Theory of Interband Raman Scattering in Semiconductors in Magnetic Fields

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A theory is developed for interband Raman scattering in a semiconductor in an external magnetic field. The process involves an interband electronic transition and an interband transition between Landau levels. Specific formulas are presented for the two-band model of a semiconductor, and numerical calculations are presented for PbTe. The scattering efficiency shows density-of-states peaks and a strong resonance when the scattered frequency equals the cyclotron frequency. The latter scattering should be experimentally observable.

The development of lasers had led to an upsurge in the study of Raman scattering from solids including, in particular, electronic Raman scattering. The electronic Raman effect involves a frequency shift which is related to an electronic excitation or deexcitation in the solid. Electronic Raman scattering by impurities has been discussed theoretically by Elliott and Loudon[1] and observed experimentally in semiconductors by Henry, Hopfield, and Luther[2] and by Wright and Moordian.[3] Scattering associated with transitions of electrons between Landau levels in semiconductors in magnetic fields has been discussed theoretically by Wolff[4] and by Yafet,[5] while experimental observations have been reported by Slusher *et al.*[6]

The foregoing examples of electronic Raman scattering involve *intraband* electronic transitions between initial and final states. In the present paper, we develop a theory of an electronic Raman process which involves *interband* transitions. For a crystal with a center of symmetry such as PbTe, interband Raman scattering between the uppermost valence band and the lowest conduction band is forbidden at the band extrema by parity considerations. However, the process can be made allowed by the application of an external magnetic field, so that an interband transition to an intermediate state is followed by an intraband transition between adjacent Landau levels to the final state, see Fig. 1. The situation is analogous to that obtaining in interband two-photon absorption in an external magnetic field.[7] We find that for the process in Fig. 1, the Raman scattering efficiency exhibits a striking resonance when the frequency of the scattered radiation equals the cyclotron frequency. The scattering efficiency associated with the resonance is estimated to be very large.

We base our development on the formulation of Yafet.[5] The differential scattering cross section per unit frequency interval can be written in the form

$$\left(\frac{d^2\sigma}{d\Omega\, d\omega}\right)_{fO} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} |A_{fO}|^2 \hbar\delta(\hbar\omega - E_f + E_O)\,, \tag{1}$$

where the second-order matrix element A_{fO} is given by

$$A_{fO} = \frac{1}{m}\sum_r \left(\frac{(f|\vec{\epsilon}_1\cdot[\vec{p}+(e/c)\vec{A}_e]|r)(r|\vec{\epsilon}_0\cdot[\vec{p}+(e/c)\vec{A}_e]|O)}{E_O - E_r + \hbar\omega_0} + \frac{(f|\vec{\epsilon}_0\cdot[\vec{p}+(e/c)\vec{A}_e]|r)(r|\vec{\epsilon}_1\cdot[\vec{p}+(e/c)\vec{A}_e]|O)}{E_O - E_r - \hbar\omega_1} \right). \tag{2}$$

Here, O, r, and f refer to the initial, intermediate, and final states of the electron; E_O, E_r, and E_f are the corresponding electronic energies; $\vec{\epsilon}_0$, ω_0 and $\vec{\epsilon}_1$, ω_1 are, respectively, the polarization vector and frequency of the incident and scattered radiation; $\omega = \omega_0 - \omega_1$; $\vec{A}_e$ is the vector potential of the external magnetic field; and m is the free-electron mass. Equations (1) and (2) are based on the electric dipole approximation. Terms quadratic in the vector potential of the radiation field do not contribute in this approximation.[5] The δ function in Eq. (1) expresses the energy-conservation condition

$$\hbar\omega_0 = \hbar\omega_1 + E_f - E_O\,. \tag{3}$$

We have calculated the scattering cross section for a two-band model of an intrinsic semiconductor having parabolic nondegenerate (except for spin) energy bands. Explicit numerical results are presented for the case of PbTe with the external magnetic field in the (001) direction.

