riers, but also the effects of dielectric polarization, electrode injection, and ionic contributions.

The identity of these photogenerated carriers cannot be established with these results. We argue, however, that the result of drift-mobility measurements[11] showing the transition probability for electron hopping to be orders-of-magnitude larger than that for holes strongly indicates that electron motion is responsible for the high-frequency dispersion. A detailed quantitative comparison with the drift-mobility results is not warranted, however, because the carriers in drift-mobility measurements traverse the whole sample, while the frequencies employed here cause the carriers to sense a more localized environment. Localized potential fluctuations[12,13] within the solid would result in the set of localized states sensed by the drifting carriers. In fact, the weaker temperature dependence of the photocarriers (estimated from the lower-frequency results to be about 0.3 eV when expressed in terms of an activation energy) relative to the 0.7-eV activation for the drift mobility is consistent with this interpretation.

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Resonant Raman Scattering in a Semiconductor Superlattice*

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We report the observation of enhancement in the Raman cross section for photon energies near electronic resonance in GaAs-$Ga_{1-x}Al_xAs$ superlattices of a variety of configurations. Both the energy positions and the general shape of the resonant curves agree with those derived theoretically based on the two-dimensionality of the quantum states in such superlattices. Polarization studies indicate a major contribution to the scattering from forbidden processes.

In this Letter, we report the results of resonant Raman scattering (RRS) on GaAs-$Ga_{1-x}Al_xAs$ superlattices (periodic structures consisting of ultrathin semiconductor layers). The structures were prepared by molecular-beam epitaxy,[1] which has been shown to produce extremely sharp boundaries and atomically smooth interfaces.[2] The one-dimensional periodic potential produces quantum states of two-dimensional (2D) character, and the choice of the layer thicknesses and the Al concentration determine their energy levels and bandwidths.[3,4] Thus, the superlattice apparently has a most intriguing electronic structure suited for RRS investigations.

Figure 1 shows schematically the periodic potential of a superlattice in the x direction with the quantum states E_{nc} and E_{nv}. The electron and hole envelope wave functions $\Psi_e(x)$ and $\Psi_h(x)$ are largely confined in the well region. From the Kronig-Penney model, we have calculated the energy levels and bandwidths, using the electron and hole effective masses, 0.1 and 0.56, respectively. The joint density of states, $\rho(E)$, is obtained with the application of the selection rule, allowing optical dipole transitions only between E_{nv} and E_{nc}. Pertinent parameters (see Fig. 1) of four superlattices, a, b, c, and d, used in the experiments are listed in Table I: the thicknesses, d_1 and d_2, for GaAs and $Ga_{1-x}Al_xAs$ layers, respectively; the Al concentration, x; the number of periods, p; the potential discontinuities, E_c and E_v, for the conduction and valence bands,

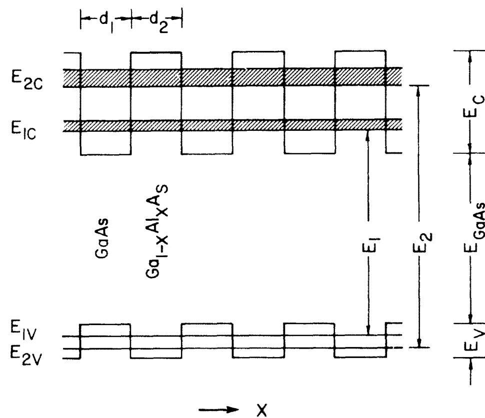

FIG. 1. A schematic energy diagram of a GaAs-$Ga_{1-x}Al_xAs$ superlattice indicating the definition of the various parameters.

respectively; the first and second transition thresholds, E_1 and E_2; and the sums of electron and hole bandwidths for E_1 and E_2, Δ_1 and Δ_2, respectively. The energy dependence of the joint density of states for each specimen is shown by dashed lines in Fig. 2. The widening of Δ_1, as it progresses from the specimen *a*, to *b*, *c*, and *d*, gives a measure of the departure from 2D behavior.

The RRS measurements were performed at room temperature with a $LiNbO_3$ parametric oscillator pumped by a pulsed dye laser. The tuning range was 7600–8500 Å, with an average available power of a few milliwatts. The peak power was kept below 50 $W/(mm)^2$ to avoid heating effect. Backscattering geometry was used with the polarization of the scattered photon parallel to that of the incident one, unless specified otherwise. The superlattice specimens were (100)-oriented, a choice dictated by the growth process.

