Dzyaloshinsky–Moriya Spin Canting in the LTT Phase of La$_{2-x-y}$Eu$_x$Sr$_y$CuO$_4$

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The Cu spin magnetism in La$_{2-x-y}$Eu$_x$Sr$_y$CuO$_4$ ($x \leq 0.17; y \leq 0.2$) has been studied by means of magnetization measurements in fields up to 14 Tesla. Our results clearly show that in the antiferromagnetic phase Dzyaloshinsky–Moriya (DM) superexchange causes Cu spin canting not only in the LTO phase but also in the structural low-temperature phases LTLO and LTT. In La$_{1.8}$Eu$_{0.2}$CuO$_4$ the canted DM-moment is about 50% larger than in pure La$_2$CuO$_4$ which we attribute to the larger octahedral tilt angle in the Eu-doped compound. We also find clear evidence that the size of the canted DM-moment does not change significantly at the structural transition at $T_{LT}$ from LTO to LTLO and LTT. The most important change induced by the transition is a significant reduction of the magnetic coupling between the CuO$_2$ planes. As a consequence, the spin-flip transition of the canted Cu spins which is observed in the LTO phase for magnetic field perpendicular to the CuO$_2$ planes disappears in the LTT phase. The shape of the magnetization curves changes from the well known spin-flip type to a weak-ferromagnet type. However, no spontaneous weak ferromagnetism is observed even at very low temperatures, which seems to indicate that the interlayer decoupling in our samples is not perfect. Nonetheless, a small fraction ($\lesssim 15\%$) of the DM-moments can be remanently magnetized throughout the entire antiferromagnetically ordered LTLO/LT phase, i.e. for $T < T_{LT}$ and $x < 0.02$. It appears that the remanent DM-moment is perpendicular to the CuO$_2$ planes. For magnetic field parallel to the CuO$_2$ planes we find that the critical field of the spin-flop transition decreases in the LTT phase, which might indicate a competition between different in-plane anisotropies. To study the Cu spin magnetism in La$_{2-x-y}$Eu$_x$Sr$_y$CuO$_4$, a careful analysis of the Van Vleck paramagnetism of the Eu$^{3+}$ ions was performed.

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I. INTRODUCTION

In rare-earth (RE) doped La$_{2-x-y}$Sr$_y$CuO$_4$ the structural transition from the low-temperature orthorhombic phase (LTO) to the low-temperature tetragonal phase (LTT) has attracted a lot of attention because it causes a suppression of the superconducting ground state in favor of an incommensurate antiferromagnetic (AF) charge and spin stripe order
d$^1$2345. On the other hand, it is found that the low-temperature (LT) transition has a considerable impact on the AF order in insulating ($x = 0$) and lightly-Sr-doped compounds ($x \leq 0.02$). Though several experimental
d$^7$8.9.10.11.12.13 and theoretical
d$^{14}$15.16.17.18.19 studies have focused on the AF regime, important details of the spin structure in the LTT phase are still a matter of debate. In particular, the question in dispute is whether or not the magnetic groundstate in the LTT phase has a canted spin structure, as is the case in the LTO phase.

In the LTO phase of La$_2$CuO$_4$ the Néel temperature ($T_N \approx 325$ K) as well as the spin structure are determined by a combination of small anisotropic contributions to the superexchange, $J \approx 135$ meV, and a weak interlayer coupling, $J_{\perp} \approx 10^{-5} \times J$. As is shown in Fig.1(a), the resulting magnetic ground state is a collinear spin structure with Cu spins almost parallel to the b-axis (spacegroup $Bmab$) but slightly canted out-of-plane (|| c) by $\sim 0.2\degree$. Since for a particular layer all spins cant into the same direction each layer carries a weak ferromagnetic (WF) moment $\parallel$ Cu$^{2+}$ Spin canting originates from the Dzyaloshinsky–Moriya (DM) antisymmetric anisotropic superexchange, $J_{DM} \sim 10^{-2} \times J$, which is directly coupled to the tilting of the CuO$_6$ octahedra by an angle of the order of 5° at low temperatures.

In the LTO phase the gain in DM exchange energy, and hence the canting angle, is maximum for Cu spins perpendicular to the octahedral tilt axis $\parallel [100]$ [cf. Fig.1(b)]. In RE-doped La$_2$CuO$_4$ at the LT-transition the octahedral tilt axis rotates azimuthally by an angle $\alpha$, whereby axes in adjacent layers rotate in opposite directions. The question that arises is, whether the spins follow the rotation of the tilt axes to maintain a maximum gain in DM exchange energy [black spins in Fig.1(b)], or whether, as was predicted theoretically, they rotate in the opposite direction, resulting in a spin structure without spin canting [white spins in Fig.1(b)].

In this paper we present a detailed study of the Cu spin magnetism in the LTO and LTT phases of La$_{2-x-y}$Eu$_x$Sr$_y$CuO$_4$ in magnetic fields up to 14 Tesla. Our results unambiguously show that DM spin canting exists in the LTT phase. Furthermore, the size of the DM moments does not change at the structural transition LTO$\leftrightarrow$LTT within the error of the experiment. These findings support a spin structure in the LTT phase where, due to the DM superexchange, the Cu spins re-
main perpendicular to the octahedral tilt axis [black spins in Fig. 1(b)]. Compared to pure La$_2$CuO$_4$, the DM moment in La$_{1.8}$Eu$_{0.2}$CuO$_4$ is about 50% larger in both the LTO and LTT phases, which we attribute to a larger octahedral tilt angle. The most important difference between LTO and LTT is a significant reduction of the interlayer coupling in the LTT phase which macroscopically results in the disappearance of the spin-flip transition and in a considerable increase of the susceptibility and magnetization at low magnetic fields. To be able to directly compare the Cu spin magnetism in Eu-doped samples with pure La$_{2-x}$Sr$_x$CuO$_4$, we have put considerable effort into the careful analysis and subtraction of the Van Vleck magnetism of the Eu$^{3+}$ ions.

The paper is organized as follows: In Sec. II we provide some more detailed background knowledge. In Sec. III we briefly discuss the investigated stoichiometries, experimental details and relevant aspects of the crystal structure. In Sec. IV we determine the Van Vleck magnetism of the Eu$^{3+}$ ions, which was necessary to uncover the Cu spin magnetism. In Sec. V we present a general overview of the Cu spin magnetism as a function of Sr and Eu doping. Most of these measurements were performed at 1 Tesla. In the subsequent Sec. VI we focus on the Dzyaloshinsky–Moriya spin canting and the in-plane gap in pure La$_{1.8}$Eu$_{0.2}$CuO$_4$. We present the high-field magnetization up to 14 Tesla for a polycrystal and a single crystal. In Sec. VII we analyze the Dzyaloshinsky–Moriya spin canting in this pure compound. Resulting phase diagrams are discussed in Sec. VIII. In Sec. VII we show results for lightly-Sr-doped La$_{1.8}$Eu$_{0.2}$CuO$_4$ polycrystals ($0 \leq x \leq 0.02$) and directly compare them with pure La$_{2-x}$Sr$_x$CuO$_4$. A discussion is given in Sec. VIII.

II. BACKGROUND

It is well known that the azimuthal rotation of the octahedral tilt axis below the LT-transition can take values in the range $0^\circ < \alpha < 45^\circ$. In contrast, the octahedral tilt angle itself changes only very little at the transition. The LTT phase with spacegroup $P4_2/nmc$ is stabilized when $\alpha = 45^\circ$, while for $\alpha < 45^\circ$ the low-temperatureless-orthorhombic phase (LTLO) with spacegroup $Pccn$ is formed, which is an intermediate phase between LTO and LTT [cf. Fig. 1(b)]. In particular in lightly-Sr-doped samples containing excess-oxygen $\delta$, the LTLO phase with $\alpha$ much smaller than $45^\circ$ is formed. By reducing the excess-oxygen concentration, $\alpha$ increases, and for $\delta \to 0$ the LTLO structure approaches LTT. In particular, for $x = 0$ and $\delta$ just slightly larger than zero it was shown that the LT-transition is better described by a sequence of transformations: a discontinuous transition LTO$\rightarrow$LTLO, followed by a continuous transition LTLO$\rightarrow$LTT.

In the LTT phase the octahedral tilt axes are oriented alternately along [110] and [1$\bar{1}$0] directions in adjacent CuO$_2$ planes (cf. Fig. 1). As a result, the Cu-O-Cu bonds of a particular layer are buckled only in one crystallographic direction and the DM superexchange is active only for spin components pointing along the direction of buckling. Hence, for the LTT phase two principle magnetic ground states are possible: one with and another without DM spin canting, depending on whether the spins stay perpendicular (black spins) or parallel (white spins) to the octahedral tilt axis. Similarly, in the LTLO phase the spins are assumed to be either perpendicular to the tilt axis or rotated by an angle of $2\alpha$ relative to the
perpendicular spin direction. Which of these spin structures (white/black spins) represents the ground state in the LTLO and LTT phase is the subject of this paper.

