R$  and the critical incident power  $P_{cr}$ .<sup>7</sup> The nonlinear dielectric constant  $\epsilon_2$  plays an important role in determining these quantities. Shorter focal lengths and smaller incident power may be realized if  $\epsilon_2$  can be made large. In fact, one may demonstrate that for large ratios of incident to critical power  $P/P_{cr}$

$$R \sim (P/P_{cr})^{-1/2} \sim \epsilon_2^{-1/2}. \quad (11)$$

It is, therefore, of interest to compare the value of  $\epsilon_2$  obtained in this paper with that of TG.<sup>7</sup> The nonlinear dielectric constant  $\epsilon_2^{TG}$  given by these authors is

$$\epsilon_2^{TG} \approx \frac{3}{8} \frac{q^2}{m^2 c_*^2} \frac{\omega_p^2}{\omega^4}, \quad (12)$$

where

$$c_* = (E_g/2m)^{1/2},$$

with  $E_g$  being the gap energy.

Using expressions (10) and (12), we have

$$\frac{\epsilon_2}{\epsilon_2^{TG}} = \frac{32}{45\pi^{1/2}} \left( \frac{v_0}{v_t} \right)^5 \left( \frac{c_*}{v_t} \right)^2 e^{-v_0^2/v_t^2}, \quad (13)$$

if the distribution function  $F_0 \sim \exp(-v^2/v_t^2)$  is assumed to be Maxwellian. This is a reasonable assumption, since we expect  $v_0$  to be larger than the thermal velocity  $v_t$ .

Substituting the data given in TG, we find that the nonlinear mechanism considered here would be stronger than that of TG (i. e.,  $\epsilon_2 > \epsilon_2^{TG}$ ) if

$$\left( \frac{v_0}{v_t} \right)^5 e^{-v_0^2/v_t^2} > 0.02, \quad (14)$$

which can be realized if, for example,  $v_0 = 3v_t$ . The nonlinear mechanism considered in this paper is, however, most effective when  $v_0 \approx 1.6v_t$ , since the left-hand side of the above inequality then attains a maximum value of 0.8.

Heating due to external constant or oscillating electric fields may locally raise the thermal speed of the conduction electrons and flatten the distribution function  $F_0$  (although some experiments do not seem to support this idea). It is then possible that even if our nonlinear mechanism is not

significant at the start, it becomes important as the interaction process continues.

### B. Parametric Excitation

In their paper<sup>8</sup> on the parametric excitation of waves in semiconductor plasmas, Gersten and Tzoar (GT) used the same nonlinear mechanism as that in their work<sup>7</sup> on self-focusing. They considered two processes of excitation, namely, the direct conversion of two identical photons into two plasmons and the stimulated downconversion of a photon to a lower frequency with the emission of two plasmons. The conclusion of GT that these processes should cause instability to occur in the plasma appears doubtful to us, however. A brief examination of their work indicates that their growth rates should in fact be merely frequency shifts. That is, the values of  $\mu$  given by Eq. (13) in GT should be imaginary, but with the magnitudes unchanged.<sup>9</sup> However, it is still meaningful to compare our results with those of GT if the above correction is made.

We first consider the direct-conversion process. Assuming  $\vec{E}(t) = \vec{E}_1(t) \cos \omega_1 t$ , where<sup>8</sup>  $\omega_1 = \omega$ , one may easily obtain the dispersion relation

$$1 - \omega_p^2/\omega^2 + \epsilon_2 E_1^2 \approx 0. \quad (15)$$

The frequency is then given by

$$\omega \approx \pm \omega_p \left(1 - \frac{1}{2} \epsilon_2 E_1^2\right). \quad (16)$$

Since  $\epsilon_2$  is real here, it is clear that there is no damping or growth to this order of approximation. The frequency shift  $\mu_1$  is

$$\mu_1 = \mp \frac{2\pi q^4 v_0^5 F_0''}{15m^3 \epsilon_0 \omega_p^3} E_1^2. \quad (17)$$

We note that if  $\mu_1^{\text{GT}}$  is the frequency shift obtained from GT, then

$$\mu_1/\mu_1^{\text{GT}} = \epsilon_2/\epsilon_2^{\text{GT}}. \quad (18)$$

Thus, our discussion concerning the relative strength of the nonlinear mechanisms in Sec. IV A also applies here.

