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# Nonmagnetic Phase Transition in $\text{La}_3\text{TiSb}_5$ Observed in $^{121}\text{Sb}$ Nuclear Quadrupole Resonance

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We measured the electrical resistivity and the  $^{121}\text{Sb}$  nuclear quadrupole resonance (NQR) spectra of  $\text{La}_3\text{TiSb}_5$ , which is a nonmagnetic reference system for a locally noncentrosymmetric antiferromagnet  $\text{Ce}_3\text{TiSb}_5$ , in order to determine its physical properties. The resistivity exhibited a hump anomaly at 120 K, suggesting a phase transition. The NQR spectra split below 110 K, whereas the nuclear spin-lattice relaxation exhibited no anomalies. These results can be explained by the incommensurate charge-density-wave (CDW) transition. This finding suggests the importance of quasi-one-dimensional characteristics and CDW instability in these systems.

Locally noncentrosymmetric systems have received considerable attention because various unusual phenomena can be realized, for example, exotic superconductivity[1, 2] and cross-correlation responses[3, 4]. Cross-correlation responses are expected to occur when odd-parity multipoles emerge in conjunction with the magnetic transitions[3, 4]. The magnetoelectric effect, that is, current-induced magnetization, is an example of an unusual response and is confirmed for two antiferromagnetic metals  $\text{UNi}_4\text{B}$ [5] and  $\text{Ce}_3\text{TiBi}_5$  [6, 7]. The origin of the magnetoelectric effect in these systems remains under active discussion. Understanding the magnetic structure is crucial to elucidate its mechanism.

Antiferromagnet  $\text{Ce}_3\text{TiBi}_5$  belongs to the series of hexagonal  $\text{RE}_3\text{TiX}_5$  systems (RE: rare-earth element; X = Sb, Bi) and these materials crystallize in a hexagonal  $P6_3/mcm$  (No. 193,  $D_{6h}^3$ ) space group symmetry[8, 9]. Rare-earth atoms form a zigzag chain along the  $c$  axis possessing no inversion symmetry.  $\text{Ce}_3\text{TiBi}_5$  exhibits a cycloid order with moments lying in the plane of the zigzag chain below  $T_N = 5.0$  K[10]. By contrast,  $\text{Ce}_3\text{TiSb}_5$  exhibits an antiferromagnetic transition at  $T_N = 5.2$  K with ordered moments along the  $c$  axis with long-period modulation[11]. Another remarkable feature of  $\text{RE}_3\text{TiX}_5$  systems is the quasi-one-dimensional chain along the  $c$  axis. This is common in the nonmagnetic reference system  $\text{La}_3\text{TiSb}_5$ , and Sb(1) and the Ti atoms are aligned along the  $c$  axis, as shown in Fig. 1. This could lead to the instability of the Fermi surface, resulting in charge-density-wave (CDW) or spin-density-wave transitions. Among the  $\text{RE}_3\text{TiX}_5$  systems,  $\text{La}_3\text{TiSb}_5$  exhibits an electrical resistivity anomaly of unknown origin at approximately 120 K[12]. Some phase transition is expected in  $\text{La}_3\text{TiSb}_5$ .

In this letter, we report the results of the electrical resistivity and  $^{121}\text{Sb}$  nuclear quadrupole resonance (NQR) measurements on  $\text{La}_3\text{TiSb}_5$  to reveal the anomalies at lower temperatures. The resistivity exhibited a hump anomaly at 120 K. The NQR spectrum began to split below the transition temperature of  $\sim 110$  K. This split was explained using a simple model that assumed spatially modulated electric field gradient

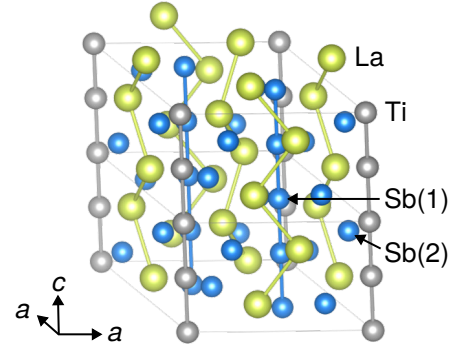


FIG. 1. (Color online) Crystal structure of  $\text{La}_3\text{TiSb}_5$  with a hexagonal  $P6_3/mcm$  space group symmetry, drawn by VESTA[13]. Two unit cells are stacked along the  $c$  axis. The lattice constants are from literature[8]. Two nonequivalent Sb sites exist and are denoted as Sb(1) for  $4d$  (3.2) position and Sb(2) for  $6g$  ( $m2m$ ). La and Sb sites lack the local inversion symmetry, whereas the Ti site is located on the inversion center.

(EFG) because of the incommensurate CDW transition. No clear anomaly was observed in the nuclear spin-lattice relaxation rate  $1/T_1$ . This finding highlights the importance of the quasi-one-dimensional character of these systems.