Early magnetization measurements on La$_{2-y}$Nd$_y$CuO$_4$ and La$_{2-y}$Sm$_y$CuO$_4$ revealed a remanent moment $M_{REM}$ in the LTT phase. This result, in combination with neutron diffraction data, led the authors of Refs. 2 and 3 to the conclusion that DM spin canting exists in the LTT phase. It happens, however, that in La$_{2-y}$Nd$_y$CuO$_4$ $M_{REM}$ increases with increasing Nd content and shows a Curie-type temperature dependence. As a result, it has been questioned whether the weak ferromagnetism emerges from a Nd-Cu interaction. Neutron diffraction experiments by Keimer et al. on La$_{2-y}$Nd$_y$CuO$_4$ indicate that at the LT-transition the Cu spins follow the azimuthal rotation of the octahedral tilt axis, i.e., the spins remain perpendicular to the tilt axis (black spins in Fig. 1). Furthermore, the authors argue that in the LTT phase the tetragonal crystal symmetry, in combination with the non-collinear spin structure, should lead to a frustrated interlayer exchange. Magnetization measurements of La$_1.8$Eu$_{0.2}$Sr$_2$CuO$_4$ polycrystals with $0 \leq x \leq 0.02$ in Ref. 10 are in general agreement with the conclusions of the neutron diffraction study. Since non-magnetic Eu$^{3+}$ was used instead of magnetic RE$^{3+}$, these measurements rule out the possibility that the observed LTT ground state with spin canting is the result of RE-Cu interactions. In contrast to these findings, recent magnetization data on a La$_{1.5}$Eu$_{0.5}$CuO$_4$ single crystal with $T_N = 265$ K were interpreted in favor of spin structures given by the white spins in Fig. 1, which means that DM spin canting is reduced in the LTLO phase and disappears in the LTT phase.

Several theoretical papers have addressed the problem of DM spin canting in the LTLO phase, and there is general agreement that the magnetic ground state depends on a delicate balance between various contributions to the in-plane and out-of-plane anisotropies, as well as the interlayer coupling. In particular, it was claimed that the in-plane spin direction depends on a competition between the DM interaction and a symmetric superexchange anisotropy, which in the case of RE-doped La$_2$CuO$_4$ is supposed to favor a LTT phase without spin canting (white arrows in Fig. 1). Although the most recent calculations have confirmed this result, there are serious discrepancies with experiment which remain to be understood. According to Shekhtman et al. in La$_2$CuO$_4$ the symmetric anisotropy would cause in-plane and out-of-plane spin-wave gaps of equal size, i.e., an Ising-type anisotropy. In contrast, neutron scattering experiments show that in La$_2$CuO$_4$ and several other layered cuprates the out-of-plane gap is larger than the in-plane gap and always of the order of 5 meV. Shekhtman et al. as well as other groups, suggest that this discrepancy follows from a contribution to the out-of-plane gap from direct exchange, which seems not to depend significantly on the ligand structure perpendicular to the CuO$_2$ plane. In particular, isostructural Sr$_2$CuO$_2$Cl$_2$ with flat CuO$_2$ planes has the same out-of-plane gap as La$_2$CuO$_4$ but a much smaller in-plane gap. This seems to show that octahedral tilting mainly tunes the in-plane-gap and, therefore, puts a very low limit on a possible contribution of the Ising-type symmetric anisotropy to both spin-wave gaps. From this perspective, one can say that the idea of the symmetric superexchange anisotropy dominating the antisymmetric DM superexchange is at least experimentally not well settled in the two dimensional (2D) spin $S = 1/2$ cuprate Heisenberg antiferromagnets.

### III. EXPERIMENT

The polycrystalline samples of La$_{2-x-y}$Eu$_x$Sr$_y$CuO$_4$ used in this study were prepared by a standard solid state reaction. Three series of samples with the following Sr and Eu concentrations have been investigated: one with fixed Eu content ($y = 0.2$) and various Sr dopings ($0 \leq x \leq 0.17$), and two others with fixed Sr concentrations ($x = 0.017$; 0.08) and various Eu dopings ($0 \leq x \leq 0.2$). For comparison, we will present results on a La$_{1.7}$Nd$_0.3$CuO$_4$ and several La$_{2-y}$Sr$_y$CuO$_4$ polycrystals. The La$_{1.8-x}$Eu$_{0.2}$Sr$_2$CuO$_4$ single crystals with $x = 0$ and 0.15 were grown using the traveling-solvent floating-zone method. The magnetization measurements were performed with two different magnetometers, a Faraday balance (4-330 K; 0-1 Tesla) as well as a vibrating-sample magnetometer (4-290 K; 0-14 Tesla). Sample masses have varied between 0.3 and 0.7 g. Some of the samples were also investigated with $\mu$SR, NQR, ESR, and x-rays. Very limited normal state magnetization data on the Eu$_{1.2}$-doped samples were previously published in Refs. 10, 12, 42. Throughout this paper, magnetization curves $M(H)$ are presented in units of $\mu_B$/Cu. To convert the data into units of G·emu/mol a factor of $1/1.79055 \times 10^{-4}$ has to be applied.

As was mentioned in Sec. 11, interstitial oxygen in samples with low Sr content has a considerable effect on the crystal structure. To remove the excess-oxygen, samples were annealed under reducing conditions. Most magnetization data on polycrystals were obtained after anneals for 3 d at 625°C in flowing N$_2$ gas. Two La$_{1.8-x}$Eu$_{0.2}$Sr$_2$CuO$_4$ specimens with $x < 0.02$, studied by x-ray diffraction, have shown a discontinuous LTO$\leftrightarrow$LTL0 transition at $T_{LTLO}$ $\sim$ 135 K and a continuous transition $LTLO \leftrightarrow$ LTT at around 60 K, which is well below $T_{LTLO}$. Later on, the magnetization of some polycrystals was remeasured after they were annealed for 1/2 h at 800°C in vacuum ($\lesssim 10^{-4}$ mbar), which results in lower values of $\delta$. These samples exhibit 10-20 K higher values of $T_N$ and very sharp structural and magnetic transitions. In the case of the polycrystal with $x = 0$, $T_N$ increased from 285 K to 316 K.

The smaller the RE ionic radius, the harder the structural ground state is pushed towards the LTT phase.
For Nd$^{3+}$ and Sm$^{3+}$-doped La$_{2-x}$Sr$_x$CuO$_4$ this has been shown by Crawford et al. in Ref. 31. Since Eu$^{3+}$ is even smaller, it induces the LTO$\leftrightarrow$LTLO transition already at a lower level of doping, and at increasing Eu content, the LTT phase is stabilized up to higher temperatures. Moreover, with respect to the average ionic radius at the La site, Eu$_0$2 is comparable to Sm$_{0.23}$ and Nd$_{0.36}$.

Since in our Eu doped $x = 0$ sample annealed at 800$^\circ$C the Néel temperature is comparable to the values in Ref. 31 (i.e. comparable $\delta$), we assume that the $T$ range below $T_{LT}$ in which the LTLO phase exists is even smaller than in La$_{1.8}$Sm$_{0.2}$CuO$_4$. Corresponding low temperature data for La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_4$ will therefore be discussed in terms of the LTT phase.

Similar arguments apply for our La$_{1.8}$Eu$_{0.2}$CuO$_4$ single crystal, which was studied first after annealing for 3 d at 625$^\circ$C in N$_2$ and a second time after annealing for 2 h at 800$^\circ$C in vacuum. It seems, though, that the oxygen content is slightly higher and less homogenous than in the La$_{1.8}$Eu$_{0.2}$CuO$_4$ polycrystal, possibly because of the longer diffusion path. Our susceptibility data indeed shows that after annealing at 625$^\circ$C, below $T_{LT}$ the crystal stays in the LTLO phase down to 4 K. After the second annealing at 800$^\circ$C it approaches the LTT phase at $T \approx 100 K$.

In our experience with La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_4$, excess-oxygen affects the LT-transition up to a Sr content as high as $x=0.08$, which is considerably higher than in pure La$_{2-x}$Sr$_x$CuO$_4$. For $x > 0.08$ it was confirmed by x-ray diffraction experiments that in as-prepared samples the LT-transition is of the LTO$\leftrightarrow$LT phase type$^{46}$. Where it is relevant, we will indicate the samples’ annealing history in the text.

**IV. ANISOTROPIC Eu$^{3+}$ VAN VLECK MAGNETISM**

An important reason for using europium is that Eu$^{3+}$ ions have a non-magnetic ground state $J = 0$, whereas other RE ions such as Nd$^{3+}$ ($J = 9/2$) and Sm$^{3+}$ ($J = 5/2$) have a magnetic ground state. Hence, by choosing Eu we avoid a possible impact of the RE–Cu interaction on the Cu spin magnetism. Eu$^{3+}$ ions exhibit Van Vleck-paramagnetism which is constant at low temperatures and only moderately changes at higher temperatures, as one can see in Fig. 2. In contrast, Nd$^{3+}$ ions exhibit a strongly temperature-dependent Curie-type susceptibility which makes it impossible to analyze the much weaker Cu-spin magnetism, as the comparison with La$_{2}$CuO$_4$ demonstrates. We mention that Sm-doped samples show a susceptibility that is even smaller than for Eu-doped samples$^{31}$.

In the following we briefly describe the analysis of the Eu$^{3+}$-Van Vleck-paramagnetism, $\chi_{VV}(\text{Eu})$, which we will then subtract from the total susceptibility to uncover the Cu-spin magnetism. The analysis of $\chi_{VV}(\text{Eu})$ was performed for $x = 0.15$, since at this Sr content the magnetic contribution of the CuO$_2$ planes is less complicated than in the AF phase at low $x$. In Fig. 3 we show the susceptibility of a polycrystal (left) and for all three direc-

**FIG. 2:** Static magnetic susceptibility ($H = 1$ Tesla) of pure and RE-doped La$_2$CuO$_4$ with RE = Nd$_{0.3}$ and Eu$_{0.2}$.

