Significant suppression of weak ferromagnetism in 

\((\text{La}_{1.8}\text{Eu}_{0.2})\text{CuO}_4\)

I. Tsukada, X. F. Sun, Seiki Komiya, A. N. Lavrov, and Yoichi Ando

Central Research Institute of Electric Power Industry,
2-11-1 Iwado-kita, Komae, Tokyo 201-8511, JAPAN

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Abstract

The magnetic structure of \((\text{La}_{1.8}\text{Eu}_{0.2})\text{CuO}_4\) has been studied by magnetization measurements of single crystals, which show antiferromagnetic long-range order below \(T_N = 265\ \text{K}\) and a structural phase transition at \(T_s = 130\ \text{K}\). At \(T_s < T < T_N\), the Cu spin susceptibility exhibits almost the same behavior as that of \(\text{La}_2\text{CuO}_4\) in the low-temperature orthorhombic phase, which indicates the existence of finite spin canting out of the \(\text{CuO}_2\) plane. At \(T < T_s\), the magnitude of the weak-ferromagnetic moment induced by the spin canting is suppressed approximately by 70%.

This significant suppression of the weak-ferromagnetic moment is carefully compared with the theoretical analysis of weak ferromagnetism by Stein et al. (Phys. Rev. B 53, 775 (1996)), in which the magnitude of weak-ferromagnetic moments strongly depend on the crystallographic symmetry. Based on such comparison, below \(T_s\) \((\text{La}_{1.8}\text{Eu}_{0.2})\text{CuO}_4\) is in the low-temperature less-orthorhombic phase with a space group of \(P\text{ccn}\). We also discuss the possible magnetic structure of the pure low-temperature tetragonal phase with space group \(P4_2/ncm\), which is relevant for rare-earth and alkaline-earth ions co-doped \(\text{La}_2\text{CuO}_4\).

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The magnetic structure of La$_2$CuO$_4$ has been extensively studied because it is considered to be a good example of two-dimensional $S=1/2$ Heisenberg antiferromagnet, whose nature is one of the central topics of quantum magnetism. La$_2$CuO$_4$, however, shows a rather classical long-range Néel order at low temperatures because of its relatively low crystallographic symmetry caused by an orthorhombic distortion in the low-temperature orthorhombic (LTO) phase and a finite inter-plane coupling. In the LTO phase, spins are aligned approximately along the $b$ axis with slight canting out of the CuO$_2$ plane, which is responsible for the weak ferromagnetism of La$_2$CuO$_4$. The schematic drawing of the magnetic structure of La$_2$CuO$_4$ in the LTO phase is shown in Fig. 1 (a). The spin canting was simply explained by antisymmetric exchange interactions of Dzyaloshinskii-Moriya (DM) type. However, it has been pointed out by Shekhtman et al., Bonesteel, and Yildirim et al. that the true origin may not be so simple, because of the importance of an additional anisotropic symmetric spin-spin interaction. This interaction was already derived by Moriya but unfortunately ignored for a long time. Kaplan was the first who noticed the importance of this interaction. He reinvestigated the role of spin-orbit interaction in super-exchange mechanism for one-dimensional $S = 1/2$ antiferromagnetic (AF) spin chains and spin rings, and found that the anisotropic symmetric interaction gives an easy-axis anisotropy to the spin system that completely compensates the easy-plane anisotropy given by the DM interaction. Although the presence of this term (which we call KSEA interaction following Zheludev et al.) has been recently established in several $S = 1/2$ antiferromagnets, Ba$_2$CuGe$_2$O$_7$, K$_2$V$_3$O$_8$, and Yb$_4$As$_3$, its role in the most interesting material La$_2$CuO$_4$ has not been experimentally revealed yet.

