Observation of modulated magnetic long-range order in $La_{1.88}Sr_{0.12}CuO_4$

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Magnetic superlattice peaks are observed in single-crystal neutron-diffraction measurements on orthorhombic $La_{1.88}Sr_{0.12}CuO_4$ at reciprocal points of $(1/2\pm\epsilon,1/2,0)$ and $(1/2,1/2\pm\epsilon,0)$ in the tetragonal notation where $\epsilon=0.126\pm0.003$. The La NMR measurement reveals a broadening of the field-swept spectrum below ~45 K corresponding to the existence of magnetic order. The remarkable softening of longitudinal sound waves along [110] is observed in the same crystal. The features observed in the neutron diffraction, NMR, and ultrasonic measurements suggest that the dynamical incommensurate spin correlation is pinned by a lattice instability toward the low-temperature tetragonal phase.
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It is well known in $La_{2-x}Ba_xCuO_4$ around $x=0.12$ that the superconducting transition temperature T_c is extremely suppressed.[1] In this concentration region of x, a structural phase transition occurs from the orthorhombic phase (OMT, the space group *Bmab*) to the low-temperature tetragonal phase (TLT, the space group $P4_2/ncm$) at $T_{d2}\sim70$ K (Ref. 2) and anomalous behavior is observed in the transport properties, such as the thermoelectric power and the Hall coefficient near T_{d2}.[3] In addition, the existence of magnetic order below ~36 K is indicated from the measurements of μSR and NMR around $x=0.12$.[4,5] In $La_{2-x}Sr_xCuO_4$, the structural phase transition to the TLT phase is not observed at least down to 4.2 K,[6] but the suppression of T_c (Ref. 7) is observed and magnetic order below ~36 K is apparent around $x=0.115$.[8–11] Recently, Tranquada *et al.* observed magnetic superlattice peaks below ~50 K by the elastic neutron scattering measurement in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ which undergoes the structural phase transition to the TLT phase at ~70 K.[12–14] They proposed the model of the magnetic structure that the modulated antiferromagnetic order is pinned and stabilized in the TLT phase but not in the OMT phase. The purpose of the present study is to search for the magnetic order in the OMT phase predicted from NMR and μSR study in a single crystal of $La_{2-x}Sr_xCuO_4$ ($x\sim0.12$), in which the magnetic structure is unknown.

A single crystal of ~7 mm in diameter and ~40 mm in length is grown by TSFZ method. The details of sample growth are given elsewhere.[15] The crystal is annealed under 1 bar of oxygen gas flowing at 900 °C for 100 h.[16] Zero-field cooled diamagnetization decreases rapidly with increasing temperature at $T\sim26$ K, reflecting the well-known anomalous suppression of T_c around $x=0.12$. Neutron scattering measurement is carried out using the KSD double-axis spectrometer installed in the JRR-3M Guide Hall at the JAERI in Tokai, Japan. The incident neutron beam has a wavelength of 1.53 Å, obtained using a PG(002) monochromator. The horizontal divergence of incident neutron beam is 12′ and the acceptance angle of scattered beam is 50′. The single crystal for the neutron measurement is sealed in an aluminum container with He gas. Field-swept spectra of ^{139}La NMR are measured with a fixed frequency of 32.57 MHz on the sample cut from the same single crystal used for the neutron measurement (~0.7×3.4×5.0 mm). The sound velocity V_s is measured by the phase comparison method with the ~22 MHz longitudinal waves generated by the Z-cut $LiNbO_3$ transducer. The sample for this measurement has a length of ~4 mm along [110], and was cut from the same single-crystal rod grown for the measurement of neutron diffraction and NMR.

Neutron diffraction studies are performed in the $(hk0)$ zone. All indexes in this report are defined as the notation in the tetragonal $(I4/mmm)$. Figure 1(a) shows the $(hk0)$ reciprocal plane and the scan direction for the neutron scattering. Figure 1(b) shows the q spectrum observed at 2.1 K by the scan along $(1/2+h,1/2,0)$, as indicated in Fig. 1(a). The sloped background is subtracted from the data by the least-squares fit with the Gaussian function plus background. With the diagonal scan along $(1/2,1/2+h,0)$, we confirmed the existence of four incommensurate peaks at $(1/2\pm\epsilon,1/2,0)$ and $(1/2,1/2\pm\epsilon,0)$ as shown in Fig. 1(a). These peak splittings around (1/2,1/2,0) reciprocal point suggest a modulated antiferromagnetic order with the incommensurability $\epsilon=0.126\pm0.003$. In the whole x region of superconducting phase of $La_{2-x}Sr_xCuO_4$, the dynamical incommensurate spin fluctuations are observed in the inelastic neutron measurement.[16] The incommensurability of the antiferromagnetic order observed in this study is really close to that of the inelastic magnetic peaks of $La_{1.88}Sr_{0.12}CuO_4$, though the incommensurability of the inelastic peaks increases monotonically with increasing Sr concentration x and tends to saturate above $x\sim0.12$.