The single-crystalline  $\text{La}_3\text{TiSb}_5$  sample was synthesized using the Sn-flux method, as in Ref. 14. La (99.9%), Ti (99.5%), Sb (99.99%), and Sn (99.999%) in molar ratios of La:Ti:Sb:Sn = 2–3:1:5:20–50 were placed in an alumina crucible and sealed in a quartz tube. The ingredients were heated to 1000°C, maintained at this temperature for 10 h, and subsequently cooled to 500°C at  $\sim -1.5^\circ\text{C/h}$ . Needle-shaped crystals were obtained. The excess Sn flux was removed by centrifugation. The sample was cleaned with dilute hydrochloric acid to remove residual Sn flux. Electrical resistivity measurements were performed on the single-crystal samples using the standard four-terminal method. The sample was crushed into a powder for NQR measurements to enhance the signal intensity. In addition, the powdered sample was sealed in epoxy

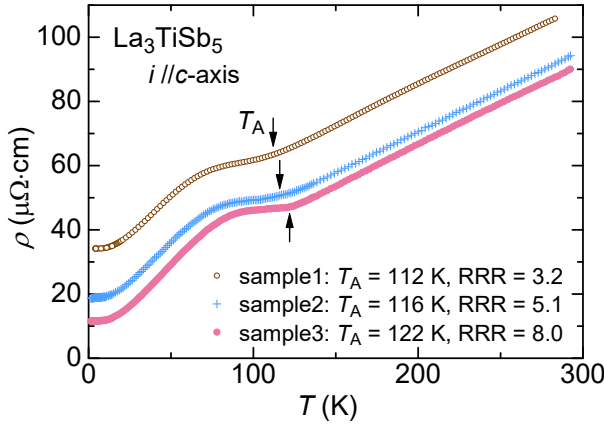


FIG. 2. (Color online) Temperature dependence on the electrical resistivity of single-crystalline  $\text{La}_3\text{TiSb}_5$  samples. The current was applied along the  $c$  axis. The hump anomaly is observed at approximately 120 K for the best sample. The vertical arrows indicate transition temperatures determined from the change in slope at the kink.

TABLE I. Quadrupole frequency  $\nu_Q$  and asymmetric parameter  $\eta$  from the band calculation and the  $^{121}\text{Sb}$  NQR experiment at the Sb(1) and Sb(2) sites in  $\text{La}_3\text{TiSb}_5$ .

|                                           | $\nu_Q$ (MHz) | $\eta$ |
|-------------------------------------------|---------------|--------|
| $^{121}\text{Sb}(1)$ (calc.)              | 66.7          | 0      |
| $^{121}\text{Sb}(2)$ (calc.)              | 15.6          | 0.4219 |
| $^{121}\text{Sb}(1)$ (NQR expr. at 300 K) | 65.3          | -      |
| $^{121}\text{Sb}(2)$ (NQR expr. at 5–7 K) | 15.9          | 0.38   |

(Stycast 1266) in a rod shape to prevent oxidization. The standard spin-echo method was used for NQR measurements. The frequency-swept spectra were obtained by summing multiple Fourier-transformed spin-echo signals at various frequencies.

Figure 2 shows the electrical resistivity of  $\text{La}_3\text{TiSb}_5$  samples along the  $c$  axis in the range of 2–290 K. A hump anomaly was observed at  $\sim 120$  K for the best-quality sample, which agrees with a previous study[12]. This suggests a reduction in the density of states (DOS) at the Fermi energy level. The samples used in this study did not exhibit zero resistivity because of the superconductivity of the Sn flux below 4 K[12, 14], although some samples exhibited a slight decrease at this temperature. The sample quality varied to some extent, and the residual resistivity ratio (RRR)  $\rho(300\text{ K})/\rho(2\text{ K})$  was between 3 and 8. Furthermore, higher RRR samples exhibited higher and sharper anomalies. Another sample from the same batch as the electrical resistivity was used for the NQR measurements.

A nucleus with  $I \geq 1$  possesses a nuclear quadrupole moment  $Q$ , and the nuclear spin Hamiltonian without a magnetic field is

$$\mathcal{H}_Q = \frac{h\nu_Q}{6} \left\{ [3I_z^2 - I(I+1)] + \frac{1}{2}\eta(I_+^2 + I_-^2) \right\}, \quad (1)$$

where  $\nu_Q$  denotes the nuclear quadrupole frequency and  $\eta$

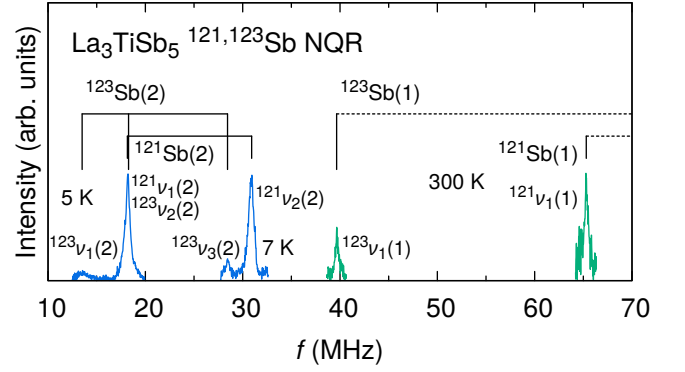


FIG. 3. (Color online)  $^{121}\text{Sb}$  and  $^{123}\text{Sb}$  NQR spectrum of  $\text{La}_3\text{TiSb}_5$ . The spectrum of Sb(1) is at 300 K, and that of the Sb(2) is at 5 K or 7 K. The expected  $^{123}\text{Sb}(2)$   $\nu_2$  line coincides with the  $^{121}\text{Sb}(2)$   $\nu_1$  line at  $\sim 18.3$  MHz. The signals of Sb(1) higher than 70 MHz were not measured in the present study. The spectra for the different frequency ranges were measured at different temperatures or with different NMR spectrometers, and the signal intensities are not comparable.