**FIG. 3:** Static magnetic susceptibility ($H = 1$ Tesla) of La$_{1.65}$Eu$_{0.2}$Sr$_{0.15}$CuO$_4$. Left: comparison of polycrystal data with averaged single crystal data as well as the susceptibility of a La$_{1.85}$Sr$_{0.15}$CuO$_4$ polycrystal. $T_{LT}$ indicates the LTO$\leftrightarrow$LT transition, $T_C$ the SC transition in the pure compound. Right: susceptibility of the single crystal for all three directions.
tions of a single crystal (right). In the left hand plot we show also the averaged susceptibility of the single crystal, which almost perfectly agrees with the polycrystal data. Compared to pure La$_{1-x}$Sr$_x$CuO$_4$ the susceptibility of the Eu$_{0.3}$-doped polycrystal is about one order of magnitude larger, due to the dominant contribution of $\chi_{VV}(\text{Eu})$. At low temperatures the susceptibility is constant, and for $T \lesssim 70$ K it decreases monotonically. In the case of the single crystal, $\chi_{VV}(\text{Eu})$ shows a strong crystal-field anisotropy, which we have already described in Ref. 42. The susceptibility for magnetic field $H \parallel c$ is much smaller than for $H \parallel ab$ and increases with increasing temperature. The structural transition LTO$\rightarrow$LTT occurs at $T_{LT} \approx 135$ K and is only visible for $H \parallel ab$-plane. It shows up due to a small $ab$-anisotropy in the LTO phase that is macroscopically eliminated in the LTT phase. As was explained in Ref. 42, the crystal direction which shows the negative (positive) step at $T_{LT}$ with decreasing temperature predominantly contains the $a$-axis ($b$-axis) and is therefore called $\chi^a$ ($\chi^b$) in terms of spacegroup $Bmab$. For $H \parallel c$, no signature at $T_{LT}$ is observed, as was also checked for crystals with $x = 0.04$, 0.08, 0.12, and 0.22 In the case of the polycrystal, the LT-transition causes only a very small anomaly, as will be discussed later (see also Ref. 48). The dotted lines in Fig. 4 are fits according to the following expression:

$$\chi_{fit}^{ab} = \chi_{LSCO}^{ab} + \chi_{VV}^{ab}(\text{Eu}, y^*)$$

where $\chi_{LSCO}$ is the susceptibility of pure La$_{2-x}$Sr$_x$CuO$_4$ and $y^*$ is the Eu-fraction as determined from the fit. For $H \parallel ab$ we have fitted the average $ab$-plane susceptibility $(\chi^a + \chi^b)/2$. To account for $\chi_{LSCO}$, we have approximated the single crystal data in Ref. 51. The Van Vleck magnetism (solid lines) was calculated using a similar approach as in Ref. 52, which we have improved so that $\chi_{VV}(\text{Eu})$ is correctly described at higher temperatures which was a major problem in Ref. 52 and 53. Details of the calculation will be presented elsewhere. The fits provide an almost perfect description of the data which suggests that we have accurate expressions for $\chi_{VV}(\text{Eu})$. Note, that this is an important ingredient for our analysis of the Cu-spin magnetism in the AF phase for $x \leq 0.02$, where the determination of $\chi_{VV}(\text{Eu})$ is considerably complicated by magnetic transitions of the Cu spins at $T_N$ and $T_{LT}$.

As an example for a sample in the AF phase, we show in Fig. 4 (a) the susceptibility $\chi$ of a La$_{1.8}$Eu$_0.2$CuO$_4$ polycrystal ($x = 0$). Two transitions are visible: the Néel order transition at $T_N = 316$ K and the LTO$\rightarrow$LTT transition at $T_{LT} = 133$ K. The solid line is the Van Vleck fit that we have subtracted to obtain $\chi - \chi_{VV}(\text{Eu})$. In Fig. 4 (b) we compare $\chi - \chi_{VV}(\text{Eu})$ with the susceptibility of La$_2$CuO$_4$. The Eu fraction $y^*$ was varied so that the curves coincide in the LTO phase at 140 K, which obviously causes coincidence in a broad temperature range of the LTO phase. This procedure yields a Eu fraction of $y^* = 0.195(5)$ which matches the nominal concentration of $y = 0.2$ within the experimental error. At the LT-transition $\chi - \chi_{VV}(\text{Eu})$ shows a step-like increase which leads to deviations of the order of $0.5 \times 10^{-4}$ emu/mol from the susceptibility of La$_2$CuO$_4$. At low temperatures both curves show a small Curie-type increase which indicates a small number of free spins. As this contribution is of same magnitude in both samples, we can exclude that Eu doping leads to a significant increase of free moments or magnetic impurities such as Eu$^{2+}$ ions. In Fig. 4 (b) we also show the susceptibility $\chi_{2DHAFF}$ of a spin $S = 1/2$ 2D Heisenberg antiferromagnet for $J = 1550$ K. To

FIG. 4: Static susceptibility ($H = 1$ Tesla) of a La$_{1.8}$Eu$_0.2$CuO$_4$ polycrystal. (a) Total susceptibility $\chi$, calculated Eu Van Vleck susceptibility $\chi_{VV}(\text{Eu})$, as well as difference $\chi - \chi_{VV}(\text{Eu})$. The dotted lines indicate the antiferromagnetic transition at $T_N$ and the structural transition at $T_{LT}$. (b) Comparison of $\chi - \chi_{VV}(\text{Eu})$ with $\chi$ of pure La$_2$CuO$_4$. Data in (b) corrected for $\chi_{DM} = -0.99 \times 10^{-4}$ emu/mol and $\chi_{VV}(\text{Cu}) = 0.43 \times 10^{-4}$ emu/mol. (□) susceptibility $\chi_{2DHAFF}$ of a $S1/2$-2D-HAF for $J = 1550$ K (after Monte Carlo data in Ref. 54).
FIG. 5: Static susceptibility ($H = 1$ Tesla) of La$_{1.8-x}$Eu$_x$Sr$_x$CuO$_4$ polycrystals for different Sr concentrations $0 \leq x \leq 0.02$ after subtraction of $\chi_{V V}(\text{Eu})$. Except for $x = 0$ (800 °C vac.) all samples were annealed at 625 °C in N$_2$ gas. Curves are shifted for clarity by $1 \times 10^{-4}$ emu/mol.

compare $\chi_{2DHAF}$ to the measured curves, the latter ones have been corrected for the core diamagnetism $\chi_{dia}$ and the Van Vleck magnetism $\chi_{V V}(\text{Cu})$ of the Cu$^{2+}$ ions.$^{26,27}$ In the LTO phase, the difference $\chi_{DM}$ between the data and $\chi_{2DHAF}$ follows from the DM spin canting. Without spin canting, the susceptibility in both the LTO and LT phase would be very close to $\chi_{2DHAF}$, as is the case at high temperatures in the high temperature tetragonal (HTT) phase with flat CuO$_2$ planes. In contrast, in the LT phase the contribution of $\chi_{DM}$ even increases, which clearly indicates that DM spin canting cannot be absent for $T < T_{LT}$.

V. OVERVIEW OF Sr AND Eu DOPING DEPENDENCE

In this section we present an overview of the evolution of $\chi - \chi_{V V}(\text{Eu})$ as a function of Eu and Sr doping. All data in this section are after subtraction of the Eu magnetism and were collected on polycrystals. In Fig. 6 we show data for La$_{1.8-x}$Eu$_x$Sr$_x$CuO$_4$ with variable Sr content $0 \leq x \leq 0.02$. Doping with Sr leads to a continuous decrease of $T_N$, while the LT-transition remains at $T_{LT} \sim 130$ K. In the LTO phase, the decrease of $T_N$ can easily be followed by the position of the Néel peak. However, the Néel peak disappears as soon as $T_N$ reaches the structural transition temperature, which happens at a Sr content $x = 0.016$. The size of the step in $\chi$ at $T_{LT}$ is about the same as long as $T_N \gtrsim T_{LT}$, but then decreases and eventually vanishes for $x = 0.02$. This doping dependence shows that the increase of $\chi - \chi_{V V}(\text{Eu})$ at the LT-transition is connected with the presence of long range AF order.

Data for $x \geq 0.018$ are shown in Fig. 6 (a) and (b). Obviously, the step in $\chi$ at $T_{LT}$ changes sign, then again decreases and finally vanishes at high Sr concentrations $x \gtrsim 0.17$. In Fig. 6(c) the data are shown on an enlarged scale after subtraction of a linear fit applied immediately above the LT-transition. In this figure one can clearly see the sign change of the anomaly at the structural transition at about $x = 0.02$.

To obtain deeper insight, we have studied the special case $x = 0.02$ in high magnetic fields. Corresponding data in Fig. 6 show that with increasing field a negative anomaly of the same kind as for $x > 0.02$ is uncovered. We assume that at $x = 0.02$ the positive jumps observed for $x < 0.02$ just compensate the negative jumps observed for $x > 0.02$. In Ref. 48 we have argued that the negative jumps for $x > 0.02$ may signal an increase of the 2D magnetic correlation length in the LT phase. A similar
jump in $\chi$ has been observed at the charge stripe order transition in the nickelates. In the nickelates, stripe order is not induced by a structural transition; i.e., the jump in $\chi$ is certainly connected to stripe order. In the cuprates the situation is less clear since static stripe order occurs at much lower temperatures than the LTO$\leftrightarrow$LTT transition. Since the negative jump seems to vanish before the structural phase boundary between the LTO and HTT phase at $x \simeq 0.25$ (for $T = T_{LT}$) is reached, the size of the jump is possibly coupled to the octahedral tilt angle.

