Optical mixing by mobile carriers in semiconductors in the presence of a dc homogeneous magnetic field*

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The effect of a magnetic field on the optical mixing by mobile carriers in semiconductors is discussed for each of the two mechanisms of nonparabolic band and energy-dependent collision time. It is shown that for both these mechanisms an external magnetic field enhances the mixed output; however, the results for the two cases are sufficiently different that a magnetic field may be used to distinguish between the two effects.

I. INTRODUCTION

The phenomenon of optical mixing of two laser beams in semiconductors such as InSb and InAs, etc. has been investigated in a recent series of experiments.[1] In this process, two monochromatic laser beams of frequencies ω_1 and ω_2 interact in a semiconductor to produce a mixed output at the difference frequencies $\omega_3 = 2\omega_1 - \omega_2$ and $\omega_4 = 2\omega_2 - \omega_1$. Measurements[1] indicated that the generated mixed output had a relatively large magnitude, and it was found to be dependent on the mobile-carrier concentration. This showed that the mobile carriers were responsible for the optical mixing, instead of the nonlinearity arising because of the polarization of the background lattice.[2]

Two mechanisms have been put forward to explain the optical mixing using the mobile carriers in the semiconductors. Wolff and Pearson[3] propose that the observed nonlinearity is due to nonparabolicity of the conduction band, an effect which is known to be relatively large in crystals such as InSb and InAs. Here the crucial point is that an electron's velocity is a nonlinear function of its momentum, which oscillates at the frequency of the applied electromagnetic fields. The nonlinear velocity-momentum relationship results in an induced current containing mixed frequency components.

The other mechanism, proposed by Kaw,[4] makes use of certain nonlinearities associated with collision processes. These nonlinear effects are due to the energy dependence of the momentum relaxation time for the conduction electrons in semiconductors, and will exist even for crystals with parabolic bands.

In this paper we discuss the effect of a constant magnetic field on the optical mixing by mobile carriers in semiconductors. The influence of the magnetic field on both of the above-mentioned mechanisms is considered. The magnetic field causes an increase in the nonlinear properties[5] of the semiconductor for both mechanisms. Hence the mixed optical output is also increased. The main limitation on the output is the collision time τ. Hence we are limited to the case when ω_1, ω_2, ω_c, $\omega_1 - \omega_c$, $\omega_2 - \omega_c$, and $\omega_3 - \omega_c$ are much larger than τ^{-1}, where ω_c is the cyclotron frequency. We find that the enhancement of the output by the magnetic field is quite different for the two mechanisms. By proper choice of ω_1, ω_2, and ω_c it is shown that the magnetic field enhancement for the nonparabolic-band case is almost six orders of magnitude, while for the energy-dependent collision-time case, for the same parameters, the enhancement is only four orders of magnitude.

In Sec. II we derive an expression for the mixed output in a semiconductor in a magnetic field using the nonparabolic mechanism. In Sec. III the same effect is considered using the energy-dependent collision-time approach, and in Sec. IV a comparison and discussion of the two results are presented.

II. NONLINEARITY DUE TO NONPARABOLICITY

For a nonparabolic semiconductor placed in a magnetic field, the equation of motion for an electron in the conduction band is given by[5]

$$\frac{d}{dt}\frac{m^*\vec{v}}{[1-(v/c^*)^2]^{1/2}} = -e\left(\vec{E}+\frac{\vec{v}}{c}\times\vec{B}\right), \tag{1}$$

where m^* is the electron's effective mass near the bottom of the conduction band and $c^* = (E_g/2m^*)^{1/2}$, where E_g is the energy gap. $\vec{E}$ and $\vec{B}$ represent the electric and magnetic fields, respectively, and are given in terms of vector potential $\vec{A}_0$ as

$$\vec{E} = -\frac{1}{c}\frac{\partial\vec{A}_0}{\partial t}, \quad \vec{B} = \nabla\times\vec{A}_0 .$$

If the external uniform magnetic field B_0 is along the z direction and the sample is irradiated with circularly polarized optical beams of frequencies ω_1 and ω_2, the vector potential can be written

$$\vec{A}_0 = \tfrac{1}{2}\vec{B}_0\times\vec{r} + \vec{A}, \tag{2}$$

where

$$\vec{A}=\frac{1}{2}\sum_{\lambda}^{1,2}[\vec{A}_\lambda e^{-i(\vec{k}_\lambda\cdot\vec{r}-\omega_\lambda t)}+(\text{complex conjugate})]\,, \tag{3a}$$

$$\vec{A}_\lambda=A_\lambda(\hat{x}+i\hat{y}). \tag{3b}$$

In Eqs. (2) and (3) $\vec{A}$ represents the vector potential due to the optical beams and $\vec{k}_\lambda$ is the wave vector. In calculating the electron dynamics we have neglected the Lorentz force of the ac magnetic field and the $(\vec{v}\cdot\nabla)\vec{v}$ term. This is justified by the fact that $c^*/c \ll 1$ (see Ref. 6).