One possible test is to determine the magnetic structure of the so-called low-temperature tetragonal (LTT) phase (space group $P4_2/ncc$), which was first reported in (La,Ba)$_2$CuO$_4$. Bonesteel discussed that the weak-ferromagnetic (WF) moment per CuO$_2$ plane should disappear in the LTT phase as is schematically drawn in Fig. 1 (b) when one takes the KSEA interaction into account. In this arrangement, spins are parallel to the rotation axes of CuO$_6$ octahedra, and therefore, they are also parallel/antiparallel to the DM vectors. This theoretical prediction became a trigger of a long debate because the magnetic structure shown in Fig. 1 (b) was different from that proposed based on neutron diffraction.
The latter magnetic structure is shown in Fig. 1 (c), where spins are perpendicular to the rotation axes of the CuO$_6$ octahedra, and thus the spin canting remains intact. Since (La,Nd)$_2$CuO$_4$ was previously believed to be in the LTT phase, it was concluded that the LTT phase favored the spin arrangement shown in Fig. 1 (c) rather than that in Fig. 1 (b). By referring to the experimentally proposed model, Koshiba et al. tried to explain the presence of WF moment in the LTT phase. They expanded the previously reported theories by assuming the magnitude of DM and KSEA interactions independently, and predicted the presence of WF moment in the LTT phase for a particular situation where the effect of DM interaction is dominant. Stein et al. also calculated the WF moment with similar assumptions as Koshiba et al. do, but concluded that the physically reasonable ratio of DM and KSEA interactions do not favor the presence of WF moment in the pure LTT phase. Therefore, the above mentioned controversy among theories and neutron experiments has not been solved yet.

In this paper we study the anisotropic magnetization of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ single crystals and discuss the magnetic structure to shed light on this problem of long debate. Since Eu$^{3+}$ ions do not give large Curie-type susceptibility in contrast to Nd$^{3+}$, (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ is a more suitable system for magnetization measurements. Although magnetic susceptibility data of polycrystalline samples have been already reported by Kataev et al., one needs the c-axis susceptibility data measured on a single crystal to evaluate the magnitude of spin canting out of the CuO$_2$ plane. We observe a significant suppression of the spin canting below the structural phase-transition temperature, which has not yet been elucidated to be a transition from the LTO to the LTT phase, or to the low-temperature less orthorhombic (LTO2, space group $Pccn$) phase. We analyze the change of WF moment across the structural phase transition, and compare the magnitude of the spin canting with that predicted by Stein et al.; the obtained results point to the LTO2 crystallographic symmetry of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ below $T_s$. Finally we will discuss the likely magnetic structure for pure LTT symmetry, and make some comments on the previous controversy in understanding the weak ferromagnetism in La$_{1.65}$Nd$_{0.35}$CuO$_4$. 


II. EXPERIMENTAL RESULTS

(La$_{1.8}$Eu$_{0.2}$)CuO$_4$ single crystals are grown by a traveling-solvent floating zone method$^{28}$ and their quality is confirmed by x-ray diffractions. The crystalline rod is carefully cut into a rectangular shape with all the faces normal to the principal axes, which is confirmed using an x-ray Laue backscattering. Then the samples are annealed in He-gas flow to remove excess oxygen. Magnetic susceptibility measurements are carried out with commercial SQUID magnetometer (MPMS, Quantum Design) in magnetic field up to $H = 70$ kOe. To avoid confusion, we follow the axis notation of the LTO phase, so that the $a$ and $b$ axes run along two orthogonal in-plane Cu-Cu diagonal directions and the $c$ axis is normal to the CuO$_2$ plane.

