THEORY OF OPTICAL MIXING BY MOBILE CARRIERS IN SEMICONDUCTORS

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In a recent series of experiments, Patel, Slusher, and Fleury[1] (PSF) have shown that conduction electrons in InAs and InSb give rise to a nonlinear interaction between the 10.6- and 9.6-μ beams from a Q-switched CO_2 laser. The main process they observe is one in which two 10.6-μ photons (each of frequency ω_1) combine to produce two photons, one at 9.6 μ (frequency ω_2) and a second at the difference frequency $\omega_3 = 2\omega_1 - \omega_2$ (wavelength 11.8 μ). A variety of measurements have indicated that, in moderately to heavily doped samples, the mixing process is caused by mobile carriers in these crystals, rather than the background lattice. The process is surprisingly strong for a nonlinear optical interaction.

In this Letter we wish to make two points concerning these experiments. We show first that nonlinearities of a classical plasma (one with a parabolic energy-momentum relation for the carriers) are far too weak to explain the observed mixing. Secondly, we propose that the observed nonlinearity is due to nonparabolicity[2] of the conduction band, an effect which is known to be relatively large in crystals such as InAs and InSb. The nonparabolicity effect is more than strong enough to explain the PSF experiments.

Many authors[3] have studied the nonlinear optical behavior of classical plasmas. They show that in such a medium, two intense optical beams mix to excite electron density oscillations at the sum or difference frequencies ($\omega_2 \pm \omega_1$ in our case). The effect is particularly large when $\omega_2 - \omega_1$ is near the plasma frequency ω_p. These driven density fluctuations can, in turn, Raman scatter a second photon of frequency ω_1, to a new frequency, $\omega_3 = 2\omega_1 - \omega_1$. The net effect is a process of the type observed by PSF. We will show, however, that it is far too weak to account for the observed mixing.

To estimate the strength of the nonlinearity described above, we use fluid (cold plasma) equations to describe the electrons. This approach is valid when all optical wavelengths are large compared with the Debye length. The fluid equations are[4]

$$\frac{\partial \vec{v}}{\partial t} + \frac{\vec{v}}{\tau} + (\vec{v}\cdot\nabla)\vec{v} - \frac{e}{m^*}(\vec{E} - \nabla\varphi) - \frac{e}{m^* c}(\vec{v}\times\vec{B}) = 0 \qquad (1)$$

and

$$\frac{\partial n}{\partial t} + \nabla\cdot(n\vec{v}) = 0. \qquad (2)$$

Here n is the electron density in the plasma and v is its velocity; $\vec{E} = \sum_\lambda \{\frac{1}{2}\vec{E}_\lambda \exp[i(\vec{q}_\lambda\cdot\vec{r} - \omega_\lambda t)]\}$ is the (transverse) electromagnetic field of the optical beams; φ is an electrostatic potential determined via Poisson's equation, $\nabla^2\varphi = 4\pi e(n - n_0)$, by the deviation of the electron density from its equilibrium value. An iterative solution of these equations shows that density perturbations are driven by $\vec{v}\times\vec{B}$ forces and are of second order in the electromagnetic fields. The equation of motion for this density perturbation is

$$\frac{\partial^2 n^{(2)}}{\partial t^2} + \left(\frac{1}{\tau}\right)\frac{\partial n^{(2)}}{\partial t} + \omega_p{}^2 n^{(2)} = \frac{e^2 n_0}{8(m^*)^2}\sum_{\lambda,\mu}\left\{\frac{(\vec{q}_\lambda + \vec{q}_\mu)^2}{\omega_\lambda \omega_\mu}(\vec{E}_\lambda\cdot\vec{E}_\mu) \times \exp\{i[(\vec{q}_\lambda + \vec{q}_\mu)\cdot\vec{r} - (\omega_\lambda + \omega_\mu)t]\}\right\}, \qquad (3)$$

where $\omega_p{}^2 = 4\pi n_0 e^2/\epsilon m^*$, and we have neglected $1/\tau$ compared with ω_λ and ω_μ.

The third-order current is given by

$$\vec{j}^{(3)} = e[n^{(0)}\vec{v}^{(3)} + n^{(1)}\vec{v}^{(2)} + n^{(2)}\vec{v}^{(1)}], \qquad (4)$$

where $\vec{v}^{(1)}$, $\vec{v}^{(2)}$, and $\vec{v}^{(3)}$ are the plasma velocity to first, second, and third order in the electromagnetic fields. However, $n^{(1)} = 0$ and it can be shown that $\vec{v}^{(3)}$ is purely longitudinal. The transverse part of $\vec{j}^{(3)}$ is thus included in

$$en^{(2)}\vec{v}^{(1)} = \frac{ie^4 n_0}{16(m^*)^3}\sum_{\lambda\mu\nu}\left\{\frac{(\vec{q}_\lambda + \vec{q}_\mu)^2(\vec{E}_\lambda\cdot\vec{E}_\mu)\vec{E}_\nu}{\omega_\lambda\omega_\mu\omega_\nu[\omega_p{}^2 - (\omega_\lambda + \omega_\mu)^2 - (1/\tau)(\omega_\lambda + \omega_\mu)]} \times \exp\{i[(\vec{q}_\lambda + \vec{q}_\mu + \vec{q}_\nu)\cdot\vec{r} - (\omega_\lambda + \omega_\mu + \omega_\nu)t]\}\right\}. \qquad (5)$$