The matrix elements required in Eq. (2) are evaluated using effective-mass wave functions carried to second order.[8] For band j and Landau level n, the wave function can be written as

$$\psi_{jn}(\vec{r}) = (2\pi)^{3/2}\{F_{jn}(\vec{r})u_j(\vec{r})$$

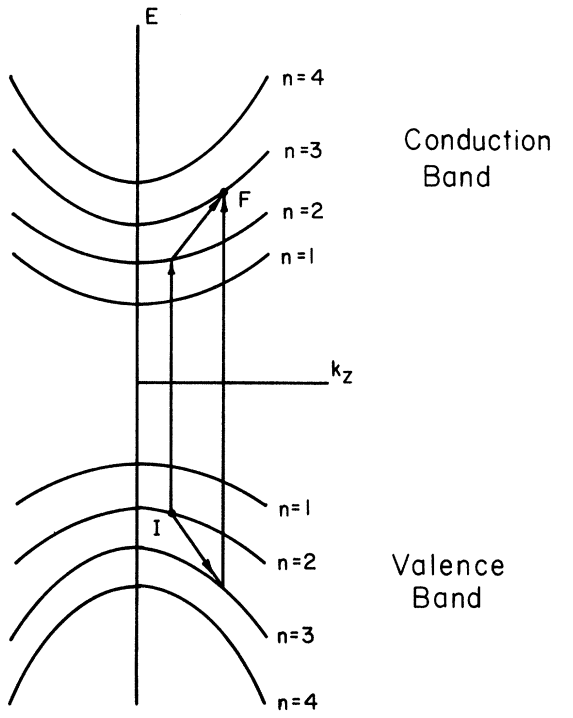

FIG. 1. Diagram of the transitions for the interband Raman scattering in a magnetic field between the $(v, 2)$ and $(c, 3)$ states showing the two possible intermediate states. All transitions occur at the same value of k_z, but are shown displaced for clarity.

$$-\sum_i \left[\frac{p_{ij}^{\alpha}}{m\hbar\omega_{ij}} \left(p_\alpha + \frac{e}{c} A_{e\alpha} \right) F_{jn}(\vec{r}) \right] u_i(\vec{r}) \Big\} , \qquad (4)$$

where $u_i(\vec{r})$ and $u_j(\vec{r})$ are the periodic parts of the Bloch functions for bands i and j at their extrema; $\hbar\omega_{ij}$ is the corresponding energy difference; $F_{jn}(\vec{r})$ is the envelope function for band j and Landau quantum number n; the Cartesian coordinate index α is summed over x, y, z; and p_{ij}^{α} is the matrix element of the αth component of momentum with respect to $u_i(\vec{r})$ and $u_j(\vec{r})$. The matrix elements of $\vec{p} + (e/c)\vec{A}_e$ with respect to the $\psi_{jn}(\vec{r})$ can be classified as either intraband or interband. Using standard procedures of effective-mass theory[9] one finds for the intraband case

$$\left(\psi_{an} \left| \vec{p} + \frac{e}{c} \vec{A}_e \right| \psi_{bn'} \right) = \left(\frac{m}{m_a^*} \right) \left(F_{an} \left| \vec{p} + \frac{e}{c} \vec{A}_e \right| F_{bn'} \right) \delta_{ab} , \qquad (5)$$

where m_a^* is the *algebraic* effective mass for band a. For the interband case, one finds

$$(\psi_{an} | \vec{p} + (e/c)\vec{A}_e | \psi_{bn'}) = (u_a | \vec{p} | u_b) \delta_{nn'} , \qquad (6)$$

where terms of order $(eH/mc\omega_{ab})$ have been neglected and H is the external magnetic field.

