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Raman spectra of $^{70}Ge/^{76}Ge$ isotope heterostructures with argon 488 and 514.5 nm excitations

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Abstract

Raman spectra of $^{70}Ge/^{76}Ge$ isotope bi-layer structures recorded with 488 and 514.5 nm Ar^+ laser excitations are reported. The relative amplitudes of the ^{70}Ge and ^{76}Ge confined optical phonon peaks differ significantly for the two excitations at $T = 10$ K, while it is almost the same for the spectra recorded at 300 K. Possible interpretations of our results are provided considering the laser penetration depths and the isotope effect on the electronic resonance Raman frequency.

Keywords: $^{70}Ge/^{76}Ge$ isotope superlattices; Raman scattering; Electronic resonance

Recent availability of isotopically pure Ge has enabled a wide variety of Raman investigation of phonons in isotope multilayer structures, in which modulation of the lattice mass can been controlled at the atomic level [1–7]. The periodic mass structures known as Ge isotope superlattices (SLs), e.g., $^{70}Ge_n/^{76}Ge_n$ isotope SLs with n being the number of mono-layers in each isotopically enriched layer, have been fabricated successfully using the state-of-the-art molecular beam epitaxy (MBE) techniques [1,2,4–7]. In contrast to the superlattices composed of heterogeneous elements such as GaAs/GaAlAs and Si/Ge, the Ge isotope SLs are unique in the sense that only the phonon properties are modulated by the mass periodicity while the macroscopic electronic structure is essentially unchanged from that of bulk Ge. Because of the negligibly small difference in the force constant and bond-polarizability between the two different isotope layers, the Raman investigation of Ge isotope SLs provide the ideal test structures for the theory of phonon dynamics in solids. In previous Ge isotope SL studies, Fuchs, et al. have calculated the zone-centered LO phonon dynamics and their Raman intensities of $^{70}Ge/^{76}Ge$ isotope SLs in the framework of the planar bond-charge (PBC) model and bond-polarizability approach [1]. Spitzer, et al. have measured the first-order Raman spectra of a series of $^{70}Ge/^{74}Ge$ isotope SLs and compared their

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results with the calculations [1,6]. The present authors have also investigated the phonons in isotope SLs using a series of $^{70}Ge/^{74}Ge$ and $^{70}Ge/^{76}Ge$ isotope SLs [4,5 and 7]. In general, good agreement between the calculated and the experimental results have been obtained in all of the above-mentioned investigations, confirming the validity of the PBC model and the high quality of the Ge isotope SLs. However, new Raman features have been discovered unexpectedly when the Ar^+ excitation was switched between 488 nm and 514.5 nm [5,6], and the possibility of the penetration depth being dependent on the excitation laser wavelength due to isotope effect has arisen.

In this paper we report on our efforts to find the answer to this puzzling result. In order to find the effect of the laser penetration depth, we have fabricated very simple $^{70}Ge/^{76}Ge$ isotope bi-layers shown in Fig. 1. Because the penetration depth of both 488 and 514.5 nm photons into Ge are estimated to be $\sim$16 nm [8], we have prepared the three samples with the thickness of the top layer (^{70}Ge) being smaller (8 nm for sample A), comparable (16 nm for sample B), and larger (32 nm for sample C) than the estimated penetration depth. The thicknesses of the buried ^{76}Ge layers are 72, 64, and 48 nm, respectively, i.e., the total thicknesses of the bi-layers are 80 nm for all three samples. The structures shown in Fig. 1 have been grown on nominally undoped natural Ge [1 0 0] substrates using molecular beam epitaxy. The growth rate has been maintained at 0.1 nm/min using PBN-Knudsen effusion cells. The temperature of the substrate during the growth was kept 350°C. The ^{70}Ge and ^{76}Ge solid sources

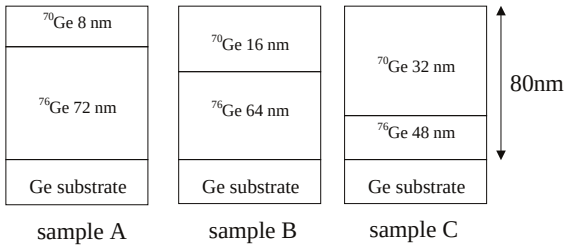

Fig. 1. Schematics of the three $^{70}Ge/^{76}Ge$ isotope bi-layers prepared for this study.

placed in the PBN-Knudsen effusion cells were isotopically enriched to 96.3% and 85.1%, respectively. The isotopic compositions of the ^{70}Ge and ^{76}Ge in each layer shown in Fig. 1 are essentially unchanged from those of the sources in the cells. The Raman measurements were performed at temperatures of 300 and 10 K using the Ar^+ 488 and 514.5 nm-lines, in the backscattering geometry. The spectral resolution was 1 cm^{-1}.

