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Optical Mixing by Mobile Carriers in Semiconductors

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Certain nonlinear optical-mixing phenomena associated with collisional effects in drifted semiconductor plasmas have been considered by means of a simple kinetic analysis. This leads to important modifications in the results of two recent papers.

INTRODUCTION

In a recent paper,[1] hereafter referred to as I, it has been shown that the nonlinearities associated with the energy dependence of the relaxation time in semiconductors can make important contributions to the optically mixed output. Thus, the current induced by the electric fields of two laser beams (frequencies ω_1 and ω_2) may have components with frequency $2\omega_1-\omega_2$, which are mainly due to collisional effects.

An error in I has recently been pointed out in another paper[2] (hereafter referred to as II), where a still more significant contribution to the current was found for frequencies larger than the inverse of the relaxation time. The purpose of the present paper is to calculate the induced current using exactly the same simplifying assumptions as those of I and II. In addition, a constant external electric field $\mathbf{E}_0$ will also be assumed to be present. The calculations will be performed by means of a kinetic analysis, whereas I and II used a moment-equation approach. The results of the present paper are compared with those of II. They are mainly equal, if the dc electric field $\mathbf{E}_0$ is zero, but differ significantly from one another, if $\mathbf{E}_0$ is sufficiently large. Thus, even the direction of the current component with frequency $2\omega_1-\omega_2$ may be different from that of II, if the current carriers have a constant nonzero drift velocity. This example illustrates clearly that results on semiconductor plasmas, derived by means of hydromagnetic equations, may be misleading. Further, we propose a new diagnostic method to determine the velocity distribution function for current carriers in semiconductors.

BASIC EQUATIONS AND RESULTS

We consider a *spatially homogeneous* electron plasma, which is situated in external electric fields. The distribution function for the carriers $F(\mathbf{v},t)$ has to satisfy the Boltzmann equation

$$\frac{\partial F}{\partial t}+\frac{q\mathbf{E}_0}{m}\cdot\frac{\partial F}{\partial \mathbf{v}}+\boldsymbol{\gamma}\cdot\frac{\partial F}{\partial \mathbf{v}}=C(F)\,, \qquad (1)$$

where $\mathbf{E}_0$ is a constant electric field,

$$(m/q)\boldsymbol{\gamma}=\mathbf{E}_1e^{i\omega_1 t}+\mathbf{E}_2e^{i\omega_2 t}+(\text{c.c.})$$

is the electric field due to externally impressed laser beams, q is the electronic charge, m is the electron effective mass, and $C(F)$ is the collision integral.

We will here divide F into two parts,

$$F=F_0+F_{12}\,,$$

where $F_0(\mathbf{v})$ is the distribution function in the absence of the field $\boldsymbol{\gamma}$. Equation (1) thus yields

$$\frac{q}{m}\mathbf{E}_0\cdot\frac{\partial F_0}{\partial \mathbf{v}}=C(F_0) \qquad (2a)$$

and

$$\frac{\partial F_{12}}{\partial t}+\boldsymbol{\gamma}\cdot\frac{\partial F_0}{\partial \mathbf{v}}+\left(\boldsymbol{\gamma}+\frac{q\mathbf{E}_0}{m}\right)\cdot\frac{\partial F_{12}}{\partial \mathbf{v}}=C(F_0+F_{12})-C(F_0)\,. \qquad (2b)$$

Equation (2b) is now multiplied by $q\mathbf{v}$ and integrated over velocity space. We obtain

$$\frac{\partial \mathbf{j}}{\partial t}-qn_0\boldsymbol{\gamma}=q\int \mathbf{v}[C(F_0+F_{12})-C(F_0)]\,d\mathbf{v}=q\int \mathbf{v}C(F_{12})\,d\mathbf{v}\,, \qquad (3)$$

where $\mathbf{j}=q\int\mathbf{v}F_{12}\,d\mathbf{v}$ is the current induced by the laser beams and where $n_0=\int F\,d\mathbf{v}$ is the (constant) density of the carriers.