Using a dimensionless velocity $\vec{u}=\vec{v}/c^*$ and substituting Eqs. (2) and (3) into Eq. (1), the equation of motion can be rewritten

$$\frac{d}{dt}\left(\frac{\vec{u}}{(1-u^2)^{1/2}}\right)=\mu\frac{d\vec{A}}{dt}-i\omega_c\vec{u}\,, \tag{4}$$

where $\mu=e/m^*c^*c$, $\omega_c=eB_0/m^*c$ is the cyclotron frequency, and the velocity $\vec{u}$ is a circularly polarized vector.

We employ a series solution method for Eq. (4). Let

$$\vec{u}=\mu\vec{U}_1+\mu^3\vec{U}_2+\mu^5\vec{U}_3+\cdots\,, \tag{5}$$

where

$$\vec{U}_1\equiv\frac{1}{2}\sum_{\lambda}^{1,2}(\vec{U}_{1\lambda}e^{-i(\vec{k}_\lambda\cdot\vec{r}-\omega_\lambda t)}+\text{c.c.}) \tag{6a}$$

and

$$\vec{U}_{1\lambda}=U_{1\lambda}(\hat{x}+i\hat{y}). \tag{6b}$$

Therefore

$$\begin{aligned}\frac{\vec{u}}{(1-u^2)^{1/2}}&=\vec{u}(1+\tfrac{1}{2}\vec{u}\cdot\vec{u}+\cdots)\\&=(\mu\vec{U}_1+\mu^3\vec{U}_2+\mu^5\vec{U}_3+\cdots)\\&\quad\times[1+\tfrac{1}{2}(\mu\vec{U}_1+\mu^3\vec{U}_2+\cdots)\\&\quad\cdot(\mu\vec{U}_1+\mu^3\vec{U}_2+\cdots)+\cdots]\,.\end{aligned} \tag{7}$$

Substituting Eqs. (5) and (7) into Eq. (4) and comparing coefficients of μ gives

$$\frac{d}{dt}\vec{U}_1=\frac{d\vec{A}}{dt}-i\omega_c\vec{U}_1\,, \tag{8}$$

from which, using Eqs. (3) and (6), we get

$$\vec{U}_{11}=\omega_1\vec{A}_1/(\omega_1-\omega_c)\,, \tag{9a}$$

$$\vec{U}_{12}=\omega_2\vec{A}_2/(\omega_2-\omega_c)\,. \tag{9b}$$

Comparing coefficients of μ^3 gives

$$\frac{d}{dt}[\vec{U}_2+\tfrac{1}{2}\vec{U}_1\cdot(\vec{U}_1\cdot\vec{U}_1)]=-i\omega_c\vec{U}_2\,. \tag{10}$$

Equation (10) clearly shows the presence of mixed frequency components due to the term $\vec{U}_1\cdot(\vec{U}_1\cdot\vec{U}_1)$, which contains the frequencies $\omega_3=2\omega_1-\omega_2$ and $\omega_4=2\omega_2-\omega_1$ in addition to ω_1 and ω_2.

Writing $\vec{U}_2$ as

$$\vec{U}_2=\sum^{1,2,3,4}\vec{U}_{2\lambda}e^{-i(\vec{k}_\lambda\cdot\vec{r}-\omega_\lambda t)}\,, \tag{11a}$$

where

$$\vec{U}_{2\lambda}=U_{2\lambda}(\hat{x}+i\hat{y})\,, \tag{11b}$$

and substituting into Eq. (10) we can get various $\vec{U}_{2\lambda}$'s. However, we are only interested in $\vec{U}_{23}$ with frequency ω_3 and obtain it as

$$\vec{U}_{23}=-\frac{\vec{A}_2}{2}\,\frac{A_1^2}{(1-\omega_c/\omega_3)(1-\omega_c/\omega_1)^2(1-\omega_c/\omega_2)}\,. \tag{12}$$