Fig. 2 (a) shows the Raman spectra recorded at 300 K of samples A, B, and C using 488 and 514.5 nm excitations, respectively. As expected, the intensity of the ^{70}Ge phonon peak (the large peak at $\sim$305 cm^{-1}) relative to that of the ^{76}Ge peak (the large peak at $\sim$293 cm^{-1}) becomes stronger as the thickness of the top ^{70}Ge is increased. When the laser penetration depth ($\sim$16 nm) is significantly larger than the ^{70}Ge layer thickness (8 nm) of sample A, the phonon spectrum is composed of both the ^{70}Ge and ^{76}Ge confined phonon modes. When the laser penetration depth ($\sim$16 nm) is significantly smaller than the ^{70}Ge layer thickness (32 nm) of sample C, the phonon spectrum is composed predominantly of the ^{70}Ge peak. Moreover, the agreement between the two sets of Raman spectra recorded with 488 and 514.5 nm excitations are very good, indicating that the penetration depths are essentially the same for the excitations.

Fig. 2(b) shows similar experimental results recorded at $T = 10$ K. As expected, the intensity of the ^{70}Ge phonon peak (the large peak at $\sim$309 cm^{-1}) relative to that of the ^{76}Ge peak (the large peak at $\sim$297 cm^{-1}) becomes stronger as the thickness of the top ^{70}Ge layer is increased for excitation at 488 nm and at 514.5 nm. However, the relative intensity of ^{70}Ge and ^{76}Ge is very different at $T = 10$ K as is seen clearly for sample A for the two excitations. The signal from the ^{76}Ge layer is enhanced drastically with respect to that from the ^{70}Ge layer when the spectrum is recorded with 514.5 nm excitation. In order to show this trend more quantitatively, the intensity of the ^{76}Ge confined phonon peaks normalized by that of ^{70}Ge, i.e. (intensity of ^{76}Ge confined phonon peak)/(intensity of ^{70}Ge confined phonon peak) is plotted as a function of the ^{70}Ge top-layer thickness in Fig. 3.

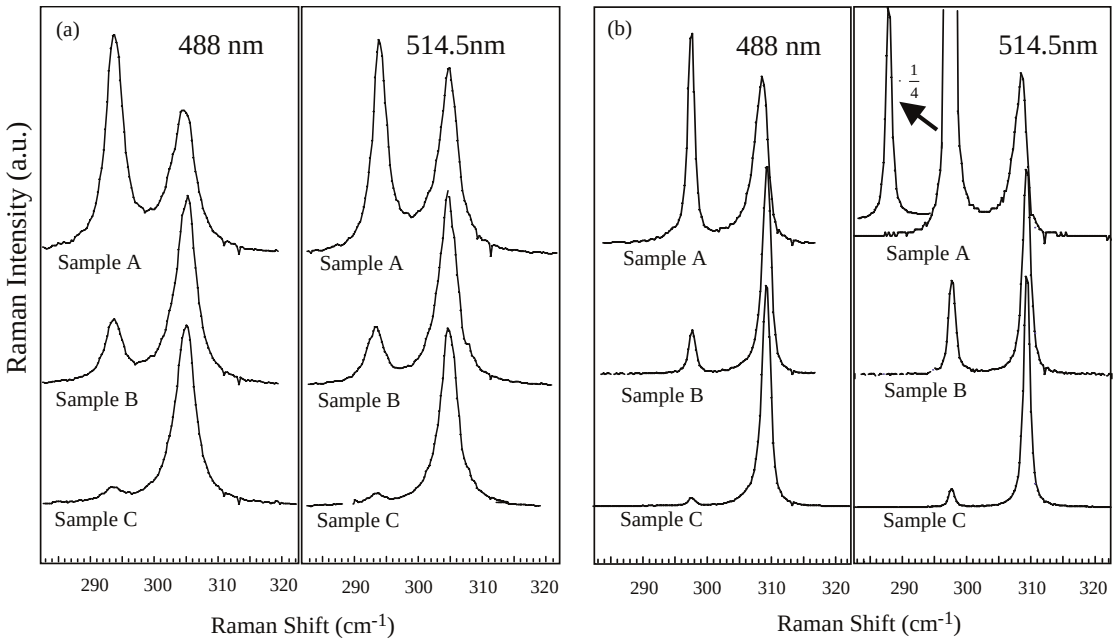

Fig. 2. Raman spectra of: (a) samples A, B, and C recorded at 300 K using 488 and 514.5 nm excitation lines, and (b) samples A, B, and C recorded at 10 K using 488 and 514.5 nm excitation lines.

