

# Low-frequency Raman scattering in thin semiconductor films

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Natural acoustic modes of thin epitaxial semiconductor films have been observed experimentally.

The vibrational properties of composite layered systems are the subject of active theoretical research,<sup>1,2</sup> but so far there has been no experimental observation of natural acoustic vibrations of thin films grown on substrates of a different material.

In an effort to observe such vibrations, we selected thin stressed and unstressed epitaxial films of Ge on Si and GaAs substrates, respectively. The films were grown in ultrahigh vacuum at a residual gas pressure below  $10^{-8}$  Pa. The structure of the films was studied simultaneously by electron diffraction. The substrate surface was cleaned by baking at 850°C before the molecular-beam epitaxy. The Raman spectra were recorded in a 90° scattering geometry on a Ramanor-U 1000 spectrometer in an arrangement of “doubly parallel” polarization; in other words, the exciting and scattered light beams were polarized in the scattering plane.

On the experimental Raman spectra of the films (Fig. 1) we observed some broad, intense peaks at frequencies above that of the Brillouin peak. As in the case of Raman scattering of microcrystals,<sup>3,4</sup> we attributed these peaks to natural acoustic vibrations, since their energies are inversely proportional to the film thickness.

Microcrystals, like films, are extended systems whose natural vibrations correspond to phonons with wave vectors considerably larger than the difference between the wave vectors of the incident and scattered light. These vibrations can therefore be seen in a Raman spectrum only as the result of a violation of a wave-vector selection rule due to a limitation imposed on the phonon wave function by the boundaries of the film or due to a spatial damping of phonons by the disorder of the crystal structure in the stressed films. The stress and disorder are indicated by the energy shift, broadening, and asymmetry of the Raman spectrum of LO phonons in these films, on both the Si and GaAs substrates. We believe that the disorder of the crystalline films may cause a localization of acoustic phonons.

The energy eigenvalues of phonons localized in a film and damped exponentially in the substrate are described by <sup>2</sup>

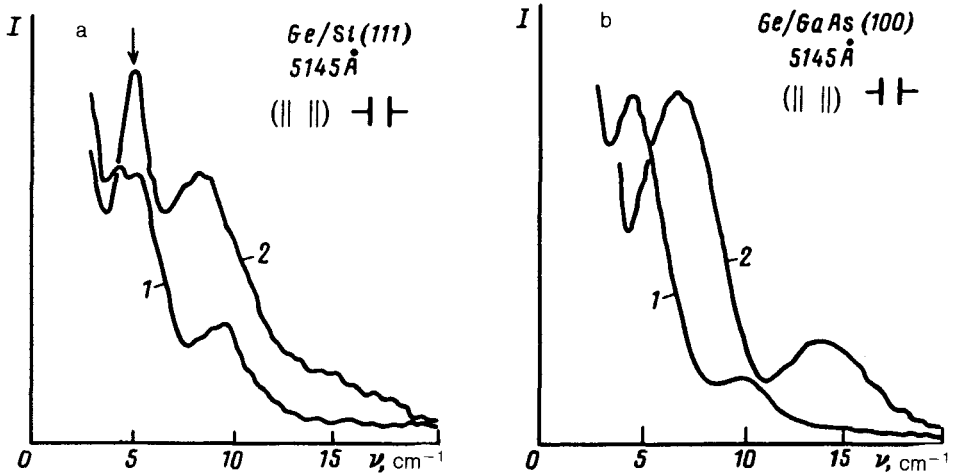


FIG. 1. Spectra of Raman scattering by natural acoustic vibrations. a: Stressed epitaxial films of Ge with thicknesses of (1) 19 and (2) 10 nm on Si substrates. b: Unstressed films of Ge with thicknesses of (1) 18 and (2) 12 nm on GaAs substrates. The arrow in frame a shows the Brillouin LA peak from the Si substrate. The Brillouin peaks from the Ge and the GaAs are at approximately 3 and 2  $\text{cm}^{-1}$ , respectively, and cannot be seen in this figure. The spectra contain the  $n = 1$  and  $n = 2$  vibrational modes, except for spectrum 2 in frame a, which exhibits only one mode, the  $n = 1$  mode.