The second process to be considered here is due to the simultaneous interaction of two beams in the plasma. A photon at frequency  $\omega_1$  is downconverted to frequency  $\omega_2 = \omega_1 - 2\omega$  with the emission of two plasmons. A beam at frequency  $\omega_2$  stimulates this process. The nonlinear dielectric constant for the present case can be derived easily in a similar manner to that of Eq. (10). Comparing the corresponding frequency shift  $\mu_2$  with that ( $\mu_2^{\text{GT}}$ ) obtained from Eq. (13b) of GT, we find that the functional

dependence of the ratio  $\mu_2/\mu_2^{\text{GT}}$  is similar to that of  $\epsilon_2/\epsilon_2^{\text{GT}}$  in Sec. IV A; hence, the conclusions obtained there are also valid for this case.

It is interesting to note that our collision model may also be used to approximate the nonlinear mechanism of TG. The latter mechanism arises because the conduction-electron Hamiltonian in the effective-mass approximation near the bottom of the conduction band is not parabolic in momentum and resembles a relativistic Hamiltonian. The same effect can be approximately achieved if we assume that the effective mass is constant below a certain speed  $v_0$  and becomes infinite for higher velocities. This assumption is, of course, equivalent to considering that electrons with speeds higher than  $v_0$  are stationary and do not participate in the interaction.

### C. Third-Order Frequency Mixing

The use of third-order frequency mixing for diagnostic purposes has recently been treated by many authors.<sup>10,11</sup> It has been demonstrated that nonlinear mixing of two monochromatic waves at frequencies  $\omega_1$  and  $\omega_2$  may be possible in a plasma due to the velocity dependence of the electron collision frequency. Furthermore, it has been shown<sup>11</sup> that the enhanced generation, while weak in general, may be comparatively strong if the mixing frequency, say,  $2\omega_1 - \omega_2$ , is close to the plasma frequency  $\omega_p$ . In Sec. IV C we reconsider this problem using the present collision model.

The current density and the electric fields are related by the Maxwell equation

$$\vec{j} + \epsilon_0 \frac{\partial \vec{E}}{\partial t} = \epsilon_0 \frac{\partial \vec{E}_{\text{ext}}}{\partial t}, \quad (19)$$

where

$$\vec{E}_{\text{ext}} = \vec{E}_1 e^{i\omega_1 t} + \vec{E}_2 e^{i\omega_2 t} + \text{c. c.}$$

is the external electric field.

The current density may be eliminated by combining Eqs. (7) and (19). One thus obtains an equation for the electric field,<sup>6</sup>

$$\frac{\partial^2 \vec{E}}{\partial t^2} + \omega_p^2 \vec{E} = \frac{\partial^2 \vec{E}_{\text{ext}}}{\partial t^2} + \frac{8\pi}{15} \frac{q^4}{m^3 \epsilon_0} v_0^5 F_0'' \times \frac{\partial}{\partial t} \left( \int^t \vec{E} dt \right)^3. \quad (20)$$

By assuming that the nonlinear term is small, Eq. (20) may be solved easily by iteration. The resulting electric field  $\vec{E}_3$  with frequency  $2\omega_1 - \omega_2$  is

$$\vec{E}_3 = - \frac{8}{15\pi^{1/2}} \frac{q^2}{m^2} \omega_p^2 (2\omega_1 - \omega_2) \frac{v_0^5}{v_i} e^{-v_0^2/v_i^2} \frac{(\vec{E}_1^2 \vec{E}_2^* + 2\vec{E}_1 \vec{E}_1 \cdot \vec{E}_2^*)}{[1 - (\omega_p^2/\omega_1^2)]^2 [1 - (\omega_p^2/\omega_2^2)] \omega_1 \omega_2 [(2\omega_1 - \omega_2)^2 - \omega_p^2]}, \quad (21)$$

where we have assumed a Maxwellian distribution. It is clear that  $\vec{E}_3$  is large if  $2\omega_1 - \omega_2$  is close to  $\omega_p$ .

Comparing the enhanced field  $\vec{E}_3$  with that ( $\vec{E}_3^S$ ) obtained by Stenflo,<sup>11</sup> who used a model in which the collision frequency  $\nu$  varies as some power of the velocity, we obtain for hard-sphere collisions

$$\left| \frac{E_3}{E_3^S} \right| \approx 2 \left( \frac{v_0}{v_t} \right)^5 \frac{2\omega_1 - \omega_2}{\nu_1} e^{-v_0^2/v_t^2}, \quad (22)$$

where  $\nu_1 = \nu(v_t^2)$ . Thus, again assuming  $v_0 \gg v_t$ , the present model predicts a greater third-order frequency generation if the mixing frequency is sufficiently larger than the collision frequency.

