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Lattice instability and elastic dispersion due to the rattling motion in the type-I clathrate $Ba_8Ga_{16}Sn_{30}$

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To investigate the off-center rattling motion and its charge-carrier dependence in type-I clathrate compounds, we carried out ultrasonic measurements on type-I $Ba_8Ga_{16}Sn_{30}$ and a reference compound, $K_8Ga_8Sn_{38}$. We found elastic softening of C_{44} originating from a lattice instability due to the off-center rattling motion of Ba atom in $Ba_8Ga_{16}Sn_{30}$. Elastic softening below 1 K suggests that the lattice instability remains at very low temperatures. We also found ultrasonic dispersion which has no mode selectivity. No-mode-selective ultrasonic dispersion in $Ba_8Ga_{16}Sn_{30}$ would be caused by a strong electron-phonon coupling. No charge-carrier dependence is observed between n-type and p-type $Ba_8Ga_{16}Sn_{30}$. The significant softening on the bulk modulus in $Ba_8Ga_{16}Sn_{30}$ contrasts to the continuous hardening in $K_8Ga_8Sn_{38}$, indicating the central role of the rattling motion in the softening.

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I. INTRODUCTION

Cage compounds, such as filled skutterudites and intermetallic clathrates, have attracted much attention because their thermal conductivity is very low due to phonon scattering by the so-called rattling motion, which is the large-amplitude atomic motion of the guest atom accommodated in polyhedral cages.[1–3] These compounds are expected to be thermoelectric materials which effectively convert waste heat to electricity.

The type-I clathrate with cubic structure (space group $Pm\bar{3}n$) consists of polyhedral cages with 12 faces (small cage) and 14 faces (large cage).[4] The guest atoms are located at the 2a site and at the 6d site in the small cage and large cage, respectively. Ga atoms preferentially occupy framework 6c sites. In the type-I clathrate $A_8Ga_{16}Ge_{30}$ (A = Ba, Sr, and Eu), nuclear density of the guest atom determined in neutron-scattering experiments reveals that the guest atom in the large cage moves from the center to an off-center position with decreasing an atomic radius.[5] In $Eu_8Ga_{16}Ge_{30}$ (EGG), the guest atom in the large cage locates in four off-center sites from the cage center. The off-center rattling motion has also been proposed in the Mössbauer and microwave absorption experiments, the Raman scattering experiment, the nuclear magnetic resonance (NMR) experiment, and the study by Zerec *et al.*[6–9] In addition, the Raman scattering experiment revealed that the guest atom moves rotationally in plane perpendicular to the fourfold axis in $Sr_8Ga_{16}Ge_{30}$ (SGG), which also has the off-center rattling motion.[10]

Type-I $Ba_8Ga_{16}Sn_{30}$ (BGS) can have n-type or p-type charge carriers, depending on a slight change of the composition ratio between Ga and Sn atoms.[11] BGS shows semiconducting behavior due to very low density of charge carriers. The temperature T dependences of thermal conductivity κ of n-type and p-type BGSs are almost the same as that of glasses and amorphous compounds.[12] In x-ray diffraction and optical conductivity experiments, the guest potential well of a Ba atom in a large cage indicates that the Ba atom locates in four sites 0.43 Å away from the cage center, suggesting the off-center rattling motion of Ba atom.[13,14] From the Raman scattering experiment, the rotational motion of the Ba atom among off-center positions is also reported in n-type and p-type BGSs.[15] On the other hand, type-I clathrate $K_8Ga_8Sn_{38}$ (KGS) with n-type carriers has Ga-Sn cages similar to those of BGS. The κ of KGS shows the characteristic behavior of a crystalline solid, and the K atom in the large cage is located at the on-center position.[16] KGS is a reference compound for n-type and p-type BGSs.

Here, depending on crystal growth conditions, BGS is known to also form the type-VIII cubic structure (space group $I\bar{4}3m$) consisting of polyhedral cages with 8 and 12 faces.[11,12] BGS undergoes no structural transition and keeps the formed structure down to low temperatures. The type-VIII BGS shows a crystalline solidlike behavior in the T dependence of κ.[12,17] In our previous work on the type-VIII BGS, we found monotonic hardening in the elastic modulus C_{44} with very weak ultrasonic frequency dependence.[18] On the other hand, the softening of C_{44} is observed in $Sr_8Ga_{16}Si_{30-x}Ge_x$ (SGSG), and the amount of C_{44} softening depends on the tendency of the off-center motion of the Sr atom.[19] These results suggest that the type-VIII BGS has on-center rattling guests.

