LIGHT SCATTERING FROM SINGLE-PARTICLE ELECTRON EXCITATIONS IN SEMICONDUCTORS

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Light scattering from screened single-particle electron excitations has been observed in GaAs, InP, and CdTe. The intensity, polarization properties, and temperature dependence of the single-particle scattering in GaAs are compared with effective-mass calculations based on nonparabolic as well as parabolic bands. The experimental results are in disagreement with present theory.

This Letter reports the observation of scattered light from screened single-particle electron excitations in GaAs, InP, and CdTe. The Raman scattering from plasmons and coupled plasmons and phonons[1] has previously been reported as well as the polarization properties[2] of the scattered light from these excitations. Both these cases dealt with the collective excitations of the electron gas in a solid. The present results yield the single-particle excitation spectrum of the electron gas. The single-particle scattering is compared with effective-mass calculations based on simple parabolic bands,[3] which involves scattering from electron density fluctuations, as well as a more recent effective-mass calculation which predicts a large enhancement due to nonparabolicity.[4] The experimental results, however, are in disagreement with both of these theories.

The scattered light was generated using a 1.06-μ neodymium-doped yttrium aluminum garnet laser which could operate with a continuous output power in excess of 10 W. The GaAs samples were single-crystal rectangular parallelepipeds with $\{100\}$ faces and were either mounted on a cold finger in contact with liquid helium or directly immersed in liquid helium pumped below its lambda point. The polarized laser beam was incident along a $\langle 100\rangle$ axis of the crystal while the scattered light was collected at 90 deg along another $\langle 100\rangle$ axis. The elastically scattered laser light was sufficiently low in intensity to permit measurements within 4 cm^{-1} of the laser line at room temperature. The InP and CdTe samples were unoriented single crystals. Spectra were recorded with incident laser powers as small as 10 mW to make certain that sample heating did not affect the spectra. The intermediate temperature of 30°K shown in Fig. 1 was estimated from the magnitude of the anti-Stokes spectrum. The other experimental methods have been described previously.[1,2]

Figure 1 shows the Raman spectrum of an n-type GaAs sample at 300, 30, and 5°K. We will concentrate on GaAs here; however, similar spectra were also observed in the other materials. The peaks labeled L_-, L_+, and TO have been described elsewhere[2] and refer to the coupled modes of the longitudinal optical phonons and the plasmons (L_-, L_+), and the transverse optical phonon (TO). The scattering from the single-particle excitation spectrum of the conduction electrons is seen at lower frequencies near the laser line. At $T = 0$°K, the single-particle excitation spectrum comes from electrons with momentum $\vec{p}$ that are excited from occupied states below the Fermi surface to unoccupied states just above the Fermi surface with momentum $\vec{p}+\vec{q}$, where $\vec{q}$ is the momentum change of the scattered electron. At $T = 0$°K, and for infinite lifetime, the existing theories[3,4] predict a continuum spectrum increasing linearly with ω to a peak near $0.7qv_F$ for the parameter values involved here and cutting off at qv_F, where v_F is the Fermi velocity. Samples with concentrations ranging from 1.5×10^{17} to 2.8×10^{18} cm^{-3} were measured. At low temperature, the single-particle spectrum, while smeared by a finite col-

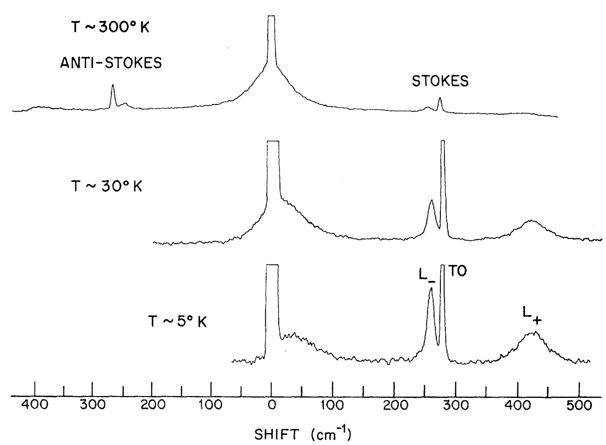

FIG. 1. The Raman spectrum of GaAs ($n = 1.4\times10^{18}$ cm^{-3}) at 300, 30, and 5°K. The scattering angle was 90° with the incident and scattered light propagating along (100) crystal axes.

lision time, did show a cutoff consistent with $\omega = qv_F$. At higher temperatures more electrons can participate in the scattering as the electron distribution becomes Maxwellian. In the high-temperature limit, the spectral shape of the scattered light should resemble[4] a Maxwellian velocity distribution, which was found to be consistent with the experimental results.

