Journal of Luminescence 87–89 (2000) 942–944

JOURNAL OF LUMINESCENCE

www.elsevier.com/locate/jlumin

Comparison of coherent and incoherent LO phonons in isotopic $^{70}Ge/^{74}Ge$ superlattices

M. Nakajima[a,*], K. Mizoguchi[a], K. Morita[b], K. Itoh[b,c], H. Harima[a], S. Nakashima[a]

[a]Department of Applied Physics, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan
[b]Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223-8522, Japan
[c]PRESTO-JST, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223-8522, Japan

Abstract

Coherent LO phonon oscillations excited by 20-fs ultrashort laser pulses have been observed in isotopic $^{70}Ge/^{74}Ge$ superlattices by a pump–probe technique. Fourier-transformed spectra of the time-domain signal were compared with incoherent LO phonon spectra measured by Raman spectroscopy. The spectra of coherent phonons are quite different from the Raman spectra with common excitation wavelength. This comparison indicates that the pump–probe experiment detects the region which is different from where Raman experiment probes. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Coherent phonons; Isotopic superlattices; Femtosecond pulse laser

Investigations of coherent phonons have been conducted in the past using materials such as molecular crystals [1], semiconductors [2,3] and semiconductor superlattices (SLs) [4,5]. However, basic properties of coherent phonons including their generation mechanisms are still unclear, especially for absorbing materials. In this study, we report on the observation of confined coherent LO phonons in isotopic $^{70}Ge/^{74}Ge$ superlattices (abbreviated hereafter as isotopic Ge SLs) by a pump–probe technique using ultrashort laser pulses. This study is interesting especially in the sense that it allows us to compare directly the coherent phonons measured here to incoherent phonons measured by Raman spectroscopy. Raman investigations of incoherent LO phonons in isotopic Ge SLs have been performed recently by Cardona and co-workers [6,7]. The isotopic SLs present an excellent model to study the vibrational properties of SLs because the phonons are subject to a periodic isotopic mass modulation while the electronic system is practically the same as that of bulk Ge. We will report here variation of the confined LO phonon spectra in ^{70}Ge and ^{74}Ge layers for different layer thicknesses, and propose the mechanism which determines the coherent phonon amplitudes.

The present samples $(^{70}Ge)_n/(^{74}Ge)_n$ were grown by molecular beam epitaxy (MBE), having periodic structures with n (= 4, 8, 16, 32) monolayers of ^{70}Ge and ^{74}Ge. The thickness of one monolayer corresponds to 0.141 nm. The high-purity Ge (0 0 1) wafers of the natural isotopic composition have been used as substrates. ^{70}Ge buffer layers of 20 nm thickness were grown prior to the SL formation. The total thickness of each SLs is 50 nm, i.e., the total periods of each SLs is different depending on the thickness n of the each layer.

A reflection-type pump–probe measurement was performed at room temperature by using a mode-locked Ti : sapphire laser. The central wavelength of the laser was about $\lambda = 800$ nm and the pulse duration was about 20 fs. The power of the pump and probe beam was adjusted to 70 and 10 mW, respectively. The delay time of the probe beam was adjusted by a variable optical delay line. The optical path of the pump beam was modulated by a shaker. The time derivative of the reflectivity change $d(\Delta R/R_0)/dt$ was measured in order to

* Corresponding author. Tel.: + 81-6-6879-7854; fax: + 81-6-6879-7856.

E-mail address: nakajima@ap.eng.osaka-u.ac.jp (M. Nakajima)

PII: S0022-2313(99)00481-0

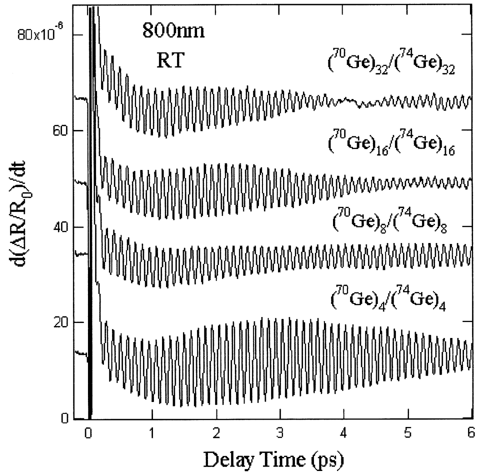

Fig. 1. Oscillatory component of the time-domain signals of isotopic $(^{70}Ge)_n/(^{74}Ge)_n$ SLs for various SL layer thicknesses at room temperature.