In ultrasonic measurements, ultrasonic frequency dependence on elastic moduli and ultrasonic attenuation, which is the so-called ultrasonic dispersion (UD), is observed at low temperatures in cage compounds, such as the filled skutterudites ROs_4Sb_{12} and RFe_4Sb_{12}[20–25] and the clathrate-like compound $R_3Pd_{20}Ge_6$ (R: rare earth).[26,27] The temperature at which UD appears corresponds to the energy scale of the low-lying optical phonon mode determined by the Raman scattering experiment.[7] In these cage compounds, UD was believed to originate from the rattling motion. These compounds also show elastic softening at very low temperatures due to the rattling motion. In the present work, we measured T dependences of elastic moduli and ultrasonic attenuation at various ultrasonic frequencies in n-type BGS, p-type BGS, and the reference

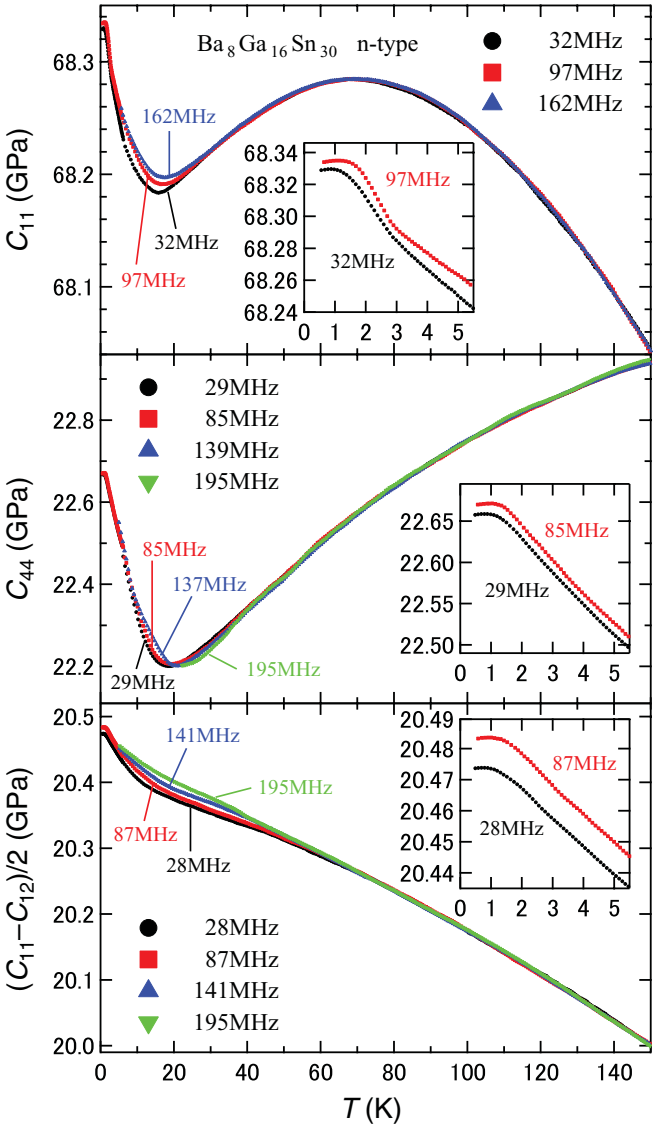

FIG. 1. (Color online) T dependences of elastic moduli C_{11}, C_{44}, and $(C_{11} - C_{12})/2$ in n-type BGS. The insets represent the same data in an expanded scale below 5.5 K.