In addition we have studied two series of samples as a function of the Eu content $y$. The Sr content was fixed at $x = 0.017$, which is in the AF phase, and at $x = 0.08$, which is in the underdoped superconducting phase. For $x = 0.017$ the LT-transition appears first at $y = 0.05$ and $T_{LT} \simeq 50$ K. With increasing Eu content, $T_{LT}$ as well as the size of the jump in $\chi$ increases rapidly. At Eu content of $y = 0.2$, $T_{LT}$ reaches the magnetic transition at $T_{N}$ which appears to be independent of the Eu content. As is well known, for small $y$ the structural transition at $T_{LT}$ is of the LTO$\leftrightarrow$LTLO type rather than of the LTO$\leftrightarrow$LTT type. We assume that the increase of the jump in $\chi$ with increasing $y$ is connected to a gradual change of the transition from LTO$\leftrightarrow$LTLO to LTO$\leftrightarrow$LTT. In the second series of samples with $x = 0.08$, the LT-transition appears first at $y = 0.07$ and $T_{LT} \simeq 62$ K as a small negative jump in $\chi$. With increasing $y$, $T_{LT}$ shifts to higher temperatures. The size of the anomaly, however, does not change significantly. Below $T_{LT}$ the slope $d\chi/dT$ shows a considerable change with increasing $y$. Although for $T \gtrsim 70$ K this may partially be due to a small inaccuracy in the subtraction of $\chi_{VV}(\text{Eu})$, we emphasize that for $T \lesssim 70$ K the slope is real, as $\chi_{VV}(\text{Eu})$ is constant. Fig. (b) also shows that the onset temperature $T_{c}$ of the superconducting transition (at 1 Tesla) is suppressed from 34 K at $y = 0$ to below 4 K at $y = 0.2$. In contrast, $\mu$SR experiments have shown that in the Eu$_{0.2}$-doped polycrystal, static magnetic (stripe) order occurs at $T_{N} \simeq 6$ K, whereas in pure La$_{2-x-y}$Sr$_{y}$CuO$_{4}$ with $x=0.08$ this transition occurs at the much lower temperature of 2.5 K. One might wonder whether the steep increase of the susceptibility of the Eu$_{0.2}$-doped sample indicates the magnetic transition. However, no such upturn is observed in our Eu$_{0.2}$-doped single crystal with $x = 0.08$, showing that polycrystals in general seem to have a larger number of magnetic defects. All critical temperatures are summarized in Fig. (c) and (d). It is obvious that in spite of the quite different Sr content, $T_{LT}(y)$ is almost identical.

VI. PURE La$_{1.8}$Eu$_{0.2}$CuO$_{4}$

In this section we present the high-field magnetization of La$_{1.8}$Eu$_{0.2}$CuO$_{4}$ ($x = 0$) obtained for a polycrystal (Sec. VI A) and for a single crystal. The polycrystal was annealed at 800°C in vacuum ($T_{o} = 316$ K). Its sus-
FIG. 9: Static susceptibility of La$_{1.8}$Eu$_{0.2}$CuO$_4$ after subtraction of $\chi_{VV}$ (Eu) and pure of La$_2$CuO$_4$ for different magnetic fields $H$. For comparison 1 Tesla data shown in all plots. Data corrected for $\chi_{dia}$ and $\chi_{VV}$ (Cu) (cf. Fig. [4]). Dashed line: susceptibility of $S_{1/2}$-2D-HAF for $J = 1550$ K (after Ref. [5]).

The susceptibility at 1 Tesla was already presented in Fig. [1]. The results for the single crystal annealed at 800°C ($T_N = 315$ K) will be presented in Sec. [VI B] and those for the crystal after annealing at 625°C ($T_N = 280$ K) in Sec. [VI C].

A. The La$_{1.8}$Eu$_{0.2}$CuO$_4$ polycrystal

In Fig. [9] we compare the susceptibility of the La$_{1.8}$Eu$_{0.2}$CuO$_4$ polycrystal after subtraction of $\chi_{VV}$ (Eu) and the susceptibility of a La$_2$CuO$_4$ polycrystal for magnetic fields up to 14 Tesla and temperatures up to 270 K. Data were corrected for $\chi_{dia}$ and $\chi_{VV}$ (Cu) (cf. Fig. [4]). In the LTO phase, with increasing $H$, a significant increase occurs for both samples. In contrast, in the LTT phase of the Eu-doped compound changes are rather weak. In the LTO phase, the influence of the field is particularly strong between 3 and 6 Tesla and follows from the first order spin-flip transition that can be induced for $H || c$. As is shown in Fig. [9] (a), at this transition the DM moments of adjacent CuO$_2$ planes, which are antiferromagnetically coupled in zero magnetic field, become ferromagnetically

FIG. 10: Magnetization $M(H)$ of La$_{1.8}$Eu$_{0.2}$CuO$_4$. (a) $M(H)$ and $M - M_{VV}$ (Eu) at 150 K. $M_{VV}$ (Eu) is the calculated Van Vleck-term. (b),(c) $M - M_{VV}$ (Eu) at different temperatures from measurements with increasing and decreasing magnetic field, respectively. $M_{SF}$ and $M_{B}$ indicate spin-flip and weak ferromagnetic contribution of DM moment, respectively.
We show a selection of measurements with increasing (○) and decreasing (●) magnetic field. (a) at $T = 150$ K in comparison with pure La$_2$CuO$_4$. Curves are shifted for clarity.

In the LTT phase, the spin-flip transition is obviously very weak or absent. It is also interesting that the jump at $T_{LT}$ drastically decreases, changes sign and almost vanishes at 14 Tesla. Since in the LTO phase the critical field $H_c$ increases with decreasing $T$, at intermediate fields of $\sim$ 4 Tesla the spin-flip in La$_2$CuO$_4$ at high temperatures has progressed to a higher degree than at low temperatures. In contrast, at maximum field the DM moments in La$_2$CuO$_4$ are ferromagnetically aligned down to 4 K (to the degree that is possible in a polycrystal). The susceptibility is smooth and increases monotonically with decreasing $T$ and, moreover, is significantly larger than $\chi_{3DHF}$. A very similar temperature dependence is observed for the Eu-doped sample at 14 Tesla. We therefore conclude that in La$_{1.8}$Eu$_{0.2}$CuO$_4$ the DM-moments are ferromagnetically aligned in both the LTO and the LTT phases, though the higher susceptibility indicates that the DM-moments are even larger than in La$_2$CuO$_4$. Furthermore, the absence of a jump at $T_{LT}$ at 14 Tesla implies that the size of the DM-moment does not change significantly at the structural transition LTO$\leftrightarrow$LTT. We note that it is very difficult to understand this result in terms of a LTT phase without DM spin canting as was suggested in Ref. [1].

In Fig. 11 we show a selection of $M(H)$ curves above and below the LT-transition. Though in principle $\chi(T)$ provides exactly the same information as $M(H)$, information about the spin-flip is accessible more directly via $M(H)$. Extracting this information is, however, a challenge. For example, in the case of the $M(H)$ curve at 150 K in Fig. 11(a), the magnetic contribution at 14 Tesla from the spin-flip amounts to 3% of the total signal, while that of Eu accounts for 92% [18]. In Fig. 11(b) and (c) we show $M - M_{VV}$(Eu) curves measured with increasing and decreasing magnetic field at temperatures above and below $T_{LT} \approx 133$ K. The dashed line in this figure represents the sum of all contributions that are approximately linear in magnetic field. The contribution of the DM moments $M_{DM}$ in a polycrystal is actually described by a function $f(M_{DM})$ [19]. In the LTO phase we observe well defined spin-flips; i.e., the magnetization shows a step like increase at the critical spin-flip field $H_c$. The critical field increases with decreasing temperature. In the LT phase the spin-flip rapidly decreases and in particular for $dH/dT < 0$, it is almost absent for $T < 130$ K. However, this figure unambiguously shows that the magnetic contribution due to the DM spin canting does not vanish in the LT phase. It rather transforms from an antiferromagnetic-type to a ferromagnetic-type magnetization of the DM moments.

The drastic changes that occur at $T_{LT}$, in particular between 135 K and 130 K, are also clearly seen in the derivative $dM/dH$ shown in Fig. 11(a). In the LT phase, the spin-flip is indicated by a pronounced maximum, which disappears in the LTT phase. This figure also shows that $M(H)$ becomes strongly hysteretic below 250 K. In contrast, in La$_2$CuO$_4$ the $M(H)$ curves stay reversible down to 150 K as is displayed in Fig. 11(b) and was reported in Ref. [20]. It is reasonable to attribute the enhanced hysteretic behavior in the Eu-doped compound to a local lattice distortion around the rare-earth site.

B. La$_{1.8}$Eu$_{0.2}$CuO$_4$ single crystal (800°C vacuum)

In the following we present the results on the La$_{1.8}$Eu$_{0.2}$CuO$_4$ single crystal after it was annealed at 800°C in vacuum. In Fig. 12 we show the total susceptibility [plot (a)] as well as the susceptibility after subtraction of the anisotropic Eu Van Vleck magnetism [plot (b)]. The solid lines in Fig. 12(a) are the calculated Eu Van Vleck contributions. The Eu fraction $y^*$ was chosen so that after subtraction of the Eu Van Vleck term the resulting curves in Fig. 12(b) are in fair agreement with the susceptibility of pure La$_2$CuO$_4$ (cf. Ref. 11). In this way we have determined the Eu content of the crystal to $y = 0.185(5)$. Note, that there is no Curie-like upturn of $\chi$ at low temperatures, documenting the absence of a significant number of magnetic impurities, defect Cu spins or Eu$^{2+}$ ions.