Here, of course, we are particularly interested in terms which vary in frequency as $\omega_3 = 2\omega_1 - \omega_2$. These currents are the source of the 11.8-μ field. To estimate the intensity of the 11.8-μ radiation it is necessary to solve the driven wave equation. We will postpone this calculation until we have estimated the corresponding third-order currents arising from nonparabolicity.

We now consider frequency mixing due to nonparabolicity. Here the crucial point is that an electron's velocity is a nonlinear function of its momentum. The momentum of each electron oscillates at the frequencies of the applied electomagnetic fields if weak magnetic forces due to these fields are ignored. However, because of the nonlinear velocity-momentum relation, the induced current contains mixed frequency components. To estimate the order of magnitude of this effect, we consider the response of a single electron to the electromagnetic fields. Following Kane,[5] we describe its motion by the Hamiltonian[6]

$$H = \left[\left(\frac{E_G}{2}\right)^2 + \left(\frac{p^2}{2m^*}\right)E_G\right]^{1/2}, \tag{6}$$

where E_G is the band gap and m^* is the mass at the band edge. The velocity is

$$\vec{v}(\vec{p}) = \frac{\partial H}{\partial \vec{p}} = \frac{(\vec{p}/m^*)}{[1 + 2p^2/m^* E_G]^{1/2}}. \tag{7}$$

Finally, the equation of motion is $\dot{\vec{p}} = e\vec{E}$, with solution

$$\vec{p} = \vec{p}_0 + ie\sum_\lambda \{(\vec{E}_\lambda/2\omega_\lambda)\exp[i(\vec{q}_\lambda\cdot\vec{r} - \omega_\lambda t)]\}$$
$$= \vec{p}_0 + \Delta\vec{p}. \tag{8}$$

When substituted into Eq. (7), this momentum generates a third-order velocity; $\vec{v}^{(3)} = \frac{1}{6}[\Delta\vec{p}\cdot\vec{\nabla}_p]^3 \times \vec{v}(\vec{p}_0)$. To find the induced current, we average $\vec{v}^{(3)}(\vec{p}_0)$ over a Fermi-Dirac distribution function (this is equivalent to a solution of the Boltzmann equation) and multiply by the electron density and charge. The result is the expression

$$\vec{j}^{(3)}\big|_{\text{nonparabolicity}} = \left\{\frac{ie^4 n_0}{8(m^*)^2 E_G}\frac{(1 + 8E_F/5E_G)}{(1 + 4E_F/E_G)^{5/2}}\right.$$
$$\left.\times \sum_{\lambda\mu\nu}\left[\frac{(\vec{E}_\lambda\cdot\vec{E}_\mu)\vec{E}_\nu}{\omega_\lambda\omega_\mu\omega_\nu}\exp\{i[(\vec{q}_\lambda + \vec{q}_\mu + \vec{q}_\nu)\cdot\vec{r} - (\omega_\lambda + \omega_\mu + \omega_\nu)t]\}\right]\right\}, \tag{9}$$

where $E_F = p_F^2/2m^*$ is the Fermi energy calculated with the band-edge mass. This nonlinear current should be compared with Eq. (5). The two expressions are similar in form, but that given in Eq. (9) is much bigger numerically when the light beams are traveling in the same direction. Even when Eq. (5) is resonant ($\omega_\lambda + \omega_\mu \simeq \omega_p$), the ratio is large—about $mc^2/\epsilon E_G \omega_p \tau$. In InSb this factor, which appears squared in the final power estimates, is never less than 10^2. Off resonance the ratio is even bigger, about 10^3. In the PSF experiments there is, of course, some power reflected from the back surface of the sample which produces opposed beams inside the crystal. These drive the plasma nonlinearity more efficiently, but the phase-matching condition is badly violated in this case.

The effect of nonparabolicity is relatively large because it is an electric field effect, whereas the classical plasma nonlinearity is driven by the oscillating magnetic fields of the light waves.[3] It should also be emphasized that the nonlinearity due to band nonparabolicity in no way involves the collective behavior of the plasma. If Lorentz forces are ignored, the induced currents in the plasma are purely transverse in the parallel beam geometry, and frequency mixing occurs only because an individual electron's response is anharmonic.