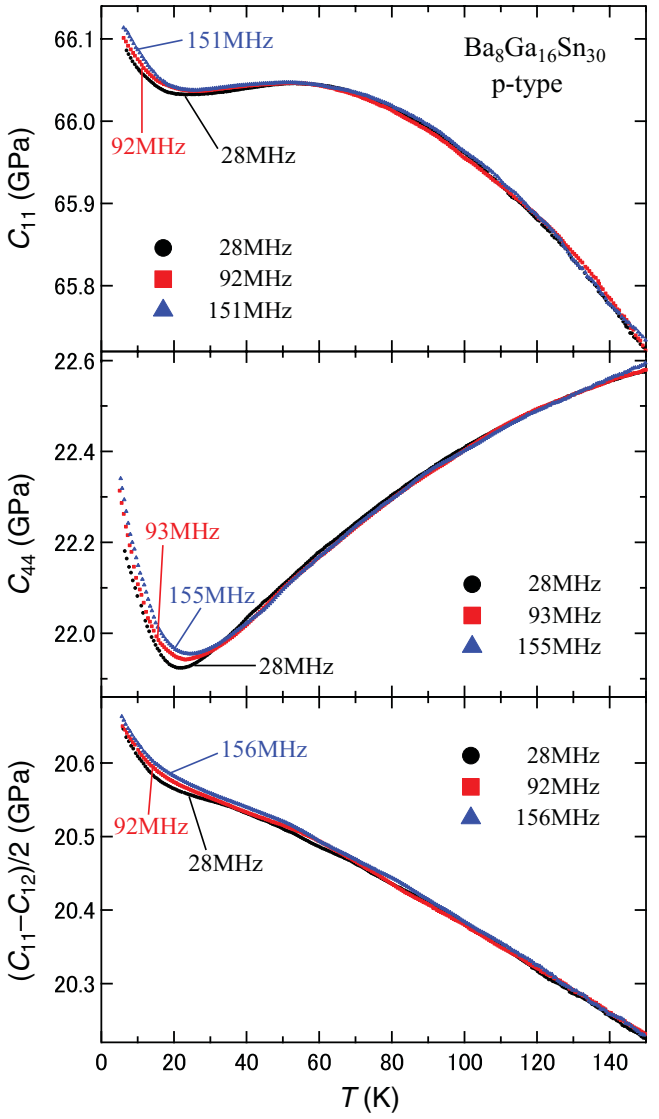

FIG. 2. (Color online) T dependences of elastic moduli C_{11}, C_{44}, and $(C_{11} - C_{12})/2$ in p-type BGS.

compound KGS to investigate the off-center rattling motion and its charge-carrier dependence.

II. EXPERIMENTAL

Single crystals of n-type BGS, p-type BGS, and KGS were grown by a self-flux method. Details of the sample preparation are described elsewhere.[12,13,16] The elastic moduli C_{11}, C_{44}, and $(C_{11} - C_{12})/2$ were measured as a function of T from 0.4 to 150 K for n-type BGS and from 4.2 to 150 K for p-type BGS and KGS using the phase-comparison pulse-echo method.[28] The elastic modulus C was calculated using $C = \rho v^2$ with room-temperature mass densities $\rho = 6.011$ g/cm^3, 5.977 g/cm^3, and 5.250 g/cm^3 for n-type BGS, p-type BGS, and KGS, respectively, where v is the sound velocity in a sample. The absolute value of v is estimated at 4.2 K by using the relation $v = 2l/t$, where l is a sample length and t is a time interval between pulse echoes. Ultrasonic attenuation α was measured as a function of T from 4.2 to 100 K using the attenuation recorder MATEC 2470B for n-type BGS. To measure the ultrasonic frequency dependence, we used the overtone of the fundamental resonance frequencies of ~30 MHz of $LiNbO_3$ transducers. The frequency range was between 24 and 220 MHz.

III. RESULTS

A. Elastic moduli in BGS

Figures 1 and 2 show T dependences of elastic moduli C_{11}, C_{44}, and $(C_{11} - C_{12})/2$ at various ultrasonic frequencies in n-type and p-type BGSs, respectively.

The longitudinal modulus C_{11} increases monotonically with decreasing T at high temperatures in both BGSs. The C_{11} saturates gradually below 70 K and turns into decrease below 60 K. The magnitude of elastic softening in the n-type BGS is larger than that in the p-type BGS. At around 20 K, C_{11} shows an upturn, and the temperature where C_{11} shows the upturn

increases with increasing ultrasonic frequency, suggesting UD in both BGSs. The C_{11} levels off below 2 K and shows slight elastic softening below 1 K, as shown in the inset of Fig. 1.

Remarkable elastic softening of the transverse modulus C_{44} is observed from high temperatures in both type-I BGS in contrast to the monotonic hardening in the type-VIII BGS reported previously.[18] This fact demonstrates that there exists substantial structure dependence of C_{44} softening between type-I and type-VIII structures. The C_{44} also shows UD accompanied by large ultrasonic attenuation at the same T range where UD is observed in C_{11}. In n-type BGS, we failed to measure C_{44} below 22 K for 195 MHz due to huge ultrasonic attenuation. The slight softening below 1 K is also observed.

