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THOMSON AND RAMAN SCATTERING BY MOBILE ELECTRONS IN CRYSTALS

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In this Letter we discuss the scattering of light by mobile electrons (or holes) in semiconducting crystals. Two processes will be considered: elastic (Thomson) scattering, and Raman scattering by electrons in a magnetic field. The elastic scattering has previously been treated[1,2] within the framework of the effective-mass theory (which is valid at low frequencies). We will show that, at higher frequencies, it is possible to achieve large enhancements of the cross section. This result may be of some importance since incoherent scattering experiments in solids are presently made difficult by the smallness of the cross section.

The Raman process we treat is one in which the electron changes its Landau-level quantum number by $\Delta n = 2$, and the outgoing radiation is shifted in frequency by twice the cyclotron frequency. For free, nonrelativistic electrons, it is easy to see that the matrix element for such a transition vanishes. However, if the electrons are relativistic (or the band structure nonparabolic) the cross section is finite and, in appropriate materials, can attain quite reasonable values.

Consider first the elastic scattering. Interest in this process has been aroused by recent papers[1,2] in which it is shown that a study of such scattering gives detailed information concerning the plasma properties of electrons in semiconductors. The multivalley semiconductor is of particular interest since it can support an acoustic mode (the so-called plasma sound wave) whose existence and dispersion relation one hopes to infer from the spectrum of the scattered light. In both the analyses referred to above, the electrons are described in the effective-mass approximation, and an individual electron is assumed to scatter with cross section

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{m_0 c^2}\right)^2 (\vec{\epsilon}_0 \cdot \alpha \cdot \vec{\epsilon}_1)^2, \tag{1}$$

where $\vec{\epsilon}_0$, $\vec{\epsilon}_1$ are polarization vectors of the incident and scattered light, and α the (dimensionless) reciprocal effective-mass tensor. The main aim of these calculations was to determine how the intensity and spectrum of scattered light is modified by Coulomb interactions between electrons. Here, on the other hand, we consider the problem of scattering by a single electron in the conduction band of an otherwise intrinsic semiconductor. This system is described by the Hamiltonian

$$H = \frac{[\vec{p} - (e/c)\vec{A}]^2}{2m_0} + V(r), \tag{2}$$

where m_0 is the free-electron mass, $\vec{A}$ the vector potential of the electromagnetic field, and $V(r)$ the crystal potential. It is well known[3] that, for completely free electrons ($V=0$), the major part of the matrix element for the scattering of light is produced by the A^2 term of Eq. (2). There is also a contribution, in second order, from the $\vec{p}\cdot\vec{A}$ term, but a near cancellation of the energy denominators in the two Feynman diagrams illustrated in Fig. 1 reduces its amplitude by a factor v/c (where v is electron velocity) compared to the A^2 term.

For the case of an electron in a crystal, the situation with respect to the intraband matrix elements of the $\vec{p}\cdot\vec{A}$ interaction is similar. However, no cancellation occurs in processes involving virtual interband transitions, and they can drastically change the scattering. A straightforward calculation shows that the total matrix element for elastic scattering by an electron of crystal momentum k is

$$M=\left(\frac{e^2}{m_0c^2}\right)\left(\frac{2\pi\hbar c^2}{\omega}\right)\left\{\vec{\epsilon}_0\cdot\vec{\epsilon}_1+\sum_n{}'\left[\frac{\langle\vec{k}0|\vec{p}\cdot\vec{\epsilon}_1|\vec{k}n\rangle\langle\vec{k}n|\vec{p}\cdot\vec{\epsilon}_0|\vec{k}0\rangle}{m_0[E_n(\vec{k})-E_0(\vec{k})-\hbar\omega_0]}+\frac{\langle\vec{k}0|\vec{p}\cdot\vec{\epsilon}_0|\vec{k}n\rangle\langle\vec{k}n|\vec{p}\cdot\vec{\epsilon}_1|\vec{k}0\rangle}{m_0[E_n(\vec{k})-E_0(\vec{k})+\hbar\omega_0]}\right]\right\}. \quad (3)$$

