

Response to "Comment on 'Optical and acoustic phonon modes in self-organized Ge quantum dot superlattices'"

[Appl. Phys. Lett. 78, 1160 (2001)]

J. L. Liu,^{a)} G. Jin, Y. S. Tang, Y. H. Luo, and K. L. Wang

Device Research Laboratory, Department of Electrical Engineering, University of California at Los Angeles, Los Angeles, California 90095-1594

D. P. Yu

Electron Microscopy Laboratory, Peking University, Beijing 100871, People's Republic of China

(Received 1 May 2000; accepted for publication 30 December 2000)

[DOI: 10.1063/1.1350590]

In a comment¹ on our recent letter,² Yu first pointed out that there was strong alloying between the Ge dots and Si barrier layers because of the appearance of Si-Ge modes as also observed in SiGe alloys. It is correct that the Ge dot samples reported in our letter have some degrees of alloying due to interdiffusion. This was due to the fact that the samples were grown at a high temperature of 600 °C. The reason for using this temperature came from our intention to control the size uniformity. It was found that an optimum temperature occurs at around 600 °C for high-uniform monomodal Ge dots on planar Si substrate.^{3,4} At lower temperatures, the uniformity becomes worse. Moreover, two kinds of dots coexisted on the Si substrate (pyramid and dome).^{3,4} Figure 1(a) shows a cross-sectional TEM image of a ten-period Ge quantum dot sample grown at 550 °C (sample 1). Vertically correlated dots are evident. The size variation of the first Ge dot layer, however, is determined to be 20% from AFM measurements [Fig. 1(b)]. The nonuniformity arises from limited diffusion at the lower growth temperature⁵ and becomes worse in the vertically correlated multilayers because vertical correlation rearranges the strain distribution, leading to the fact that the upper layer dots are larger than those in the lower layers [Fig. 1(a)]. On the other hand, at a higher temperature around 600 °C, the dot uniformity is much improved and the size variation decreases to, for example 7%–8% in our samples in the letter.² Such uniformity may be desirable, but the interdiffusion between the Ge dots and Si spacers reduces quantum confinement effects and is undesirable for optical applications. Thus, there is always a tradeoff between the uniformity and interdiffusion.

Now let us take a look at the Raman spectrum of the sample 1 [Fig. 1(c)]. This figure includes a spectrum for a ten-period Ge dot superlattice grown at 500 °C (sample 2). One can easily see Si-Ge vibration modes at around 400 cm⁻¹ from both samples. The relative strength $I_{\text{Si-Ge}}/I_{\text{Ge-Ge}}$ decreases as the temperature decreases. Another phenomenon is that there is a weak feature (indicated by an arrow in the figure) between Si-Ge mode and Si-Si mode for sample 1. This is due to localized Si-Si motion in the neighborhood of one or more Ge atoms.⁶ Such kinds of localized Si-Si optical modes (Si-Si_{loc}) are often observed in SiGe alloys

and interface-diffused superlattices. A detailed image describing these can be found in Ref. 6. Thus, the higher the growth temperature, the more Si-Ge bonds per unit volume due to interdiffusion, and thus the relatively stronger Si-Ge mode and localized Si-Si mode intensity.

It is also necessary to add some clarifications to low frequency "acoustic-phonon-like" modes observed in our Ge dot samples.² First, these modes were not multiphonon modes as the Raman scattering was nonresonant. Second, they were not associated with a superlattice where low frequency zone edge phonon modes have been folded into the zone center because the Si barriers were too thick to account for the breathing and torsional modes of both samples B and C. In order to assign these modes to be confined phonon modes in quantum dots, one may expect that the frequencies