Figure 2 (a) shows the magnetic susceptibility of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ measured along the $c$ axis ($\chi_c$) at $H = 5$ kOe. The peak structure is identified as a Néel transition at $T_N = 265$ K similar to that of Eu-free La$_2$CuO$_4$.$^{2,29}$ In addition to this peak, we observe a rapid increase in $\chi_c$ below $T = 130$ K as was reported for polycrystalline samples.$^{27}$ Before discussing $\chi_c$ in detail, we should consider the contribution from Eu$^{3+}$ ions. The magnitude of $\chi_c$ of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ is several times larger than that of Eu-free La$_2$CuO$_4$,$^{2,29}$ which is likely to be due to the Van-Vleck contribution of Eu$^{3+}$ ions. This Van-Vleck contribution is anisotropic as can be seen from the data of $\chi_c$ and $\chi_{110}$ (susceptibility measured along the [110] direction) shown in Fig. 2 (b). $\chi_{110}$ exhibits a large-scale increase upon cooling from room temperature to 80 K similar to that observed in Eu$_2$CuO$_4$.$^{30}$ A slight and broad hump around 265 K in $\chi_{110}$ comes from the Néel transition. On the other hand, such large-scale increasing behavior upon cooling is not found in $\chi_c$ of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ in contrast to $\chi_c$ of Eu$_2$CuO$_4$ where the Van-Vleck contribution of Eu$^{3+}$ ions gives a similar increase of $\chi_c$ also down to approximately 150 K.$^{30}$ We speculate that the different oxygen coordinations around Eu$^{3+}$ ion, rock-salt type in (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ and fluorite type in Eu$_2$CuO$_4$, are the origin of the different Van-Vleck contributions. In the following discussion, we assume that the temperature dependence of $\chi_c$ is mostly coming from the magnetic response of CuO$_2$ planes.

Our sample shows somewhat lower $T_N$ in comparison with polycrystalline (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ (Ref. [27]) or single-crystalline La$_2$CuO$_4$ (Ref. [29]), which suggests that a small amount of excess oxygen may remain in our sample. However, this does not change the...
magnetic structure of AF long-range order, and we can safely neglect its effect in the present context. Below $T_N$, $\chi_c$ decreases down to $T_s = 130$ K, which coincides with the reported LTO $\rightarrow$ LTO2/LTT structural phase-transition temperature$^{31,32}$ and then steeply increases until $T = 110$ K; such increase is not seen in $\chi_{110}$. The size of this increase is approximately $5 \times 10^{-5}$ emu/mol. This is larger than that observed in polycrystalline samples$^{27}$ which is apparently because of averaging of $\chi_{110}$ and $\chi_c$. This increase at $T_s$ is most likely related to the change of inter-plane magnetic couplings as will be discussed later.

In order to clarify whether the magnitude of WF moment is changed across the structural phase transition, we measure the high-field susceptibility, which is shown in Fig. 3 (a). The temperature dependence of $\chi_c$ at $H = 70$ kOe ($\chi_c^{70k}$) is notably different from that at $H = 5$ kOe ($\chi_c^{5k}$) below $T_N$; it increases continuously instead of forming a peak structure at $T_N$, shows a broad maximum around 170 K, and then decreases toward 50 K. Below 20 K there is a hysteresis between zero-field cool (ZFC) and field cool (FC) data as conventional ferromagnets with a finite coercive force show, which suggests the presence of magnetic domains. It should be noted that such a hysteresis is absent in La$_2$CuO$_4$, and only lightly hole-doped (La$_{2-x}$Sr$_x$)CuO$_4$ exhibits hysteretic behavior between FC and ZFC process.$^{29}$ It is likely that the substitution of Eu$^{3+}$ for La$^{3+}$ induces a structural disorder that causes a domain structure in our sample.

The field dependence of the magnetization at $T = 50$ K [inset of Fig. 3 (a)] clearly shows the WF transition around $H_c = 30$ kOe. The transition width is large in comparison with that of La$_2$CuO$_4$.$^{33}$ These data give us two important clues to reveal the magnetic structure of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$. One is a finite but significantly suppressed WF moment and the other is a decrease of the critical field from that of La$_2$CuO$_4$. To see the suppression of WF moment more clearly, we plot $\Delta\chi_c = \chi_c^{70k} - \chi_c^{5k}$, which is shown in Fig. 3 (b). Below $T_N$, $\Delta\chi_c$ follows the temperature dependence typical for the evolution of spontaneous magnetization. Just above $T_s$, $\Delta\chi_c$ reaches almost $1.5 \times 10^{-4}$ emu/mol, and its extrapolation to the lower temperatures gives $\Delta\chi_c \approx 1.7 \times 10^{-4}$ emu/mol at $T = 0$ K. This value corresponds to the spontaneous magnetization of 12 emu/mol, which would be the WF moment if the LTO structure were maintained down to $T = 0$ K. In the real sample, however, the structural phase transition occurs, and $\Delta\chi_c$ is reduced approximately to $5 \times 10^{-5}$ emu/mol ($M_{WF}$). This value gives the magnetization of 3.5 emu/mol, consistent with the vertical shift of two dotted lines in the inset of Fig. 3 (a). Therefore, the magnitude of WF moment is
suppressed almost by 70%. Moreover, the decrease of the critical field indicates a reduction in the inter-plane coupling. The critical field of the WF transition is mainly determined by the competition between AF inter-plane coupling and Zeeman effect $H_c \times M_{WF}$ which is lowered approximately by 5 times. Thus, the AF inter-plane coupling is reduced as well.