denotes the asymmetric parameter. These parameters are expressed by the local EFG  $V_{ij}$  at the nuclear site:  $\nu_Q = 3eQV_{zz}/[2I(2I-1)\hbar]$  and  $\eta = (V_{xx} - V_{yy})/V_{zz}$  when the principal axes of the EFG are selected along the  $x$ ,  $y$ , and  $z$  axes, such that  $|V_{zz}| \geq |V_{yy}| \geq |V_{xx}|$ . Two isotopes of Sb exist:  $^{121}\text{Sb}$  ( $I = 5/2$ ) and  $^{123}\text{Sb}$  ( $I = 7/2$ ). For  $^{121}\text{Sb}$ , two NQR transition lines  $\nu_1$  and  $\nu_2$  are expected for each site. The three-fold symmetry at the Sb(1) site ensures  $\eta = 0$ , which leads to simple resonant frequencies  $\nu_1 = \nu_Q$  and  $\nu_2 = 2\nu_Q$  for the  $\pm 1/2 \leftrightarrow \pm 3/2$  and  $\pm 3/2 \leftrightarrow \pm 5/2$  transitions. In contrast,  $\eta$  is nonzero for the Sb(2) site because of the lower symmetry, resulting in an  $\eta$ -dependent ratio between  $\nu_1$  and  $\nu_2$ . The  $^{123}\text{Sb}$  gives three lines per site, and the resonant frequencies are related to those of  $^{121}\text{Sb}$  because these nuclei are located at the same crystal site.

Figure 3 shows the  $^{121}\text{Sb}$  and  $^{123}\text{Sb}$  NQR spectra at the selected temperatures. The experimental  $\nu_Q$  value for  $^{121}\text{Sb}$  was determined from the  $\pm 1/2 \leftrightarrow \pm 3/2$  transition line for Sb(1) at  $^{121}\nu_1(1) = ^{121}\nu_Q(1) (\simeq 65.3\text{ MHz at } 300\text{ K})$ . Sb(2) parameters were evaluated based on the frequency of  $\nu_1(2)$  and  $\nu_2(2)$  lines ( $\sim 18$  and  $31\text{ MHz at } 5\text{--}7\text{ K}$ , respectively) using numerical diagonalization of the Hamiltonian in Eq. (1). Table I lists the quadrupole parameters of Sb(1) and Sb(2) for the  $^{121}\text{Sb}$  nucleus obtained through band calculation and the NQR experiment. The band calculation result is in good agreement with the experimental values. The ratio of  $\nu_Q$  between  $^{121}\text{Sb}$  and  $^{123}\text{Sb}$  nuclei was  $^{121}\nu_Q(1)/^{123}\nu_Q(1) = 1.64$  for Sb(1) site at 300 K, being consistent with the expected value from the quadrupole moments[15]:  $^{121}\{Q/[2I(2I-1)]\}/^{123}\{Q/[2I(2I-1)]\} = 1.65$ . The signal intensity of  $^{123}\text{Sb}$  is weaker than that of  $^{121}\text{Sb}$  due to the smaller gyromagnetic ratio and natural abundance, although it is unexpectedly weaker compared to the typical relative intensity with that of  $^{121}\text{Sb}$ . Hereafter, the NQR result for the  $^{121}\text{Sb}$  nucleus is shown.