The third-order velocity component of frequency ω_3 is written

$$\vec{u}^{(3)}=\tfrac{1}{2}\mu^3(\vec{U}_{23}e^{-i(\vec{k}_3\cdot\vec{r}-\omega_3 t)}+\text{c.c.})\,. \tag{13}$$

The amplitude of the third-order current $\vec{J}^{(3)}$ of frequency component ω_3 is hence given by

$$\begin{aligned}\vec{J}^{(3)}&=-Ne\vec{v}^{(3)}\\&=+\frac{Ne^4}{2m^{*3}c^3c^{*2}}\,\frac{A_1^2\vec{A}_2}{(1-\omega_c/\omega_3)(1-\omega_c/\omega_1)^2(1-\omega_c/\omega_2)}\\&=\frac{e^4N}{m^{*2}E_g}\,\frac{E_1^2\vec{E}_2\omega_3}{(\omega_3-\omega_c)(\omega_1-\omega_c)^2(\omega_2-\omega_c)}\,,\end{aligned} \tag{14}$$

where N is the carrier concentration, $\vec{E}_1=\omega_1\vec{A}_1/c$, and $\vec{E}_2=\omega_2\vec{A}_2/c$.

To estimate the intensity of the ω_3 beam generated by the nonlinear mixing, one must solve the driven wave equation

$$\nabla^2\vec{E}_3-\frac{1}{c^2}\frac{\partial^2\vec{D}_3}{\partial t^2}=\frac{4\pi}{c^2}\,\frac{\partial\vec{J}^{(3)}}{\partial t}\,,$$

where

$$\vec{D}_3=\epsilon(\omega_3)\vec{E}_3\,. \tag{15}$$

Substituting Eq. (14) into Eq. (15), one finds that the amplitude of $\vec{E}_3$ grows linearly with interaction distance l, and if the absorption is neglected one gets for the magnitude of $\vec{E}_3$

$$E_3=\frac{2\pi e^4NlE_1^2E_2\omega_3^2}{c^2m^{*2}E_gk_3(\omega_3-\omega_c)(\omega_1-\omega_c)^2(\omega_2-\omega_c)}\,. \tag{16}$$

As long as l is smaller than the photon mean free path or the dephasing length, E_3 will represent electric field of the generated radiation at ω_3.

III. NONLINEARITY DUE TO ENERGY-DEPENDENT COLLISION TIME

Consider a semiconductor with a parabolic conduction band placed in a uniform magnetic field $\vec{B}_0$. The equation of motion for an electron of effective mass m^* in the conduction band can be written

$$\frac{\partial \vec{v}}{\partial t}+\frac{\vec{v}}{\tau}=-\frac{e}{m^*}\left(\vec{E}+\frac{\vec{v}}{c}\times\vec{B}_0\right) , \tag{17}$$

where a finite momentum relaxation time τ is considered and $\vec{E}$ represents the electric field due to the optical beams incident on the crystal. The ac magnetic field has been neglected.[6] As before, the electric field can be expressed

$$\vec{E}=\frac{1}{2}\sum_{\lambda}^{1,2}[E_\lambda e^{-i(\vec{k}_\lambda\cdot\vec{r}-\omega_\lambda t)}+(\text{complex conjugate})] . \tag{18}$$

The momentum relaxation time τ is a function of the energy. Now in the presence of the strong electric field, the carrier temperature T_e becomes different from the lattice temperature T and one can express the energy dependence of the relaxation time τ as[4]

$$\tau=\tau_0(T_e/T)^{n/2} ,$$

where τ_0 is the relaxation time corresponding to the lattice temperature and n is an integer. At the temperatures at which the experiment is usually performed (e.g., liquid N_2), the dominant scattering is by ionized impurity, for which $n=3$. Under these conditions, we also have

$$(T_e-T)/T\ll 1 ,$$

so that

$$\tau^{-1}\simeq\tau_0^{-1}[1-\tfrac{1}{2}(T_e/T-1)] . \tag{19}$$

For the carrier temperature, one can write[4] the energy balance equation

$$\frac{dT_e}{dt}=\frac{2}{3Nk}(\vec{J}_r\cdot\vec{E})-\frac{T_e-T}{\tau_\epsilon} , \tag{20}$$

where $\vec{J}$ is the current density ($\vec{J}_r$ denotes its real part), N is the carrier concentration, k is the Boltzmann constant, and τ_ϵ is the energy relaxation time for carriers. Equations (17)–(20) give a complete solution of the problem. The first-order steady-state solution of Eq. (17) is directly given as

$$\vec{v}_1=-\frac{e\vec{E}_1\tau}{m^*[1+i(\omega_1-\omega_c)\tau]}e^{-i(\vec{k}_1\cdot\vec{r}-\omega_1 t)} , \tag{21a}$$

$$\vec{v}_2=-\frac{e\vec{E}_2\tau}{m^*[1+i(\omega_2-\omega_c)\tau]}e^{-i(\vec{k}_2\cdot\vec{r}-\omega_2 t)} . \tag{21b}$$