III. DISCUSSION

A. Theoretical predictions of the magnetic structure

Let us briefly survey several theoretical works on the magnetic structure of rare-earth doped La$_2$CuO$_4$. In order to analyze the experimental data, we need information of the WF moment not only in the LTO and LTT phases but also in the LTO2 phase. To our knowledge, Coffey et al.\textsuperscript{7} was the first who discussed possible magnetic structures of La$_2$CuO$_4$ in the LTT phase. They proposed two kinds of ground state: one with a canted moment per CuO$_2$ plane and one without. After Shekhtman et al.\textsuperscript{10} pointed out the importance of the KSEA term upon discussing the magnetic structure of LTO phase, Bonesteel\textsuperscript{12} tried to include this term in calculations of the magnetic ground state of the LTT phase, and concluded that the canted moment per CuO$_2$ plane should be zero (Fig.1(b)) if the KSEA term is properly incorporated into the spin Hamiltonian. In the Bonesteel’s paper, a structural difference between the LTO2 and LTT phases has been carefully considered; a spin canting angle out of the CuO$_2$ plane has been derived to be proportional to the angle $\kappa$ that is the angle between the Cu-O-Cu bond direction and the rotation axis of CuO$_6$ octahedron ($\kappa = \pi/4$, $0 < \kappa < \pi/4$, and $\kappa = 0$, for the LTO, LTO2, and LTT phases, respectively). Viertiö and Bonesteel\textsuperscript{22} then discussed the relation between the interplanar coupling and the weak ferromagnetic transition in (La$_{2-x}$Nd$_x$)CuO$_4$ using the same assumptions as in Ref. 12.

The theoretical calculation done by Stein et al.\textsuperscript{23} is basically the same as that done by Bonesteel and co-workers\textsuperscript{12,22}, but they have evaluated the WF moment more rigorously.\textsuperscript{23} They calculated the $\kappa$ dependence (it is denoted as $\alpha$ in their original paper) of the WF moment under several different conditions, which is reproduced in Fig. 1. The vertical axis $M_F/(SD/2J)$ is the WF moment normalized by the magnitude of spin, Heisenberg interaction and DM interaction. The only tunable parameter, $\Omega$, qualitatively gives the relative magnitude of the DM and KSEA interactions; $\Omega = 1$ corresponds to the situation
without KSEA interaction (Heisenberg + DM), and $M_F/(SD/2J) = 1$ regardless of $\kappa$, while $\Omega = 0$ corresponds to the situation where the KSEA interaction completely compensates the easy-plane anisotropy given by the DM interaction as was derived by Bonesteel\textsuperscript{12} (Heisenberg + DM + KSEA). Intermediate values of $\Omega$ correspond to the situation that the easy-plane anisotropy caused by the DM interactions still survives even under the presence of KSEA interactions. Since the case discussed by Koshibae et al. is included in this parameter region, Stein’s calculation may reproduce all the possible situations by tuning $\Omega$, and thus, it is most convenient for analyzing our experimental data.