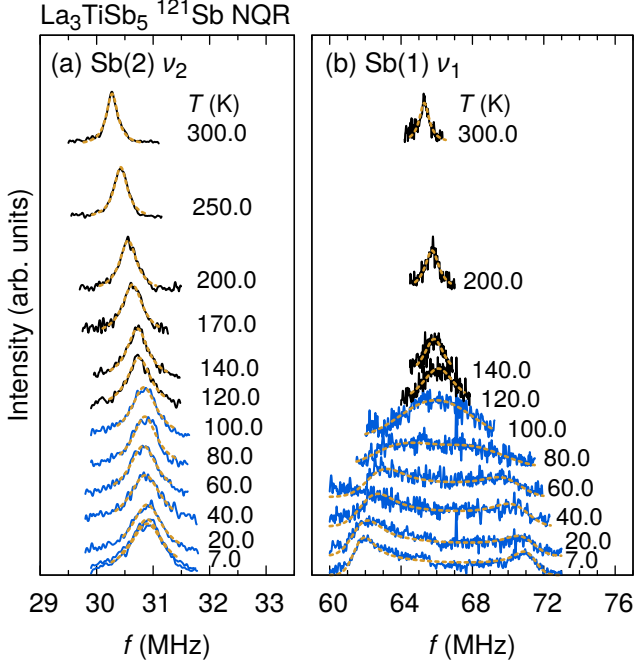


FIG. 4. (Color online) Temperature dependence of  $^{121}\text{Sb}$  NQR spectrum of (a) the  $^{121}\nu_2(2)$  transition at the Sb(2) site and (b)  $^{121}\nu_1(1)$  transition at the Sb(1) site in  $\text{La}_3\text{TiSb}_5$ . The spectra are shifted vertically. The dashed lines show fitting results with a Lorentzian function in the entire temperature range for (a) and at 120 K and above for (b); and a simulated spectral shape for the CDW state for 100 K and below for (b) (see text). The Lorentzian functions were obtained using the least-squares method, and the calculated spectra for the CDW state were selected manually to reproduce the experimental results.

Figure 4 shows the NQR spectra at (a) Sb(2)  $^{121}\nu_2(2)$  and (b) Sb(1)  $^{121}\nu_1(1)$  between 7–300 K. The NQR spectra at  $^{121}\nu_1(1)$  broadened below 100 K and split into two peaks at lower temperatures. This demonstrates a phase transition with symmetry reduction. In contrast, the spectra at the  $^{121}\nu_2(2)$  line showed a small anomaly at approximately 100 K. No clear splits were observed.

The site dependence of the NQR split can be explained by the difference in the local structure between two sites. Sb(1) site forms a quasi-one-dimensional chain along the  $c$  axis possessing the CDW instability, whereas Sb(2) forms a zigzag chain. The nuclear relaxation shown later suggests the non-magnetic transition. Therefore, the split peak in Fig. 4(b) was analyzed using a spatially modulated quadrupole frequency caused by the CDW transition

$$\nu_Q(x) = \nu_0 + W \sin qx, \quad (2)$$

where  $W$  and  $q$  denote the amplitude and wavenumber of the modulation, respectively. The nuclear position along the  $q$  vector is denoted by  $x$ . If the wavelength  $2\pi/q$  cannot be expressed as a rational multiple of the lattice constant,  $qx$  is uniformly distributed between zero and  $2\pi$ , which results in a continuous distribution of  $\nu_Q(x)$  between  $\nu_0 - W$  and  $\nu_0 + W$ . Then, the NQR intensity  $I(f)$  at frequency  $f$  is given

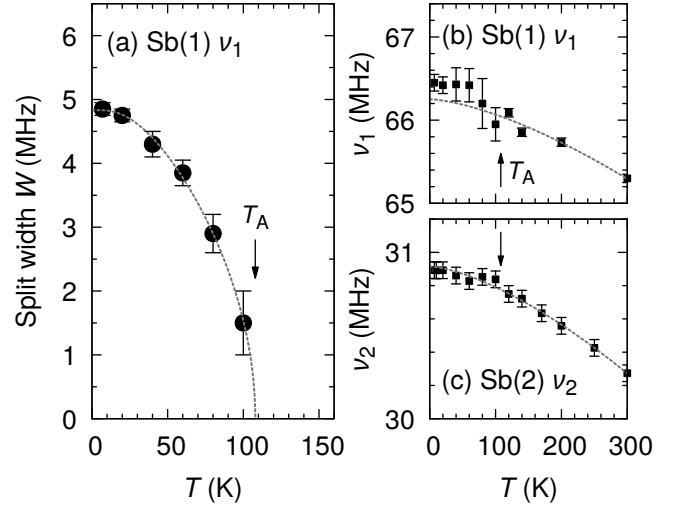


FIG. 5. (a) Temperature dependence of the  $^{121}\text{Sb}$  NQR split width for Sb(1) site. The dashed line shows the fitting result with the BCS-like gap function obtained using the least-squares method. (b) The temperature dependence of the resonant frequency  $\nu_1$  for Sb(1). (c) The temperature dependence of the resonant frequency  $\nu_2$  for Sb(2). The dashed lines in (b) and (c) are the fitting result with an empirical temperature dependence  $\nu_Q(T) = \nu_Q(0)(1 - \alpha T^{3/2})$  above  $T_A$ .

by  $I(f)df \propto d\theta$ , where  $\theta = qx$  and  $f = \nu_0 + W \sin \theta$ . The NQR spectral shape is described as follows[16]:

$$I(f) = \begin{cases} A [W^2 - (f - \nu_0)^2]^{-1/2} & (|f - \nu_0| < W), \\ 0 & (|f - \nu_0| \geq W), \end{cases} \quad (3)$$

where  $A$  denotes a constant. This yields a double-peak spectrum at  $f = \nu_0 \pm W$  with a finite intensity between two peaks. The asymmetric parameter  $\eta$  is fixed to zero for simplicity in this model. This can be determined by comparing frequencies between  $^{121}\nu_1(1)$  and  $^{121}\nu_2(1)$ .