Here $\langle\vec{k}0|\vec{p}\cdot\vec{\epsilon}_1|\vec{k}n\rangle$ is the matrix element of the operator $\vec{p}\cdot\vec{\epsilon}_1$ between Bloch states of wave vector $\vec{k}$ in bands 0 and n; $E_0(\vec{k})$ and $E_n(\vec{k})$ are the corresponding energies; and the sum is over all bands except the conduction band. The first term within brackets in this formula arises from the A^2 term of the electron-photon interaction; the other two are second-order contributions of the $\vec{p}\cdot\vec{A}$ term, corresponding to the diagrams shown in Fig. 1. In the limit $\omega_0\to 0$, $\vec{k}=0$, the bracketed expression in Eq. (3) becomes $\vec{\epsilon}_0\cdot\alpha\cdot\vec{\epsilon}_1$ and the resulting cross section is that given by Eq. (1). At higher frequencies, on the other hand, the resonant denominators can produce important changes in the scattering. The form of this frequency variation will usually depend in detail on the band structure, but can be explicitly evaluated for a two-band model of the semiconductor (this is a fair approximation in a number of important cases[4]).

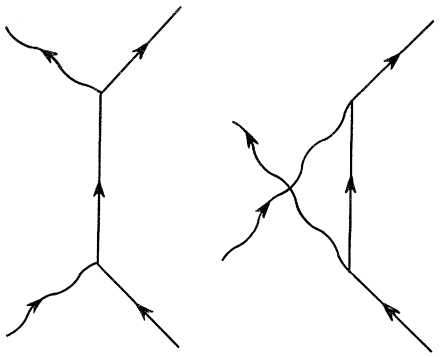

FIG. 1. Feynman diagrams for light scattering via the $\vec{p}\cdot\vec{A}$ interaction.

The expression for the cross section of an electron at $k=0$ is then

$$\frac{d\sigma}{d\Omega}=\left(\frac{e^2}{m_0c^2}\right)^2\left[\frac{E_G^{\,2}}{E_G^{\,2}-(\hbar\omega_0)^2}\right]^2 \times\left[(\vec{\epsilon}_0\cdot\alpha\cdot\vec{\epsilon}_1)-\left(\frac{\hbar\omega_0}{E_G}\right)^2(\vec{\epsilon}_0\cdot\vec{\epsilon}_1)\right]^2, \quad (4)$$

where E_G is the direct band gap. In materials with small effective masses this cross section is bigger than that at zero frequency by nearly a factor $\{E_G^{\,2}/[E_G^{\,2}-(\hbar\omega_0)^2]\}^2$.

To observe the cross-section enhancement implied by Eq. (4) it is clearly desirable to use light frequencies such that $\hbar\omega_0\sim E_G$. This condition necessitates a direct band-gap semiconductor. On the other hand, the calculations of Refs. 1 and 2 indicate that the most interesting collective effects are to be expected in multicomponent plasmas. The only common direct-gap, multivalley semiconductors are the lead salts,[5] PbS, PbSe, and PbTe. Of these, PbTe has the advantage of having a relatively large longitudinal-to-transverse effective-mass ratio ($m_l/m_t\sim 10$), and thus might be expected to show a particularly well-defined acoustic mode.[2] In PbSe the mass ratio is lower ($m_l/m_t\sim 2$), but the gap energy can be turned over a considerable range with pressure.[6] This fact might be of importance in adjusting E_G to the frequency of one of the high-powered infrared lasers.[7]

We now turn to the Raman scattering of an electron in a magnetic field. Here the A^2 term

in the electron-phonon interaction gives no contribution in the dipole approximation, and the whole matrix element must arise from $\vec{p}\cdot\vec{A}$ terms. The electric-dipole selection rules insure that in a two-photon process the Landau level quantum number of the electron changes either by $\Delta n = 0$ (elastic scattering) or $\Delta n = \pm 2$ (Stokes and anti-Stokes scattering). The same Feynman diagrams contribute to the Raman processes as do to the elastic scattering (see Fig. 1) and, once again, they cancel for free, nonrelativistic electrons. The cancellation is a direct consequence of the fact that the Landau levels are equally spaced. This spacing, in turn, reflects the fact that a nonrelativistic electron in a magnetic field behaves as a harmonic oscillator. One does not, of course, expect frequency mixing when light interacts with a perfectly harmonic system. However, if the electron's properties are slightly anharmonic (as will occur if the conduction band is nonparabolic) the cancellation is upset and the Raman process has a finite matrix element.

