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OPTICAL MIXING BY MOBILE CARRIERS IN SEMICONDUCTORS*

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It is shown that certain nonlinearities associated with the energy dependence of the carrier-momentum relaxation time in semiconductors make contributions to the optically mixed output, which may be larger than the ones considered by earlier workers.

Recently Patel, Slusher, and Fleury[1] have shown in some experiments with InSb and InAs that conduction electrons can be responsible for the nonlinear optical mixing of two laser beams in semiconductors. The relatively large magnitude of the generated mixed output and its dependence on the mobile carrier density are some of the features of this type of generation which rule out[2] an explanation based on the usual optical nonlinearities arising because of the polarization of the background lattice.[3] Wolff and Pearson[2] have put forward a theory of optical mixing by mobile carriers in semiconductors, which is based on the nonparabolicity of the band structure. It is the aim of the present Letter to show that certain nonlinaerities associated with collisional processes may make even larger contributions to the generated mixed output. These nonlinear effects are associated with the energy dependence of the carrier-momentum relaxation time and will exist even for semiconductors with parabolic bands.[4]

We start with the equation of motion for an average carrier, which may be written as

$$\frac{\partial \vec{V}}{\partial t}+\frac{\vec{V}}{\tau}=-\frac{e\vec{E}}{m^*}, \tag{1}$$

where $\vec{E}=\vec{E}_1\exp(i\omega_1 t)+\vec{E}_2\exp(i\omega_2 t)$ is the total electric vector[5] due to externally impressed laser beams, e is the electronic charge, m^* the electron effective mass, and τ, the momentum relaxation time, is a function of the energy ϵ, of the form

$$\tau=a\epsilon^{n/2}, \tag{2}$$

where n is an integer. For dominant acoustic photon scattering $n=-1$, and for dominant ionized impurity scattering (which will be so[6] for InSb at the low temperatures considered by Patel, Slusher, and Fleury[1]) $n=3$. We have neglected the $(\vec{V}\cdot\nabla)\vec{V}$ and $\vec{V}\times\vec{B}$ terms in Eq. (1) because their contribution to the mixed output is several orders of magnitude smaller.[2] In the presence of the two strong electric vectors, the carrier temperature T_e is different from the lattice temperature T; one can write an energy-balance equation for the average carrier, viz.,

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

where $\vec{J}$ is the current density (the subscript r denotes its real part), N is the carrier concentration, k the Boltzmann constant, and τ_ϵ is the energy relaxation time for the carriers. The current density $\vec{J}$ is related to the directed carrier velocity $\vec{V}$ by the relation

$$\vec{J}=-Ne\vec{V}. \tag{4}$$

Now, Eq. (2) may be expressed as

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

where τ_0 is the momentum relaxation time corresponding to the lattice temperature. If $(T_e - T)/T \ll 1$, i.e., if the electrons are only "warm," then

$$\tau^{-1} \simeq \tau_0^{-1}[1 - \tfrac{1}{2}n(T_e/T - 1)]. \quad (5)$$

Equations (1) and (3)-(5) together give a complete solution of the problem. We solve this set of equations as follows. First, the two fundamental components of $\vec{V}$ are directly written from Eq. (1) as

$$\vec{V}_1 = -\frac{e\vec{E}_1\tau}{m^*(1 + i\omega_1\tau)}, \quad (6a)$$

$$\vec{V}_2 = -\frac{e\vec{E}_2\tau}{m^*(1 + i\omega_2\tau)}. \quad (6b)$$

Using (3), (4), and (6a, b) one obtains the following expressions for the $2\omega_1$ and $\omega_1 - \omega_2$ frequency components of T_e/T:

$$\frac{T_{e1}}{T} = -\frac{ie^2E_1^2}{6m^*kT\omega_1^3\tau}\exp(2i\omega_1 t) \quad (7a)$$

and

$$\frac{T_{e2}}{T} = -\frac{ie^2E_1E_2}{3m^*kT\omega_1^3\omega_2^2\tau}\left[\frac{\omega_1^2 + \omega_2^2}{\omega_1 - \omega_2}\right] \times \exp[i(\omega_1 - \omega_2)t], \quad (7b)$$

where E_1 and E_2 have been assumed to be in the same direction and ω_1, ω_2, and $\omega_1 - \omega_2 \gg \tau_\epsilon^{-1}$ (both these conditions are met with in the experiments of Ref. 1). Using (5), one can now obtain expressions for the time-dependent components of τ^{-1} which, when substituted back into Eq. (1), yield the desired mixed frequency components at $2\omega_1 \pm \omega_2$, etc., in the carrier velocity. We restrict ourselves to the component with frequency $\omega_3 = 2\omega_1 - \omega_2$; using Eq. (4), one obtains for the corresponding current density component

$$J_3 = \left(\frac{n}{3}\right)\frac{iNe^4E_1^2E_2}{8m^{*2}kT\omega_1\omega_2\omega_3}\left[\frac{2(\omega_1^2 + \omega_2^2)}{(\omega_1 - \omega_2)\omega_2} - 1\right] \times \frac{1}{(\omega_1\tau)^2}. \quad (8)$$