The fitting results obtained using Eq. (3) are represented by the dashed line in Fig. 4(b) for spectra at 100 K or below. For a better fit to the experimental data, the above function was broadened using a Gaussian function and is multiplied by a linearly varying weight function. This simple model reproduces the experimental Sb(1) NQR spectrum at temperatures below 100 K. The analyzed split width  $W$  is shown in Fig. 5(a). This temperature dependence appeared to be an order parameter with a second-order phase transition and was analyzed using the following approximate BCS-type gap function[17]:

$$W(T) = W(0) \tanh \left[ a(T_A/T - 1)^{1/2} \right], \quad (4)$$

where  $W(0)$ ,  $a$ , and  $T_A$  denote fitting parameters. Although the selection of this model is arbitrary, it approximately reproduces the experimental result, as indicated by the dashed lines in Fig. 5(a). The transition temperature was evaluated to be  $T_A \approx 110$  K. This value was close to 120 K, where an electrical resistivity anomaly was observed in Ref. 12 and in



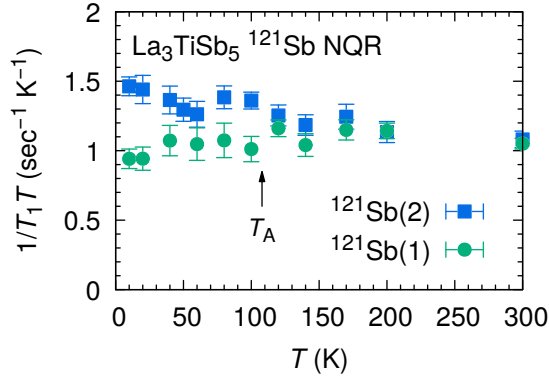


FIG. 6. (Color online) Nuclear spin-lattice relaxation rate divided by temperature ( $1/T_1T$ ) obtained by  $^{121}\text{Sb}$  NQR in  $\text{La}_3\text{TiSb}_5$ . The circle and square symbols represent the results at Sb(1) and Sb(2) sites, respectively. The vertical arrow shows the transition temperature determined by the Sb(1) line split.

the present study (Fig. 2). The relative split width regarding the quadrupole frequency is  $W(0)/\nu_0 \approx 0.073$  for  $T \rightarrow 0$ . In contrast, the increase in the spectral width in  $\nu_2(2)$  line is less than 0.15 MHz below  $T_A$  estimated from the change in the half width at half maxima, yielding  $W(0)/\nu_0 \leq 0.005$ . Because the NQR anomaly is more pronounced in Sb(1), which forms a quasi-one-dimensional chain, this result supports the CDW transition originating from Fermi surface nesting. The NQR result, combined with the electrical resistivity, was consistent with the incommensurate CDW transition.

The anomaly at  $T_A$  was also observed in the temperature dependence of  $^{121}\nu_1(1)$  or  $^{121}\nu_2(2)$  for Sb(1) and Sb(2), as shown in Fig. 5(b) and (c). The frequencies gradually increased as the temperature decreased, which is ascribed to the lattice shrinkage. This can be explained by empirical temperature variations in the NQR frequency[18]  $\nu_Q(T) = \nu_Q(0)(1 - \alpha T^{3/2})$  above  $T_A$ , and the fitted result is indicated by the dashed lines. The deviation from this curve is pronounced in  $^{121}\nu_1(1)$  below  $T_A$ , and a small kink is observed in  $^{121}\nu_2(2)$  at  $T_A$ . These results suggest a lattice constant change accompanying the CDW ordering.

Figure 6 shows the nuclear spin-lattice relaxation rate divided by temperature  $1/T_1T$  for Sb(1) and Sb(2). The relaxation rate probes the dynamics of the magnetic and electric origins. The nuclear relaxation curve  $M(t)$  was analyzed using

$$\frac{M(\infty) - M(t)}{M(\infty) - M(0)} = R(t), \quad (5)$$

where  $R(t)$  denotes a normalized relaxation curve consisting of multiple exponential functions. In the analysis, only the magnetic fluctuations were taken into consideration for the nuclear relaxation process, and the quadrupole relaxation mechanism was neglected.  $^{121}\nu_1(1)$  line corresponds to a  $\pm 1/2 \leftrightarrow \pm 3/2$  transition and  $R(t)$  is described as

$$R(t) = \frac{3}{28} \exp\left(-\frac{3t}{T_1}\right) + \frac{25}{28} \exp\left(-\frac{10t}{T_1}\right). \quad (6)$$

For  $^{121}\nu_2(2)$ , a finite  $\eta$  altered the relaxation curve from that of the  $\pm 3/2 \leftrightarrow \pm 5/2$  transition, and thus, we adopt the following function[19] obtained from numerical calculations for  $\eta = 0.38$ :

$$R(t) = 0.3875 \exp\left(-\frac{3t}{T_1}\right) + 0.6125 \exp\left(-\frac{9.0875t}{T_1}\right). \quad (7)$$

The relaxation curves exhibited a single component in two sites.  $1/T_1$  at the Sb(1) site was measured at the lower peaks below  $T_A$ . Overall,  $1/T_1T$  is nearly constant over the entire temperature range at these sites, indicating that  $\text{La}_3\text{TiSb}_5$  is a normal metal with small magnetic fluctuations. Because magnetic fluctuations are absent in Sb(1), which exhibits an NQR line split, the ordered phase is nonmagnetic. This value is somewhat larger for Sb(2) than for Sb(1) below  $T_A$ , which may be due to the change in the magnetic fluctuations or hyperfine coupling constant caused by the ordering.