The scans through one of the magnetic superlattice peaks at temperatures of 2.1, 40, and 60 K are shown in Fig. 2. The solid lines are the results of the least-squares fit. The peak

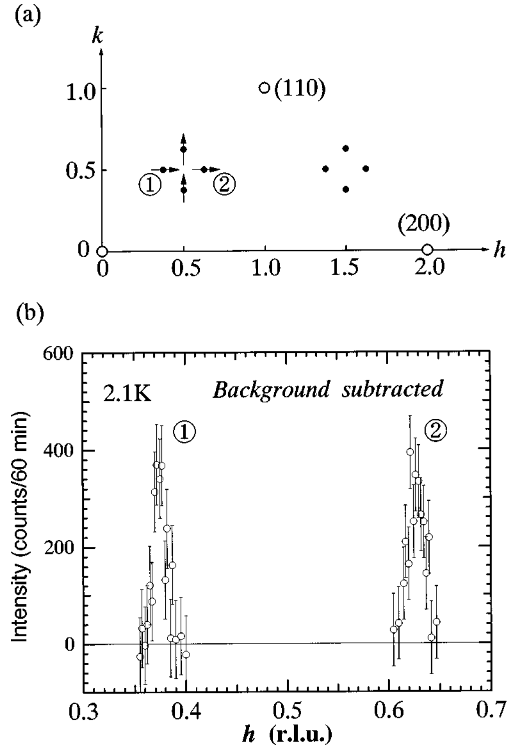

FIG. 1. (a) The diagram of $(hk0)$ reciprocal plane. The scan direction on the neutron diffraction is $(1/2+h,1/2,0)$ as indicated by the arrow. Closed circles represent the reflection points of magnetic superlattice peaks. Open circles correspond to Bragg peaks. (b) Scan through magnetic superlattice peaks along the direction indicated in (a) at 2.1 K in $La_{1.88}Sr_{0.12}CuO_4$. The wavelength of the incident neutron is 1.53 Å.

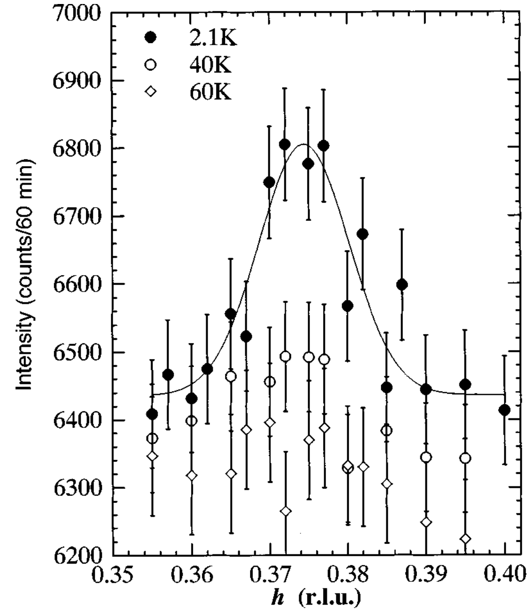

FIG. 2. Scans at 2.1, 40, and 60 K through the magnetic superlattice peak along the $(1/2+h,1/2,0)$ in $La_{1.88}Sr_{0.12}CuO_4$. The solid lines are the results of a curve fit with the Gaussian function plus background.

intensity dramatically decreases with increasing temperature up to 40 K and merges to the background at 60 K.