The matrix elements involving the envelope functions F on the right-hand side of Eq. (5) can be evaluated[8] using a convenient gauge for a magnetic field in the (001) direction. We shall ignore the z component in Eq. (5) so that the Landau quantum number n changes by ± 1. The result for the second-order matrix element A_{fO} as applied to the two-band model can be written in the form

$$A_{fO} = -(\tfrac{1}{2}s)^{1/2} \begin{pmatrix} (n+1)^{1/2} \\ n^{1/2} \end{pmatrix} \times \left[(\vec{\epsilon}_0 \cdot \vec{P}) \epsilon_1^{\mp} \left(\frac{1}{m_c^*(\omega_1 \pm \omega_c)} + \frac{1}{m_v^*(\omega_1 \mp \omega_v)} \right) - (\vec{\epsilon}_1 \cdot \vec{P}) \epsilon_0^{\mp} \left(\frac{1}{m_c^*(\omega_0 \mp \omega_c)} + \frac{1}{m_v^*(\omega_0 \pm \omega_v)} \right) \right] , \qquad (7)$$

where $s = eH/\hbar c$, m_c^* and m_v^* are the algebraic effective masses of the conduction and valence bands, ω_c and ω_v are the cyclotron frequencies given by $eH/m_c^* c$ and $eH/|m_v^*|c$, respectively, $\vec{P}$ is $(u_c|\vec{p}|u_v)$, $\epsilon_0^{\mp} = \epsilon_{0x} \mp i\epsilon_{0y}$, and $\epsilon_1^{\mp} = \epsilon_{1x} \mp i\epsilon_{1y}$. In obtaining Eq. (7) we have made use of the energy-conservation condition, Eq. (3). The second-order transitions represented by Eq. (7) correspond to the excitation of an electron from the nth Landau level of the valence band to either the $(n+1)$th or $(n-1)$th Landau level of the conduction band. The upper symbols in Eq. (7) correspond to the former case and the lower symbols to the latter case. In each case there are two intermediate states involved corresponding to transitions to final states by conduction electrons and by holes, respectively. The situation for the transition from the nth valence-band level to the $(n+1)$th conduction-band level is shown diagrammatically in Fig. 1.

The differential scattering cross section can now be calculated with the aid of Eq. (7). Rather than present the general result, we specialize to the case $\vec{\epsilon}_0 = (1, 0, 0)$ and $\vec{\epsilon}_1 = (1, 0, 0)$. We further assume that the two bands considered interact strongly, so that $|m_v^*| = m_c^* = m^*$ and hence, $\omega_c = \omega_v$. Dividing the differential scattering cross section by the volume, we obtain $S(\omega)$, the scattering efficiency per unit length per unit frequency per unit solid angle in the form

$$S(\omega) = \frac{2}{\pi^2} \left(\frac{e^2}{mc^2} \right)^2 \frac{\omega_1}{\omega_0} \left(\frac{P}{\hbar} \right)^2 \left(\frac{m^*}{\hbar} \right)^{1/2} \left(\frac{\omega_c^2}{(\omega_1^2 - \omega_c^2)} + \frac{\omega_c^2}{\omega_0^2 - \omega_c^2} \right) \sum_{n=1}^{n_{\max}} [n(\omega_0 - \omega_1 - \omega_G - 2n\omega_c)^{-1/2}] , \qquad (8)$$

where $\hbar\omega_G$ is the energy gap between valence and conduction bands. The interband momentum matrix element P can be eliminated by using the following relation for the two-band model:

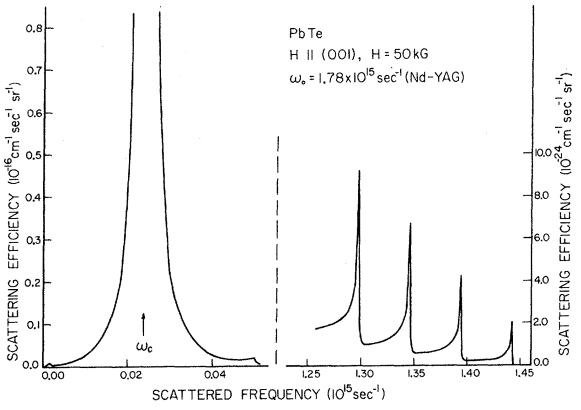

FIG. 2. Spectrum of scattered radiation for the interband Raman scattering in PbTe in a magnetic field of 50 kG. Note the change in scale of the ordinate for the left-hand and right-hand sides of the figure. The peak value of the scattering efficiency at the resonance at ω_c is 735×10^{-16} cm^{-1} sec^{-1} sr^{-1}.

$$2P^2/m\hbar\omega_G = m/m^* \ . \tag{9}$$

The scattering efficiency can now be calculated from Eqs. (8) and (9) and available information about m^* and $\hbar\omega_G$.

