Theory of Resonant, Far-Infrared Generation in InSb

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The spin-flip Raman interaction causes a nonlinear spin-resonance process in semiconductor crystals. This mechanism is responsible for a large, resonant coefficient for difference-frequency generation in InSb.

One of the important goals of quantum optics is the development of a tunable, coherent, reasonably intense source of far-infrared radiation. Several experiments have been reported[1] in which visible or near infrared laser beams were mixed in nonlinear materials to generate far infrared (IR). To date, however, these processes have not been sufficiently efficient or controllable to be used as spectroscopic sources. The problem is tantalizing, because the recent development of the spin-flip Raman laser in InSb[2] has made available an excellent, tunable source of near-infrared radiation, whose difference frequency from the pump of the laser spans the far-IR region. The difficulty has come in finding a suitable nonlinear material to mix the two beams. The aim of this Letter is to point out that the spin system of an n-type InSb crystal may be an excellent medium for this purpose. In fact, we suspect that spin-flip lasers emit far-infrared radiation, provided they are operated in such a manner that the spin-resonance frequency is above the electron plasma frequency.

A number of workers have studied the coupling of light to electron spins in semiconductors. In narrow-gap materials—especially n-InSb—this interaction can be quite strong. The Raman gain for the spin-flip process in InSb is the largest known. Two factors combine to make it efficient —a large cross section and an exceedingly small linewidth. The theory of the spin-flip Raman process has been worked out in detail for the InSb case.[3] It yields an effective Raman interaction (after summing over virtual, interband processes) of the form

$$H_R \cong \frac{ie^2}{4m_s c^2}\frac{\hbar\omega_1 E_G}{E_G^2-(\hbar\omega_2)^2}\vec{\sigma}\cdot\vec{A}(\omega_1)\times\vec{A}(\omega_2)^* + \text{c.c.} \quad (1)$$

Here $E_G \simeq 0.25$ eV is the band gap; $m_s = 2m/|g| \simeq 0.04m$ is the spin mass; $\vec{A}(\omega_1)$ and $\vec{A}(\omega_2)$ are the vector potentials of the two pump beams,[4] with frequencies ω_1 and ω_2. The matrix elements of H_R give the spin-flip scattering rate. Several experiments[5] have confirmed that H_R has the form indicated in Eq. (1).

It is useful to compare Eq. (1) with the standard coupling of a spin to a magnetic field,

$$H_{\text{Zeeman}} = (e\hbar/2mc)\vec{\sigma}\cdot\vec{B}. \quad (2)$$

We see that the vector product $\vec{A}(\omega_1)\times\vec{A}(\omega_2)^*$ in Eq. (1) acts as an *effective magnetic field* which drives the spin system at the difference frequency $\omega_3 = \omega_1 - \omega_2$—thereby inducing a form of nonlinear spin resonance. This nonlinear process is the crucial mechanism in the method we propose for generating far IR. Thus the three-proton mixing process we are discussing is directly related with the Raman nonlinearity. If the spin-resonance frequency (ω_s) is tuned to the difference frequency (ω_3), the spin system is driven on resonance by the interaction of Eq. (1). Moreover, because the electron spin-resonance linewidths in InSb are small, even modest fields can drive the spin precession to appreciable amplitude. Subsequently, this precessing magnetic moment radiates energy at frequency ω_3. In effect, the spin system acts as a resonant, nonlinear medium for the photon-mixing process. The far-IR radiation is created by a magnetic dipole transition. Hence, the nonlinear medium need not be acentric for this process to be allowed. This point is an important one since, though InSb lacks inversion symmetry, the Hamiltonian which leads to Eq. (1) is centrosymmetric.