B. Magnetic structure of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$

Based on the Stein’s calculations, we can discuss the source of the 70% suppression of the WF moment across $T_s$. Figure 4 teaches us that the WF moment would be the same in the LTO, LTO2, and LTT phases when only the DM interaction is effective. This is however inconsistent with our observation. We do observe the suppression of the WF moment, and this fact itself indicates that the KSEA interaction actually works in (La$_{1.8}$Eu$_{0.2}$)CuO$_4$. It should be noted that such quantitative comparison of the WF moment across $T_s$ can be done only by magnetization measurements, while a neutron diffraction measurement is not sensitive enough to judge whether the spin canting is present or not. Thus, it is not surprising that previous neutron experiments failed to detect this change.

Figure 4 also gives the range of $\Omega$ and $\kappa$ for our sample. One can easily notice that the critical line, $\Omega = 0.5$, is terminated at $M_F/(SD/2J) = 1/\sqrt{2} \approx 0.70$ at $\kappa = 0$. This means that if one observes the WF moment less than 70% of that in the LTO phase, $\Omega$ must be smaller than 0.5. The WF moment of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ below $T_s$ is only $\approx 30\%$ of that just above $T_s$ as we indicate by the position of $M_F/(SD/2J) \approx 0.3$ in Fig. 4. Therefore, to produce such small WF moment, $\kappa$ should be limited to the region $0 < \kappa < \pi/20$. This suggests that our sample is in the LTO2 phase even though the rotation angle is close to that for the LTT phase.

It should be pointed out that our identification of the structural symmetry is different from the reported symmetry for (La,Eu)$_2$CuO$_4$\textsuperscript{31,32} which was suggested to be the LTT phase at the lowest temperature. However, to our knowledge, there has been no conclusive structural analysis for the low-temperature phase of (La,Eu)$_2$CuO$_4$. Since the symmetry below $T_s$
was not important in many studies on (La,Eu)$_2$CuO$_4$, peoples have not paid attention to the difference between the LTO2 and LTT phases. What we found in the present work is that the spin canting out of the CuO$_2$ plane is quite sensitive to the structural difference between these phases that is the reason why we can distinguish the LTO2 phase from the LTT phase. We also emphasize that Ω is definitely smaller than 0.5 for (La,Eu)$_2$CuO$_4$. This is consistent with the conclusion by Stein et al.\textsuperscript{23} where they estimated Ω to be 0.10 from several microscopic parameters.

As a result, we arrive at a conclusion that the most probable magnetic structure for LTO2 (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ should be as shown in Fig. 5 where spins are parallel to neither [100] nor [110] directions. This model implies a reduction of the effective inter-plane coupling, which explains the increase of $\chi_c$ below $T_s$. The inter-plane coupling is determined by a subtle balance of the exchange interactions mainly between spins at (0, 0, 0) and (a/2, 0, c/2) and that between spins at (0, 0, 0) and (0, b/2, c/2). Since the above two interactions are not equivalent in the LTO phase\textsuperscript{35}, the effective inter-plane coupling remains finite. This inequivalence should decrease according to the symmetry change from the LTO to the LTO2 phases, and finally disappear in the LTT phase. The effective coupling between spins at the neighboring layers then decreases, and spins become more susceptible to the magnetic field applied along the $c$ axis. A more detailed discussion on the effect of inter-plane coupling has been already given by Viertio and Bonesteel.\textsuperscript{22} It is most likely that the reduction of the effective inter-plane coupling is the main reason why $\chi_c$ increases below $T_s$. It should be noted that the Van-Vleck susceptibility of Eu$^{3+}$ ions can also contribute to the rapid increase of $\chi_c^{5k}$ below $T_s$, because the oxygen coordination around Eu$^{3+}$ ions is modified across this temperature. However, this effect is sufficiently small as is suggested by the $c$-axis susceptibility data of (La$_{1.05}$Eu$_{0.2}$Sr$_{0.15}$)CuO$_4$\textsuperscript{36} and, therefore, one may safely attribute the large increase of $\chi_c^{5k}$ below $T_s$ to the response from CuO$_2$ planes.