A few general remarks may be made about the scattering predicted by Eq. (8). As expected, the scattering efficiency vanishes in zero external magnetic field. The dependence on magnetic field is essentially quadratic, so high fields will be favorable. Since the process depends on the initial transition being an interband transition, no free carriers are required, and the effect should be observable in intrinsic materials. The scattering should be particularly marked near the singularities of Eq. (8), namely, the density-of-states singularities and the resonance singularities. Concerning the latter, only the resonance involving the scattered radiation is important, because the incident radiation frequency must be greater than the band gap and $\omega_c < \omega_G$.

We have made a specific calculation for the case of PbTe with the external magnetic field parallel to the (001) direction. For this situation the two-band model is reasonably appropriate. The singularities in the scattering efficiency have been smoothed out by introducing a phenomenological relaxation time corresponding to a mobility[10] of 500 000 cm^2/V sec. The precise value of this relaxation time is not important. The energy band gap[11] was taken to be 0.190 eV, appropriate to 4.2 °K. The effective mass[12] was taken to be $0.037m$ appropriate to conduction electrons with magnetic field parallel to the (001) direction. The calculated frequency spectrum for the scattered radiation is given in Fig. 2 for an external field of 50 kG and an incident radiation frequency of 1.06 μ (Nd-YAG laser). For scattered frequencies near the incident frequency, the density-of-states peaks are evident. The magnitude of the scattering is relatively small in this region, the integrated scattering efficiency over one of the peaks being of the order of 10^{-11} cm^{-1} sr^{-1}. For a skin depth of 10^{-5} cm the effective integrated scattering efficiency becomes on the order of 10^{-16} sr^{-1}, which is quite small. Furthermore, self-absorption will cut down the scattered light until the scattered photon energy drops below the band gap.

A different situation obtains in the region where $\omega_1 \approx \omega_c$. Here, the frequency shift is very large, and the scattering efficiency is very large as can be seen from the left-hand side of Fig. 2. For intrinsic material without free carriers the scattered radiation will not be self-absorbed. The scattering efficiency integrated over the resonance is on the order of 0.01 cm^{-1} sr^{-1}. To appreciate the very large cross section associated with the resonant scattering, it is useful to compare this scattering efficiency with that for other electronic Raman processes. For phosphorous-doped silicon, with 1.6×10^{16} impurities/cm^3, Wright and Mooradian[3] have estimated that the Raman efficiency/unit length/unit solid angle is the order of 6×10^{-9}. For InP with 10^{16} electrons/cm^3, Yafet[5] estimates the same quantity to be 2×10^{-7} for intraband Raman scattering between Landau levels. Thus, the scattering resonance associated with the interband process considered in this work gives a scattering efficiency/unit length/unit solid angle several orders of magnitude larger than the two processes just discussed. For a skin depth of 10^{-5} cm, we get an effective integrated scattering efficiency of 1×10^{-7} sr^{-1}. This resonance scattering at the cyclotron frequency should be experimentally observable and might provide a tunable infrared source. In planning an experiment it is desirable to use an incident laser frequency fairly close to the intrinsic absorption edge so that the skin depth is maximized and competing absorption from higher interband transitions is eliminated.

For a more general situation than the two-band model considered here, there will be resonances at the cyclotron frequencies of both electrons and holes. In a material such as InSb, the complications of the valence band will introduce additional structure into both the density-of-states peaks and the resonance peaks.

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