Experimental results of Raman intensity vs incident photon energy are shown in Fig. 2. The

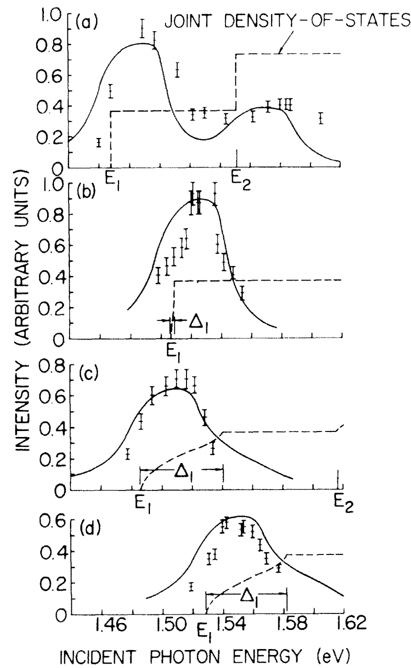

FIG. 2. Raman intensity vs incident-photon energy for four superlattice configurations as described in Table I. Solid curves are the calculated intensities. The energy dependence of joint density of states is shown in dashed lines.

phonon involved is the LO phonon of GaAs at 36.5 meV, the only one observed in the present experiments. The absence of the TO phonons is consistent with the simple selection rule given by Loudon.[5] The phonons associated with Ga_{1-x}-Al_xAs are not evident either, because electrons and holes are primarily confined in GaAs regions. The scatter of data points, typically ±10%, results from statistical noise due to the low Raman count and the large background of photoluminescence. Within this spread, however, the data are quite reproducible: The experimental data in Fig. 2 contain points taken in different runs.

TABLE I. Parameters for the four superlattice configurations used. The first four parameters are from growth conditions, while the last four from calculations with the Kronig-Penney model. Δ_1 is the combined bandwidth for the lowest quantum level, others having been defined in Fig. 1.

Specimen	GaAs d_1 (Å)	$Ga_{1-x}Al_xAs$ d_2 (Å)	Al x	Number of periods p	E_c (eV)	E_v (eV)	E_1 (eV)	E_2 (eV)	Δ_1 (meV)	Δ_2 (meV)
a	100	100	0.25	100	0.22	0.04	1.468	1.551	0	0
b	52	66	0.25	130	0.22	0.04	1.507	1.66	2	32
c	39	33	0.11	200	0.103	0.018	1.486	1.615	53	⋯
d	35	28	0.25	200	0.22	0.04	1.523	1.742	54	⋯

This reproducibility makes it possible to compare intensities for various cases, as shown in the figure, where the same scale is used for the ordinates. The specimens in Fig. 2 were undoped with Sn up to a few times 10^{17} cm^{-3} have shown essentially the same results. We might add also that no signal was observed for homogeneous GaAs in the range of its energy gap.

In a 2D electronic system, the resonant enhancement of Raman scattering, both allowed and forbidden, can be formulated in a similar manner as Martin's treatment in the three-dimensional (3D) case.[6] Considering only scattering between quantum states of the same index according to the selection rule mentioned earlier, we have carried out the 2D summations in the third-order perturbation expressions through the exact Green's function formalism. Following the definition of the scattering amplitude $F(E_i, \vec{q})$ of Ref. 6, we obtain

$$F_{e,h}(E_i, \vec{q}) \sim [M(q_x)]_{e,h} \frac{\ln[u-(u^2-1)^{1/2}]}{[(2\mu\omega_0/\hbar)^2-2q_t^2(k_i^2+k_s^2)+q_t^4]^{1/2}}, \tag{1}$$

where $\vec{q}$ is the phonon wave vector with q_t and q_x referring to components in the layered plane and perpendicular to it,

$$u=\frac{q_t^2-k_i^2-k_s^2}{2k_ik_s} \quad \mathrm{Im}k_{i,s}>0,$$

$$\hbar^2k_i^2/2\mu=E_i-E_n-\hbar\omega_0-i\Gamma,$$

$$\hbar^2k_s^2/2\mu=E_i-E_n-i\Gamma,$$

where E_i, $\hbar\omega_0$, μ, and Γ are the incident photon energy, phonon energy, reduced mass, and the damping factor, respectively, and

$$[M(q_x)]_{e,h}=\int\Psi_e(x_e)\Psi_h(x_h)\exp(iq_xx_{e,h})\Psi_e^*(x_e)\Psi_h^*(x_h)dx_e dx_h.$$

