Three statistically independent measurements of ξ were made. The spectrum of Fig. 2 gives $2\xi = -11.5(8.6)\%$; the spectrum of the enriched absorber in Fig. 1 gives $2\xi = -8.0(2.3)\%$; and another enriched absorber taken over a higher velocity range gives $2\xi = -13.3(3.9)\%$. The weighted mean is $2\xi = -9.5(1.9)\%$. Fits were made with ξ constrained to equal zero. Besides providing a poorer fit, these constrained fits produced a small and otherwise unexplainable shift of the resonance away from zero velocity to $+0.92(14)$ μm/sec. With the quoted dispersion term, this zero shift exactly disappears. Using our measured shift in Eq. (2) and assuming $\epsilon = 1$ give $f_2 = 1.4(3)\%$ for the fraction of the photoelectric effect that is $E2$ in character at 13.3 keV in Ge.

The value of ξ has recently been explicitly calculated for the 13.3-keV ^{73}Ge transition by Goldwire and Hannon.[16] They find $2\xi = -6.8\times 10^{-2}$, but this does not include the contribution to ξ from the L- and M-shell electrons, which they expect will increase their value by about 15–20%. Thus the large interference dispersion term that we find for this $E2$ transition in ^{73}Ge is in good agreement with theory.

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[1]H. de Waard and G. J. Perlow, Phys. Rev. Lett. 24, 566 (1970).
[2]C. Sauer, E. Matthias, and R. L. Mössbauer, Phys. Rev. Lett. 21, 961 (1968).
[3]R. S. Raghavan and Loren Pfeiffer, Phys. Rev. Lett. 32, 512 (1974).
[4]G. Kaindl, D. Solomon, and G. Wortmann, Phys. Rev. B 8, 1912 (1973).
[5]Loren Pfeiffer, Nucl. Instrum. Methods 140, 57 (1977).
[6]Loren Pfeiffer, R. S. Raghavan, C. P. Lichtenwalner, and A. G. Cullis, Phys. Rev. B 12, 4793 (1975).
[7]*Mössbauer Effect Data Index, 1974*, edited by J. G. Stevens and V. E. Stevens (Plenum, New York, 1975).
[8]S. M. Sze and J. C. Irvin, Solid State Electron 11, 599 (1968).
[9]K. B. Wolfstirn, J. Phys. Chem. Solids 16, 279 (1960).
[10]R. R. Hewill and B. F. Williams, Phys. Rev. 129, 1188 (1963).
[11]T. C. Wang, Phys. Rev. 99, 566 (1955).
[12]G. T. Trammell and J. P. Hannon, Phys. Rev. 180, 337 (1969).
[13]J. Hannon, private communication. This is a generalization of an expression appearing in Ref. 11.
[14]G. Kaindl and D. Solomon, Phys. Lett. 32B, 364 (1970).
[15]F. E. Wagner, B. D. Dunlap, G. M. Kalvius, H. Schaller, R. Felscher, and H. Spieler, Phys. Rev. Lett. 28, 530 (1972).
[16]H. C. Goldwire, Jr., and J. P. Hannon, to be published.

Resonant Brillouin Scattering of Excitonic Polaritons in Gallium Arsenide

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We report the first experimental observation of resonant Brillouin scattering of excitonic polaritons in a semiconductor. The usual symmetric Brillouin doublet becomes an asymmetric multiplet when the incident light energy is scanned through the $n = 1$ exciton resonance and allows a direct measurement of the polariton dispersion curve. In GaAs we determine a longitudinal-transverse splitting of 0.08 ± 0.02 meV and a translational mass $M_{ex} = (0.6 \pm 0.1)m_0$ for the [100] heavy exciton. The excitions couple preferentially to LA phonons.

It has been proposed that Brillouin scattering in semiconductors should undergo, like Raman scattering, a resonant enhancement when the incident light energy $\hbar\omega_0$ approaches that of electronic excitations in the crystal. The resonance behavior at the fundamental gap energy is now well established.[1] Resonant Brillouin scattering (RBS) of *excitonic polariton* states, however, has not yet been observed experimentally. A recently developped theory of RBS[2] predicts a multiplet of Brillouin lines near the exciton resonance with line separations and intensities depending strongly on

$\hbar\omega_0$ because of polariton dispersion. This Letter reports the first experimental observation of RBS via $n=1$ exciton-polariton states in a direct-gap semiconductor.