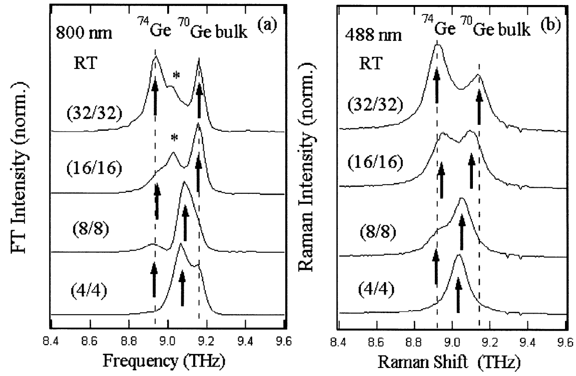

Fig. 2. (a) Fourier transform spectra of time-domain signals in Fig. 1. (b) Raman spectra of isotopic Ge SLs excited at 488 nm at room temperature. The vertical broken lines indicate the frequencies of LO phonon mode in bulk ^{70}Ge and ^{74}Ge.

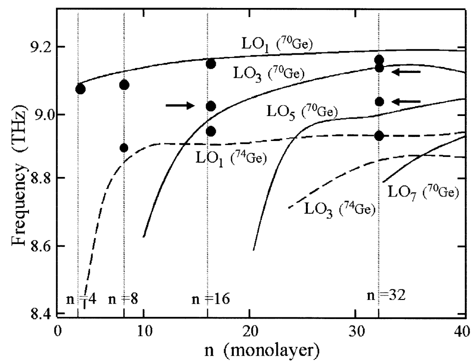

Fig. 3. The solid and broken lines denote calculations for LO_m frequencies (m = odd) in ^{70}Ge and ^{74}Ge layers, respectively. Observed peak frequencies are shown by filled circles against n, the number of monolayers in $(^{70}Ge)_n/(^{74}Ge)_n$ SLs.

obtain only the oscillatory component. The Raman scattering measurement was carried out at room temperature and at 10 K in a quasi-back-scattering geometry by using an Ar-ion laser at 488 and 514.5 nm, and a cw Ti : sapphire laser at 800 nm.

Fig. 1 shows time derivatives of the reflectivity change $d(\Delta R/R_0)/dt$ in the isotopic Ge SLs with different periodic structures. The beatings observed indicate that multiple phonon modes are excited in all four samples. To analyze the time-domain signals and compare with the Raman spectra, we have obtained Fourier transform (FT) spectra of the time-domain signals as depicted in Fig. 2(a). The vertical broken lines stand for frequencies of LO phonon modes in bulk ^{70}Ge (9.16 THz) and ^{74}Ge (8.94 THz) [8,9]. The frequencies for $n = 32$ are in agreement with those for the bulk modes. A peak for bulk ^{70}Ge due to the buffer layer appears clearly for the $n = 4$ sample. The arrows denote confined LO phonon modes in ^{70}Ge and ^{74}Ge layers, as observed previously by Raman scattering [6]. As n increases, the intensity of the confined mode in ^{74}Ge layers is enhanced as compared with that in ^{70}Ge layers, and the confined mode in ^{70}Ge layers shifts to higher frequency. Higher-order confined LO phonons from ^{70}Ge layers as marked by asterisks, denoted here as $LO_m(^{70}Ge)$ with m being the order, are observed in the spectra for $n = 16$ and 32 between ^{70}Ge and ^{74}Ge confined LO phonon modes [6]. Raman spectra observed for $\lambda = 488$ nm are shown in Fig. 2(b) for comparison. The Raman spectra are almost the same as the FT spectra in Fig. 2(a) in the sense that the frequency and intensity ratio of the confined LO modes are extremely similar to each other.

In order to see the frequency variation of the confined LO phonons in more detail, the peaks in the FT spectra were decomposed into multiple Lorentzian components, and the constituent peak frequencies were compared with a calculation based on a linear chain model [10] using appropriate frequencies and masses for the Ge isotopes [8,9]. In Fig. 3, the LO phonon frequencies are plotted against n by filled circles. The solid and broken lines denote calculations for LO_m (m = odd) in ^{70}Ge and ^{74}Ge layers, respectively. Although pronounced anticrossings between different branches of optical modes occur at around $n = 12$, 20, and 30 [6], this effect is not included in our calculation. The confined LO branches with even m are omitted here because a Raman intensity calculation based on the linear chain model combined with the bond polarizability model shows that their contribution can be well neglected. The calculation shows that at $n = 16$, for example, the three confined mode frequencies observed agree fairly well with those for LO_1, LO_3 (for

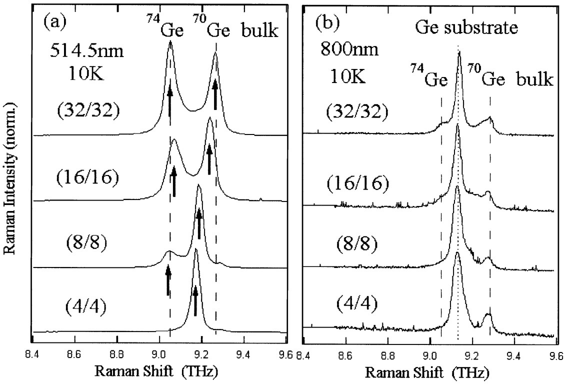

Fig. 4. Raman spectra of isotopic Ge SLs at $T = 10$ K excited with (a) 514.5 nm and (b) 800 nm. The broken lines indicate the frequencies of LO phonon mode in bulk ^{70}Ge and ^{74}Ge. The dotted line shows for frequencies of LO phonon mode in bulk Ge substrate.