The last equality in (3) follows from the fact that electron-electron collisions, which conserve momentum, do not contribute to the right-hand side of (3) whereas the remainder of the collision integral C, which is due to scattering of electrons by molecules, phonons, impurities, etc., is linear.

Following I and II we now assume that ω_1 and ω_2, as well as their sum and difference frequencies, are much larger than any collision frequency of interest, i.e.,

$$\frac{\partial F_{12}}{\partial t}\gg C(F_{12}) \quad\text{and}\quad \frac{\partial F_{12}}{\partial t}\gg\frac{q\mathbf{E}_0}{m}\cdot\frac{\partial F_{12}}{\partial \mathbf{v}}\,. \qquad (4)$$

[1] P. K. Kaw, Phys. Rev. Letters **21**, 539 (1968).
[2] B. S. Krishnamurthy and V. V. Paranjape, Phys. Rev. **181**, 1153 (1969).

The function F_{12} in the right-hand side of (3) can then approximately be replaced by the solution of

$$\frac{\partial F_{12}}{\partial t}+\boldsymbol{\gamma}\cdot\frac{\partial F_0(\mathbf{v})}{\partial \mathbf{v}}+\boldsymbol{\gamma}\cdot\frac{\partial F_{12}}{\partial \mathbf{v}}=0 \tag{5a}$$

or

$$F_{12}=F_0(\mathbf{v}-\mathbf{u})-F_0(\mathbf{v}), \tag{5b}$$

where

$$\mathbf{u}=\int^t \boldsymbol{\gamma}\,dt=\frac{q}{m}\left(\frac{\mathbf{E}_1e^{i\omega_1 t}}{i\omega_1}+\frac{\mathbf{E}_2e^{i\omega_2 t}}{i\omega_2}+(\text{c.c.})\right). \tag{5c}$$

Accordingly, we write Eq. (3) in the form

$$\frac{\partial \mathbf{j}}{\partial t}-qn_0\boldsymbol{\gamma}=q\int \mathbf{v}C[F_0(\mathbf{v}-\mathbf{u})-F_0(\mathbf{v})]\,d\mathbf{v}. \tag{6}$$

The different time-dependent components of $\mathbf{j}$ can now be determined from Eq. (6) and our problem is thus in principle already solved.

In accordance with our limitation to large frequencies we now assume that the absolute value of the vector $\mathbf{u}$ is much smaller than that of $\mathbf{v}$ for most velocities of interest. If, for simplicity, we further assume that all electric fields and currents are parallel to the x axis ($\mathbf{E}=E\mathbf{e}_x$, $\mathbf{u}=u\mathbf{e}_x$, $\mathbf{j}=j\mathbf{e}_x$, etc.), the Taylor expansion of $F_0(\mathbf{v}-\mathbf{u})$ yields

$$F_0(\mathbf{v}-\mathbf{u})-F_0(\mathbf{v})\approx -u\frac{\partial F_0}{\partial v_x}+\tfrac{1}{2}u^2\frac{\partial^2 F_0}{\partial v_x^2}-\tfrac{1}{6}u^3\frac{\partial^3 F_0}{\partial v_x^3}. \tag{7}$$

Following I and II we focus our attention on the current component $\mathbf{j}_3$, which corresponds to the frequency $2\omega_1-\omega_2$. Accordingly, by means of (5c) we write

$$u^3=-\frac{3iq^3}{m^3}\frac{E_1^2E_2^*}{\omega_1^2\omega_2}e^{i(2\omega_1-\omega_2)t}+\text{terms with frequencies}$$
$$\pm\omega_1,\pm\omega_2,\pm3\omega_1,\pm3\omega_2,\pm(\omega_1\pm2\omega_2),\pm(2\omega_1+\omega_2),$$
$$\omega_2-2\omega_1, \tag{8}$$

where the star over E_2 stands for its complex conjugate. Equations (6)–(8) then yield

$$\frac{\partial \mathbf{j}_3}{\partial t}=i(2\omega_1-\omega_2)\mathbf{j}_3=\frac{iq^4E_1^2E_2^*}{2m^3\omega_1^2\omega_2}e^{i(2\omega_1-\omega_2)t}\times\int \mathbf{v}C\left(\frac{\partial^3F_0}{\partial v_x^3}\right)d\mathbf{v}. \tag{9}$$

Equation (9), which is the basic result of this paper, corresponds to (8) in I and to (8) in II.

