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Sm atomic dynamics in a charge density wave compound $SmNiC_2$

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Abstract ^{149}Sm nuclear resonant inelastic scattering was carried out in a charge density wave compound $SmNiC_2$. We have investigated temperature dependences of the Sm partial phonon density of states and recoil-free fraction at the Sm site and the average sound velocity estimated from the Sm partial density of states. The Sm partial density of states exhibits temperature dependence, suggesting that the phonon modes between 20 and 25 meV may correlate with the charge density wave. Temperature dependence of the recoil-free fraction is difficult to prove the correlation with either the charge density wave or ferromagnetic ordering. The average sound velocity obtained by the Sm partial phonon density of states exhibits temperature dependence, agreeing qualitatively with very recent elastic constant measurements.

Keywords Nuclear resonant inelastic scattering · Charge density wave · Ferromagnetism · Recoil-free fraction · Average sound velocity

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1 Introduction

Charge density wave (CDW) is one of the interesting phenomena in solid state physics. When compounds exhibit a CDW transition [1], a long-period structural transition can be observed below the transition temperature. This transition reflects by a structural instability accompanied with Fermi surface nesting. Whereas crystallographic investigation provides knowledge of structural modulation with a long-period atomic ordering, investigation of phonons in CDW compounds provides nature of structural instability such as dimensionality of a CDW transition, identification of atomic motions correlated with the CDW transition and so on. If another interaction competes with a CDW transition, what happens in structural instability connected with the transition? This is one of the motivations in the present study.

$SmNiC_2$ has an orthorhombic structure with the space group of $Amm2$. Electrical resistivity in $SmNiC_2$ exhibits a kink below 150 K, suggesting a CDW transition, and a discontinuous change at about 17.7 K with a hysteresis, suggesting a first-order transition [2]. Electrical resistivity drops at about 17.7 K with decreasing temperature. The transition suggested by electrical resistivity is caused by a ferromagnetic ordering (FMO) [3]. The FMO was also confirmed by X-ray magnetic circular dichroism (XMCD) experiment using Sm $L_{2,3}$-edges and Ni K-edge, which is a useful tool to investigate ferromagnetism using synchrotron radiation X-ray [4]. In addition, structural investigation was carried out in the CDW and FMO phases of $SmNiC_2$ [5]. The results of the structural investigation clarified that superlattice reflections appeared only in the CDW phase, suggesting structural modulation with long-period atomic ordering, and these reflections disappeared in the FMO phase.

We investigated Sm atomic dynamics in $SmNiC_2$ by ^{149}Sm nuclear resonant inelastic scattering (NRIS) at several temperatures. We have found temperature dependences of NRIS spectra and the parameters obtained by the spectral analyses based on Lipkin's sum rule [6]. Temperature dependence of the Sm partial phonon density of states (Sm PDOS) implies that the optical modes around 23 meV correlates with both CDW and FMO transitions. Temperature dependence of the average sound velocity qualitatively agrees with the very recent results of the elastic constant measurements [7]. The temperature dependence of the recoil-free fraction obeys the Debye model as is applied in conventional Mössbauer spectroscopy.

2 Experimental procedure

^{149}Sm NRIS measurements were carried out at BL09XU of SPring-8 in Japan. ^{149}Sm NRIS spectra of $SmNiC_2$ were measured at several temperatures. Energy resolution for the NRIS measurements was 1.5 meV resolution at ^{149}Sm Mössbauer resonance energy of 22.502 keV [8, 9], which was obtained by Si(4 4 0)-Si(16 8 8) nested-type high resolution monochromator [10]. Measured sample was prepared by crushing a piece of single crystalline sample into powder. Sm partial phonon density of states (Sm PDOS) was extracted with the same method as was reported previously [11].

3 Experimental results and discussion

Sm PDOS obtained by ^{149}Sm nuclear resonant inelastic scattering (NRIS) measurements at 300 K is shown in Fig. 1. Inset of Fig. 1 shows temperature dependence of the Sm PDOS magnified between 20 and 25.5 meV. Peak energy of the Sm PDOS in this energy region

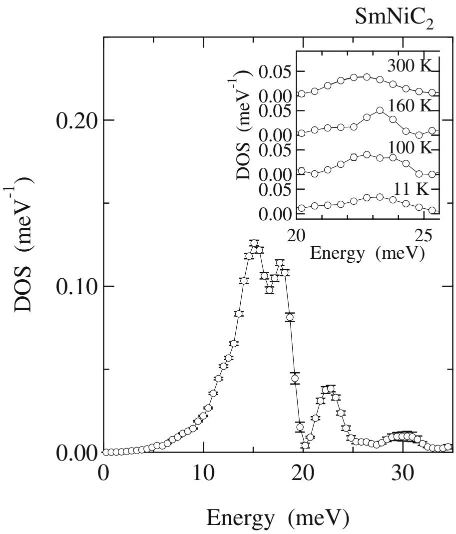

Fig. 1 Sm partial phonon density of states in $SmNiC_2$ at 300 K. The inset shows temperature dependence of the density of states between 20 and 25.5 meV

shifts in higher side towards T_{CDW} and seems to stay below T_{CDW} with decreasing temperature. This suggests that the phonon modes in this energy region correlate with the CDW transition, but do not with the FMO strongly.