$$2\{1 - (\beta_1/\beta_2)[t_2/(t_2 - 1)]\} \sinh(q_1 a L) + (\beta_I/\beta_1) \times \cosh[q_1 a(L - 1/2)] [\sinh(q_1 a/2)]^{-1} = 0, \quad (1)$$

where  $t_m = \xi_m + i(1 - \xi_m)^{1/2}$ ,  $\xi_m = 1 - (M_m/2\beta_m)\omega^2$ ,  $m = 1$  refers to the film,  $m = 2$  refers to the substrate,  $\beta_I = (\beta_1 + \beta_2)/2$  is the force constant acting between interfacial atoms,  $L$  is the number of atomic layers in the film,  $a$  is the distance between layers in the film, and  $q$  is the wave-vector eigenvalue of the phonon. We seek the eigenfrequencies in the form  $\omega = 2(\beta_1/M_1)^{1/2} \sin(pLa/2)$ , where  $p = p_1 + ip_2$  is a complex wave vector. We introduce  $p_1 La \equiv x$  and  $p_2 La \equiv y$ . We then have the following expressions for the real and imaginary parts of the eigenvalue of the phonon wave vector:

$$\frac{\beta_1}{2\beta L} x = \frac{\tan x(1 - \tanh^2 y)}{\tan^2 x + \tanh^2 y}, \quad \frac{\beta_1}{2\beta L} y = \left(\frac{M_1 \beta_1}{M_2 \beta_2}\right)^{1/2} + \frac{\tanh y}{\sin^2 x + \cos^2 x \tanh^2 y}, \quad (2)$$

where  $1/\beta = 1/\beta_1 + 1/\beta_2 - 2/\beta_I$ . In the case at hand, an analytic solution of Eqs. (2) can be derived by virtue of the small parameter  $\beta_1/2\beta L \approx 10^{-4}$ . We find the following expressions for the energy eigenvalues of the vibrational modes and their half-widths from Eqs. (2);

$$\omega_n = n \frac{v}{La} \pi \left(1 + \frac{\beta_1}{2\beta L}\right) \approx n \frac{v}{La} \pi, \quad \Gamma = \frac{v}{La} \operatorname{arctanh} \left(\frac{m_2 \beta_2}{m_1 \beta_1}\right)^{1/2} \approx 0.9 \frac{v}{La}, \quad (3)$$

where  $v$  is the longitudinal sound velocity. It can be seen from Eqs. (3) that the half-widths of the phonon modes are independent of the energy and that the vibra-

tional spectrum of the film consists of equidistant modes like the energy levels in a one-dimensional quantum well. Note that the light interacts most intensely with the lowest-index phonon modes, since the spectrum of phonon wave vectors becomes Lorentzian near the center of the Brillouin zone upon a violation of the wave-vector selection rule.

Corresponding peaks have apparently been observed<sup>5,6</sup> in the low-frequency Raman scattering by ultrathin  $\text{Ge}_m\text{Si}_n$  superlattices. The low-frequency part of the spectrum of such lattices is insensitive to the layer structure and corresponds to the vibration of a single layer with average parameters. Lockwood *et al.*<sup>5,6</sup> interpreted these peaks as resonant phonon modes which arise from an interaction of quasilocal modes in the superlattice and the continuum of acoustic modes of the substrate. We believe that the interaction with a substrate leads to simply a blurring of a resonance.

The film thicknesses estimated from the positions of the experimental energy peaks and Eqs. (3) are slightly smaller (by  $\approx 2$  nm) than the thicknesses found by direct measurements. One possible reason for this discrepancy is that the film surface was covered by an oxide, i.e., was not a free surface, so the peaks were shifted toward higher energies.<sup>5,6</sup> Another possibility is that the constant of the interaction of the light with the vibrations depends on the frequency; the shift of the peaks in this case might be 20%.

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<sup>2</sup>B. Sylla and L. Dobrzynski, *J. Phys. Cond. Matt.* **1**, 1247 (1989).

<sup>3</sup>N. N. Ovsyuk, E. B. Gorokhov, V. V. Grishchenko, and A. P. Shebanin, *Pis'ma Zh. Eksp. Teor. Fiz.* **47**, 248 (1988) [*JETP Lett.* **47**, 298 (1988)].

<sup>4</sup>N. N. Ovsyuk, L. M. Gorokhov, V. V. Grishchenko, and A. P. Shebanin, *Pis'ma Zh. Eksp. Teor. Fiz.* **48**, 626 (1988) [*JETP Lett.* **48**, 678 (1988)].

<sup>5</sup>D. J. Lockwood, M. W. C. Dharma-wardana, G.C. Aers, and J.-M. Baribeau, *Appl. Phys. Lett.* **52**, 2040 (1988).

<sup>6</sup>M. W. C. Dharma-wardana, D. J. Lockwood, G. C. Aers, and J.-M. Baribeau, *J. Phys. Cond. Matt.* **1**, 2445 (1989).

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