The absence of an anomaly in  $1/T_1T$  may be unexpected, given the drastic NQR line split and resistivity hump. The  $1/T_1T$  behavior in the CDW state varies for different materials. No detectable changes have been reported in the CDW state of  $2\text{H-NbSe}_2$ [20] as in  $\text{La}_3\text{TiSb}_5$ , whereas an enhancement of  $1/T_1T$  was observed in  $\text{BaTi}_2\text{Sb}_2\text{O}$ [21] and  $\text{SrPt}_2\text{As}_2$ [22] toward  $T_{\text{CDW}}$ . Site-dependent  $1/T_1T$  has been reported for  $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$ [23] and  $\text{CsV}_3\text{Sb}_5$ [24, 25]. The small change in  $^{121}\text{Sb}$   $1/T_1T$  of  $\text{La}_3\text{TiSb}_5$  suggests that  $1/T_1T$  is mainly caused by the electrons not directly involved in the CDW transition, and that the reduction of DOS accompanying the transition does not strongly affect  $1/T_1T$ . It is interesting to obtain  $1/T_1T$  at the La site to clarify the site dependence of the CDW anomaly.

Although the simple CDW model expressed in Eq. (2) reproduced the experimental results, the NQR spectrum deviates slightly from the expected spectral shape in Eq. (3): the lower-frequency peaks were stronger, and an edge structure was observed at approximately 66 MHz at lower temperatures, as shown in Fig. 4(b). Therefore, the actual CDW modulation may be more complicated than expected. The complex spectrum may be related to the presence of Sb(1) and Ti chains with different symmetries. Sb(1) lacks local inversion symmetry, whereas Ti is located at the inversion center. These factors can cause successive CDW transitions similar to those of  $\text{LaAgSb}_2$ [26] and  $\text{SrPt}_2\text{As}_2$ [27–29]. Further studies, including x-ray or neutron diffraction experiments, may reveal the details of the CDW structure in  $\text{La}_3\text{TiSb}_5$ .

Finally, we discuss the isostructural systems. CDW instability may also exist in  $\text{Ce}_3\text{AX}_5$  systems ( $A = \text{Ti, Zr}$ ;  $X = \text{Sb, Bi}$ ). However, only antiferromagnetic transitions have been reported thus far[9, 12, 14, 30–33]. In  $\text{Ce}_3\text{TiSb}_5$  and  $\text{Ce}_3\text{TiBi}_5$ , the magnetic structures are somewhat different.  $\text{Ce}_3\text{TiSb}_5$  exhibits a long-period modulation of the ordered moment along the easy axis (in plane) with double transitions[11]. In contrast, the cycloid structure has been reported for  $\text{Ce}_3\text{TiBi}_5$ , where the moment can be along the hard ( $c$ ) axis[10]. This difference may be partly due to the various mechanisms involved in the magnetic structures of these

systems. The CDW instability inferred in this study can also be classified into these mechanisms. Future experimental and theoretical studies will unravel the interplay between magnetism and CDW instability.

In summary, electrical resistivity and  $^{121}\text{Sb}$  NQR measurements were performed on  $\text{La}_3\text{TiSb}_5$ , and the NQR spectrum of Sb(1) split below  $T_A \simeq 110$  K. This corresponds to a hump anomaly in the electrical resistivity. The broad double-peak spectrum can be explained by the incommensurate CDW structure. The nuclear spin-lattice relaxation rate shows no anomalies across the transition, indicating the non-magnetic ordering. The CDW instability, owing to its quasi-one-dimensional character, can affect the magnetism of the systems where the magnetoelectric effect is expected.