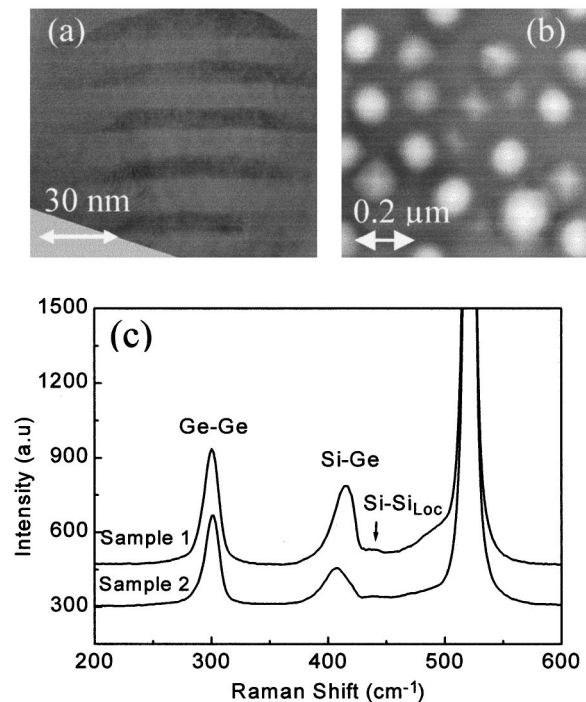


FIG. 1. (a) Cross sectional TEM of the ten-period Ge quantum dot superlattice sample grown at 550 °C (sample 1). (b) AFM image of the first Ge dot layer of the sample 1. (c) Raman spectra of samples 1 and 2 (grown at 500 °C). Si-Ge modes at around 400 cm⁻¹ are evident for both samples. The Si-Si_{loc} mode is observed for sample 1 but very weak for sample 2.

^{a)}Electronic mail: jliu@ee.ucla.edu

of these modes scale as (dot radius)⁻¹. It is well known that such a dependence came from the celebrated Lamb's theory.⁷ Nevertheless, the application of this theory to some nanocrystals (for example, Si and Ge) is far from satisfactory.^{8,9} The first reason is the oversimplified assumption that the nanocrystal is sphere and elastically isotropic, whereas Si and Ge are highly elastically asymmetrical and in our case, the Ge dots have a dome shape.² The second is that Lamb's theory treats free-standing nanocrystals, in other words, it does not take into account the important effects of the surrounding matrix (Si spacers and/or diffused interfaces in our case) to the acoustic phonons.⁹ The third is that the assumption of an elastic continuum in the Lamb's theory may not be valid for nanocrystals with very small size (for example, 15–20 Å in our case). Furthermore, suppose that the four low-frequency peaks are the higher orders of acoustic phonons for sample C, then the first-order pure compressional (breathing) or shear (torsional) mode should be 22.5 cm⁻¹. Similarly for sample B, the breathing or torsional mode should be 24.9 cm⁻¹. Strictly speaking, the difference between the two frequencies is small, indicating that the size difference between sample B and sample C (namely, the height difference is 5 Å, and still with 7%–8% nonuniformity) is too small to distinguish the breathing and torsional

modes of both samples and to see any dependence. On the whole, the exact mechanism for the appearance of these low frequency peaks remains unclear as clearly stated in our letter.² More quantum dot samples with larger dot sizes and reasonably good uniformity are needed for this purpose.

In summary, Yu gave an insightful comment on our letter which raises several concerned issues for self-assembly of Ge quantum dots on Si for optical applications. Acoustic phonon assignment to quantization of dots remains a challenge.

This work was supported by a U.S. DOD/ONR MURI Project on thermoelectrics (Dr. John Pazik).

¹P. Y. Yu, Appl. Phys. Lett. **78**, 1160 (2001), preceding paper.

²J. L. Liu, G. Jin, Y. S. Tang, Y. H. Luo, K. L. Wang, and D. P. Yu, Appl. Phys. Lett. **76**, 586 (2000).

³O. G. Schmidt, C. Lange, and K. Eberl, Appl. Phys. Lett. **75**, 1905 (1999).

⁴G. Jin, J. L. Liu, Y. H. Luo, and K. L. Wang (unpublished).

⁵J. Drucker and S. Chaparro, Appl. Phys. Lett. **71**, 614 (1997).

⁶M. I. Alonso and K. Winer, Phys. Rev. B **39**, 10056 (1989).

⁷H. Lamb, Proc. London Math. Soc. **13**, 189 (1882).

⁸M. Fujii, Y. Kanzawa, S. Hayashi, and K. Yamamoto, Phys. Rev. B **54**, 8373 (1996).

⁹J. Zi, K. M. Zhang, and X. D. Xie, Phys. Rev. B **58**, 6712 (1998).