The transverse modulus $(C_{11} - C_{12})/2$ increases monotonically with decreasing T in both BGSs. The UD accompanied by large ultrasonic attenuation and the slight softening below 1 K are also observed in $(C_{11} - C_{12})/2$. We found UD with no mode selectivity of ultrasound in the type-I clathrate $Ba_8Ga_{16}Sn_{30}$ for the first time. The temperature where UD appears is consistent with the rattling energy of the guest mode by the Raman scattering experiment.[15] On the other hand, no qualitative difference is observed between n-type and p-type BGSs. The lack of charge-carrier dependence in BGS is also reported by Suekuni *et al.*,[12,15] though $Ba_8Ga_{16}Ge_{30}$ (BGG) shows clear charge-carrier dependence (for instance, the Raman scattering and thermal conductivity experiments).[29,30]

The softening of elastic moduli continues down to 0.4 K. We also measured T dependences of C_{44} and $(C_{11} - C_{12})/2$ below 6 K in magnetic fields up to 12 T with a superconducting magnet. The application of magnetic fields does not affect this softening at all, indicating that the softening does not originate from a magnetic origin.

B. Elastic moduli in KGS

Figure 3 shows T dependences of elastic moduli C_{11}, C_{44}, and $(C_{11} - C_{12})/2$ at various ultrasonic frequencies in KGS.

For easier viewing of each data curve, the data for the overtone are increased with a constant value. All moduli increase with decreasing T. The C_{11} and C_{44} saturate gradually below 20 K. No frequency dependence is observed in KGS in contrast to n-type and p-type BGSs with clear UD.

C. Ultrasonic attenuation in BGS

Both n-type and p-type BGSs show a change in ultrasonic attenuation at the same T range where UD is observed in elastic moduli. Figures 4(a) and 4(b) show T dependences of ultrasonic attenuation α corresponding to C_{44} and $(C_{11} - C_{12})/2$ modes, respectively, at various ultrasonic frequencies in n-type BGS.

The α for the fundamental resonance frequency increases below 20 K and peaks at around 8 K. The temperature and magnitude of the attenuation peak increase with increasing ultrasonic frequency. These results also indicate no-mode-selective UD in BGS.