To estimate the strength of the process described above, we consider the geometry illustrated in Fig. 1. An n-type InSb crystal is pumped with two infrared beams whose difference frequency ω_3 equals the spin-resonance frequency ω_s. The polarizations are orthogonal, with one parallel to the dc magnetic field. The cross product $\vec{A}(\omega_1)\times\vec{A}(\omega_2)^*$ creates an "effective" magnetic field in the y direction which couples to the spins via Eq. (1). The subsequent motion is a standard problem in spin resonance, whose solu-

tion near resonance is

$$\langle\sigma_x(\omega_3)\rangle \cong \frac{e^2}{m_s c^2}\frac{\hbar\omega_1 E_G}{E_G^2-(\hbar\omega_2)^2}\frac{A(\omega_1)A(\omega_2)\exp[i(k_1-k_2)y]\langle\sigma_z\rangle}{2\hbar(\omega_3-\omega_s+i\gamma/2)}. \tag{3}$$

Here γ is the linewidth (full width at half-maximum) of the spin transition; k_1 and k_2 are the wave vectors (inside the crystal) of the two pump beams. In most cases the dc magnetic field will be strong enough to completely polarize the electrons, i.e., $\langle\sigma_z\rangle = 1$.

The spin magnetization can be written in the form

$$M_x(\omega_3) = C(\omega_3)E_{\omega_1}E_{\omega_2}\exp[i(k_1-k_2)y], \tag{4}$$

where

$$C(\omega_3) = n_0\mu^*\frac{e^2}{m_s c^2}\frac{\hbar\omega_1 E_G}{E_G^2-(\hbar\omega_1)^2}\frac{c^2}{\omega_1\omega_2}[2\hbar(\omega_3-\omega_s+i\gamma/2)]^{-1}. \tag{5}$$

n_0 is the electron density and μ^* is the magnetic moment of electrons in InSb. We have written Eq. (4) in terms of electric fields (rather than vector potentials) to facilitate comparison with more conventional optical nonlinearities. The nonlinear magnetization described by Eq. (4) is the source of the far-IR wave:

$$\partial^2 H_x(\omega_3)/\partial y^2 + \epsilon\omega_3^2 H_x(\omega_3)/c^2 = -4\pi\epsilon_3\omega_3^2 M_x(\omega_3)/c^2. \tag{6}$$

To solve, we write $H_x(\omega_3) = h(y)e^{ik_3 y}$, where $h(y)$ is a slowly varying envelope function. Equation (6) then takes the form

$$2ik_3\,dh/dy + i\,\mathrm{Im}(\epsilon)\omega_3^2 h/c^2 = -(4\pi\epsilon_3\omega_3^2/c^2)C(\omega_3)E_{\omega_1}E_{\omega_2}e^{i\Delta k y}, \tag{7}$$

where $\Delta k = k_1 - k_2 - k_3$ is the phase mismatch. With the boundary condition $h(0) = 0$, the solution to Eq. (7) is

$$h(y) = 4\pi[k_3/(2\Delta k - i\alpha)]C(\omega_3)E_{\omega_1}E_{\omega_2}(e^{i\Delta k y} - e^{-\alpha y/2}); \tag{8}$$

α is the absorption coefficient, at frequency ω_3, of the InSb crystal.

It is interesting to compare the IR powers expected from the nonlinear spin-resonance process described above with those produced by the conventional bound-electron nonlinearity. In the experiments of Nguyen and Bridges,[6] n-InSb crystals containing 2.2×10^{15} electrons/cm^3 are pumped with 9.6 and 10.6-μm CO_2 laser beams to generate far-IR radiation. A sharp resonance in the output, which we attribute to the spin process, occurs when $\omega_3 = \omega_s$. From its width one finds $\gamma \simeq 4\times10^{10}$ rad/sec $\simeq 0.2$ cm^{-1}. With these values we can now estimate the effective nonlinear coefficient for the spin-mixing process from Eq. (5). It is important in this calculation to note that Eq. (6) contains a dielectric constant on the right-hand side. This factor does not appear in the more conventional situation,[1] where the wave equation for the *electric* field is driven by a nonlinear *polarization*. As a consequence, the effective nonlinear susceptibility is $\sqrt{\epsilon_3}\,C(\omega_3)$. This value must be compared with the conventional nonlinear susceptibility which, in the geometry of the Nguyen-Bridges experiment, is $(2/\sqrt{3})d_{14}$. Thus, the calculated strength of the resonance is

$$Q \simeq 3\epsilon_3|C(\omega_3)|^2/4d_{14}^2 \simeq 200. \tag{9}$$

The measured value[6] is $Q = 88$. Agreement is reasonable, considering the many uncertainties in our estimate.