DISCUSSION AND COMPARISONS WITH PREVIOUS WORKS

In order to make a detailed comparison with the results derived in II (and I) we will now define a momentum transfer collision frequency[3] $\nu(v^2)$ from the relation

$$\int \mathbf{v}C(F_a)d\mathbf{v}=-\int \mathbf{v}\nu F_a\,d\mathbf{v}, \tag{10}$$

where F_a is an arbitrary distribution function.

Equation (9) can then be rewritten in the form

$$\mathbf{j}_3=-\frac{q^4E_1^2E_2^*}{2m^3\omega_1^2\omega_2(2\omega_1-\omega_2)}e^{i(2\omega_1-\omega_2)t}\int \mathbf{v}\nu\frac{\partial^3F_0}{\partial v_x^3}d\mathbf{v}. \tag{11}$$

In the moment-equation approach it is usually assumed that $\nu(v^2)$ approximately can be replaced by $\nu(\langle v^2\rangle)$, where $\langle v^2\rangle$ is the mean-square velocity, i.e.,

$$\langle v^2\rangle=n_0^{-1}\int v^2F\,d\mathbf{v}. \tag{12}$$

It is then possible to introduce a momentum relaxation time $\tau=[\nu(\langle v^2\rangle)]^{-1}$ (see, e.g., II). Accordingly, if we replace ν in Eq. (11) by

$$\nu\approx\tau^{-1}+(v^2-\langle v^2\rangle)\frac{\partial\tau^{-1}}{\partial\langle v^2\rangle}, \tag{13}$$

we easily find that Eq. (11) in the present work and Eq. (8) in II are identical. However, as will be illustrated below, an approximation like (13) may be dangerous.

Let us, for simplicity, now suppose that ν varies with energy according to some arbitrary power law, i.e., ν is proportional to v^{-n}. By means of three partial integrations of (11) we then obtain

$$j_3=\frac{q^4E_1^2E_2^*n}{2m^3\omega_1^2\omega_2(2\omega_1-\omega_2)}e^{i(2\omega_1-\omega_2)t}\int\left(-3+6(n+2)\frac{v_x^2}{v^2}-(n+2)(n+4)\frac{v_x^4}{v^4}\right)\frac{\nu}{v^2}F_0\,d\mathbf{v}. \tag{14}$$

We have here limited ourselves to cases where n is smaller than unity in order to ensure convergence of (14) and to avoid run-away phenomena.[4]

We shall now consider (14) for different limiting cases. Thus, we assume that $F_0(=F_{\text{is}})$ is almost isotropic in velocity space (which means that the drift velocity is considerably smaller than the thermal velocity). Accordingly, it is then possible to replace v_x^2/v^2 and v_x^4/v^4 in (14) by $\frac{1}{3}$ and $\frac{1}{5}$, respectively. On the other hand, if we assume that $F_0(=F_{\text{an}})$ is strongly

[3] I. P. Shkarofsky, T. W. Johnston, and M. P. Bachynski, *The Particle Kinetics of Plasmas* (Addison-Wesley Publishing Co. Inc., London, 1966), Chap. 3; E. M. Conwell, *High Field Transport in Semiconductors* (Academic Press Inc., New York, 1967), Chap. 5.

