Response to "Comment on 'Raman scattering from a self-organized Ge dot superlattice' " [Appl. Phys. Lett. 75, 3572 (1999)]

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In the Comment[1] on our recent letter,[2] Kolvol and Tanaka pointed out that the observed Raman peak at 301 cm^{-1} had nothing to do with Ge quantum dots but came from the Si substrate. We disagree with their comment. Here is our response.

We recognize that there should be a Raman 303 cm^{-1} line under a proper configuration for Si, which arises from Si acoustic phonons.[3] However, a peak at around 303 cm^{-1} (301 cm^{-1} in our case) does not mean that the peak must come from Si acoustic phonons. In order to prove our assignment, Raman scattering measurements were performed on the dot sample and an identical Si substrate using the same experimental Raman system with an identical data collection time. In addition, different polarization configurations according to selection rules were used to better distinguish the signals from the dot sample and the Si substrate. Figure 1 shows the observed results. The spectrum from the sample (top solid curve) was recorded in the $001(100,010)00\bar{1}$ backscattering geometry. This configuration was chosen to minimize the acoustic phonon peak at around 303 cm^{-1} from Si substrate. The spectrum from the Si substrate (bottom solid curve) was recorded in the $001(110,110)00\bar{1}$ backscattering configuration in order to enhance the Si acoustic phonon peak. A peak at 301 cm^{-1} from the sample in the top solid curve is about six times stronger than the Si acoustic phonon peak at 303 cm^{-1} from the substrate in the bottom solid curve. The strain on multilayered Si induced by the formation of Ge dots changes the symmetry of the localized Si (around the dots). Because of this effect, the 303 cm^{-1} Si acoustic phonon peak may show up even though the sample is under the $001(100,010)00\bar{1}$ configuration. Thus the observed 301 cm^{-1} Raman line from the dot sample may include the contribution from the Si acoustic phonons. Limited work on this issue[4] seems to indicate that the intensity of the Si acoustic phonon peak does not change significantly with and without the existence of strain. The only dominant signal in our case is from the Ge–Ge mode. In addition, the appearance of the Si–Ge mode at 403 cm^{-1} (top solid curve in Fig. 1) suggests imperfect Si–Ge interfaces due to high growth temperature and/or the formation of Ge dots. Otherwise, this peak should be forbidden in the $001(100,010)00\bar{1}$ backscattering configuration. This also supports the existence of the Ge–Ge mode. In fact, the only concern here may be that which parts, SiGe wetting layers or Ge dots, mostly contribute to the Ge–Ge mode. Existing work[5] indicated that the Ge–Ge mode of a sample with Ge dots was significantly more intense than that of a sample without dots (both contain SiGe wetting layers). In addition, we can also distinguish the signals from wetting layers or Ge dots of our sample by considering selection rules. In Fig. 1, the dotted spectrum is collected on the sample in the $001(100,100)00\bar{1}$ backscattering configuration. In this configuration, the signals from the Ge wetting layers should be forbidden according to the selection rules.[6] However, the Ge–Ge mode does not change significantly compared with that in the top solid curve in Fig. 1. All of the above allow us to conclude that the observed 301 cm^{-1} Raman line in our dot sample is mainly due to Ge–Ge vibrations in the dots rather than from the acoustic mode from the Si substrate.

The confusion also comes from the fact that the observed Ge–Ge mode is more or less similar to (although not identical) to the standard acoustic mode of Si, as also can be seen in Kolobov and Tanaka's Comment.[1] The peak shape similarity is not surprising, and may be due to the weak phonon confinement and/or strong interface roughness effect.[5] The weak confinement in our case comes from the Ge–Si interdiffusion process during the growth, which makes the interfaces between the Ge dots and Si spacers not as abrupt. This analysis allows us to acknowledge that the statement "the strong phonon confinement" in our letter[2] may be questionable.

In fact, similar Raman peaks at around 303 cm^{-1}, which

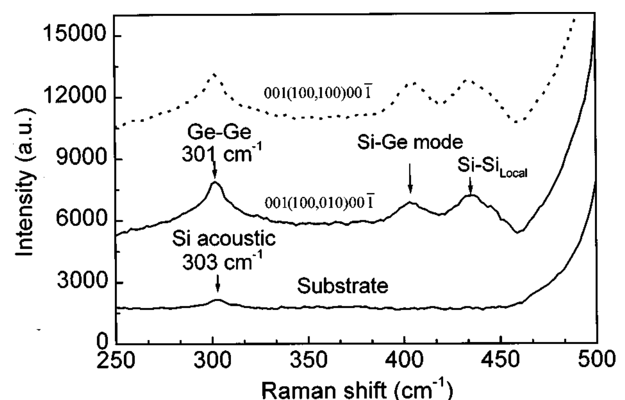

FIG. 1. Raman spectra of the sample and the substrate. The measurements were performed consecutively on a Renishaw Raman Imaging 2000 microscope. The accumulation time for the three spectra was identical. Different scattering configurations are used as specified in the text. A peak at 301 cm^{-1} in the spectrum of the sample (top solid curve) due to Ge–Ge vibrations is much stronger than a Si acoustic phonon peak at 303 cm^{-1} in the spectrum of the substrate (bottom solid curve).

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are due to Ge–Ge vibrations, have been observed recently in the Ge quantum dots and wires by many groups.[4,5,7–11] These Raman peaks shift with changing dot or wire size due to the phonon confinement effect.[4,5,7–9]

To summarize our Response, we stress that the observed peak at 301 cm^{-1} in our recent letter[2] is mainly due to Ge–Ge vibrations in the dots. It is not appropriate to increase the accumulation time during the scattering measurement on a Si substrate alone and then to assign the Ge–Ge peak from the dot sample to the Si acoustic phonon peak from the substrate.

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