^{70}Ge, with arrow) and LO_1 (for ^{74}Ge). The calculation also shows that at $n = 32$ the modes at 9.03 and 9.13 THz (with arrows) correspond to LO_3 and LO_5 for ^{70}Ge, respectively.

Spitzer et al. [6] observed Raman spectra of isotopic Ge SLs, and compared the result with the calculation based on the planar bond-charge model and the bond polarizability approach. They have found that the optic phonon modes in isotopic Ge SLs are confined in the two different constituent layers. Since the Raman intensity depends on the atomic displacements, the resemblance of Raman and FT spectra indicates that coherent phonons are also confined in ^{70}Ge or ^{74}Ge layers of the isotopic SLs, and that the amplitude of the coherent phonon is governed by the displacement of atoms in a constituent layer.

The FT spectra with $\lambda = 800$ nm are similar to the Raman spectra with $\lambda = 488$ nm as described above, although the difference in the laser penetration depth δ into Ge is large between $\lambda = 800$ and 488 nm [11]. In order to clarify the dependence of spectra on δ, Raman measurements were performed at 10 K by using two different excitation wavelengths $\lambda = 514.5$ and 800 nm as shown in Fig. 4(a) and (b). The confined LO phonon modes appear clearly for $\lambda = 514.5$ nm, while for $\lambda = 800$ nm the spectra are dominated by the signals from the Ge substrate (9.13 THz) and the ^{70}Ge buffer layer (9.28 THz). This is explained by the difference of penetration depth: They are estimated as $\delta = 200$ and 17 nm for $\lambda = 800$ and 514.5 nm, respectively [11]. Thus, the Raman scattering with $\lambda = 800$ nm probes both the SLs and the substrate, while that with $\lambda = 514.5$ nm mainly probes the SLs. Surprisingly, the series of spectra of coherent phonons taken with $\lambda = 800$ nm (Fig. 2(a)) is similar to those of Raman spectra taken with a much shorter wavelength $\lambda = 514.5$ nm (Fig. 4(a)). This means that the pump–probe experiment with $\lambda = 800$ nm is sensitive to the region close to the surface only. Since the generation of the coherent phonons in Ge will require excitation of high density of carriers, the region where the coherent phonons are generated is restricted to the outer surface region. In order to understand why the pump–probe experiment is sensitive to the region close to the surface only, it is desirable to compare the coherent phonons in superlattices with various thicknesses.

In summary, we have observed coherent confined LO phonon oscillations in isotopic Ge SLs by the pump–probe technique at excitation wavelength of 800 nm. The Fourier-transformed spectra of the oscillations were compared directly with Raman spectra recorded at various excitation wavelengths. The Fourier-transformed spectra are found to be very similar to the Raman data taken with a shallow penetration depth of laser, i.e., the observed pump–probe signals derives from the near-surface region only. We clarified that the amplitude of the confined coherent LO modes is dominated by the atomic displacement of Ge atoms in the constituent layers.

References

[1] Y.-X. Tan, E.B. Gamble, K.A. Nelson, J. Chem. Phys. 83 (1985) 5391.
[2] T. Pfeifer, T. Dekorsy, W. Kütt, H. Kurz, Appl. Phys. A 55 (1992) 482.
[3] W.A. Kütt, W. Albrecht, H. Kurz, IEEE J. Quantum Electron. 28 (1992) 2434.
[4] K. Mizoguchi, K. Matsutani, M. Hase, S. Nakashima, M. Nakayama, Physica B 249–251 (1998) 887.
[5] A. Bartels, T. Dekorsy, H. Kurz, K. Köhler, Phys. Rev. Lett. 82 (1999) 1044.
[6] J. Spitzer, T. Ruf, M. Cardona, W. Dondl, R. Schoner, G. Abstreiter, E.E. Haller, Phys. Rev. Lett. 72 (1994) 1565.
[7] H.D. Fuchs, P. Molinàs-Mata, M. Cardona, Superlattices Microstruct. 13 (1993) 12661.
[8] H.D. Fuchs, C.H. Grein, C. Thomsen, M. Cardona, W.L. Hansen, E.E. Haller, K. Itoh, Phys. Rev. B 43 (1991) 4835.
[9] H.D. Fuchs, C.H. Grein, M. Cardona, W.L. Hansen, K. Itoh, E.E. Haller, Solid State Commun. 82 (1992) 225.
[10] B. Jusserand, M. Cardona, in: M. Cardona, G. Güntherodt (Eds.), Light Scattering in Solids V, Springer, Berlin, 1989, pp. 49–152.
[11] R.F. Potter, in: E.D. Palik (Ed.), Handbook of Optical Constants of Solids, Academic Press, New York, 1985, p. 465.