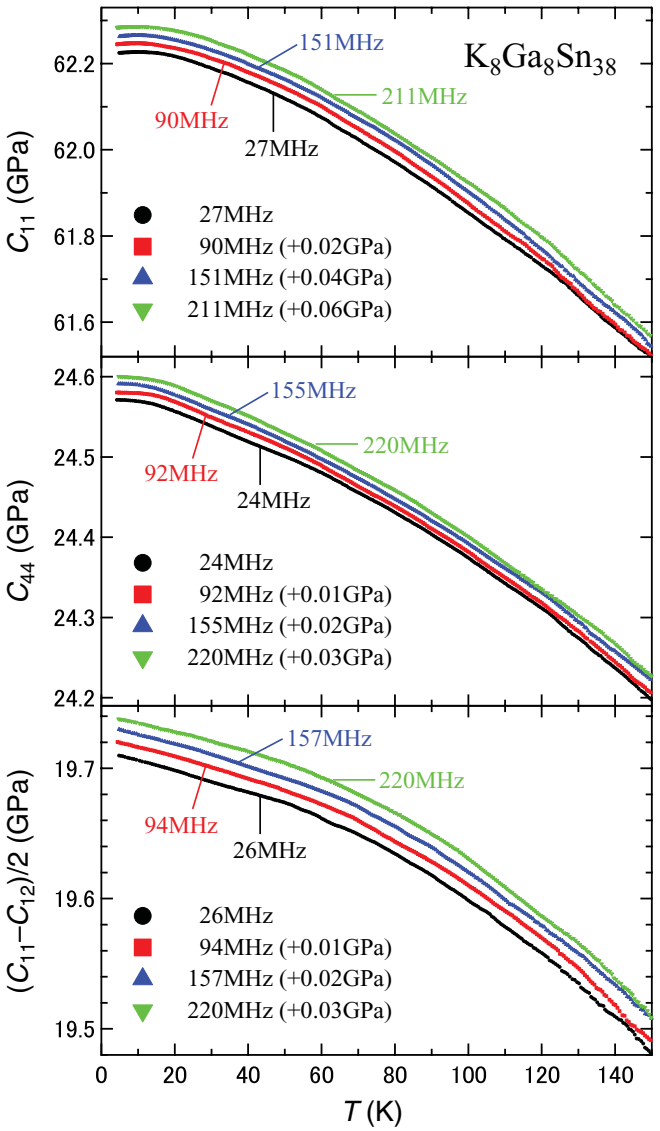

FIG. 3. (Color online) T dependences of elastic moduli C_{11}, C_{44}, and $(C_{11} - C_{12})/2$ in KGS. Because the data for overtone show an identical T dependence to the fundamental one, we plotted the data by adding a constant value so that each data curve could be easily seen.

IV. DISCUSSION

A. Elastic softening of C_{44}

The transverse modulus C_{44} shows the largest elastic softening from high temperatures in n-type and p-type BGSs. The C_{44} is the linear response to a strain with the irreducible representation Γ_5 or T_{2g} in the cubic symmetry. In contrast, monotonic elastic hardening is observed in $(C_{11} - C_{12})/2$, which is the linear response to a stain with the irreducible representation Γ_3 or E_g. On the other hand, no elastic softening is observed in KGS. The Ba atom in the large cage is located off-center in four sites from the cage center and moves rotationally among off-center positions in BGS, in contrast to KGS in which the K atom locates at the on-center position.[13,15,16] In our previous study on the elastic moduli of SGSG, we found a clear correlation between the off-center motion of Sr atom and the softening of C_{44}.[19] These results

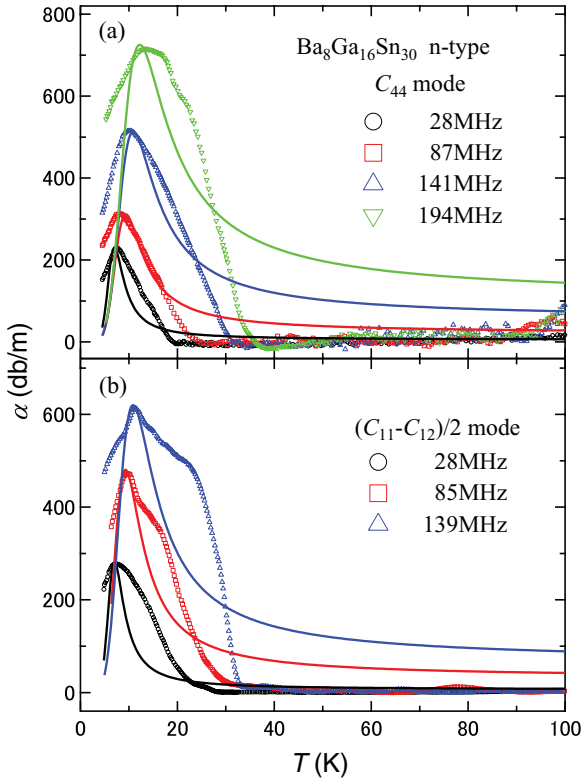

FIG. 4. (Color online) T dependences of ultrasonic attenuation α corresponding to (a) C_{44} and (b) $(C_{11} - C_{12})/2$ modes in n-type BGS. Solid curves in (a) and (b) are calculated results with the Debye-type ultrasonic attenuation.

indicate that the softening of C_{44} originates from the off-center rattling motion of Ba atom in BGS.

The Raman scattering experiment revealed that the energy of the guest atom's vibration mode softens with decreasing T, and its difference between the modes with T_{2g} and E_g emerges in a compound which has the off-center rattling motion.[10,15] The T_{2g} vibration energy is lower than the E_g vibration energy. The guest atom in the large cage moves rotationally among off-center positions. If the guest atom stops at one of the minimum positions in the off-center potential, the crystal symmetry should be lowered. Since the T_{2g} vibration energy is the lowest, a lattice instability corresponding to the T_{2g} strain is expected in a compound which has the off-center rattling motion. In n-type and p-type BGSs, the T_{2g} vibration energy softens with decreasing T and is the lowest in the Raman scattering experiment.[15] Thus, both BGSs show the softening of C_{44} due to the lattice instability corresponding to the T_{2g} strain.