The two transitions at $T_N$ and $T_{LT}$ are clearly visible in all three crystal directions (Fig. 12(b)). The LT-transition shows a temperature hysteresis of 10 K, i.e., $T_{LT} = 134$ K for increasing $T$ and 124 K for decreasing $T$. The Néel peak and the jump at $T_{LT}$ are largest for $H \parallel c$. For $H \parallel ab$ we find a similar in-plane anisotropy as for La$_{1.65}$Eu$_{0.25}$Sr$_{0.15}$CuO$_4$ in Fig. 3(b), which largely follows from the crystal field anisotropy of $\chi_{VV}$(Eu) in the
FIG. 12: Static susceptibility ($H = 1$ Tesla) of La$_{1.8}$Eu$_{0.2}$CuO$_4$ for magnetic fields perpendicular ($\chi^c$) and parallel to the CuO$_2$ planes ($\chi^{ab}$, $\chi^{b*}$). (a) total signal from measurements with decreasing temperature. (—) anisotropic Eu Van Vleck susceptibility. (•) polycrystal average. (b) after subtraction of $\chi_{VV}^{c}$ (Eu) for increasing (•) and decreasing (○) temperature. (+) Average of the two $ab$-measurements with decreasing $T$. In the shaded temperature range a mixed phase of LTO, LTLO, and LTT may exist.

LTO phase. Largely means that (in contrast to $x = 0.15$) for $x = 0$ one has to take into account that a small fraction of the $ab$-anisotropy comes from the CuO$_2$ planes. The appearance of the $ab$-anisotropy in the LTO phase clearly indicates that the crystal is partially detwinned. Below $T_{LT}$ the $ab$-anisotropy abruptly decreases to a finite value and then decreases continuously with decreasing temperature. For $T \lesssim 100$ K the $ab$-susceptibility eventually becomes isotropic within the error of the experiment, i.e. the structure becomes LTT. (Note that the remaining anisotropy of $\sim 0.1 \times 10^{-4}$ emu/mol amounts to $\sim 0.5\%$ of the total signal, only.) The finite $ab$-anisotropy in the intermediate temperature range $100$ K $< T < T_{LT}$ (shaded in) can have different reasons. One is that the transition actually consists of a sequence of transitions: LTO $\rightarrow$ LTLO $\rightarrow$ LTT. On the other hand one has to take the temperature dependence of the LTO phase fraction into account.

At this point we mention that after the first annealing procedure at 625$^\circ$C the crystal was detwinned to a much higher degree, as one can see from the apparently larger $ab$-anisotropy in Fig. 13(a) (which we will discuss in detail later). Moreover, also the Néel peak in Fig. 13 shows a much stronger $ab$-anisotropy than in Fig. 12(b), indicating a lower degree of twinning, too. From pure La$_2$CuO$_4$ it is well known that the Néel peak is largest for $H \parallel c$ and $H \parallel b$ and that there is no peak for $H \parallel a$. From these facts we infer that, in the LTO phase, $\chi_{VV}^{c}$ (Eu) $> \chi_{VV}^{b}$ (Eu). Note, that the average in-plane susceptibility (+) in Fig. 12(b) does not show a significant anomaly at $T_{LT}$. Obviously the jump occurs only for $H \parallel c$, which indicates that it is connected to the DM spin canting.

To study the DM spin canting we have performed mea-
measurements in high magnetic fields with \( H \parallel c \). Fig. 13 shows the total susceptibility, the Eu Van Vleck term and the susceptibility after subtraction of this term. Note that for \( H \parallel c \) the ratio between the Cu spin magnetism and the magnetic background due to the Eu ions is much more favorable than in the polycrystal. Moreover, the jump at \( T_{LT} \) is significantly larger, since the signal is not averaged over all directions. It is as well about twice as large as in Ref. [11] for a single crystal with \( T_N = 265 \text{ K} \), underlining the strong impact of the excess-oxygen on the intrinsic properties of the stoichiometric system (and hence the importance of annealing under reducing conditions).

The left panel of Fig. 14 shows that there is a strong field dependence in the LTO phase due to the spin-flip transition for \( H \leq 6 \text{ Tesla} \), which is similar to the polycrystal. In the LTT phase, the field dependence is remarkably weak. The jump at \( T_{LT} \) decreases, but does not change sign, and finally vanishes around \( 8 \text{ Tesla} \). As is shown in the right panel at higher fields, \( H > 6 \text{ Tesla} \), \( \chi^c - \chi^V(V) \) decreases in the LTO as well as the LTT phase. The smooth temperature dependence of \( \chi^c - \chi^V(V) \) at 14 Tesla, in particular around \( T_{LT} \), again shows that DM spin canting exists in the LTT phase and that the size of the DM moment does not change at the transition.

In Fig. 14, we show a selection of \( M^c(H) \) curves for \( H \parallel c \) for increasing as well as decreasing field. In the LTO phase we observe well defined spin-flips, while below \( T_{LT} \) they become weaker and finally vanish. Similar to the polycrystal, the shape of the \( M^c(H) \) curves transforms from an antiferromagnetic-type to a weak ferromagnetic-type. Moreover, at low temperatures \( M^c(H) \) curves become strongly hysteretic and show a significant remanent moment.

The inset shows non-shifted data for increasing field. The dashed line approximates the linear magnetic background \( H \chi_0^c \). Apparently, upon decreasing the temperature the \( M^c(H) \) curves in the LTT phase are the envelope of the curves at higher temperature; i.e., the contribution of the DM moments neither vanishes nor decreases. Note that in the LTO phase the step in \( M^c(H) \) due to the spin-flip is significantly larger than in the case of the polycrystal, since in the crystal for \( H \parallel c \) the full DM moment can be aligned (see next paragraph). Another thing to mention is that the slope \( dM^c/dH \) at low magnetic fields significantly increases for \( T < T_{LT} \), which explains the jump in \( \chi(T) \) at \( T_{LT} \). Below \( \sim 100 \text{ K} \), however, \( dM^c/dH \) becomes temperature independent, i.e. it does not steepen further as is typical for a classical ferromagnet. In particular at very low temperatures \( M^c(H) \) is almost linear up to \( \sim 6 \text{ Tesla} \) before it bends in a ferromagnetic fashion. In contrast, in La$_2$-yNd$_y$CuO$_4$ at 2 K a ferromagnetic-type curvature is already visible in \( M(H) \) curves with a maximum field as low as 1.2 Tesla, indicating that in this case it indeed seems to be the large \( 4f \) moment of Nd$^{3+}$ that shows a Brillouin-type magnetization.

In Fig. 15, we show the deviations \( \Delta M(H) \) from linearity for the \( M(H) \) curves at 150 K of the single crystal and of the Eu-doped, as well as pure, La$_2$CuO$_4$ polycrystals. It is obvious that the single crystal shows the largest step at the spin-flip transition. In an ideal single crystal, the spin-flip causes a discontinuous increase of \( \Delta M \) by \( M^F \), where \( M^F \) is the antiferromagnetically-ordered fraction of the DM moment. In a polycrystal, \( \Delta M \) increases continuously and converges to \( M^F \) in the high field limit. If we multiply the data of the Eu-doped polycrystal by a factor of 2 we indeed obtain a fair agreement with the single crystal data. In the case of the La$_2$CuO$_4$ sample we have to apply a factor of three. Consequently, in the Eu-doped sample the size of the DM-moments is about 50% larger. At 150 K a more precise analysis yields \( M^F \approx 2.45 \times 10^{-3} \mu_B \) for the Eu-doped single crystal, \( 2.6 \times 10^{-3} \mu_B \) for the Eu-doped polycrystal and \( 1.8 \times 10^{-3} \mu_B \) for the La$_2$CuO$_4$ polycrystal (details in Sec. VI.D and VI.E). It is well known
that Eu-doping causes the high temperature structural transition HTT→LTO to shift to higher temperature, which means that the octahedral tilt angle at low temperature becomes larger. For a Eu content of $y = 0.2$ the HTT→LTO transition shifts from 525 K in pure La$_2$CuO$_4$ to about 700 K. To be more quantitative we have compared data from literature for orthorhombic strain and octahedral tilt angles for pure and RE-doped La$_2$CuO$_4$ [8,9,30,31,46,69,70,71]. Thereafter, the apex oxygen tilt angle $\phi$ for Eu$_{0.2}$-doping compares to that for Nd$_{0.35}$-doping, and, in the temperature range $T_{LT} < T < 300$ K, is about $1.0(5)^\circ$ larger than in pure La$_2$CuO$_4$; i.e., at 300 K $\phi$ increases from $4.2(5)^\circ$ to $5.2(5)^\circ$ and at $T_{LT}$ (Eu$_{0.2}$) $\simeq 135$ K from $5.1(5)^\circ$ to $6.0(5)^\circ$. It is reasonable to attribute the larger DM-moments to the enlarged octahedral tilt angle. Note, however, that the relative change of $\phi$ is small compared to that of the DM-moment, if we assume that $M_{DM} \propto \phi^{1.14-20}$ Though we have no definite explanation for this discrepancy, we think that it might follow from a non-linear relation between $M_{DM}$ and $\phi$ or from a doping induced disorder of the octahedral tilts.

In Ref. 55 it was reported that in La$_{2-x}$Nd$_x$CuO$_4$ and La$_{2-y}$Sm$_y$CuO$_4$ magnetization curves exhibit a remanent moment below $T_{LT}$. However, it was not clear whether these remanent moments stem from the polarized 4f-moments of the Nd and Sm ions due to an interaction with the Cu spins. Corresponding results for our Eu-doped samples are summarized in Fig. 16 where we show the remanent moment $M_{REM}$ at $H = 0$ after application of 14 Tesla at 4 K as a function of increasing temperature for the single crystal (left), the polycrystal (right) and an oriented powder (middle) made from the polycrystal. In all three cases we observe a remanent moment which decreases with increasing $T$ and disappears at $T_{LT}$. The results on single crystal and oriented powder clearly show that $M_{REM}$ is perpendicular to the CuO$_2$ planes, which supports the idea that $M_{REM}$ is caused by ferromagnetically aligned DM-moments. Note, that the alignment of the powder was successful to about 80\% which is the reason for the small remanent moment for $H \parallel ab$. At 4 K $M_{REM}$ in single- and polycrystal amount to about 10-20\% of $M_{DM}$, only.