A continuously tunable cw dye laser in the near infrared[3] was used for excitation. With an intracavity quartz etalon, a linewidth of less than 0.1 Å with 1 mW total power incident on the sample was achieved. The experiments were performed on high-purity vapor-phase and liquid-phase epitaxial samples of GaAs with total shallow-impurity concentrations $N_D+N_A\simeq 2\times10^{14}-5\times10^{15}$ cm^{-3} and 77-K mobilities $\mu_n\simeq 1-2\times10^5$ cm^2/Vsec ($\mu_p\simeq 7-9\times10^3$ cm^2/Vsec for the n- (p-) type layers. The samples were held in a variable-temperature flowing helium gas cryostat. Excitation power densities were changed in the range 10–300 mW/cm^2 by focusing. We checked sample heating and found it to be negligible. The scattered light from the as-grown sample surfaces with [100] orientation was collected in backward direction by a $f/3$ lens and focused into a 1-m double-grating spectrometer. A photon counting system with a cooled GaAs-cathode photomultiplier and logarithmic count rate output was used. From the measured quantum efficiency of the whole optical setup including photomultiplier, spectrometer, etc., the backward-scattered light efficiency η of the sample (defined as the number of outgoing photons per sterad per incoming photon) was determined quantitatively.[4]

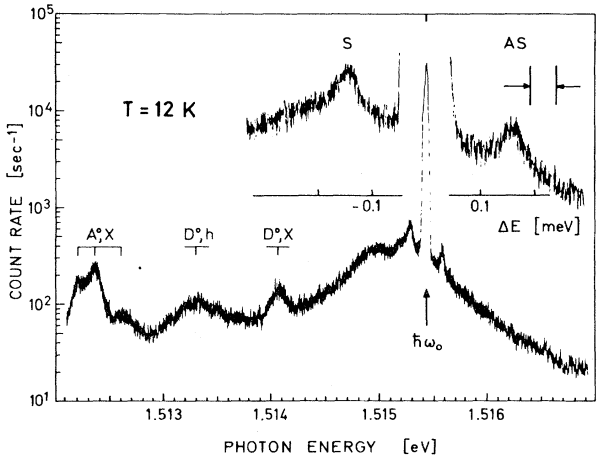

FIG. 1. Resonant Brillouin scattering (RBS) spectrum of GaAs ([100], backscattering) for given excitation light energy $\hbar\omega_0$ slightly above E_L. The light intensity is given on a logarithmic scale. The inset shows the Stokes (S) and anti-Stokes (AS) RBS lines on an expanded scale. The broad band around 1.515 eV is due to $n=1$ exciton-polariton luminescence. The other luminescence peaks are labeled according to the initial state of the corresponding electronic transitions.

A typical RBS spectrum at $T=12$ K is presented in Fig. 1, together with an inset displaying the sharp Brillouin peaks on an expanded scale. The apparent width of laser and Brillouin lines is due to instrumental resolution. When $\hbar\omega_0$ is scanned through the exciton resonance, Stokes and anti-Stokes components (S, AS) strikingly reflect the dispersion of the $n=1$ excitonic polariton levels through their varying and asymmetric Brillouin shifts. Close to resonance (i.e., $\hbar\omega_0\simeq E_L$, the longitudinal exciton energy at $K=0$) we observed a multiplet of lines with as many as six resolved components which are due to the different possible transitions between both polariton branches, as predicted by Brenig, Zeyher, and Birman.[2] Figure 2(a) shows a theoretical polariton dispersion curve[5] along the direction of the wave vector of the incident light, and illustrates the various one-phonon backscattering Stokes processes. Our measured energy shifts ΔE for all distinct S and AS lines are given in Fig. 2(b) as a function of $\hbar\omega_0-E_T$ (E_T is the transverse exciton energy). The observed variation of the Stokes scattered light efficiency η is shown in Fig. 2(c).

The results of Fig. 2(c) demonstrate that the intraband processes $1\rightarrow1'$ and $2\rightarrow2'$ dominate over interband scattering except in a small ($\simeq$0.3-meV wide) range above E_L, where we attribute the additional peaks to $1\rightarrow2'$ (S) and $2\rightarrow1'$ (AS) scattering. Some further smaller structures in the spectra could not be identified unambiguously. For $\hbar\omega_0$ well below (above) E_L, but still within an exciton Rydberg from E_L, we observe the common symmetric Brillouin doublet approaching asymptotically an energy shift $\Delta E=0.17\pm0.01$ meV [$0.16(5)\pm0.01$ meV]. Assuming the background dielectric constant to be $\epsilon_0=12.55$,[6] we obtain a sound velocity $u_s=c\Delta E(2\hbar\omega_0\sqrt{\epsilon_0})^{-1}=(4.8\pm0.2)\times10^5$ cm/sec. This value for u_s agrees perfectly with the longitudinal sound velocity in the [100] direction, as derived from the elastic constants for GaAs given in the literature.[7] The transverse sound velocity for propagation in [100] direction and arbitrary polarization is much smaller, $u_s^t=3.36\times10^5$ cm/sec.[7] We therefore conclude that RBS of polaritons in GaAs occurs predominantly through LA phonon coupling. In the following analysis we assume that in the energy range and geometric configuration studied here the polaritons couple exclusively to dispersionless LA phonons of sound velocity[7] 4.81×10^5 cm/sec.