Although Stenflo's paper is concerned explicitly with the ionospheric plasma, the results are also applicable to semiconductor plasmas. In fact, the similarity between different types of plasmas has recently promoted the possible application of knowledge developed in one plasma to the other, for example, the possibility of using parametric excitation to determine ionospheric conditions.<sup>12</sup>

#### V. CONCLUSION

We have, in an unconventional manner, considered nonlinear effects originating from the velocity dependence of the electron collision frequency. Our results are applied to problems in semiconductor-plasma physics and compared with previous works on the same subjects. It is shown that the nonlinear mechanism proposed here may be of importance in some of these problems. To describe the effects of collisions, we used a very simple, yet somewhat realistic model, such that the mathe-

tical complexity usually associated with collision operators is decisively avoided. It is thus possible to obtain useful qualitative information even with only an extremely small number of calculations.

The Harp model, which we have used here, can also be refined to better approximate the actual velocity dependence of the collision frequency. For example, in the case of a good Ramsauer gas one may write

$$\nu = \begin{cases} 0, & v_1^2 < v^2 < v_0^2 \quad \text{and} \quad v_2^2 < v^2 \\ \infty, & v^2 < v_1^2 \quad \text{and} \quad v_0^2 < v^2 < v_2^2 \end{cases}$$

where  $v_{0,1,2}$  correspond to speeds at which the frequency changes rapidly. The basic difficulty is, of course, the possible uncertainty in the determination of  $v_0, v_1$ , etc. It is also possible, although mathematically much more complicated, to replace the infinity in Eq. (3) by a finite constant collision frequency. The results are not significantly changed, however, if that collision frequency is larger than the other frequencies involved in the problem. In a similar manner one can extend the Harp model to include other plasmas.

In this paper we have neglected effects such as plasma inhomogeneity and velocity-space anisotropy, which can be important in experiments. It should also be pointed out that nonlinear excitations owing to collisional effects are not dominant in general, as other mechanisms, such as nonlinearities owing to nonparabolic energy-momentum relations,<sup>13</sup> may be more important in some plasmas. However, we believe that the basic physical picture obtained from our results should be useful in many further experimental and theoretical investigations.

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<sup>5</sup>The mass  $m$  is assumed to be constant. We take this opportunity to stress that there exist alternative and important causes for the nonlinearities considered here, e.g., the nonparabolicity effect discussed later. In fact, there is a raging and still unresolved controversy in the literature as to which is the dominant effect in particular situations.

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1650 (1971).

<sup>9</sup>Let us, for example, rederive (13a) of Ref. 8. Following the notations of GT, we write the current density as  $\vec{j} = q \int \vec{v} f_0 (\vec{p} - q \int^t \vec{E} dt) d\vec{p}$  where  $\vec{v} = c_* \vec{p} [(m c_*)^2 + p^2]^{-1/2}$ . By means of a Taylor expansion we then obtain  $\vec{j} \approx nq^2/m \int^t \vec{E} dt - (nq^4/2m^3 c_*^2) (\int^t \vec{E} dt)^3$  where  $n = \int f_0 d\vec{p}$  is the conduction-electron number density. A similar equation was also derived in Ref. 7. It corresponds to Eq. (7) of the present paper. Replacing  $\vec{j}$  by  $-\epsilon_0 \partial \vec{E} / \partial t$ , we then obtain, except for a factor  $\sqrt{-1}$ , an expression like (13a) in Ref. 8.

<sup>10</sup>For example, see B. Y. Lao and M. M. Litvak, J. Appl. Phys. **42**, 3357 (1971); M. S. Sodha, P. K. Dubey, S. K. Sharma, and P. K. Kaw, Phys. Rev. B **1**, 3426 (1970), and the many basic references therein such as the one by P. K. Kaw, Phys. Rev. Lett. **21**, 539 (1968).

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<sup>13</sup>For example, C. K. N. Patel, R. E. Slusher, and P. A. Fleury, Phys. Rev. Lett. **17**, 1011 (1966); P. A. Wolff and G. A. Pearson, Phys. Rev. Lett. **17**, 1015 (1966).