Assuming ω_1, ω_2, $\omega_1-\omega_2$, ω_c, $\omega_1-\omega_c$, and $\omega_2-\omega_c\gg\tau_\epsilon^{-1}$, one can use the above equations in Eq. (20) to obtain the $2\omega_1$ and $\omega_1-\omega_2$ frequency components of T_e as

$$T_{e1}=\frac{-ie^2E_1^2}{6m^*k\omega_1(\omega_1-\omega_c)^2\tau}e^{-i(2\vec{k}_2\cdot\vec{r}-2\omega_1 t)} , \tag{22a}$$

and

$$T_{e2}=\frac{-ie^2E_1E_2}{3m^*k\tau(\omega_1-\omega_2)}\frac{(\omega_1-\omega_c)^2+(\omega_2-\omega_c)^2}{(\omega_1-\omega_c)^2(\omega_2-\omega_c)^2}e^{-i[(\vec{k}_1-\vec{k}_2)\cdot\vec{r}-(\omega_1-\omega_2)t]} . \tag{22b}$$

Using Eqs. (22a), (22b), and (19) in Eq. (17) one obtains various mixed frequency components at frequencies $(2\omega_1\pm\omega_2)$, $(2\omega_2\pm\omega_1)$, etc. in the carrier velocity. The current density $\vec{J}$ is related to the carrier velocity $\vec{v}$ by the relation

$$\vec{J}=-Ne\vec{v} . \tag{23}$$

We are interested only in the frequency component $\omega_3=2\omega_1-\omega_2$ and obtain the corresponding current-density component as

$$\vec{J}^{(3)}=\frac{n}{3}\frac{iNe^4E_1^2\vec{E}_2}{4m^{*2}kT\omega_3}\left(\frac{2[(\omega_1-\omega_c)^2+(\omega_2-\omega_c)^2]}{(\omega_1-\omega_c)(\omega_2-\omega_c)(\omega_1-\omega_2)}-\frac{1}{\omega_1}\right)\frac{1}{\omega_2-\omega_c}\frac{1}{(\omega_1-\omega_c)^2\tau_\epsilon^2} . \tag{24}$$

Using the driven wave Eq. (15) as in Sec. II, we obtain the amplitude of E_3 with frequency ω_3

$$E_3=\frac{2\pi}{c^2}\frac{n}{3}\frac{Ne^4E_1^2E_2}{4m^*kT}\frac{l}{k_3}\left(\frac{2[(\omega_1-\omega_c)^2+(\omega_2-\omega_c)^2]}{(\omega_1-\omega_c)(\omega_2-\omega_c)(\omega_1-\omega_2)}-\frac{1}{\omega_1}\right)\frac{1}{(\omega_2-\omega_c)}\frac{1}{(\omega_1-\omega_c)^2\tau_\epsilon^2} . \tag{25}$$

Here E_3 is the electric field of the radiation of frequency ω_3.

IV. DISCUSSION

We now make an estimate of the enhancement of the nonlinear effect by using actual numerical values for various frequencies. The expressions given by Eqs. (16) and (25) show that if it were not for the limitation of

$$(\omega_1-\omega_c),\ (\omega_2-\omega_c),\ \text{and}\ (\omega_3-\omega_c)\gg\tau^{-1} ,$$

we could get extremely large mixed outputs. However the above conditions restrict us to values of ω_c away from resonance with all of ω_1, ω_2, and ω_3.

We take ω_1 and ω_2 to correspond to the 10.6-

and the 9.6-μm lines, respectively, for the CO_2 laser. This gives $\omega_3 = 2\omega_1 - \omega_2$ to correspond to the 11.8-μm line. These values are the same as used by Patel *et al.*[1] in their experiment. The calculations are performed for InSb which has a low electronic effective mass $m^* = \frac{1}{60} m_e$ (where m_e is the free-electron mass) and relatively small energy gap $E_g = 0.234$ eV. This yields $c^*/c = \frac{1}{270}$, thus justifying the neglect of the ac magnetic field in Eq. (2).