We applied the present results to Linpin's sum rule to discuss more details [6]. This sum rule enables us to discuss element-specific thermodynamical properties under assumption of harmonic potential. To estimate the experimental error, we also estimate the same parameters obtained by the sum rule from the Sm PDOS. The content of the sum rule is represented as follows:

$$\int S^i(\epsilon)d\epsilon = 1 - f, \tag{1}$$

$$\int (\epsilon - E_R)S^i(\epsilon)d\epsilon = 0, \tag{2}$$

$$\int (\epsilon - E_R)^2 S^i(\epsilon)d\epsilon = 4E_R E_K, \tag{3}$$

$$\int (\epsilon - E_R)^3 S^i(\epsilon)d\epsilon = \frac{E_R}{m}\hbar^2 F_K, \tag{4}$$

where $S^i(\epsilon)$ denotes an inelastic part of a NRIS spectrum, f recoil-free fraction, E_R recoil energy, E_K kinetic energy, and F_K average force constant. m denotes mass of a probe nucleus and $\hbar$ is the reduced Planck constant. Among them, we discuss temperature dependence of the recoil-free fraction. The temperature dependence is shown in Fig. 2. This is reproduced very well with the Debye model assuming the Debye temperature θ_D of 275 K. This means that the recoil-free fraction is insensitive to neither the CDW nor FMO transition.

Structural instabilities related to the CDW and FMO phase are also suggested by another parameter. When the obtained PDOS $D(\epsilon)$ in such low energy region is proportionate to square of energy, the prove atoms in the NRIS measurements have evident contribution to the acoustic modes. Figure 3 shows the plots of the Sm PDOS against square of phonon

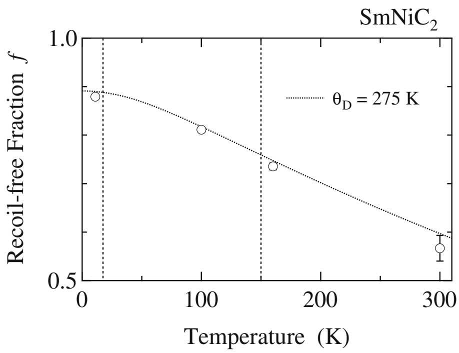

Fig. 2 Temperature dependence of the recoil-free fraction associated with Sm atoms in $SmNiC_2$. The open circles denote the datum points obtained by Lipkin's sum rule [6]

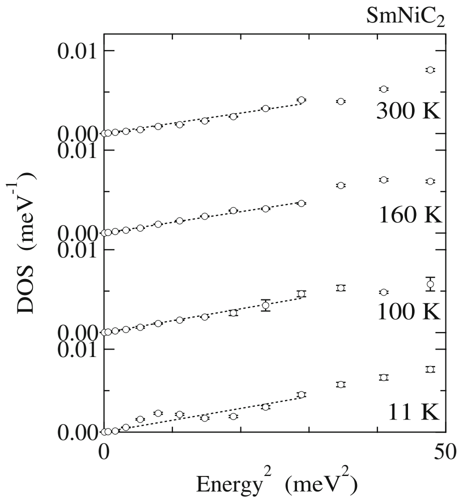

Fig. 3 Plots of the Sm PDOS against square of energy at various temperatures. The open circles denote the datum points of the Sm PDOS. The solid lines denote the fitting results

energy. The average sound velocity v_D in $SmNiC_2$ can be estimated from the slope of the Sm PDOS, because relationship between the Sm PDOS and v_D is given as

$$D(\epsilon) = \frac{m}{M} \frac{\epsilon^2}{2\pi^2 \hbar^3 \rho {v_D}^3}, \tag{5}$$

where M is the average nuclear mass and ρ is the density of number of atoms per unit volume in the measured sample [12]. Note that, concerning ρ, temperature variation of the unit cell volume was considered when the average sound velocity was estimated from the Sm PDOS [2]. Figure 4 shows temperature dependence of v_D in $SmNiC_2$. Average sound velocity at 100 K in the CDW phase is slower than that at 300 K. In addition, that at 11 K in the FMO phase is also slower than that at 100 K. These facts agree qualitatively with temperature dependence of elastic constants obtained by ultrasonic measurements, because the softening of the elastic constants was observed in more than one directions at both CDW and FMO transitions [7].

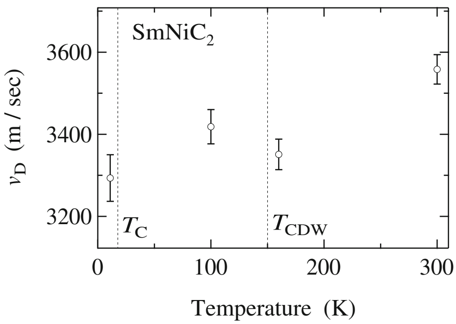

Fig. 4 Temperature dependence of average sound velocity v_D estimated from Sm PDOS

4 Summary

We carried out ^{149}Sm NRIS measurements of $SmNiC_2$ at various temperatures. Unlike investigation of phonon dispersion relations using inelastic neutron or X-ray scattering, the results of the NRIS measurements are difficult to discuss dimensionality of the CDW in $SmNiC_2$. However, analyses based on the sum-rule of the NRIS and the Sm PDOS provide the following results:

1) Temperature dependence of the optical mode between 20 and 25 meV may correlate with the CDW transition.
2) Temperature dependence of the recoil-free fraction is insensitive to the CDW transition as well as the FMO one.
3) Temperature dependence of average sound velocity agrees qualitatively with the results of sonic wave measurements.

These findings demonstrates that the Sm atomic motion plays an important role of successive transitions in $SmNiC_2$ in the viewpoint of phonons.

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