In Fig. 17 we display $M_{REM}$ of the single crystal at 40 K as a function of a negative counter-field after +14 Tesla was applied at 4 K. Each time after the negative counter-field was increased, the remaining $M_{REM}$ was measured at zero field. The initial idea of this measurement was to check if there is a particular counter-field at which the remanent moment can be inverted. Obviously this is not the case. $M_{REM}$ is inverted monotonically and -14 Tesla has to be applied to fully invert the remanent moment initially induced by +14 Tesla.

**FIG. 15:** Deviations $\Delta M(H)$ of the magnetization from linearity at 0-2 Tesla in La$_{1.8}$Eu$_{0.2}$CuO$_4$ single- and polycrystal as well as in La$_2$CuO$_4$ polycrystal at $T = 150$ K. For comparison we show also scaled polycrystal data.

**FIG. 16:** Remanent moment $M_{REM} = M(H = 0)$ versus temperature of La$_{1.8}$Eu$_{0.2}$CuO$_4$ after application of 14 Tesla at $T = 4$ K for $H \parallel$ and perpendicular to CuO$_2$ plane for (a) single crystal, (b) oriented powder, and (c) polycrystal.

**FIG. 17:** Remanent moment $M_{REM} \parallel c$ of La$_{1.8}$Eu$_{0.2}$CuO$_4$ after application of +14 Tesla at $T = 4$ K as a function of a negative counter-field at $T = 40$ K.
However, it is important to notice that if the resistance yield a more plane Eu
$
\chi_{ab}^V\propto \chi_{ab}^{V}\propto \chi_{ab}^{V}
$
directions and increasing \(\nu\) as well as decreasing \(\nu\) temperature. (+) Average of the two \(ab\)-measurements with decreasing \(T\). (b) for \(H\) up to 14 Tesla parallel to the two \(ab\)-directions as a function of increasing \(T\).

C. \(\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4\) single crystal (625 °C N2)

In Fig. 18(a) we display \(\chi - \chi_{V}V\) at 1 Tesla for the \(\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4\) single crystal after its first anneal-

![Graph](image)

FIG. 18: Static susceptibility of \(\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4\) after subtraction of \(\chi_{V}V\). (a) for \(H = 1\) Tesla for all three crystal directions and increasing (+) as well as decreasing (-) temperature. (+) Average of the two \(ab\)-measurements with decreasing \(T\). (b) for \(H\) up to 14 Tesla parallel to the two \(ab\)-directions as a function of increasing \(T\).

The susceptibility data the magnetization shows a weak negative curvature for \(H \parallel a^*\) and a positive curvature with clear magnetic transitions for \(H \parallel b^*\). Our data for \(H \parallel b^*\) behave qualitatively similarly to a measured, as well as calculated, spin-flop for \(\text{La}_2\text{CuO}_4\) in Fig. 3 and Fig. 9(a) of Ref. 67, respectively. We mention that the measured transition is much broader than the calculated one. One reason for this is certainly the excess-oxygen. In Ref. 67 it is argued that the broadening can occur because the magnetic field is not exactly parallel to the \(b\)-axis. In the LTLO phase we observe a magnetic transition which we attribute to a spin-flop. In \(\text{La}_2\text{CuO}_4\) the spin-flop occurs for \(H \parallel b\) and is a measure of the in-plane spin-wave gap $\Delta^*$. Corresponding $M(H)$ curves in Fig. 19 yield a more detailed picture of this transition. It is sufficient to focus on plot (b) where we show the deviations from linearity of the $M(H)$ curves in plot (a). In accordance with

![Graph](image)

FIG. 19: Spin-flop in \(\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4\). (a) magnetization $M^{a*}$ and $M^{b*}$ of \(\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4\) after subtraction of Eu magnetism for different temperatures in the LTO and LTLO phase. (b) deviation of $M^{a*}$ and $M^{b*}$ from linearity. (c) derivative $dM^{b*}/dH$ at different temperatures. Curves are shifted for clarity. (d) spin-flop field $H_{c1}$ versus temperature.
that below $T_{LT}$ the critical field $H_c$ strongly decreases and becomes hysteretic at lower $T$ as can be seen from the derivative $d(M^c - M_{V}^c(Eu))/dH$ and the determined phase diagram in Fig. 19. Since $H_c$ is a measure of the in-plane gap, our results suggest that in the LTLO phase the effective in-plane gap decreases, which is in contradiction to the increase reported in Ref. 3 (details in discussion).

**D. Analysis of spin canting in LTO and LTT phase**

For a more quantitative analysis of $M_{DM}$ we have analyzed the $M^c(H)$ curves of the single crystal (after it was annealed at 800 °C). In Fig. 20 we show the fit results for two $M^c(H)$ curves measured above (left) and below (right) the LT-transition according to the function:

$$M^c - M_{V}^c(Eu) = H \cdot \chi_0 + M_{SF}^{c} + M_B^c$$  \hspace{1cm} (1)

where $H \chi_0$ represents all terms that are linear in field, $M_{SF}^{c}$ describes the spin-flip of the antiferromagnetically ordered part of the DM-moments $M_{DM}^{AF}$, and $M_B^c$ the contribution of that part of the DM-moment $M_{DM}^{AF}$ which behaves like a weak ferromagnet. $M_{SF}^{c}$ is described by a step of the size of $M_{DM}^{AF}$ with a gaussian distribution $\Delta H_c$ of the critical field $H_c$:

$$M_{SF}^{c} = M_{DM}^{AF} \frac{1}{\sqrt{2\pi \Delta H_c}} \int_0^H \exp\left(-\frac{(H-H_0)^2}{2\Delta H_c^2}\right) dH$$  \hspace{1cm} (2)

and $M_B^c$ by a Brillouin function for spin 1/2:

$$M_B^c = M_{DM}^{WF} \tanh(kH/T)$$  \hspace{1cm} (3)

where $k$ is a fit parameter. The $M^c(H)$ curves for sixteen temperatures were fitted simultaneously. The linear term $H \chi_0$ was confined to vary linearly as a function of $T$. All other fit parameters $M_{DM}^{AF}, M_{DM}^{WF}, H_c, \Delta H_c$, and $k$ were varied independently for each $M^c(H)$ curve. Overall the fits provide a good description of the data (solid lines in Fig. 20). The comparison in Fig. 20 shows that at 150 K (LTO) the spin-flip contribution $M_{SF}^{c}$ is significantly larger than the weak ferromagnetic contribution $M_B^c$. In contrast, at 100 K (LTT) it is $M_B^c$ which dominates over $M_{SF}^{c}$ and the critical field $H_c$ of the residual spin-flip has shifted to lower values.

In Fig. 21 we plot the temperature dependence of $M_{DM}^{AF}, M_{DM}^{WF}$ and $M_{DM}^{AF} + M_{DM}^{WF}$ we have extracted from $M^c(H)$ curves measured with increasing and decreasing field. At the transition into the low-temperature phase $M_{DM}^{AF}$ drastically decreases, while $M_{DM}^{WF}$ increases. However, the sum of both terms, within the error of our analysis, increases monotonically. In particular, there is no drastic change of the DM moment at the structural transition itself:

$$[M_{DM}^{AF} + M_{DM}^{WF}](LTO)_{T \geq T_{LT}} \approx [M_{DM}^{AF} + M_{DM}^{WF}](LTO)_{T \geq T_{LT}}$$

At $T = 4$ K we find a DM moment of $\simeq 4 \times 10^{-3} \mu_B/Cu$ which is about 50% larger than in pure La$_2$CuO$_4$, in agreement with our comparison of $M(H)$ at 150 K in Fig. 15. Obviously DM spin canting does not disappear.
in the LTT phase. There is, however, a significant shift of weight from $M_{AF\parallel}^0$ to $M_{AF\perp}^0$.

The shaded regions in Fig. 22 mark the temperature range $100 \, K < T < T_{LT}$ where, based on the susceptibility in Fig. 22(b), the crystal, after the discontinuous decrease at $T_{LT}$, still shows a clear $ab$-anisotropy. It is reasonable to assume that in this region volume fractions of LTO, LTLO, and LTT coexist (cf. Sec. IIII). In this temperature range the remaining spin-flip is clearly visible for increasing as well as decreasing field. In contrast, no well defined spin-flip is observed for $T < 100 \, K$. Here, weak signatures ($M_{AF\parallel}^0 < 1 \times 10^{-3} \mu_B/Cu$) are detected only for increasing field while there is no sign ($M_{AF\parallel}^0 = 0$) of a spin-flip for decreasing field (see also Fig. 14). We do not believe that the spurious spin-flip for increasing field represents a bulk property of the LTT phase. We rather think that it is connected to a LTO or LTT minority phase. This interpretation is supported by the temperature dependence of the critical field $H_c$ in Fig. 22. While $H_c$ drops sharply for $100 \, K < T < T_{LT}$, it again starts to increase for $T < 100 \, K$. First of all, this would be a very unusual temperature dependence if it were intrinsic to LTT. Moreover, at low temperatures $H_c$ reaches $\sim 6$ Tesla, which is roughly the zero-temperature approximation for the LTO phase. At the same time, the width $\Delta H$ increases from 1 Tesla in the LTO phase to about $\sim 2.5$ Tesla, underlining that this transition is not at all well defined. Hence, our main conclusion with regard to the bulk properties of La$_{1.8}$Eu$_{0.2}$CuO$_4$ is that spin-flips with well defined $H_c$ occur only in the LTO phase, whereas no spin-flip exists in the LTT phase.