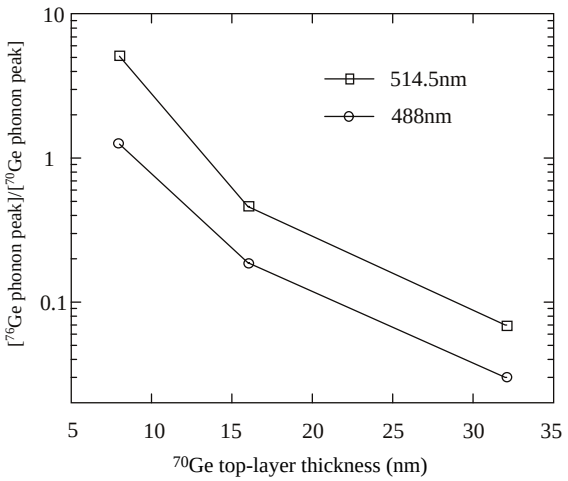

Fig. 3. The relative intensity of the ^{76}Ge confined phonon peaks normalized by that of the ^{70}Ge confined phonon peaks are plotted as a function of the ^{70}Ge top-layer thickness.

Our results clearly show that the penetration depth can change depending on the excitation laser wavelength. The penetration depth can be estimated using the absorption coefficient α calculated from the temperature dependent dielectric function of Ge [8]. At room temperature, the penetration depths of 488 and 514.5 nm excitations estimated from α^{-1} are $\sim$17.8 and 17.2 nm, respectively. At 100 K, the estimated penetration depths of 488 and 514.5 nm excitations are $\sim$16.5 and 16.4 nm, respectively. Intuitively, the penetration depth should not change much below 100 K since the temperature dependence of the electronic band structure is not strong enough to affect the dielectric property of Ge so significantly. Our 10 K result, therefore, cannot be explained solely by the difference in the penetration depth estimated from α at 100 K. There may be other physical reasons for the large change in the penetration depth at 10 K. One may claim that the enhancement of the ^{76}Ge confined phonon peaks at 10 K for the excitation 514.5 nm is due to the laser being in resonance with specific critical points of the electronic band structures of ^{76}Ge but not with those of ^{70}Ge, i.e., the ^{76}Ge Raman peak is enhanced by the resonant Raman scattering. Critical points in the band-structure of ^{76}Ge are in resonance with the energy of the 514.5 nm line

(2.41 eV), while it should not be in resonance with the same critical points of ^{70}Ge due to the subtle isotope effect on the band-structure. One of the candidates for the critical point in resonance is E_1 and $E_1 + \Delta_1$. However, the energy difference between ^{76}Ge and ^{70}Ge of E_1 and $E_1 + \Delta_1$ critical points is only 0.005 eV [9] such that a small level of isotope effect is not likely to lead to a large difference in the penetration depth between ^{76}Ge and ^{70}Ge. If isotope effect is not responsible for the large change in the laser penetration depth between 488 and 514.5 nm excitations, it is natural to assume that the change in the penetration depth between 488 nm and 514.5 nm excitations is equal for all the stable isotopes of Ge, i.e., there is no isotope effect on the penetration depth of 488 and 514.5 nm excitations. We can confirm this by reversing the stacking order of the ^{70}Ge and ^{76}Ge isotope layers of the samples shown in Fig. 1. By placing ^{76}Ge layers in front of ^{70}Ge layers, it is possible to check if the excitation energy dependence of the Raman spectra is related to the isotope effect or not. Such experiments are in progress by the present authors.

There are other interesting features in Fig. 2(b) besides those discussed above. For example, the full-widths at the half-maximum of the ^{70}Ge confined phonon peak becomes larger in sample A than in sample C. The frequency of this phonon peak also shifts. These results are most likely caused by a finite size effect of ^{70}Ge layers [10]. Understanding of these details will be achieved by systematic investigations involving a variety of Ge isotope bi-layers in the near future.

In summary, we have successfully fabricated a series of ^{70}Ge/^{76}Ge isotope bi-layers in order to investigate the excitation frequency dependence of the Raman spectra. A significant difference in the laser penetration depth has been confirmed between 488 and 514.5 nm excitations. Our preliminary analysis has suggested that the difference in the penetration depth is not induced by the isotope effect.

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References

[1] J. Spitzer, et al., Phys. Rev. Lett. 72 (1994) 1565.
[2] E. Silveira, et al., Phys. Rev. B 56 (1997) 2062.
[3] H.D. Fuchs, et al., Superlatt. Microstruct. 13 (1993) 447.
[4] K. Morita, et al., Thin Solid Films 369 (2000) 405.
[5] K. Morita, et al., in: N. Miura, T. Ando (Eds.), Proceedings of the 25th International Conference on the Physics of Semiconductors, Springer, Berlin, 2000, p. 877.
[6] J. Spitzer, Ph.D. Thesis, University of Stuttgart, 1994.
[7] M. Nakajima, et al., Phys. Rev. B 63 (2001) 161304.
[8] L. Viña, et al., Phys. Rev. B 30 (1984) 1979.
[9] S. Zollner, et al., Phys. Rev. B 45 (1983) 3376.
[10] I.H. Campbell, et al., Solid State Commun. 58 (1986) 739.