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- [1] S. Sumita and Y. Yanase, Superconductivity in magnetic multipole states, *Phys. Rev. B* **93**, 224507 (2016).
- [2] J. Ishizuka and Y. Yanase, Odd-parity multipole fluctuation and unconventional superconductivity in locally noncentrosymmetric crystal, *Phys. Rev. B* **98**, 224510 (2018).
- [3] S. Hayami, H. Kusunose, and Y. Motome, Toroidal order in metals without local inversion symmetry, *Phys. Rev. B* **90**, 024432 (2014).
- [4] Y. Yanase, Magneto-electric effect in three-dimensional coupled zigzag chains, *J. Phys. Soc. Jpn.* **83**, 014703 (2014).
- [5] H. Saito, K. Uenishi, N. Miura, C. Tabata, H. Hidaka, T. Yanagisawa, and H. Amitsuka, Evidence of a new current-induced magnetoelectric effect in a toroidal magnetic ordered state of  $\text{UNi}_4\text{B}$ , *J. Phys. Soc. Jpn.* **87**, 033702 (2018).
- [6] M. Shinozaki, G. Motoyama, M. Tsubouchi, M. Sezaki, J. Gouchi, S. Nishigori, T. Mutou, A. Yamaguchi, K. Fujiwara, K. Miyoshi, and Y. Uwatoko, Magnetolectric effect in the antiferromagnetic ordered state of  $\text{Ce}_3\text{TiBi}_5$  with Ce zig-zag chains, *J. Phys. Soc. Jpn.* **89**, 033703 (2020).
- [7] M. Shinozaki, G. Motoyama, T. Mutou, S. Nishigori, A. Yamaguchi, K. Fujiwara, K. Miyoshi, and A. Sumiyama, Study for current-induced magnetization in ferrotoroidal ordered state of  $\text{Ce}_3\text{TiBi}_5$ , *Proceedings of the International Conference on Strongly Correlated Electron Systems (SCES2019)*, JPS Conf. Proc. **30**, 011189 (2020).
- [8] G. Bolloré, M. J. Ferguson, R. W. Hushagen, and A. Mar, New ternary rare-earth transition-metal antimonides  $\text{RE}_3\text{MSb}_5$  (RE = La, Ce, Pr, Nd, Sm; M = Ti, Zr, Hf, Nb), *Chem. Mater.* **7**, 2229 (1995).
- [9] G. Motoyama, M. Sezaki, J. Gouchi, K. Miyoshi, S. Nishigori, T. Mutou, K. Fujiwara, and Y. Uwatoko, Magnetic properties of new antiferromagnetic heavy-fermion compounds,  $\text{Ce}_3\text{TiBi}_5$  and  $\text{CeTi}_3\text{Bi}_4$ , *Physica B: Condensed Matter* **536**, 142 (2018).
- [10] N. Gauthier, R. Sibille, V. Pomjakushin, O. S. Fjellvåg, J. Fraser, M. Desmarais, A. D. Bianchi, and J. A. Quilliam, Magnetic structure of  $\text{Ce}_3\text{TiBi}_5$  and its relation to current-induced magnetization, *Phys. Rev. B* **109**, L140405 (2024).
- [11] C. Ritter, A. K. Pathak, R. Filippone, A. Provino, S. K. Dhar, and P. Manfrinetti, Magnetic ground states of  $\text{Ce}_3\text{TiSb}_5$ ,  $\text{Pr}_3\text{TiSb}_5$  and  $\text{Nd}_3\text{TiSb}_5$  determined by neutron powder diffraction and magnetic measurements, *J. Phys.: Cond. Matter* **33**, 245801 (2021).
- [12] S. H. D. Moore, L. Deakin, M. J. Ferguson, and A. Mar, Physical properties and bonding in  $\text{RE}_3\text{TiSb}_5$  (RE = La, Ce, Pr, Nd, Sm), *Chem. Mater.* **14**, 4867 (2002).
- [13] K. Momma and F. Izumi, *VESTA3* for three-dimensional visualization of crystal, volumetric and morphology data, *J. Appl. Cryst.* **44**, 1272 (2011).
- [14] M. Matin, R. Kulkarni, A. Thamizhavel, S. K. Dhar, A. Provino, and P. Manfrinetti, Probing the magnetic ground state of single crystalline  $\text{Ce}_3\text{TiSb}_5$ , *J. Phys: Cond. Matter* **29**, 145601 (2017).
- [15] N. Stone, Table of nuclear electric quadrupole moments, *At. Data Nucl. Data Tables* **111-112**, 1 (2016).
- [16] R. Blinc, Magnetic resonance and relaxation in structurally incommensurate systems, *Phys. Rep.* **79**, 331 (1981).
- [17] K. Ishida, H. Mukuda, Y. Kitaoka, Z. Q. Mao, Y. Mori, and Y. Maeno, Anisotropic superconducting gap in the spin-triplet superconductor  $\text{Sr}_2\text{RuO}_4$ : Evidence from a Ru-NQR study, *Phys. Rev. Lett.* **84**, 5387 (2000).
- [18] J. Christiansen, P. Heubes, R. Keitel, W. Klinger, W. Loeffler, W. Sandner, and W. Witthuhn, Temperature dependence of the electric field gradient in noncubic metals, *Z. Phys. B* **24**, 177 (1976).
- [19] J. Chepin and J. H. Ross, Jr., Magnetic spin-lattice relaxation in nuclear quadrupole resonance: the eta not=0 case, *J. Phys.: Condens. Matter* **3**, 8103 (1991).
- [20] K. Ishida, Y. Niino, G.-Q. Zheng, Y. Kitaoka, K. Asayama, and T. Ohtani,  $^{93}\text{Nb}$  NQR study in layered superconducting  $2\text{H-NbSe}_2$ , *J. Phys. Soc. Jpn.* **65**, 2341 (1996).
- [21] S. Kitagawa, K. Ishida, K. Nakano, T. Yajima, and H. Kageyama, *s*-wave superconductivity in superconducting  $\text{BaTi}_2\text{Sb}_2\text{O}$  revealed by  $^{121/123}\text{Sb}$ -NMR/nuclear quadrupole resonance measurements, *Phys. Rev. B* **87**, 060510 (2013).
- [22] S. Kawasaki, Y. Tani, T. Mabuchi, K. Kudo, Y. Nishikubo, D. Mitsuoaka, M. Nohara, and G.-q. Zheng, Coexistence of multiple charge-density waves and superconductivity in  $\text{SrPt}_2\text{As}_2$  revealed by  $^{75}\text{As}$  - NMR/NQR and  $^{195}\text{Pt}$  - NMR, *Phys. Rev. B* **91**, 060510 (2015).
- [23] Z. Li, W. H. Jiao, G. H. Cao, and G.-q. Zheng, Charge fluctuations and nodeless superconductivity in quasi-one-dimensional  $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$  revealed by  $^{125}\text{Te}$ -NMR and  $^{181}\text{Ta}$ -NQR, *Phys. Rev. B* **94**, 174511 (2016).
- [24] C. Mu, Q. Yin, Z. Tu, C. Gong, H. Lei, Z. Li, and J. Luo, *S*-wave superconductivity in kagome metal  $\text{CsV}_3\text{Sb}_5$  revealed by  $^{121/123}\text{Sb}$  NQR and  $^{51}\text{V}$  NMR measurements, *Chinese Phys. Lett.* **38**, 077402 (2021).
- [25] J. Luo, Z. Zhao, Y. Z. Zhou, J. Yang, A. F. Fang, H. T. Yang, H. J. Gao, R. Zhou, and G.-q. Zheng, Possible star-of-david pattern charge density wave with additional modulation in the kagome superconductor  $\text{CsV}_3\text{Sb}_5$ , *npj Quantum Materials* **7**, 30 (2022).
- [26] C. Song, J. Park, J. Koo, K.-B. Lee, J. Y. Rhee, S. L. Bud'ko, P. C. Canfield, B. N. Harmon, and A. I. Goldman, Charge-density-wave orderings in  $\text{LaAgSb}_2$ : An x-ray scattering study, *Phys. Rev. B* **68**, 035113 (2003).
- [27] A. Imre, A. Hellmann, G. Wenski, J. Graf, D. Johrendt, and A. Mewis, Inkommensurabel modulierte Kristallstrukturen und Phasenumwandlungen – Die Verbindungen  $\text{SrPt}_2\text{As}_2$  und  $\text{EuPt}_2\text{As}_2$ , *Z. Anorg. Allg. Chem.* **633**, 2037 (2007).
- [28] A. F. Fang, T. Dong, H. P. Wang, Z. G. Chen, B. Cheng, Y. G. Shi, P. Zheng, G. Xu, L. Wang, J. Q. Li, and N. L. Wang, Single-