The integrated scattering cross section for electrons in a simple parabolic conduction band,[3] i.e., for $(\perp,\perp)$ scattering from electron density fluctuations, is given by

$$\frac{d\sigma(\perp,\perp)}{d\Omega} \approx 0.25 n r_0^2 \left[\frac{E_g^2}{E_g^2-(\hbar\omega_i)^2}\right]^2 \times \left(\frac{\hbar q v_F}{E_F}\right)\left(\frac{q}{q_{FT}}\right)^4 \qquad (1)$$

for a degenerate distribution at $T = 0°K$, unity scattering volume, and an infinite relaxation time. Here n is the electron concentration, $r_0^2 = (e^2/m^*c^2)^2$ is the Thomson electron cross section, m^* the electron effective mass, q_{FT} the Fermi-Thomas momentum, E_F the Fermi energy, E_g the energy gap, and ω_i the frequency of the incident photon. The $(\perp,\perp)$ signs refer to the polarization of the incident and scattered light relative to the plane formed by the wave vectors of the incident and scattered light, respectively. The theory predicts zero scattering cross sections for the other polarizations. The resonant enhancement factor

$$\left[\frac{E_g^2}{E_g^2-(\hbar\omega_i)^2}\right]^2 \qquad (2)$$

in Eq. (1) has been predicted by Wolff[5] for a two-band parabolic model.

The cross sections of the mixed longitudinal modes appear to be satisfactorily understood.[2] The $(\perp,\perp)$ polarization components, which may be used as a relative comparison, are given by

$$\frac{d\sigma}{d\Omega d\omega} = r_0^2\left[\frac{E_g^2}{E_g^2-(\hbar\omega_i)^2}\right]^2 \frac{\hbar q^2 \epsilon_\infty \omega_p^4}{4\pi e^2\omega^4}\,\mathrm{Im}\frac{1}{\epsilon(\omega)}, \qquad (3)$$

at $T = 0°K$, where $\omega_p = (4\pi n e^2/m^*\epsilon_\infty)^{1/2}$ is the plasma frequency, ϵ_∞ the optical dielectric constant, and $\epsilon(\omega)$ is the total longitudinal dielectric constant given by

$$\epsilon(\omega) = \epsilon_\infty\left[1-\frac{\omega_p^2}{\omega(\omega-i/\tau)}\right] + \frac{(\epsilon_0-\epsilon_\infty)\omega_t^2}{\omega_t^2-\omega^2}, \qquad (4)$$

Table I. Integrated single-particle scattering cross sections for GaAs, $n = 1.4\times10^{-3}$, 90° scattering angle. Both experimental and theoretical cross sections are normalized to the integrated intensity in the $(\perp,\perp)$ configuration of the L_+ mode.

	Polarization	Nonparabolic theory $(\times 10^{-2})$	Experiment
5°K	$\perp,\perp$	0.70×10^{-2}	0.22 ± 0.1
	$\perp,\parallel$	0.70×10^{-2}	0.55 ± 0.1
	$\parallel,\perp$		
	$\parallel,\parallel$	0.60×10^{-2}	0.48 ± 0.1
300°K	$\perp,\perp$	3.0×10^{-2}	3.8 ± 0.5
	$\perp,\parallel$	2.0×10^{-2}	4.7 ± 0.5
	$\parallel,\perp$		
	$\parallel,\parallel$	1.9×10^{-2}	3.5 ± 0.5

where ϵ_0 is the static dielectric constant, ω_t is the transverse optical phonon frequency, and τ is a "phenomenological" electron collision time. The ratio of the single-particle to plasmon cross section calculated from (1) and (3) for GaAs with $n = 1.4\times10^{18}$ cm^{-3} is 200 times smaller at 5°K than the experimentally determined ratio given in Table I.