One material in which nonparabolicity effects are particularly large is InSb. To estimate the Raman cross section of an electron in the conduction band of this crystal, we carry out the effective-mass transformation to fourth order and obtain the Hamiltonian

$$H' = \left[\frac{\pi^2}{2m^*} - \vec{\mu}\cdot\vec{B}\right] + \frac{c}{E_G}(\vec{\pi}\cdot\vec{\mu}\times\vec{F}) - \frac{1}{E_G}\left[\frac{\pi^2}{2m^*} - \vec{\mu}\cdot\vec{B}\right]^2 + \frac{e^2F^2}{2m^*E_G^{\,2}}, \quad (5)$$

where $\vec{\pi} = [\vec{p} - (e/c)\vec{A}]$, $\vec{\mu}$ is the magnetic moment of the electron, and $\vec{F}$ the electric field. We have used the two-band model[4] in obtaining H'. This approximation is made for simplicity and is not a crucial one. H' would have the same form, though with slightly altered coefficients, in a more exact treatment. It should be emphasized, however, that this expansion for H' is a power series in both the quantities $(\pi^2/2m^*E_G)$ and $(\hbar\omega_0/E_G)$. Thus, Eq. (5) can only be used to calculate the Raman cross section for photon frequencies somewhat less than E_G. When $\hbar\omega_0 \sim E_G$ the band-decoupling scheme no longer converges, and it is necessary to use exact valence- and conduction-band wave functions to calculate the cross section.

The π^4 term in H' produces an unequal Landau level spacing and upsets the matrix-element cancellation which occurs for parabolic bands. In addition, it gives rise to new electron-photon coupling terms of the form p^2A^2, $p^2(\vec{p}\cdot\vec{A})$, and $(\vec{p}\cdot\vec{A})^2$. Of these, only the last need be considered in lowest order. The A^2 term gives no Raman effect in the dipole approximation, and the contribution of the second is reduced by the energy denominator cancellation. The $(\vec{p}\cdot\vec{A})^2$ term produces a Raman effect in first order and is not subject to the cancellation. The total Raman matrix element turns out to be

$$\left(\frac{e^2}{m^*c^2}\right)\frac{2\pi\hbar c^2}{(\omega_0\omega_1)^{1/2}}\left(\frac{\epsilon_{0x}-i\epsilon_{0y}}{\sqrt{2}}\right)\left(\frac{\epsilon_{1x}-i\epsilon_{1y}}{\sqrt{2}}\right)\left(\frac{2}{E_G}\right) \times\left[\frac{\omega_0(\omega_0-2\omega_c)}{(\omega_0-\omega_c)^2}\right]\frac{\langle n+2\,|\,\pi^+\,|\,n+1\rangle\langle n+1\,|\,\pi^+\,|\,n\rangle}{m^*} \quad (6)$$

for a transition in which an electron is excited from the nth to $(n+2)$nd Landau levels. In this formula ω_c is the cyclotron frequency, $\pi^+ = (\pi_x + i\pi_y)/\sqrt{2}$, and the dc magnetic field has been taken in the z direction. We have neglected the contributions of terms involving μ to this matrix element since the spin mass in InSb is about three times larger than the cyclotron mass. Assuming that the electron starts in its lowest Landau level ($n = 0$), one computes a Raman cross section