To estimate the intensity of the 11.8-μ beam generated by the nonlinear mixing, one must solve the driven wave equation

$$\nabla^2\vec{E}^{(3)} - \frac{\epsilon}{c^2}\frac{\partial^2\vec{E}^{(3)}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial\vec{j}^{(3)}}{\partial t}. \tag{10}$$

PSF and Giordmaine[7] have noticed that the four-photon process under consideration here is essentially phase matched. Under these circumstances the amplitude of $\vec{E}^{(3)}$ grows linearly with interaction distance l if absorption is

neglected. One finds that

$$\frac{E_{\omega_3}}{E_{\omega_2}}=\frac{3\pi n_0 e^2 l}{2E_G q_3}\left(\frac{\omega_3}{\omega_2}\right)\frac{1+8E_F/5E_G}{(1+4E_F/E_G)^{5/2}}\left(\frac{eE_{\omega_1}}{m^*\omega_1 c}\right)^2. \quad (11)$$

Here E_{ω_1}, E_{ω_2}, and E_{ω_3} are the amplitudes of the three beams, which have been assumed collinear and of parallel polarization. To facilitate comparison with PSF, it is also convenient to rewrite this expression in terms of a nonlinear susceptibility and the beam powers. The formula which results is the same as the equation in their Table I, with $\chi^{(3)}$ defined as

$$\chi^{(3)}=\left[\frac{n_0 e^4}{4(m^*)^2 E_G \omega_1^2 \omega_2 \omega_3}\right]\left[\frac{1+8E_F/5E_G}{(1+4E_F/E_G)^{5/2}}\right]. \quad (12)$$

As shown in PSF, numerical values calculated from this formula are in reasonable agreement with experiment. The formula also gives the correct ratio of susceptibilities for InAs and InSb. This ratio is more accurately known experimentally than are the absolute values of $\chi^{(3)}$. Finally, Eq. (12) accounts for the measured variation of P_{ω_3} with electron density in InAs, for densities above 10^{16}/cc. This power is proportional to the square of $\chi^{(3)}$ and varies less rapidly than n_0^2 because of the second term in Eq. (12), which reduces the nonlinearity at higher doping levels. Crudely speaking, this reduction can be thought of as arising from an increase in carrier mass with doping.

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[3]Two recent references are G. G. Comisar, Phys. Rev. 141, 200 (1966); N. Bloembergen and Y. R. Shen, ibid. 141, 298 (1966). An extensive bibliography of earlier work is given in the paper by Bloembergen and Shen.

[4]L. Spitzer, Physics of Fully Ionized Gases (Interscience Publishers, Inc., New York, 1962).

[5]E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957).

[6]The use of a single-band Hamiltonian to describe the response of an electron to a perturbation is only correct when the perturbing frequencies are small compared with $E_G/\hbar$. This criterion is not particularly well satisfied in our case. Thus one should expect corrections of order $(\hbar\omega_1/E_G)^2 \lesssim \frac{1}{4}$ to succeeding formulas.

[7]J. A. Giordmaine, private communication.

ISOSPIN MIXING IN DEUTERON REACTIONS

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The reaction $C^{12}(d, \alpha)B^{10}$ has been employed recently[1] to study the isospin impurity of N^{14} as a function of excitation energy, since the deuteron, the alpha particle, and the C^{12} nucleus are in $T=0$ states. It has been suggested[2] that since the deuteron is distorted by the electric field of the target, some admixture of $T=1$ states will be produced and will contribute to the observed isospin impurity. A crude upper limit of 10% was obtained[2] for the similar reaction $Ca^{40}(d, \alpha)K^{38}$ by assuming that the neutron and proton are completely uncoupled in the nuclear Coulomb field.

An adiabatic approximation has been applied by the author[3] to evaluate the polarization potential acting on a deuteron at any distance from a fixed point charge. In this Letter we will show that this previous work[3] implies a value for the isospin impurity carried into the reaction by the distorted deuteron, and we will present numerical results.

Expanded in relative partial waves, the deuteron wave function has the form[3]

$$\psi(\vec{r},\vec{x})=\varphi(r)\left[1+\sum_{L=0}^{\infty} F_L(x,r)P_L(\cos\theta)\right], \quad (1)$$

where $\vec{x}$ is the center-of-mass coordinate of the deuteron measured from the target nucleus, the relative coordinate $\vec{r}=\vec{r}_n-\vec{r}_p$, the deuteron ground state is approximated by

$$\varphi(r)=(\gamma/2\pi)^{1/2}e^{-\gamma r}/r, \quad (2)$$