E. Phase diagrams of La$_{1.8}$Eu$_{0.2}$CuO$_4$

In Fig. 28 we compare the temperature dependence of $M_{AF\parallel}^0$, $M_{AF\perp}^0$, and $M_{AF\parallel}^0 \times H_c$ for the La$_{1.8}$Eu$_{0.2}$CuO$_4$ single and polycrystal as well as the La$_2$CuO$_4$ polycrystal. The $M(H)$ curves of the polycrystals had to be fitted separately for every temperature, since, due to the more complex fit function, simultaneous fits were not stable. Also, $H_{\chi_0}$ was varied independently. For the sake of consistency, we have refitted the single crystal data accordingly. For $M_{AF\parallel}^0$ in Fig. 28(a) we find a convincing agreement between the La$_{1.8}$Eu$_{0.2}$CuO$_4$ single and polycrystal. Moreover, the comparison with La$_2$CuO$_4$ shows the much larger values for $M_{AF\parallel}^0$ in the LTO phase of the Eu-doped compounds. Again, we emphasize that $M_{AF\parallel}^0$ is the AF ordered part of the DM moment only, and not a measure for the full DM moment.

$H_c$ in Fig. 28(b) was determined from the average for measurements with increasing and decreasing fields. In the LTO phase, $H_c$ increases with decreasing $T$ and is significantly higher in the single crystal than in either of the polycrystals. A possible explanation may involve finite size effects in the polycrystals due to a limited magnetic correlation length in the crystallites. On the other hand, we find for the single crystal that, with increasing oxygen content, the spin-flip transition broadens, and the maximum in $dM_c/dH$ associated with $H_c$ effectively shifts to higher fields. Therefore, the crystal’s relatively high $H_c$ values in the LTO phase might to some extent follow from a finite and possibly inhomogeneous oxygen excess. Further experiments are needed to clarify this problem.

Below $T_{LT}$, the spin-flip field decreases, for the single crystal in particular. In the polycrystal, $M(H)$ changes so drastically below $T_{LT}$ that the average $H_c$ can only be followed down to 110 K. In this temperature range of $\sim 25 \, K$ below $T_{LT}$, $H_c$ drops only slightly. Since the structural transition in the polycrystal is much sharper than in the single crystal, we infer that the polycrystal approaches the LTT phase at a higher temperature than the single crystal (cf. Sec. IIII); i.e., the polycrystal is closer to showing the intrinsic properties of the LTT phase. The residual spin-flip, therefore, can be associated with a minority phase LTO and/or LTLO whose volume fraction rapidly decreases with decreasing $T$. The fact that $H_c$ below $T_{LT}$ remains almost as high as in the LTO phase supports this interpretation.

In Fig. 28(c) we show the temperature dependence of the effective interlayer coupling:

$$J^*_1(T) = M_{AF\parallel}^0(T) \cdot H_c(T)/S^2 \quad(5)$$

which has been introduced and discussed in detail in Ref. 51. $J^*_1$ is a measure for the strength of the AF interlayer coupling at a particular temperature and only in the limit $T \to 0$ identical with the interlayer superexchange constant $J_1$. It is apparent from Eq. 5 that $J^*_1(T)$ is an implication of $M_{AF\parallel}^0(T)$ and $H_c(T)$. In the LTO phase of the Eu-doped samples $J^*_1$ is well-defined and is significantly higher than in pure La$_2$CuO$_4$. Below
the shaded region might be much smaller than suggested by Fig. 23(c), since \( M_{DM}^{AF} \) has to be normalized with the volume fraction.

Let us go back to the strong reduction of \( M_{DM}^{AF} \) below \( T_{LT} \). This observation shows us that the \( M(H) \) curves in the LTT phase do not comply with a simple shift of a spin-flip of unchanged magnitude to a lower critical field. This means that there is no simple, uniform reduction of the interlayer coupling \( J^*_\perp \). On the other hand we do not observe spontaneous weak ferromagnetism which indicates that \( J^*_\perp \) is not zero, either. In the case \( J^*_\perp = 0 \) we would expect that the full DM moment can be ferromagnetically aligned by a relatively small field which would lead to a large initial slope \( dM/dH \). We mention that Viertiö and Bonesteel have calculated \( M(H) \) curves for different interlayer coupling mechanisms in the LTO and LTT phases and that weak ferromagnetism in the LTT phase is one of their solutions. Obviously, our measured \( M(H) \) curves do not show this type of behavior (as well as none of the other types). Nevertheless, the fact that in the LTT phase \( dM/dH \) at low fields is significantly larger than in the LTO phase indicates that \( J^*_\perp \) is reduced (cf. Fig. 14 and 10). Thus, we believe that the LTT phase in our sample is characterized by a broad distribution of \( J^*_\perp \) with the center of mass shifted to lower values than in the LTO phase. This idea is supported by the measurement of \( M_{REM} \) in Fig. 17 which shows that, on the one hand, there is no well defined critical field to invert \( M_{REM} \), and on the other hand, that the required field to fully invert \( M_{REM} \) is rather large. Nonetheless, it still remains unclear to us whether a broad distribution of \( J^*_\perp \) represents the intrinsic properties of an ideal strain-free LTT phase.

VII. LIGHTLY Sr DOPED \( \text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4 \)

A. Comparison with \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \)

In this section we study the influence of light Sr doping (\( x \leq 0.02 \)) on the Cu spin magnetism in the LTT phase. For this purpose, we directly compare in Fig. 24 the susceptibility \( \chi \) of pure and \( \chi - \chi_{\text{V(V)Eu}} \) of Eu-doped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) for polycrystalline samples with similar Sr content and \( T_N \). The Eu fraction \( y^* \) was varied, so that \( \chi - \chi_{\text{V(V)Eu}} \) and \( \chi \) match at high temperatures (for \( x = 0 \) at 150 K). The determined \( y^* \) values agreed with the nominal value \( y = 0.2 \) within the experimental error of \( \Delta y \approx 0.005 \). Furthermore, all data sets were corrected for \( \chi_{\text{dia}} + \chi_{\text{V(V)Cu}} \), so that they can be compared to \( \chi_{2DHAF} \), which we have approximated by the dotted lines. As mentioned before, the difference between the data and \( \chi_{2DHAF} \) is caused by the DM spin canting.

In the LTO phase, the pairs of curves show a fair agreement in the paramagnetic phase as well as in the AF phase. In contrast, the differences between the LTT and
LTO phase at Sr content $x = 0$ become even larger with increasing $x$. This result shows that DM spin canting is present in the LTT phase of Sr-doped samples, as well. Eventually, for $x \gtrsim 0.017$ ($T_N \lesssim T_{LT}$) the differences between LTT and LTO start to decrease.

As a consequence, for $x \gtrsim 0.017$ the susceptibility does not decrease below $T_N$, indicating that the DM moments in adjacent layers do not arrange AF. To study the interlayer coupling in Sr-doped La$_{1-x}$Eu$_x$CuO$_4$ in more detail, we have performed $M(H)$ measurements on samples with $x = 0.01$ and $x = 0.018$, both annealed at 625 °C in N$_2$. For $x = 0.01$ we observe basically the same behavior as for $x = 0$; i.e., spin-flips for $T_{LT} < T < T_N$, strongly decreasing spin-flips for $T < T_{LT}$ and eventually no spin-flips for $T \lesssim T_{LT}$. More interesting is the case of $x = 0.018$ with $T_N \lesssim T_{LT}$ (see Fig. 24). Here we observe weak ferromagnetic-type $M(H)$ curves for all temperatures. The inset shows some of the curves after subtraction of the slope between 6.5 and 8 Tesla, assuming that at these fields the contribution of the DM moment has saturated. The weak ferromagnetic behavior is obvious, though the magnetic field scale to align the DM moments is quite high, consistent with the absence of spontaneous weak ferromagnetism.

**B. Remanent moment**

In analogy to Fig. 16 the remanent moment of the Sr-doped samples was measured in zero field after a field of 14 Tesla was applied at 4 K. Fig. 24 shows $M_{REM}$ for different Sr concentrations $0 \leq x \leq 0.02$ as a function of increasing temperature. With increasing Sr content $M_{REM}$ decreases systematically and disappears at the critical concentration $x = 0.02$. $M_{REM}$ also decreases as was shown in Ref. 61, the effective interlayer coupling in pure La$_{2-x}$Sr$_x$CuO$_4$ strongly decreases with increasing Sr content and for $x = 0.017$ is already quite weak. In the Eu-doped compounds, the LT-transition causes a further reduction of the interlayer coupling. As with increasing temperature and for all $x < 0.02$ vanishes at $T_{LT}$. Obviously, a remanent moment is a feature of the entire AF ordered LT phase. From the comparison in the inset it is evident that in the LTO phase, except for very low temperatures, no remanent moment is observed. Here, $M_{REM}$ is related to the spin-freezing regime for $T \lesssim 25$ K. 59,61,77,78,79,80 The spin freezing regime might also explain the upturn of $M_{REM}$ for $T \lesssim 25$ K in the Eu-doped samples with $x > 0.13$. 89

**C. Phase diagram of La$_{1.8-x}$Eu$_x$Sr$_x$CuO$_4$**

From our measurements on La$_{1.8-x}$Eu$_x$Sr$_x$CuO$_4$ we have constructed the phase diagram in Fig. 27. For this phase diagram we have remeasured the polycrystals after they were annealed at 800 °C in vacuum, which led to slightly higher Néel temperatures than in Fig. 13. In the LTO phase, the Sr doping dependence of $T_N$ is practically identical for pure and Eu-doped La$_{2-x}$Sr$_x$CuO$_4$. For $x > 0.016$ no Néel peaks are observed in the LT phase. However, $\mu$SR experiments have shown that in the LT phase AF order disappears at roughly the same critical Sr content $x \approx 0.02$ as in La$_{2-x}$Sr$_x$CuO$_4$. 89 The most important difference between the LTO and LT phase is the loss of a well defined interlayer coupling $J^*_L$. The weakening of the effective interlayer coupling progresses with decreasing temperature and increasing Sr content. Accordingly, well defined spin-flip transitions are observed only in the LTO phase, while in the LT phase the spin-

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**FIG. 24: Comparison of $\chi$ at 1 Tesla of La$_{1.8-x}$Eu$_x$Sr$_x$CuO$_4$ after subtraction of $\chi_{VV}$ (E4) and pure La$_{2-x}$Sr$_x$CuO$_4$ for different Sr concentrations $x < 0.02$. Data also corrected for $\chi_{dia}$ and $\chi_{VV}$ (Cu) (cf. Fig. 12). The arrows for $x = 0$ and 0.017 indicate the contribution of the DM-Moments $\chi_{DM}$ in the LTO and LT phase. The dashed line approximately represents $\chi_{2DHAF}$.**
flip disappears. Moreover, in the entire AF ordered LTT phase a small fraction of the Dzyaloshinsky–Moriya spin canting can be remanently magnetized, underlining the presence of DM moments as well as their weak ferromagnetic character.