The polarization properties of the single-particle scattering are shown in Fig. 2. In contrast to the theory for the parabolic-band scattering, which predicts the scattered light to be polarized only in the same direction as the incident light, i.e., $(\perp,\perp)$, all possible polarization combinations of the incident and scattered light were

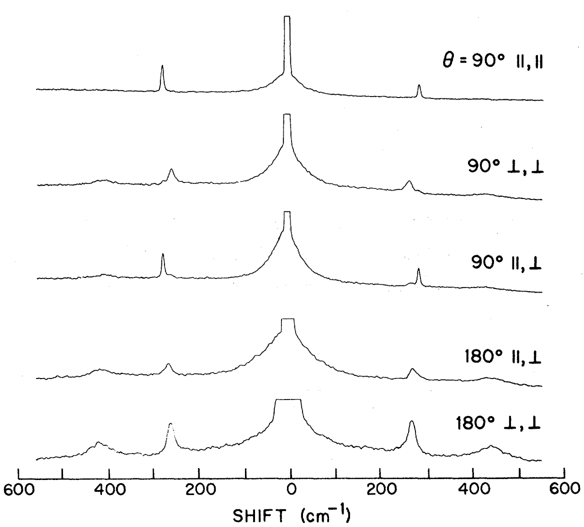

FIG. 2. Polarized Raman spectrum of GaAs ($n = 1.4\times10^{18}$ cm^{-3}) at 300°K. The $\parallel$ and $\perp$ signs refer to the polarization of the incident and scattered light in the scattering plane.

observed. Contributions to the single-particle cross section from the electro-optic effect would give rise to $(\parallel, \perp)$ and $(\perp, \parallel)$ polarization components but not to a $(\parallel, \parallel)$ component. However, this effect is too small to account even for the observed $(\parallel, \perp)$ and $(\perp, \parallel)$ scattering.

The scattering at an angle of 180° was also measured (Fig. 2) to determine the angular dependence. The single-particle cross section varies as q^4 in the parabolic model, which would mean a factor of 4 increase in amplitude at fixed ω/qv_F in going from a 90° to 180° scattering angle. The screened LO phonon at 252 cm^{-1}, whose intensity should show[6] a factor of 2 increase in going from 90° to 180°, was used to calibrate the relative intensities for the $(\parallel, \perp)$ polarization configuration. No significant increase in the amplitude at fixed ω/qv_F was found for the single-particle scattering at 180° relative to that at 90°.

Wolff[4] has recently calculated within the effective-mass approximation the contribution to the single-particle electron-scattering cross section arising from a nonparabolic conduction band and finds a significant departure from what is predicted by the theory involving a simple parabolic conduction band.

For $\omega_i \ll E_g$, the scattering cross section is given by[4]

$$\frac{d^2\sigma}{d\omega d\Omega} = \left(\frac{\omega_s}{\omega_i}\right) \int_{-\infty}^{\infty} e^{i\omega t} \langle H^\dagger(t) H(0)\rangle \frac{dt}{2\pi}, \tag{5}$$

where ω_s is the frequency of the scattered radiation, and

$$H = \left(\frac{e}{c}\right)^2 \sum_i \vec{e}_0 \cdot \frac{\partial^2 E}{\partial \vec{p}_i \partial \vec{p}_i} \cdot \vec{e}_i \exp(i\vec{q}\cdot\vec{r}_i). \tag{6}$$

Here $\vec{e}_0$ and $\vec{e}_i$ are the polarization vectors of the incident and scattered light, and E is the energy of the electron, which Wolff took to be of the form

$$E(p) = \frac{p^2}{2m^*} - \frac{1}{E_g}\left(\frac{p^2}{2m^*}\right)^2. \tag{7}$$

The integrated cross sections of the single-particle scattering for the $(\perp, \perp)$ polarization configuration for a degenerate distribution at $T = 0°K$ and infinite relaxation time is given by[4]

$$d\sigma(\perp, \perp)/d\Omega \approx 0.36 n r_0^2 (E_F/E_g^2) \hbar q v_F. \tag{8}$$

The nonparabolic theory, while predicting the existence and lack of angular variation of all the observed polarized components, gives between one and two orders of magnitude smaller cross section than that observed.[7] In this comparison, the resonant enhancement factor was assumed to be the same as for the plasmons. The relative intensities among the various polarizations also fall outside the experimental error as indicated in Table I.

Conclusions.—The scattered light from single-particle excitations of electrons in solids has been observed for the first time. The strength of this scattering relative to the plasmon line is found to be between one and two orders of magnitude larger than that predicted by present theory.

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