Comment on "Confined-to-Propagating Transition of LO Phonons in GaAs/$Al_xGa_{1-x}As$ Superlattices Observed by Picosecond Raman Scattering"

In an intriguing, recent Letter Kim *et al.* [1] reported a nearly discontinuous change in the hot-phonon generation efficiency in GaAs/$Al_xGa_{1-x}As$ superlattices as a function of the alloy layer thickness L_b. These results were interpreted in terms of a confined-to-propagating transition which, according to the authors, occurs when the barrier width becomes thinner than $L_b = 20$ Å for $x = 0.4$, and $L_b = 11$ Å for $x = 1.0$. The results for alloy barriers raise the fascinating possibility that at some critical concentration, x, the alloy becomes too diluted to present any barrier to well phonons, thereby explaining the sharp transition observed by Kim *et al.*

In order to verify the likelihood of such a transition, we have calculated the phonon structure of a $(GaAs)_6$-$(Al_xGa_{1-x}As)_6$ superlattice for $x = 0$ to $x = 1$ in increments of 0.1. We followed the method of Chang, Ren, and Chu [2] and used an 11-parameter rigid-ion model [3] to describe the phonon structure of the parent compounds, GaAs and AlAs. Since the model is three dimensional, the alloy disorder is treated exactly. The force constants used are very realistic; in addition, the results reported in this Comment are largely independent of their exact values. Our superlattice structure has $L_b = 17$ Å < 20 Å and a well thickness $L_z = 17$ Å (smaller than $L_z = 100$ Å for all samples in Ref. [1]), so that a confined-to-propagating transition, if it exists, might be expected at $x \geq 0.4$.

Figure 1 shows our calculated results. We plot two indicators of phonon localization: the fractional squared eigenvector ratio (see caption) of the LO_1 phonon in the GaAs layer, and the energy separation between zone center and zone edge modes along the superlattice growth direction. A significant separation of the modes implies dispersion along the superlattice axis, signaling the presence of propagating phonons. Figure 1 indicates that no sharp, delocalizing transition takes place as the alloy concentration is reduced. We obtain a smooth curve that approaches 0.5 as $x \rightarrow 0$. The well phonons remain almost perfectly localized for concentrations as low as $x = 0.1$. For $x = 0.4$, 98% of the phonon amplitude is localized in the well. Even if the barrier width is reduced to half the value quoted by Kim *et al.* (not shown in Fig. 1), the degree of localization remains unchanged. *Therefore, one cannot invoke delocalization to explain the large changes in the electron-phonon matrix elements needed to account for an order-of-magnitude change in the hot-phonon generation efficiency.*

For AlAs barriers, we also find that the well phonons should be perfectly localized even at the lowest nominal thickness quoted by Kim *et al.*, but here the experimental results could be explained in terms of sample imperfections. This is not the case for the alloy barriers, where the well phonons are found to remain localized even when assuming unreasonably large errors in the barrier thickness.

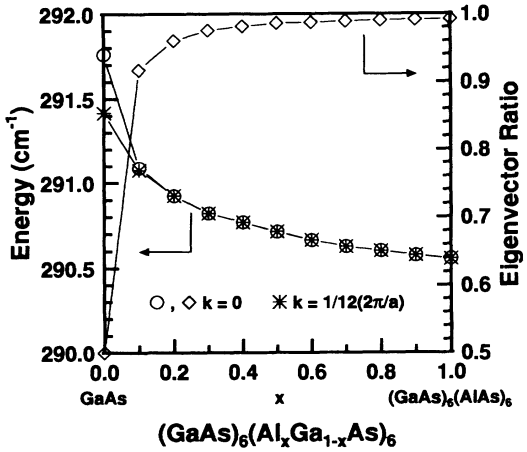

FIG. 1. Indicators of phonon localization in $(GaAs)_6$-$(Al_xGa_{1-x}As)_6$ superlattices for the first confined mode calculated using a supercell of 240 atoms. Mode energy versus alloy composition for zone center and zone edge are plotted against the left y axis. "Eigenvector ratio" represents the sum of squared eigenvectors in the GaAs layer divided by the sum of squared eigenvectors in both layers. This ratio versus alloy composition is plotted against the right y axis. Both the separation of the energy of the modes and the "eigenvector ratio" indicate that, even at low concentrations, the first confined phonon is localized.

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[1] D. S. Kim, A. Bouchalkha, J. M. Jacob, J. F. Zhou, J. J. Song, and J. F. Klem, Phys. Rev. Lett. **68**, 1002 (1992).
[2] Y. C. Chang, S. F. Ren, and H. Chu, Superlattices Microstruct. **9**, 383 (1991).
[3] K. Kunc, M. Balkanski, and M. A. Nusimovici, Phys. Rev. B **12**, 4346 (1975).

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