It is clear from Eq. (5) that the resonant, spin-induced nonlinearity can be enhanced if the pump frequencies are near the band gap. This enhance-

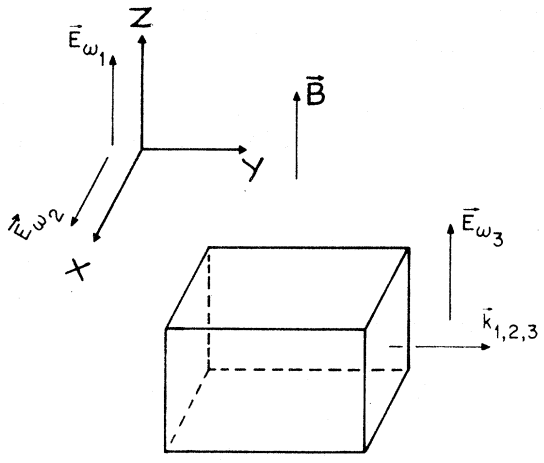

FIG. 1. Geometry used in calculation of mixing.

ment has been well documented in spin-flip laser work.[2] The experiments of Brueck and Mooradian[2,5] suggest that $C(\omega_3)$ might be increased by an order of magnitude with CO laser pumping. Smaller linewidths may also be attainable. These considerations lead us to expect, in an optimal situation, an effective nonlinear coefficient for the spin process more than 100 times larger than the conventional one. The spin-mixing process is strong, despite the fact that it involves a magnetic dipole transition, because it is *doubly* resonant under such circumstances. The difference frequency ω_3 is at the spin resonance frequency ω_s; the pump frequency ω_1 is near the band gap of the InSb crystal. Each of these resonances enhances the spin precession and the subsequent radiation of far-IR energy.

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Energy-Momentum Relation for Weakly Coupled Optical Polarons

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We have investigated the energy-momentum relationship for optical polarons by taking into consideration the phonon–cross-exchange diagrams. Our result is similar to that of the improved Tamm-Dancoff method, except that the whole curve shifts downward. Therefore, the ground-state energy is lower than the upper bound obtained by the variational method.

The motion of an electron in a polar crystal has always been an interesting problem in solid-state physics. Pekar[1] introduced the concept of the polaron, a quasiparticle composed of the combination a conduction electron and a longitudinal optical phonon of an ionic crystal. The first field-theoretic approach was given by Fröhlich, Pelzer, and Zienau.[2] They devised a model based on macroscopic dielectric theory, and established the following Hamiltonian:

$$H=\sum_{\vec{k}}(\hbar^2k^2/2m)C_{\vec{k}}^{\dagger}C_{\vec{k}}+h\Omega\sum_{\vec{q}}a_{\vec{q}}^{\dagger}a_{\vec{q}}+H'. \quad (1)$$

H' is the interaction Hamiltonian given by

$$H'=i\sum_{\vec{q},\vec{k}}V_{\vec{q}}(a_{\vec{q}}^{\dagger}-a_{-\vec{q}})C_{\vec{k}+\vec{q}}^{\dagger}C_{\vec{k}}, \quad (2)$$

where

$$V_{\vec{q}}=(4\pi\alpha/V)^{1/2}|\vec{q}|^{-1}. \quad (3)$$

The above equation is expressed in energy units of $\hbar\Omega$ and momentum units of $(\hbar/2m\Omega)^{1/2}$. Here $C_{\vec{k}}$ ($C_{\vec{k}}^{\dagger}$) is the electron destruction (creation) operator with momentum $\vec{k}$; Ω, the optical phonon frequency; $a_{\vec{q}}$ ($a_q^{\dagger}$), the destruction (creation) operator of a longitudinal-optical phonon of wave vector $\vec{q}$; α, the dimensionless coupling constant given by

$$\alpha=\frac{e^2}{2}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right)\left(\frac{2m}{\Omega\hbar^3}\right)^{1/2}; \quad (4)$$

e, the electron charge; m, the conduction-band mass; and ϵ_∞ (ϵ_0), the optical (static) dielectric constant.

There are many ways to investigate the dispersion relation of optical polarons,[3,4] one of which is the Green's-function technique. Recall that the one-particle electron Green's function G is