[4] L. Stenflo, Plasma Phys. **10**, 801 (1968).

anisotropic in velocity space (the drift velocity is much larger than the thermal velocity), it is possible to replace both v_x^2/v^2 and v_x^4/v^4 in (14) by 1. By means of (14) we thus obtain

$$\frac{j_{3\,\mathrm{is}}}{j_{3\,\mathrm{an}}}=-\frac{1}{5}\frac{3-n}{1+n}\left(\int v^{-2}\nu F_{\mathrm{is}}\,d\mathbf{v}\bigg/\int v^{-2}\nu F_{\mathrm{an}}\,d\mathbf{v}\right) \quad (15)$$

as well as

$$\frac{j_{3\,\mathrm{is}}}{j_{3\,\mathrm{II}}}=\frac{(3-n)(1-n)}{15}\frac{\langle v^2\rangle\tau}{n_0}\int\frac{\nu}{v^2}F_{\mathrm{is}}\,d\mathbf{v}, \quad (16)$$

where $j_{3\,\mathrm{II}}$ denotes the expression for j_3, which was derived in II.

If, in addition, we assume that F_{is} is (an undrifted) Maxwellian, the right-hand side of (16) reduces to (8/15) $(\frac{3}{2})^{n/2+1}\pi^{-1/2}\Gamma[(5-n)/2]$, where Γ is the γ function. The current component $j_{3\,\mathrm{II}}$ should thus in general be corrected by a factor of order unity. This factor, which is due to an approximation like that of (13), may even be significantly different from unity, if F_{is} is slightly non-Maxwellian for small velocities. It is more remarkable, however, to note from Eq. (15) that *the sign of* $j_{3\,\mathrm{an}}$ *in general is different from that of* $j_{3\,\mathrm{is}}$ (or $j_{3\,\mathrm{II}}$). Accordingly, it is evident that the sign of j_3 can be changed when the applied constant electric field $\mathbf{E}_0$ is so large that the magnitude of the drift velocity is comparable to the thermal velocity. Such anisotropies in velocity space occur in many semiconductor plasmas of interest,[5] where experimental values for j_3 thus can give information about the degree of anisotropy. The numerical data may also contribute to the development of a somewhat more reliable description of the distribution function.

In conclusion, it may be worthwhile to point out that hydromagnetic equations yield results which are comparatively insensitive to details of the scattering processes and to the form of the distribution function. Thus, the numerical coefficients derived in I and II are not correct and can even be significantly wrong[6] if the distribution function is highly anisotropic in velocity space. The kinetic approach to the problem, which also is very simple and straightforward, avoids some basic limitations of the moment-equation approach, however, and predicts different results.

Following I and II we have here made four basic assumptions; namely, that the fields are homogeneous in space, ionization as well as recombination phenomena can be neglected, the conduction band is parabolic, and the frequencies ω_1 and ω_2 are much larger than any other characteristic frequency of the system. If these assumptions are not valid, it should be possible to derive similar, although much more complicated, results by means of more elaborate methods.[7] A detailed treatment of resonant wave coupling phenomena[8] may also be of interest.

[5] See, e.g., L. Stenflo, Proc., IEEE **54**, 1970 (1966); **55**, 1088 (1967).

[6] It should be stressed, however, that the authors of I and II did not intend to consider the case where F_0 has a nonzero drift velocity.

[7] See, e.g., P. Das, Phys. Rev. **138**, A590 (1965); M. S. Sodha and P. K. Kaw, Proc. Phys. Soc. (London) **88**, 373 (1966); I. P. Shkarofsky, Plasma Phys. **10**, 169 (1968); P. K. Kaw, J. Appl. Phys. **40**, 793 (1969); M. S. Sodha and G. P. Gupta, Plasma Phys. **11**, 473 (1969); M. S. Sodha and S. Sharma, J. Phys. C **2**, 914 (1969).

[8] A. Jarmén, L. Stenflo, H. Wilhelmsson, and F. Engelmann, Phys. Letters **28A**, 748 (1969); H. Wilhelmsson, L. Stenflo, and F. Engelmann, J. Math. Phys. (to be published).