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{m^*c^2}\right)^2 2\left(\frac{\hbar\omega_c}{E_G}\right)^2\left(\frac{\omega_1}{\omega_0}\right)\left[\frac{\omega_0(\omega_0-2\omega_c)}{(\omega_0-\omega_c)^2}\right]^2 \times(\epsilon_{0x}^{\,2}+\epsilon_{0y}^{\,2})(\epsilon_{1x}^{\,2}+\epsilon_{1y}^{\,2}), \quad (7)$$

where $\omega_1 = \omega_0 - 2\omega_c$ is the frequency of the scattered radiation. Once again, it should be stressed that this formula is only correct when $(\hbar\omega_0/E_G)$ is smaller than unity. To investigate the behavior of the Raman cross section as $\hbar\omega_0 \to E_G$, we have also carried out a calculation for the simple two-band (Dirac) model which describes the band structure of Bi,[8] and possibly some semiconductors. The matrix elements and selection rules for this model are known[9] and the computation is straightforward. The resulting cross section is essentially identical to that of Eq. (7), except enhanced by the factor $\{E_G^{\,2}/[E_G^{\,2}-(\hbar\omega_0)^2]\}^2$ which appears in Eq. (4).[10] From these results we conclude that the Raman scattering of electrons in semiconductors will, under fairly general circum-

stances, have a cross section of the order of that given by Eq. (7) (with possible enhancement for $\hbar\omega_0 \sim E_G$).

Finally, we make estimates of the scattered powers that might be attained in an experiment to detect the Raman effect. For this purpose, InSb seems an ideal crystal (since sizable values of $\hbar\omega_C/E_G$ can be achieved with reasonable magnetic fields), and a recently developed CO_2 laser[7] (100 W cw at 10 μ) a good source. For example, in InSb a field of 30 kG gives a cyclotron energy $\hbar\omega_C \simeq 0.02$ eV, and the Raman cross section [from Eq. (7)] is about 4×10^{-24} cm^2/sr. The Raman-scattered power from a 1-cc sample containing 10^{16} electrons is then about 10^{-6} W/sr at a wavelength of ~15 μ. This power should be detectable in the presence of the elastically scattered background at 10 μ. There is even a possibility, with focused beams and a somewhat more heavily doped sample, of achieving stimulated Raman emission. If such a situation could be reached it would provide an intense, magnetically tunable source of infrared radiation.

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MAGNETOACOUSTIC EFFECTS WITH FINITE OMEGA-TAU*

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The magnetoacoustic effect is now widely employed as a tool for studying the geometry of Fermi surfaces.[1] Since the technique gives extremal dimensions of orbits, it can be used to resolve questions concerning details of band structure in metals. The effect is observed when the electron mean free path l is greater than the sound wavelength λ, or $ql > 1$, where $q = 2\pi/\lambda = \omega/v_S$, ω and v_S being the sound frequency and velocity.

For high-purity metals now available $l \sim 0.1$ cm and this condition is easily satisfied. In fact, for 100-Mc/sec sound, $ql \geq 300$ in the cadmium employed in some of our experiments. Under such conditions many oscillations appear in a plot of attenuation versus magnetic field, and one can even study beats between oscillations produced by different extremal orbits.

We show here that $ql \geq 300$ causes an effect related to cyclotron resonance which may easily be confused with beats between two series of geometric oscillations having nearly equal periods. The effect arises from the fact that $v_S/v_F \sim 1/300$ in typical metals, where v_F is the electron Fermi velocity, so that $ql \geq 300$ implies $\omega\tau \gtrsim 1$, where $v_F\tau = l$. Cyclotron resonance will occur when $\omega = n\omega_C$, where ω_C is the cyclotron frequency of the electrons.[2] In addition, when $\omega_C = \omega$ the sound field will move a distance $\lambda/2$ by the time the electrons go half-way around the orbit so that a relative minimum in attenuation will occur for an orbit diameter ordinarily corresponding to a maximum for geometric resonance, and vice versa.

Figure 1 shows the normalized compressional-wave attenuation $S_{11} = \alpha/(Nm/\rho v_S\tau)$, where α