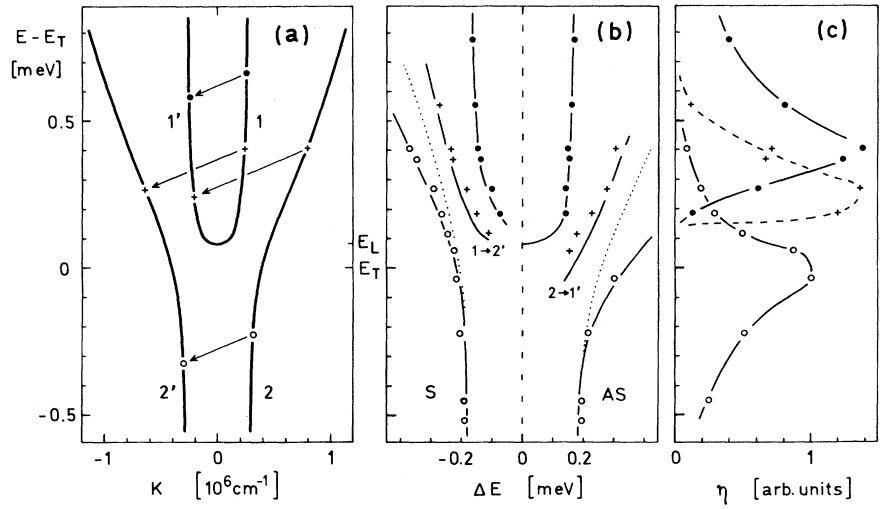

FIG. 2. (a) Polariton dispersion curve in the two-band model of Ref. 5 along the direction of the incident light. The various possible *intraband* ($2\rightarrow 2'$, $1\rightarrow 1'$) and *interband* ($1\rightarrow 2'$, $2\rightarrow 1'$) backscattering processes between the polariton branches involving emission of one acoustical phonon of energy $\Delta E = \hbar u_s (K - K')$ are indicated. (b) Measured and calculated Brillouin shifts ΔE as a function of incident light energy $\hbar\omega_0 - E_T$ (Stokes shifts, left; anti-Stokes shifts, right). Polaritons are coupled to dispersionless LA phonons. Parameters for the calculated curves (full lines) $E_T = 1.5150$ eV; $M_{ex} = 0.6 m_0$; $E_{LT} = 0.08$ meV; $u_s = 4.81 \times 10^5$ cm/sec. For comparison, a calculated curve for $M_{ex} = 0.3 m_0$ is also shown (dotted line). (c) Measured resonance behavior of the scattering efficiency η for Stokes intra- and inter-band scattering as a function of $\hbar\omega_0 - E_T$. Lines connect the experimental points for clarity.

The two basic parameters which describe the polariton dispersion curve in a two-band model with nondegenerate electron and hole bands[5] —longitudinal-transverse splitting E_{LT} and exciton translational mass M_{ex}—can be directly determined by fitting theoretical Stokes and anti-Stokes shifts [full lines in Fig. 2(b)] to the experimental data points. Both parameters influence the theoretical curves in a characteristic manner: M_{ex} determines the asymptotic behavior of the $2 \rightarrow 2'$ branch in the region of spatial dispersion ($\hbar\omega_0 \gtrsim E_L$), whereas E_{LT} gives the separation in energy of the intraband branches of Fig. 2(b). Because of the valence-band degeneracy in zinc-blende-type crystals one has to expect, in general, *two* different exciton dispersion curves, i.e., light- and heavy-exciton bands.[8] Our experimental RBS data, however, could be fitted consistently with only one exciton translational mass $M_{ex} = (0.6 \pm 0.1) m_0$, which is close to the heavy-exciton mass of $0.75 m_0$ calculated by Kane[8] for small kinetic energies.[9] A reason for this preferential heavy-exciton contribution to RBS could be the higher density of final states available in the heavy-exciton band. Figure 2(a) represents the calculated polariton dispersion curve in the two-band model,[5] taking the parameters $E_T = 1.5150$ eV, $E_{LT} = 0.08$ meV, $M_{ex} = 0.6 m_0$, $\epsilon_0 = 12.55$, which gave the best fit to the data of Fig. 2(b). To illustrate the sensitivity of this fit with respect to spatial dispersion, we also included a theoretical curve for $M_{ex} = 0.3 m_0$ in Fig. 2(b) (dotted curve).