Expanding in powers of q_x and q_t in the resonance region where $2|E_i-E_n|q_t^2/\mu \ll \omega_0^2$ and assuming that $d_1q_x/\pi \ll 1$, we obtain for the allowed and forbidden parts of the Raman tensor,

$$R_A(E_i) \sim \chi(E_i)-\chi(E_i-\hbar\omega_0), \tag{2a}$$

$$R_F(E_i, \vec{q}) \sim (1/q)[\chi(E_i)-\chi(E_i-\hbar\omega_0)]\left[q_t^2\frac{(m_e-m_h)\hbar}{2\omega_0m_em_h}+q_x^2\left(\frac{\partial^2}{\partial q_x^2}\{[M(q_x)]_e-[M(q_x)]_h\}\right)_{q_x=0}\right], \tag{2b}$$

where χ is the electronic susceptibility. The usual selection rules apply for the allowed part, while the forbidden part is large only if the polarization of the scattered photon is parallel to that of the incident one.[6] The energy dependence in Eq. (2) is determined by the 2D nature of the electronic states, and is contained in the susceptibilities. This would remain the case even if the phonons are slightly modified in the thin GaAs layers—a sum of plane waves would then be substituted for the single q_x in $[M(q_x)]$. The resonant enhancement displays the well-known logarithmic singularities of the 2D susceptibilities if $\Gamma=0$.[7] Also, contrary to the 3D situation, the energy dependence in the resonance region is the same for both allowed and forbidden scatterings. The slight deviation from the 2D character in the superlattice is taken into account through the density of states in the expression of the susceptibilities:

$$\chi(E_i)-\chi(E_i-\hbar\omega_0) \sim \int\frac{\rho(E)dE}{(E-E_i-i\Gamma)(E-E_i-\hbar\omega_0-i\Gamma)}.$$

Thus, the Raman intensity, apart from a constant, can be calculated from $\rho(E)$ for a given superlattice configuration, even without the detailed knowledge of the electron-phonon interaction involved in the scattering process.

The theoretical curves in Fig. 2 are obtained from numerically calculated $\rho(E)$. The damping factor is estimated to be ~10 meV from transport measurements.[3] The contribution of each well is numerically summed, taking into account the effect of absorption. The absorption coefficient, for which the superlattice is treated as a homogeneous material,[8] is obtained at each energy through its proportionality to the density of states,[9] with the same proportional constant as in the ordinary 3D GaAs.[10] Referring to Fig. 2, for example, the resulting values are 4900, 8300, 13 700, and 15 700 cm^{-1} in (a), (b), (c), and (d), respectively, in the region above $E_1+\Delta_1$; 9800 cm^{-1}

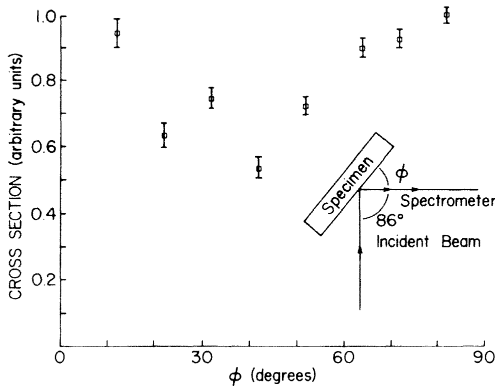

FIG. 3. Angular dependence of the Raman cross section for superlattice specimen (b) in Fig. 2, with the angle defined in the inset.

in (a) beyond $E_2+\Delta_2$. A multiplicative constant, fixed by matching the calculated and the observed peaks in Fig. 2(b), is used throughout with no additional adjustable parameters. The overall agreement is seen to be quite satisfactory both in resonant energies and in intensities.

In polarization studies, we have found the breakdown of the usual selection rule[5]; the intensity of scattered photon polarized parallel to the incident one is invariably much stronger than that perpendicular to it, indicating the involvement of forbidden processes. Furthermore, the angular dependence of the cross section exhibits a minimum when $\vec{q}$ is aligned along the x direction, as shown in Fig. 3. The magnitude of the intensity at $q_t=0$ ($\Phi\sim45°$ in Fig. 3) indicates a significant contribution from q_x-induced scattering, probably of the form of the second term in Eq. 2(b). This term, which is inherent in a superlattice, results from the generally nonidentical enevelope wave functions of $\Psi_e(x)$ and $\Psi_h(x)$, and it is similar in its origin to that of field-induced scattering.[11] The term involving q_t^2 in Eq. 2(b), on the other hand, could account for the observed angular dependence in Fig. 3, if the polaron radius could be somewhat enhanced by the electron-hole interaction in a 2D system. Of course, this does not rule out the possibility that the angular dependence arises from matrix elements not explicitly included in Eq. 2, for example, the electron-photon interaction.

In summary, we have illustrated the salient features of RRS in a man-made, 2D semiconductor structure. To our knowledge this is the first time that the strong enhancement of RRS arising from the 2D nature of the electronic states has been demonstrated.

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