In this study, we can discuss only qualitatively the size of the upper limit of the ordered Cu moment μ_{Cu} because of the extremely large extinction effect which reduces dramatically the intensity of the nuclear Bragg reflection in the $(hk0)$ zone and because of the lack of detailed information on the magnetic structure. Since the present neutron scattering measurement was performed without analyzing the scattered neutron energy, the detected intensity contains the inelastic scattering contribution in principle. In the present case, however, the scattered intensity is expected to be dominantly elastic in origin since the narrow peak width is very narrow and comparable to the instrumental limit. In other words, the long-range magnetic structure developed at low temperature is expected to be static. It seems that the dynamical magnetic correlation, which is observed as the inelastic magnetic peaks, is stabilized by some ''pinning'' mechanism because the inelastic and the elastic reflections are observed at the same position. Tranquada *et al.* proposed the model that the stripe structure is pinned along the characteristic buckling pattern of the CuO_2 plane of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ in the TLT phase but not in the OMT phase. Let us discuss the appearance of the magnetic order and the change of the crystal structure. There is a possibility that TLT-like distortion is one of the contributions toward the pinning mechanism in $La_{2-x}Sr_xCuO_4$ by analogy with the case of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. The crystal structure at 2.1 K is orthorhombic as determined by the Rietveld analysis of the neutron powder diffraction data and no evidence for the decrease of the orthorhombicity is observed.[17]. For this reason, we carried out the ultrasonic measurement using the single crystal to detect the infinitesimal change or instability of the crystal structure. Figure 3 shows the temperature dependence of the longitudinal sound velocity V_s of $(C_{11}+C_{12}+2C_{66})/2$ mode along the [110] direction and the derivative of V_s with respect to T. This mode is sensitive to the structural phase transition to the TLT phase. The hardening is observed below ~20 K and may correspond to the upturn of V_s observed in polycrystals.[18] The remarkable feature is the softening below ~45 K (Fig. 3). This temperature dependence of V_s suggests that the precursor of the structural phase transition appears below ~45 K. It seems that TLT-like distortion which appeared as a precursor of the structural phase transition results in the pinning mechanism of the dynamical magnetic correlation observed as the inelastic magnetic peaks. In other words, it is suggested that the ordered spins couple with the lattice system in this case.[11] The impurity and dislocation of the specimen should be considered as other possibilities. Actually, the magnetic superlattice peaks are observed in $La_{1.86}Sr_{0.14}Cu_{0.988}Zn_{0.012}O_{4-\delta}$.[19] If these contributions are dominant, static magnetic peaks have to be observed in all regions of x where the incommensurate peaks of inelastic components appear. The magnetic order at low temperature is observed, however, in a narrow range around $x=0.12$ by NMR measurements, so that we expect the magnetic reflection in the same region. From this consideration, it seems that the contribution from the impurity and dislocation is not essential in $La_{2-x}Sr_xCuO_4$.

We also performed the NMR measurement to examine its consistency with the result of the neutron measurement. Figure 4 shows the La NMR spectrum and the temperature dependence of the spectrum linewidth observed in the same single crystal used in the neutron scattering measurements.

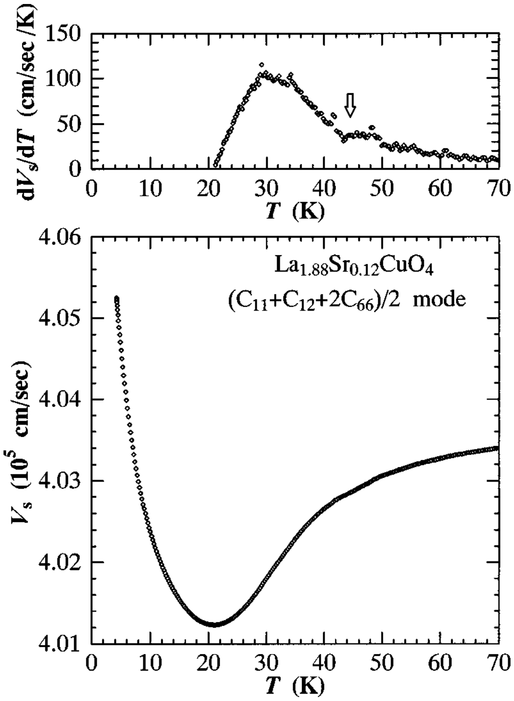

FIG. 3. Bottom: The temperature dependence of the sound velocity in the longitudinal waves along the [110] direction which corresponds to $(C_{11}+C_{12}+2C_{66})/2$ mode in $La_{1.88}Sr_{0.12}CuO_4$. Top: The temperature dependence of dV_s/dT. The arrow indicates the temperature where the remarkable softening occurs.