In SGG, the energy of the T_{2g} mode levels off and merges onto the energy of the E_g mode at low temperatures.[10] The softening of C_{44} stops at the same T range in SGG. We pointed out that the guest atom at the off-center position tends to be restored to the on-center position at low temperatures, and thus the lattice instability disappears in SGG.[19] In contrast, the energy of the T_{2g} mode shows no leveling off and continues to soften with decreasing T in n-type and p-type BGSs.[15] This result suggests that the guest atom in the large cage locates in the off-center positions down to low temperatures.

TABLE I. Fitting parameters τ_0 ($\times 10^{-11}$ s) and E (K) and the ratio τ_0^{-1}/E ($\times 10^{-2}$) in BGS and the filled skutterudites $LaFe_4Sb_{12}$ and $LaOs_4Sb_{12}$.[21,23]

	BGS	$LaFe_4Sb_{12}$	$LaOs_4Sb_{12}$
τ_0	6.0	0.2	5.0
E	32	350	127
τ_0^{-1}/E	2.50	6.86	0.76

The C_{44} continues to soften down to 0.4 K except for the T range of UD, indicating that the lattice instability remains at very low temperatures in BGS. On the other hand, C_{11} and $(C_{11} - C_{12})/2$ also show slight elastic softening below 1 K. Since the off-center rattling motion over the guest potential well dies out at very low temperatures, it may turn into a quantum tunneling state of the guest atom through the potential well. To investigate the origin of the softening below 1 K, further ultrasonic experiments below 0.4 K are requisite.

B. Ultrasonic dispersion

Both n-type and p-type BGSs show no-mode-selective UD in elastic moduli and α. To obtain a time scale of the rattling motion, we carried out the theoretical fitting for only α corresponding to the C_{44} and $(C_{11} - C_{12})/2$ modes because a background stiffness of elastic modulus C_{44} is unclear due to the softening. We assumed the Debye-type relaxation for α:

$$\alpha = A\frac{\omega^2\tau}{1+\omega^2\tau^2}, \tag{1}$$

where A is a scaling factor, τ is the relaxation time, and ω is the angular frequency of the ultrasonic wave. We applied the Arrhenius-type relation to τ:

$$\tau = \tau_0 \exp(E/T), \tag{2}$$

where E is the activation energy. Solid curves in Fig. 4 are calculated results using Eqs. (1) and (2), where A is assumed to be a constant. The α at various ultrasonic frequencies is qualitatively reproduced by common values of $\tau_0 = 6.0 \times 10^{-11}$ s and $E = 32$ K for both modes. The values of τ_0 and E in BGS and well-known filled skutterudite compounds are listed in Table I.

Hereafter, we discuss the origin of no-mode-selective UD in BGS from the viewpoints of the thermal-relaxation process of Ba atom over the guest potential well and an electron-phonon interaction. As mentioned in Sec. I, Ba atom in the large cage locates in off-center four sites from the cage center and moves rotationally among off-center positions in BGS.[15] Since UD in BGS is reproduced by the Debye-type relaxation with the Arrhenius-type relation, it may originate from thermal-relaxation process of Ba atom over the guest potential well. However, in type-I EGG, which also has the off-center rattling motion, no obvious UD is observed in our preliminary ultrasonic measurements.

On the other hand, no-mode-selective UD is also observed in the filled skutterudite RFe_4Sb_{12},[23–25] though the guest atom locates at the cage center at low temperatures in filled skutterudite compounds.[31] In inelastic x-ray and neutron