C. Implication for the magnetic structure in the LTT phase

Finally we briefly discuss the likely magnetic structure of the LTT phase. Our results indicate that $\Omega < 0.5$. This parameter implies that the WF moment should disappear in the LTT phase ($\kappa = 0$). Thus the magnetic structure of the LTT phase is expected to be consistent with that shown in Fig. 1(b), but is inconsistent with that shown in Fig. 1.
(c). A natural next step is to find a system that exhibits a true LTT symmetry without doped carriers, in which we may expect a perfect disappearance of the effective inter-plane couplings. Unfortunately, suitable compound possessing a true LTT symmetry and AF long-range order has not been found yet. However, we believe that we can find a better system other than (La,Eu)$_2$CuO$_4$ in near future, which will be useful for the study of pure two-dimensional $S=1/2$ AF spin systems.

IV. SUMMARY

To summarize, we have investigated the magnetic susceptibility of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$. The antiferromagnetic long-range order typical for the LTO phase establishes below $T_N = 265$ K, where a finite spin canting out of the CuO$_2$ plane appears. In contrast to Eu-free La$_2$CuO$_4$, (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ shows an additional structural phase transition at $T_s = 130$ K, below which the spin canting is suppressed by 70%. All the results are consistently explained by the theory given by Stein et al.,$^{23}$ in which the KSEA interactions are taken into account together with the DM interactions. According to this theory, the spin arrangement for the LTO2 phase is determined. Furthermore, we observe a reduction of the effective inter-plane coupling below the structural transition temperature, which is probably one of the keys to understand the various anomalies found in the LTT phase of (La,Nd,Sr)$_2$CuO$_4$.

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FIG. 1: (a) Magnetic structure of the LTO phase, where arrows show the spins on Cu$^{2+}$ ions, and dashed lines show rotation axes of the CuO$_6$ octahedra. (b,c) Two magnetic structures proposed for the LTT phase, without spin canting (b) and with spin canting (c).
FIG. 2: (a) Temperature dependence of $\chi_c$ of (La$_{1.8}$Eu$_{0.2}$)CuO$_4$ at $H = 5$ kOe. The structural phase-transition temperature $T_s$ is taken from published data$^{31,32}$. (b) A comparison of $\chi_c$ and $\chi_{110}$ at $H = 5$ kOe.
FIG. 3: (a) Temperature dependence of $\chi_c$ at magnetic fields below $H_c$ (5 kOe) and above $H_c$ (70 kOe). $\chi_c$ at $H = 70$ kOe ($\chi_{c70k}^c$) shows a hysteresis between ZFC and FC data below 20 K. Inset to (a): Field dependence of the magnetization along the $c$ axis at $T = 50$ K. Two dotted lines are linear fits to the low- and high-field magnetizations. (b) Temperature dependence of $\chi_{c70k}^c - \chi_{c5k}^c$. 

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FIG. 4: Theoretical calculation of the WF moment by Stein et al.\textsuperscript{23} Horizontal axis is the angle between the Cu-O-Cu bond direction and the rotation axis of CuO$_6$ octahedra: $\kappa = 0$, $0 < \kappa < \pi/4$, and $\kappa = \pi/4$ correspond to the LTT, LTO2, and LTO phases, respectively. Vertical axis is the WF moment normalized by the magnitude of spin, Heisenberg interaction, and DM interaction. The only tunable parameter, $\Omega$, qualitatively gives the relative magnitude of the DM and KSEA interactions. For any $\Omega$ values, the WF moment in the LTO2 phase is smaller than that in the LTO phase. $\Omega = 0.5$ line gives the boundary: The WF moment completely disappears in the LTT phase when $\Omega < 0.5$, while the system recovers the WF moment in the LTT phase when $\Omega > 0.5$. The line for $\Omega = 0$ gives the situation that formerly discussed by Bonesteel.\textsuperscript{12} The shaded band shows the position of the WF moment observed in this work.
FIG. 5: Magnetic structure for the LTO2 phase of \((\text{La}_{1.8}\text{Eu}_{0.2})\text{CuO}_4\), which is considered as an intermediate state between the LTO (Fig. 1(a)) and the LTT (Fig. 1(b)) phases. In this state, a finite spin canting remains, but its angle is largely suppressed from that in the LTO phase.