Anharmonicity of GaAs optical vibrations in GaAs/AlAs superlattices

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Structural features due to an anharmonicity of GaAs optical vibrations have been observed on the temperature dependence of the optical vibrational modes localized in GaAs layers. The Grüneisen parameters of localized optical vibrations far from the center of the Brillouin zone have been determined experimentally for the first time.

Research on the anharmonicity of optical vibrations in GaAs crystals by Raman scattering[1,2] and IR spectroscopy[3] has previously been limited to the Γ point in the Brillouin zone, because the interaction of phonons with light occurs at negligible small wave numbers ($q \approx 0$). This limitation has now been eliminated by progress in molecular-beam epitaxy, which is presently capable of synthesizing periodic structures with thin alternating layers (of GaAs and AlAs, for example). The quality is high, and the boundaries are sharp. If the dispersion curves for phonons in the materials making up a superlattice do not overlap, the phonons are localized within the layers, and the spectrum becomes discrete: $q_m = \pi m/(na)$, where a is the thickness of a monolayer, n is the number of monolayers of the corresponding layer, and m is equal to the difference $1 - n$. The selection rules allow the excitation of odd-m localized modes by IR light.

A localization of transverse optical (*TO*) phonons in the layers of GaAs/AlAs superlattices was recently detected[4] in IR reflection spectra.

In the present letter we are reporting a study of the temperature dependence of localized GaAs modes in GaAs/AlAs superlattices. The results provide evidence that the optical vibrational modes of GaAs are anharmonic.

We studied $(GaAs)_n/(AlAs)_m$ superlattices with $n = m = 17$ monolayers in the GaAs and AlAs layers, with 50 periods. These superlattices were grown at the Institute of Semiconductor Physics, Russian Academy of Sciences, by molecular-beam epitaxy on GaAs substrates oriented in the [100] direction. The reflection spectra were recorded in the region of GaAs *TO* vibrations over the temperature range 4–300 K with the help of a Bruker IFS-113V IR Fourier spectrometer. The reflection measurements were carried out at an approximately normal angle.

The frequencies (ω_j) of the localized *TO* modes were determined at various temperatures by the method described in Ref. 4. Figure 1 shows the temperature dependence of the *TO* modes localized in the GaAs layers, along with their derivatives with respect to the temperature, $d\omega_j/dT$. The dashed line is a temperature dependence measured in a bulk GaAs crystal. It can be seen from this figure that the temperature

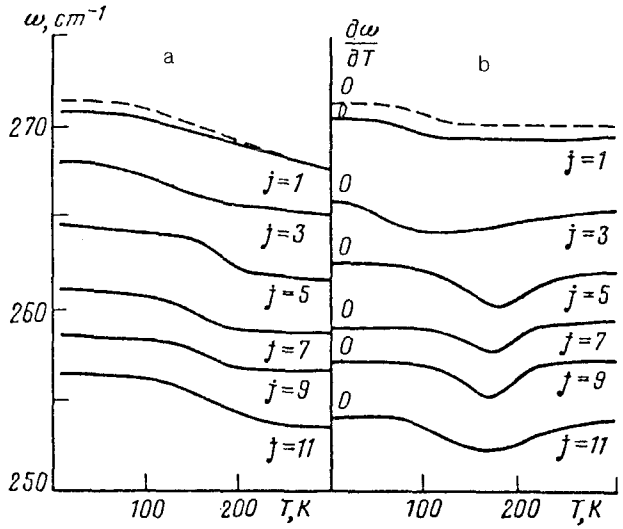

FIG. 1. Solid lines—Temperature dependence of (a) the frequencies and (b) the derivatives of the frequency with respect to the temperature, $\partial\omega_j/\partial T$, for transverse vibrational modes localized in the GaAs layers; dashed lines—temperature dependence of (a) the frequency of a bulk GaAs *TO* phonon and (b) its derivative with respect to the temperature (b). The index j specifies the localized mode.

dependence changes in shape as the index of the localized mode increases. Starting with the third localized mode, a clearly expressed minimum appears in the plot of the derivative, at $T = 160$ K. The temperature-induced change in the frequencies of the localized modes found here is a consequence of anharmonic interactions, which give rise to an eigenenergy that shifts the frequency of a free phonon.[6] Two components turn out to be important. The first stems from thermal expansion, and the second from a phonon-phonon interaction. We believe that the decrease in the frequencies of the localized modes seen at $\simeq 160$ K is caused primarily by thermal expansion, while the subsequent growth is caused by an interaction of optical phonons with virtual acoustic phonons. The probability for such a process may be higher in superlattices than in crystals, since (on the one hand) the momentum selection rules are violated by the localization of the optical phonons and (on the other) the folding of acoustic photons gives rise to new acoustic branches which promote a phonon–phonon interaction.

We worked from the temperature dependence $\omega_j(T)$, to calculate the Grüneisen parameters for each localized mode as a function of the temperature (Fig. 2). These parameters were found from

$$J_j(T) = -\frac{1}{3\alpha(T)}\frac{d\omega_j(V,T)}{dT}\frac{1}{\omega_{0j}}, \tag{1}$$

where ω_{oj} are the frequencies of the jth localized mode at $T = 4$ K, and α is the coefficient of linear expansion, which we took from Ref. 5. The Grüneisen parameters calculated from (1) include the component due to thermal expansion and also the

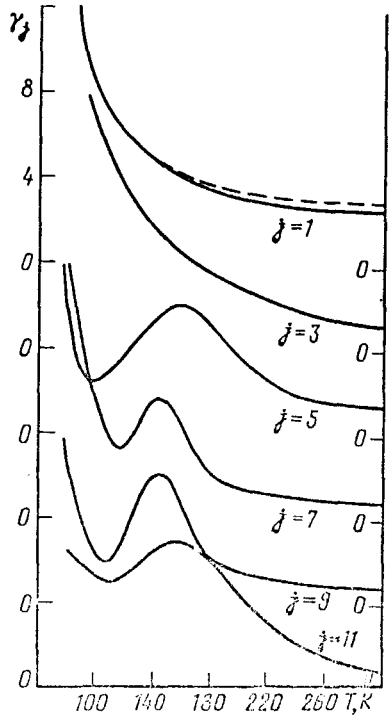

FIG. 2. Solid lines—Temperature dependence of the Grüneisen coefficient for optical vibrational modes localized in the GaAs layers; dashed line—temperature dependence of the Grüneisen coefficient for a bulk GaAs *TO* phonon. The index j specifies the localized mode.

component due to the phonon–phonon interaction.

In summary, we have studied the temperature dependence of the frequencies of transverse optical vibrations localized in the GaAs layers of GaAs/AlAs superlattices. We have calculated the Grüneisen parameters for each localized mode. We have found that the changes in the Grüneisen parameters for the higher-index modes are not monotonic. The reason is an anharmonicity of the optical vibrations in GaAs.

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