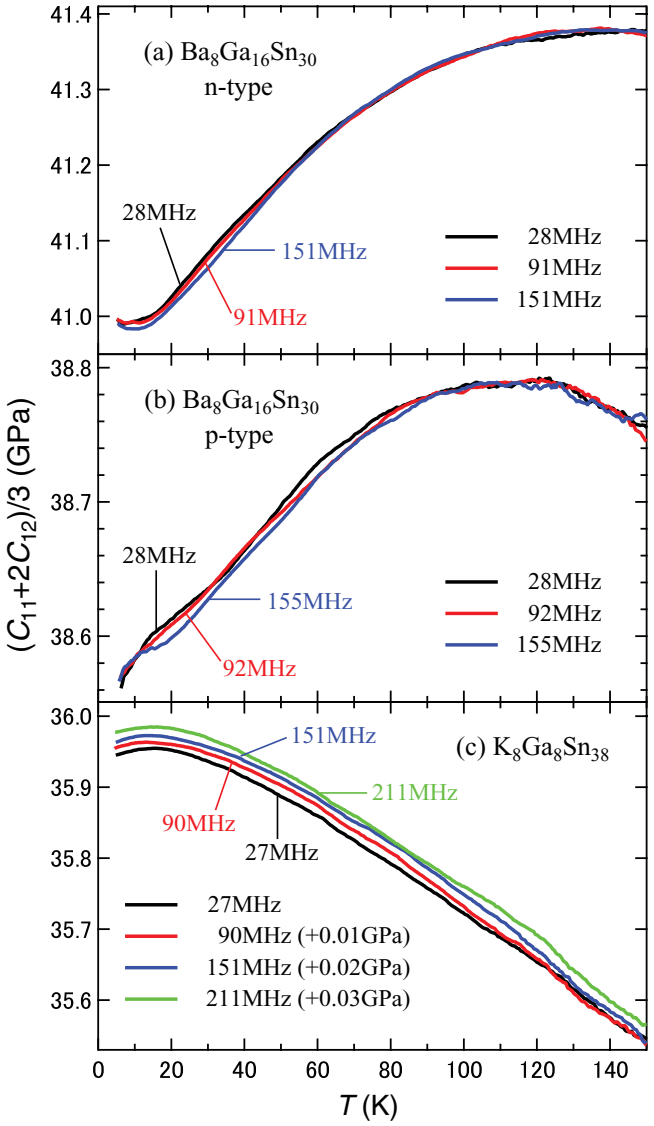

FIG. 5. (Color online) T dependences of bulk modulus $C_B = (C_{11} + 2C_{12})/3$ in (a) n-type BGS, (b) p-type BGS, and (c) KGS. The data of KGS for the overtone are increased with a constant value.

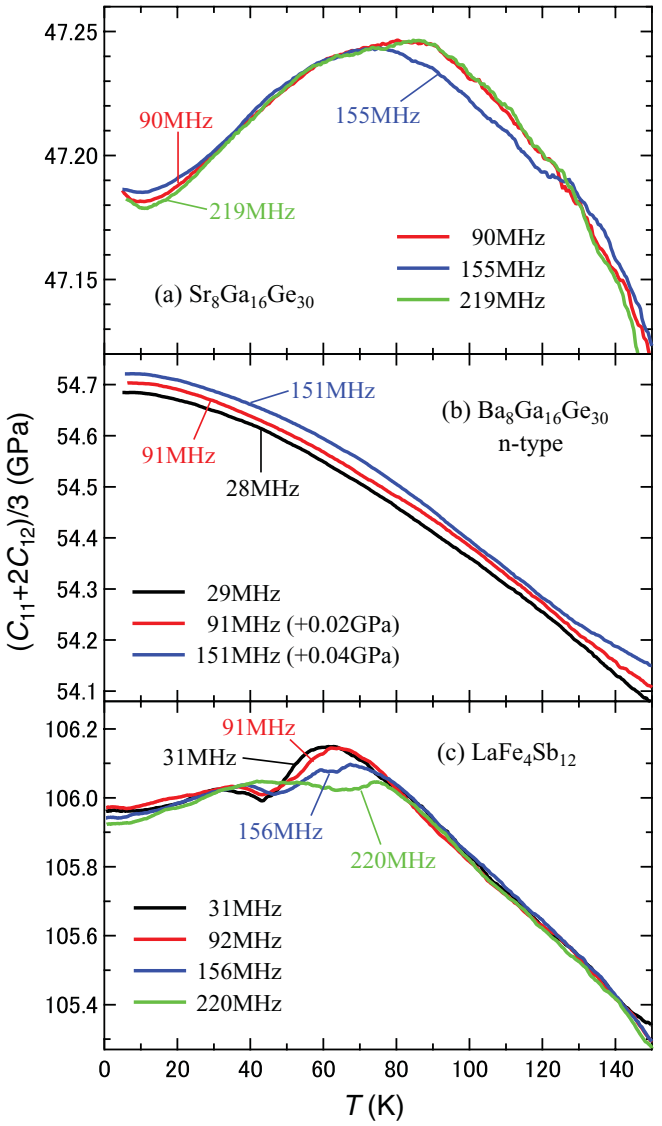

FIG. 6. (Color online) T dependences of C_B in (a) SGG, (b) n-type BGG, and (c) the filled skutterudite $LaFe_4Sb_{12}$. The data of n-type BGG for the overtone are increased with a constant value.