- crystal growth and optical conductivity of  $\text{SrPt}_2\text{As}_2$  superconductors, *Phys. Rev. B* **85**, 184520 (2012).
- [29] L. Wang, Z. Wang, H.-L. Shi, Z. Chen, F.-K. Chiang, H.-F. Tian, H.-X. Yang, A.-F. Fang, N.-L. Wang, and J.-Q. Li, Two-coupled structural modulations in charge-density-wave state of  $\text{SrPt}_2\text{As}_2$  superconductor, *Chin. Phys. B* **23**, 086103 (2014).
- [30] M. Tsubouchi, G. Motoyama, J. Gouchi, K. Miyoshi, S. Nishigori, K. Fujiwara, T. Mutou, and Y. Uwatoko, Temperature-pressure magnetic phase diagram of  $\text{Ce}_3\text{TiBi}_5$ , *Proceedings of the International Conference on Strongly Correlated Electron Systems (SCES2019)*, JPS Conf. Proc. **30**, 011102 (2020).
- [31] G. Motoyama, M. Shinozaki, M. Tsubouchi, M. Kuninaka, S. Nishigori, K. Miyoshi, K. Fujiwara, J. Gouchi, and Y. Uwatoko, Magnetic properties of new antiferromagnetic compound of  $\text{Ce}_3\text{ZrBi}_5$ , *Proceedings of the International Conference on Strongly Correlated Electron Systems (SCES2019)*, JPS Conf. Proc. **30**, 011180 (2020).
- [32] M. Shinozaki, G. Motoyama, S. Nishigori, A. Yamaguchi, Y. Yamane, T. Mutou, K. Fujiwara, M. Manago, K. Miyoshi, and A. Sumiyama, Electrical resistivity measurements of antiferromagnetic compound  $\text{Ce}_3\text{TiSb}_5$  under pressure, *J. Phys.: Conf. Ser.* **2164**, 012040 (2022).
- [33] K. Nakagawa, M. Shinozaki, G. Motoyama, S. Nishigori, K. Fujiwara, M. Manago, and K. Miyoshi, Single crystal growth of  $\text{Ce}_3\text{ZrSb}_5$  and characterization of the physical properties, *Proceedings of the 29th International Conference on Low Temperature Physics (LT29)*, JPS Conf. Proc. **38**, 011083 (2023).