Denoting the corresponding expression obtained by Wolff and Pearson[2] as J_{3WP} and setting $n = 3$ (for dominant ionized impurity scattering in InSb at 80°K), one obtains

$$r = \frac{J_3}{J_{3WP}} = \frac{E_G}{kT}\left(\frac{1}{\omega_1\tau}\right)^2\left[\frac{2(\omega_1^2 + \omega_2^2)}{(\omega_1 - \omega_2)\omega_2} - 1\right] \times \left[\frac{(1 + 4E_F/E_G)^{5/2}}{1 + 8E_F/5E_G}\right]$$

which may be approximated as

$$r \simeq \frac{E_G}{kT}\left(\frac{1}{\omega_1\tau}\right)^2\frac{4\omega_1}{(\omega_1 - \omega_2)}\left[\frac{(1 + 4E_F/E_G)^{5/2}}{1 + 8E_F/5E_G}\right], \quad (9)$$

where we have made use of the fact that $\omega_1 \simeq \omega_2$ (but $\omega_1 - \omega_2 \gg \tau_\epsilon^{-1}$), E_F is the Fermi energy, and E_G is the band gap. For InSb at 80°K with an electron concentration of 10^{17} cm^{-3}, we use the following data obtained from Hilsum[7]:

$$\omega_1\tau \simeq 30, \quad E_F/E_G \simeq 0.22, \text{ and } E_G/kT = 33.$$

Further, using for ω_1 and ω_2 the frequencies corresponding to 9.6 and 10.6 μ, respectively, one obtains

$$r \simeq 5.$$

Thus, we find that under the conditions of experiment in Ref. 1, this nonlinear effect may be larger than that considered by Wolff and Pearson[2] by a factor of 5 or so. Unfortunately, this makes the agreement between the theory and experiment even worse.

Our nonlinear effect is critically dependent on the dependence of carrier-momentum relaxation time on carrier energy; thus for $n = 0$, Eq. (8) shows that J_3 vanishes.

We have not taken account of the distribution of carrier velocities in the above treatment; this should be included to get the correct numerical coefficient in Eq. (8). Results of such an analysis will be reported later.

Finally, we note that our nonlinear effect exists even for semiconductors with parabolic bands; this, therefore, offers a simple method for isolating the two effects.

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ELECTRON SPIN-ECHO MEASUREMENTS OF E_1' CENTERS IN CRYSTALLINE QUARTZ

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A standard spin-echo technique has been employed to determine the electronic phase-memory time of radiation-produced E_1' centers in crystalline quartz at 4.2, 77, and 300°K. The phase-memory time was found to be independent of temperature.

Radiation-produced E_1' centers in crystalline quartz have been the subject of considerable study,[1] and the spin-lattice relaxation time of this center has been reported to vary from the order of minutes at 4.2°K to a value of approximately 0.2 msec at 300°K.[2] These relatively long spin-lattice relaxation times make this defect a prime candidate for the observation of electron spin echoes at liquid-nitrogen or ambient temperatures. Spin echoes have been observed at these temperatures, and the spin-echo technique has been utilized to determine the phase-memory time of this material at 4.2, 77, and 300°K.

The experimental arrangement which is employed in these investigations involves a standard X-band superheterodyne spectrometer with i.f. detection at 45 MHz plus a 10-W traveling wave tube (TWT) which has been inserted after the signal klystron. With reference to Fig. 1, the signal klystron is left to run cw while two identical microwave pulses are produced by gating the grid of the TWT. The microwave pulse width and peak power are then varied to give the maxium echo signal on the oscilloscope. Related parameters (which vary slightly with temperature) are the following: peak pulse power incident on the cavity, 0.8 W; pulse width, 1 μsec; loaded-cavity $Q \sim 1000$; effective cavity volume, ~5 cm^3; operating frequency, ~9.5 GHz; gyromagnetic ratio, 1.76×10^7 G^{-1} sec^{-1} ($g=2$). These values correspond to the two 120° pulses which are required to produce the maximum echo signal.[3] Spin-echo signals at various temperatures are shown in Fig. 2.

By varying the separation between the microwave pulses a decay envelope for the echoes is obtained from which one can estimate the conventional $1/e$ phase-memory time T_2. Similarly, the value of T_1 was determined by varying the repetition rate of the echo-producing pulses until the amplitude of the second echo was $1-1/e$ of the initial echo. These values were found to be in agreement with those determined by Castle et al.[2] Values of T_1 and T_2 are listed in Table I along with the spin-lattice relaxation time and

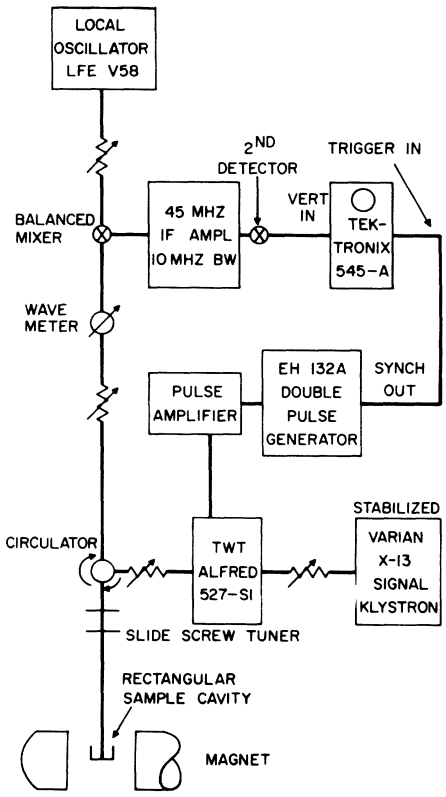

FIG. 1. Block diagram of spin-echo spectrometer.