scattering experiments, a low-lying flat optical phonon branch due to the rattling motion exists in the phonon dispersion curves of cage compounds.[32,33] From the results of ultrasonic measurements and band calculation of $LaFe_4Sb_{12}$ using the theory by Hattori and Miyake,[34] we pointed out that no-mode-selective UD is caused by the coupling between an acoustic phonon and low-lying optical phonon interacting with electrons. The tendency of UD to appear is directly related to the strength of electron-phonon coupling.[23,34] We assessed an index of the strength of electron-phonon coupling constant, g, in BGS, $LaFe_4Sb_{12}$, and $LaOs_4Sb_{12}$ from the ratio τ_0^{-1}/E: g increases as τ_0^{-1}/E increases. As listed in Table I, the ratio τ_0^{-1}/E of BGS is the same order as that of $LaFe_4Sb_{12}$ (no-mode-selective UD) and is quite larger than that of $LaOs_4Sb_{12}$ (mode-selective UD). Here, the electron-phonon coupling is proportional to $g^2 N_F$ in the theory, where N_F is the number of electronic density of states at E_F.[23,34] Although N_F is small in BGS by the band calculation based on the full-potential linearized augmented plane wave (FLAPW) method,[35] no-mode-selective UD in BGS would be caused by a larger electron-phonon coupling constant.

C. Softening of bulk modulus

The modulus C_{11} shows elastic softening below 60 K in contrast to $(C_{11} - C_{12})/2$ without softening in n-type and p-type BGSs. From these results, elastic anomaly is expected in bulk modulus $C_B = (C_{11} + 2C_{12})/3$ which increases monotonically with decreasing T in general. We estimated C_B using experimental data of C_{11} and $(C_{11} - C_{12})/2$ for n-type BGS, p-type BGS, and KGS, as shown in Fig. 5.

The C_B in n-type and p-type BGSs show substantial softening below ~120 K, in contrast to KGS, which does not show obvious softening. For other examples of C_B, C_B in

SGG, *n*-type BGG, and the filled skutterudite $LaFe_4Sb_{12}$ are shown in Figs. 6(a), 6(b), and 6(c), respectively.

The softening of C_B is observed below 80 K in SGG, in contrast to *n*-type BGG, which does not show softening. In *n*-type BGG, the Ba atom in the large cage locates at the on-center position, and a crystalline solidlike behavior is observed in the T dependence of κ.[5,10,30] On the other hand, C_B shows slight softening below 40 K in $LaFe_4Sb_{12}$, as shown in Fig. 6(c). $LaFe_4Sb_{12}$ has the rattling motion, and the isomorphic compound $LaFe_3CoSb_{12}$ shows a glasslike behavior in the T dependence of κ.[1,3] Kaneko *et al.* reported that the guest atom locates at the cage center at low temperatures in the filled skutterudite compounds during their neutron diffraction experiments.[31] We found that the softening of C_B tends to appear in the compounds which have the rattling motion; however, off-center rattling of the guest is not necessarily required. The rattling motion plays a central role for the softening of C_B.

We point out a possible origin for the softening of C_B from the viewpoint of anharmonicity of the guest atom. In the theoretical study by Yamakage and Kuramoto, cage atoms feel anharmonicity by an interaction between the guest atom and cage atoms, though an interaction among cage atoms is harmonic in cage compounds.[36] The sound velocity v decreases with decreasing T due to the anharmonicity of the guest atom, which affects cage atoms indirectly, in the theory. As described in Sec. II, the elastic modulus is proportional to v^2. The softening of C_B may originate from the anharmonicity of the guest atom.

V. CONCLUSION

We performed ultrasonic experiments in the type-I clathrate BGS with off-center rattling guests and KGS with on-center rattling guests. The elastic softening of C_{44} is observed in *n*-type and *p*-type BGSs in contrast to the hardening in KGS, indicating that the softening originates from the lattice instability by the off-center rattling motion. Since the moduli continue to soften below 1 K, the lattice instability remains down to very low temperatures. We clarified no-mode-selective UD in BGS originating from the strong electron-phonon coupling. The lack of charge-carrier dependence in BGS, shown by the results of the Raman scattering and thermal conductivity experiments, is observed in contrast to clear charge-carrier dependence in BGG. We also found an anomalous softening of C_B due to the rattling motion in the cage compounds.

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