Comment on "Resonance Raman Scattering by Confined LO and TO Phonons in GaAs-AlAs Superlattices"

In a recent Letter,[1] Sood, Menéndez, Cardona, and Ploog have analyzed the frequencies of the GaAs-like optical phonon modes in GaAs/AlAs superlattices as those of an isolated GaAs thin slab. This analysis had been used previously in other papers.[2] We want to point out in this Comment that the quantized frequencies obtained in this way are different from the ones obtained from the dispersion relation of a superlattice,[3] *even* in the limit where the frequency difference between the optical modes of the two bulk compounds is very large.

Let us consider a superlattice made on n_1 monolayers of GaAs and n_2 monolayers of AlAs. The Kronig-Penney linear-chain calculation presented in Ref. 3 leads to the following dispersion relation $\omega(q)$ for the longitudinal modes propagating along the superlattice growth axis:

$$\cos[qa(n_1+n_2)] = \cos(n_1k_1a)\cos(n_2k_2a) - \alpha\sin(n_1k_1a)\sin(n_2k_2a)$$

with

$$\alpha = \frac{1-\cos(k_1a)\cos(k_2a)}{\sin(k_1a)\sin(k_2a)}.$$

k_i is the wave vector, eventually complex, at frequency ω in medium i, q the superlattice wave vector along the growth direction, and a the common monolayer thickness in GaAs and AlAs (2.83 Å). If we take k_2 infinite and imaginary, i.e., a vanishing penetration depth in the AlAs barrier, the frequencies of the modes confined in GaAs identify with the bulk GaAs ones at wave vector:

$$k_1 = m\pi/(n_1+1)a, \quad 1 \leq m \leq n_1, \tag{1}$$

instead of

$$k_1 = m\pi/n_1a, \tag{2}$$

which is the formula used by Sood *et al.*,[1] a formula deduced from a continuous-medium description of an isolated clamped slab of thickness $d_1 = n_1a$. This difference can be very well understood if one realizes that the interfacial arsenic atom is still moving and that the first pinned atom is the first aluminum atom in the AlAs layers. Actually, our approximation perfectly reproduces the exact frequencies of the modes confined in AlAs. As a result of the vicinity of the acoustical branch which prevents k_2 from taking very large imaginary values, the GaAs modes are not so perfectly confined, especially for large values of m. In this case the correction to formula (2) is slightly less than 1 (typically 0.8).

The use of (2) instead of (1) can lead to large differences for small GaAs layer widths and especially for large values of m. Moreover, in our formula, m takes n_1 values without q reaching the zone boundary. Use of this relation in the comparison between Raman-scattering results on superlattices[1,3] and neutron-scattering results[4] on bulk GaAs reduces the discrepancy reported in Ref. 1, which, however, remains significant. This discrepancy could be attributed, for instance, to the effect of slightly graded interfaces.[5]

As a conclusion, we stress that the optical vibrations in GaAs/AlAs are confined modes rather than clamped slab modes: Their energies, though independent of the barrier thickness, clearly depend on its nature through boundary conditions.

Finally we would like to point out that the expression "folded optical phonons," if not well adapted to the GaAs/AlAs case, is meaningful in the more complex and interesting case of GaAs/GaAlAs superlattices.[3] In this sytem, the bulk optical phonon bands partially overlap and the superlattice dynamics continuously evolves from a confined behavior (n_1 dependence) to a propagative one (folded dispersion curves, n_1+n_2 dependence).

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[1]A. K. Sood, J. Menéndez, M. Cardona, and K. Ploog, Phys. Rev. Lett. **54**, 2111 (1985).

[2]See, for instance, C. Colvard *et al.*, Phys. Rev. B **31**, 2080 (1985).

[3]B. Jusserand, D. Paquet, and A. Regreny, Phys. Rev. B **30**, 6245 (1984).

[4]J. L. T. Waugh and G. Dolling, Phys. Rev. **132**, 2410 (1963).

[5]B. Jusserand, F. Alexandre, D. Paquet, and G. Le Roux, Appl. Phys. Lett. **47**, 301 (1985).