The resonant behavior of the measured RBS efficiency η for the Stokes processes on $\hbar\omega_0 - E_T$ is given in Fig. 2(c). Brenig, Zeyher, and Birman[2] have shown theoretically that the detailed shape of these resonance curves is intimately related to the so-called additional boundary conditions (ABC's). The ABC's determine the amplitude ratio of both types of polaritons when they can propagate simultaneously in the crystal, i.e., for $\hbar\omega_0 \gtrsim E_L$ and finite M_{ex}. At present, the limited dynamical range of our measurements and the problem of relating the measured η (which is affected by volume effects like reabsorption and multiple scattering[4] to calculated cross sections do not allow a definite statement on the validity of the different proposals for the choice of ABC's that have been made in literature. The rather abrupt change from $2\rightarrow 2'$ to $1\rightarrow 1'$ intraband scattering near E_L [see Fig. 2(c)] strongly favors, in our opinion, the choice of Agranovich and Ginzburg[10] (ABC 2 in the notation of Ref. 2).

In conclusion, the outcome of the first RBS experiment on excitonic polaritons in a semiconductor demonstrates that in GaAs (i) $K \simeq 0$ excitons

couple predominantly to LA phonons, (ii) the polariton dispersion curve can be measured with high accuracy (despite the remarkable smallness of E_{LT}), allowing for a precise determination of E_{LT} and the heavy-exciton mass M_{ex}, (iii) the long-standing controversy on ABC's can be decided by careful analysis of RBS which is a more sensitive probe than other optical techniques, e.g., reflectivity measurements.[11]

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[1]A. S. Pine, in *Light Scattering in Solids*, edited by M. Cardona (Springer, Berlin, 1975), Chap. 6, and references therein.

[2]W. Brenig, R. Zeyher, and J. L. Birman, Phys. Rev. B 6, 4617 (1972).

[3]A. Donzel and C. Weisbuch, Opt. Commun. 17, 153 (1976).

[4]In the region of excitation light energies $\hbar\omega_0$ discussed here, the dye-laser beam is absorbed almost totally ($\geq 90\%$) within the epitaxial layer of thickness 10 μm, i.e., we are dealing with a "semi-infinite" crystal.

[5]J. J. Hopfield and D. G. Thomas, Phys. Rev. 132, 563 (1963),

[6]G. E. Stillman, D. M. Larsen, C. M. Wolfe, and R. C. Brandt, Solid State Commun. 9, 2245 (1971).

[7]J. R. Drabble, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1966), Vol. 2, Chap. 5.

[8]E. O. Kane, Phys. Rev. B 11, 3850 (1975); M. Altarelli and N. O. Lipari, unpublished.

[9]The exchange interaction could modify the results of Ref. 8 by mixing heavy and light excitons. This has not yet been investigated theoretically.

[10]V. M. Agranovich and V. L. Ginzburg, *Spatial Dispersion in Crystal Optics and the Theory of Excitons* (Interscience, London, 1966), Chap. 3.

[11]F. Patella, F. Evangelisti, and M. Capizzi, Solid State Commun. 20, 23 (1976).

COMMENTS

Comment on the Comparison of Observed Two-Electron–One-Photon Transition Energies with Calculated Values*

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We describe a method for comparing observed two-electron–one-photon transition energies with theoretical values in order to determine the L-shell vacancy configuration at the time of x-ray emission. This method reduces the dependence on the particular computer code used in the calculation. Present calcuations indicate degree of L-shell ionization in the Fe and Ni data of Wölfli *et al.* than that found by previous investigators.

Interest in two-electron–one-photon transitions ($K\alpha\alpha^h$) has been generated by the recent work of Wölfli *et al.*[1] in which such transitions were observed in Fe and Ni following heavy-ion–atom collisions. In particular, several efforts have been made to compare the observed energies of these transitions with theoretical predictions.[2-7] The observed transition energies have been shown to be consistent with predictions based on Hartree-Fock calculations of atomic energy levels if the transitions are electric dipole ($E1$) transitions[3-7] in which a $2s$ electron and a $2p$ electron fill the two $1s$ vacancies. Complicating the comparison with theory, however, is the fact that considerable multiple L-shell ionization may exist in the emitting atom, thereby shifting the $K\alpha\alpha^h$ x-ray peak to higher energies. Since the average state of L-shell ionization at x-ray emission is