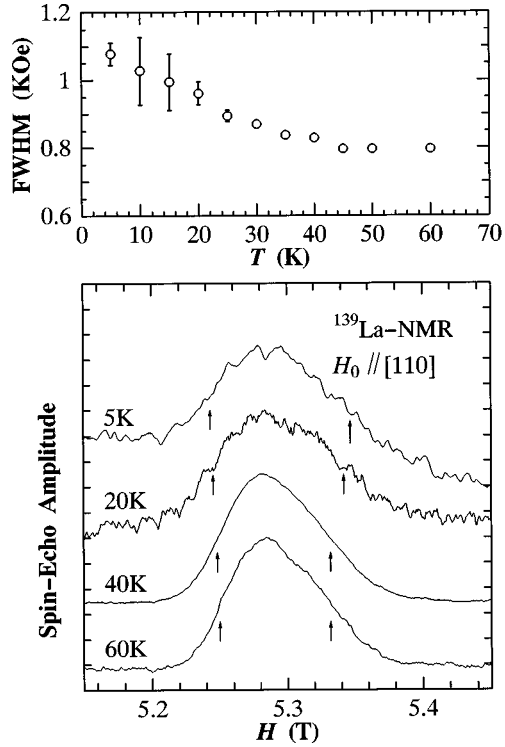

FIG. 4. Bottom: The field-swept spectra corresponding to the transition between $I_Z=\pm 1/2$ of La nuclei in $La_{1.88}Sr_{0.12}CuO_4$ at several temperatures. Top: The temperature dependence of the resonance linewidth of La NMR spectra.

The linewidth is defined as the FWHM. External field is applied to the direction $H \|$ [110], which is the direction of ordered Cu-3d spin in La_2CuO_4.[20] The broadening of the linewidth is observed below ~45 K and is consistent with the disappearance of the magnetic superlattice peaks at 60 K, and the Néel temperature T_N is determined to be ~45 K. The spectrum obtained at 5 K is simulated well by the calculation that the spectrum at 60 K is split by the distribution of internal fields from each sublattice in modulated antiferromagnetic state. In case of the c-axis aligned powder samples, the complex shape of the spectrum with a residual powder pattern causes ambiguity in estimating the hyperfine field.[9,11] In our measurement on the single crystal, the shape of the spectrum is symmetric so that the FWHM is more precisely determined. As a result, the hyperfine field at La site at 5 K is evaluated as ~140 Oe from the half of the increase of the linewidth, and is equal to that of c-axis aligned samples. This coincidence of the value of the hyperfine field confirms the accuracy of the NMR study previously reported in c-axis aligned samples. If we tentatively assume the hyperfine coupling constant $A_{La}=1.7$ KOe/μ_B which is the reported value of the end member compound La_2CuO_4,[21] the ordered moment of Cu-3d spin is estimated to be $\mu_{Cu}\sim 0.08\mu_B$. This value is smaller than that of La_2CuO_4 but is extremely larger than the value estimated from the present neutron measurement. The reason for this discrepancy is not clear at this stage.

We finally discuss the difference of $La_{2-x}Sr_xCuO_4$ from the case of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, in which the magnetic and charge order begin to appear at the structural phase transition to the TLT phase. We previously reported on the relation between the crystal structure and the change of the electronic state.[22] In $La_{2-x}Sr_xCuO_4$ around $x=0.12$, the change of the electronic state, which is observed as the anomalous behavior of the transport properties, is not directly caused by the structural phase transition to the TLT phase. If the anomaly of the transport properties found in $La_{2-x}Sr_xCuO_4$ ($x\sim 0.12$) originates from the charge ordered state, the stripe charge order observed in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ should appear in $La_{2-x}Sr_xCuO_4$ ($x\sim 0.12$). However, the conclusive evidence of the stripe model, the charge peak, has not been detected. Therefore, it is not clear as to whether or not the ''1/8'' anomaly in $La_{2-x}Sr_xCuO_4$ could be explained completely by the stripe model.

In summary, we performed neutron diffraction, NMR, and ultrasonic measurements in single crystal $La_{1.88}Sr_{0.12}CuO_4$. The magnetic superlattice peaks at 2.1 K are observed in the neutron scattering at $(1/2\pm\epsilon,1/2,0)$ and $(1/2,1/2\pm\epsilon,0)$ with $\epsilon=0.126\pm 0.003$, and are symmetrically located around (1/2,1/2,0) reciprocal point. The existence of these peaks and the NMR spectral features give evidence of the modulated antiferromagnetic long-range order and the magnetic superlattice below $T_N\sim 45$ K in the orthorhombic phase. The softening of the sound velocity below ~45 K, which is close to T_N, suggests the coupling between the ordered spin and the lattice system in $La_{1.88}Sr_{0.12}CuO_4$.

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