The carrier concentration N is taken to be 9×10^{15} cm^{-3}, corresponding to the plasma frequency $\omega_p = 1.04\times10^{12}$ rad/sec. Our enhancement results are obtained for $\omega_c = 0.8\omega_1$. This corresponds to a magnetic field of 135 kG, a value obtainable in the laboratory. Using these parameters, we calculate the coherence length $l_c = \pi/(2k_1 - k_2 - k_3)$ to be 0.21 mm. The optical-mixing theory requires that the crystal length be smaller than the coherence length. Therefore, if the experiment be performed with a crystal of thickness 0.1 mm, our results for E_3 are valid. Taking $\omega_p\tau = 13$ for our crystal sample,[7] we find the photon mean free path to be well above the sample length. Also using this value of $\omega_p\tau$, we see that $(\omega_1 - \omega_c)\tau$, $(\omega_2 - \omega_c)\tau$, and $(\omega_3 - \omega_c)\tau$ are all large in comparison with unity.

Using the above interaction length, an estimate is now made of the minimum power obtained for the mixed frequency. This will obviously happen in the absence of the magnetic field. As in Ref. 1, the powers of the ω_1 and ω_2 frequency beams are taken to be $P_1 = 10^3$ and $P_2 = 10^2$ W, respectively, and the focal area 10^{-3} cm^2. Using Eq. (16) we obtain the power for the mixed frequency ω_3 to be $P_3 = 1.2\times10^{-4}$ W for the case $\omega_c = 0$. The experiments in Ref. 1 show that this power can easily be measured in the laboratory. Of course, in the absence of the magnetic field, the photon mean free path and the coherence length will become much larger, and the condition of interaction length l's being smaller than either of these values will always be satisfied.

We now calculate the enhancement of the nonlinear effect by the magnetic field. Denoting the power P_3 obtained by nonparabolic mechanism as P_3^{NP}, the ratio of the power with the magnetic field present, and in the absence of the field, is obtained as

$$\rho^{NP} = \frac{P_3^{NP}(\omega_c = 0.8\,\omega_1)}{P_3^{NP}(\omega_c = 0)} = \left(\frac{\omega_3\omega_1^2\omega_2}{(\omega_3 - \omega_c)(\omega_1 - \omega_c)^2(\omega_2 - \omega_c)}\right)^2 = 7.2\times10^5\ ,$$

showing a large increase in the mixed output owing to the effect of the magnetic field.

Denoting the corresponding power for the energy-dependent collision time mechanism by P_3^{ED}, we obtain the magnetic-field-enhancement ratio

$$\rho^{ED} = \frac{P_3^{ED}(\omega_c = 0.8\,\omega_1)}{P_3^{ED}(\omega_c = 0)} = \frac{\{2[(\omega_1 - \omega_c)^2 + (\omega_2 - \omega_c)^2]/(\omega_1 - \omega_2)(\omega_1 - \omega_c)(\omega_2 - \omega_c) - 1/\omega_1\}^2(\omega_2\omega_1^2)^2}{[2(\omega_1^2 + \omega_2^2)/(\omega_1 - \omega_2)\omega_1\omega_2 - 1/\omega_1]^2[(\omega_2 - \omega_c)(\omega_1 - \omega_c)^2]^2} = 9.6\times10^3\ .$$

We note that for the nonparabolic mechanism, for the parameters chosen above, the enhancement by the magnetic field is almost two orders of magnitude larger than for the energy-dependent scattering mechanism. At present, it is generally believed[8] that for narrow-gap semiconductors (such as InSb) the dominant nonlinear effects arise from the nonparabolic energy-momentum relations. We would thus expect to verify experimentally the importance of this nonlinear mechanism for InSb by measuring the output ratio of the mixed frequency and comparing it with our ρ^{NP} calculated here. On the other hand, for large-gap semiconductors such as Ge or Si, it is accepted[8] that the energy-dependent collision frequency is the more important nonlinear mechanism. This can also be verified experimentally by the measurements of the ratio of the frequency outputs without and with the magnetic field.

We wish to point out that the magnetic field enhancement can be increased by choosing ω_c closer to resonance with ω_3 for materials having their nonlinearity arising from the nonparabolic mechanism. However, here we are restricted from doing that by the limitation that the interaction length l must be smaller than the coherence length, which decreases sharply when $\omega_c \to \omega_3$.

In conclusion, we have shown in this paper that a dc magnetic field, besides enhancing the mixed frequency output, also offers a possible way to verify theoretical estimates on the importance of the two nonlinear mechanisms for optical mixing in semiconducting plasmas.

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