VIII. DISCUSSION

As mentioned in the introduction, the question of whether in the LTT phase Cu spins exhibit Dzyaloshinsky–Moriya spin canting has been a matter of controversy. Our data unambiguously show the presence of DM spin canting in the LTT phase, supporting a spin structure given by the black spins in Fig. II. Although this is in qualitative agreement with neutron diffraction data, there is a significant difference with respect to the temperature dependence of the size of the DM moments. According to Keimer et al. the in-plane gap in deoxygenated La$_2$CuO$_4$ ($T_N = 325$ K) and La$_{1.65}$Nd$_{0.35}$CuO$_4$ ($T_N = 316$ K) is about the same in the LTO phase at 100 K, but increases by about 60% in the LTLO phase of the Nd doped compound. (With $\alpha \approx 40^\circ$ the sample is very close to LTT.) Keimer et al. argue that this increase is due to an increase of the DM interaction in the LTLO phase. In contrast, our data indicate a stronger DM interaction in both phases, LTO and LTLO, which we have attributed to the larger octahedral tilt angle (cf. Sec. VII).

Although the increase of the in-plane gap in the LTT phase is consistent at first glance with a larger DM moment in La$_{1.8}$Eu$_{0.2}$CuO$_4$, it is inconsistent with the decrease of the spin-flop field $H_{c1}$ that we have observed in the LTLO phase of the single crystal with $T_N = 280$ K. Since $H_{c1}$ is a measure of the in-plane gap, it actually indicates a decrease of this gap in the LTLO phase, which is just the reverse of the neutron diffraction result. Further experiments are necessary to clarify this point. We mention that Keimer et al. have determined the

FIG. 26: Remanent moment $M_{REM}$ of La$_{1.8-x}$Eu$_x$Sr$_x$CuO$_4$ for different Sr concentrations $x \leq 0.02$ as a function of temperature. Inset: comparison with pure La$_2$CuO$_4$ for $x \sim 0.01$.

FIG. 27: Magnetic and structural phase diagram of La$_{1.8-x}$Eu$_x$Sr$_x$CuO$_4$. Data for pure La$_2$CuO$_4$ are shown for comparison. For $x = 0.02$ the cluster spin glass transition at $T_{CSG}$ is indicated as well. Solid lines are guides to the eyes.
anisotropy \( \alpha_{DM} \) due to DM superexchange from the in-plane gap, which is justified if the DM superexchange is by far the dominant source. Theoretical papers, however, have pointed out that the in-plane anisotropy may be composed from multiple finite contributions which may compete, in particular, in the LTT phase.\cite{14, 15, 16, 19, 22} Tetragonal \( \text{Sr}_2\text{CuO}_2\text{Cl}_2 \) with flat \( \text{CuO}_2 \) planes, for example, has the same collinear spin structure as \( \text{La}_2\text{CuO}_4 \) but no spin canting (no DM interaction), which shows that in this system the spin direction is determined by other contributions to the in-plane anisotropy.\cite{23} Though we do not oppose the particular idea of a symmetric superexchange anisotropy, our evidence of DM spin canting in the LTT phase raises serious concerns about its quantitative relevance (cf. Sec. 11). Nevertheless, it is certainly a reasonable attempt to discuss the decrease of \( H_N \) (spin-flop) in terms of a competition between the (enhanced) DM interaction, which prefers a configuration with spins perpendicular to the octahedral tilt axis, and other contributions which support other spin directions, i.e. parallel to the tilt axis or parallel to the \( b \) or \( a \)-axis (cf. Fig. 1).

When we compare our results for the \( \text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4 \) single crystal (\( T_N = 315 \) K) with the data in Ref.\cite{11} for a less deoxygenated crystal with \( T_N = 265 \) K, we find that for \( H \parallel c \) the jump at \( T_{LT} \) in our crystal, as well as the Néel peak, is about twice as large, showing the strong impact of intercalated oxygen. When analyzing our data, e.g. in Fig. 13, in a similar way as in Ref.\cite{11} (i.e., subtracting the susceptibility \( \chi'(H = 8T) \) from \( \chi'(H = 1T) \)) we find that the difference in the LTT phase is basically zero. While this means that in the LTT phase \( M'(H) \) is approximately linear up to 8 Tesla, as can be seen in Fig. 14 it definitely does not proof a reduction or the absence of DM spin canting, contrary to the argument in Ref.\cite{11}.

Let us assume for a moment that DM spin canting is absent. Then the fact that in the LTT phase the effective interlayer coupling is reduced, would bring this system very close to a perfect \( S = 1/2 \) 2D-HAF. In this case the susceptibility of the CuO\(_2\) planes in \( \text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4 \) should be very close to \( \chi_{2DHAF} \) of non-interacting CuO\(_2\) planes, as is the case for \( \text{La}_2\text{CuO}_4 \) in the high temperature tetragonal phase or in \( \text{Sr}_2\text{CuO}_2\text{Cl}_2 \). Since \( \chi_{2DHAF} \) is smaller than the susceptibility of the CuO\(_2\) planes in the LTO phase, one would expect that \( \chi \) decreases at the transition LTO\( \rightarrow \)LTT.\cite{24} This is in sharp contrast to the fact that \( \chi \) actually increases (cf. Fig. 21). Clearly, in the absence of DM spin canting, a decrease of the interlayer coupling cannot account for an increase of \( \chi \) of the size observed in \( \text{La}_{1.8}\text{Eu}_{0.2}\text{Sr}_2\text{CuO}_4 \), as was suggested in Ref.\cite{11}. Hence, in the LTT phase Cu spins must be canted.

It is generally assumed that the interlayer coupling in the LTO phase of \( \text{La}_2\text{CuO}_4 \) benefits from the orthorhombic strain, since the perfect frustration of the interlayer superexchange in a tetragonal body-centered structure is lifted.\cite{11, 81, 82, 83} It is believed that this is the reason for \( T_N \) in \( \text{La}_2\text{CuO}_4 \) being higher than in \( \text{Sr}_2\text{CuO}_2\text{Cl}_2 \), though one has to keep in mind that in \( \text{Sr}_2\text{CuO}_2\text{Cl}_2 \) the distance between the CuO\(_2\) planes is significantly larger.\cite{25} Now, in \( \text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4 \) the orthorhombic strain around \( T_N \) is even bigger than in \( \text{La}_2\text{CuO}_4 \), and indeed we find a stronger interlayer coupling (cf. Fig. 25(c)). A significant change of \( T_N \), however, is not observed. We assume that the interlayer coupling has to change by orders of magnitude to cause a substantial shift of \( T_N \). This is also consistent with considerations about what drives the magnetic transition in 2D-H and 2D-XY systems in Ref.\cite{57, 78, 88, 89, 90}.

In the LTT phase, a reduction of the interlayer coupling is expected because of the tetragonal symmetry. The non-collinear spin structure is consistent with a further weakening of the interlayer coupling (cf. Fig. 1). In such a system, the next-nearest-layer coupling might be crucial to establish a static AF order and might result in two loosely coupled subsystems of CuO\(_2\) planes.\cite{26} In the case of perfect decoupling of adjacent layers, we would expect spontaneous weak ferromagnetism of the DM moments which we have not observed. On the other hand, even if the magnetic decoupling in the LTO and LTT phases is not perfect, the spin lattice should be less rigid than in the LTO phase, leading to an increase of magnetic fluctuations. Indications for stronger fluctuations below \( T_{LT} \) have indeed been found in ESR,\cite{39} NQR,\cite{39} and \( \mu\)SR\cite{39} relaxation experiments on \( \text{La}_{1.8}\text{Eu}_{0.2}\text{Sr}_2\text{CuO}_4 \) polycrystals (\( 0 \leq x \leq 0.02 \)). Furthermore, measurements of the internal static magnetic field by means of \( \mu\)SR indicate a slight decrease of \( H_{int}^{SR} \) at \( T_{LT} \).\cite{20} Note, however, that a change of \( H_{int}^{SR} \) at the muon site can also result from slightly different muon positions in the LTO and LTT phases. NQR measurements show a quadrupolar broadening of the NQR-lines in the LTO phase and an even much stronger magnetic broadening in the LTT phase. The broadening in the LTO phase was explained with a distribution of the electric field gradient due to the local lattice distortions caused by Eu doping.\cite{12, 13} These lattice distortions might be responsible for the field hysteresis in the \( M(H) \) curves (cf. Fig. 11). The strong magnetic broadening in the LTT phase was explained with a distribution of the internal magnetic field \( H_{int}^{NQR} \) at the La site.\cite{12, 13} Since spin structure and octahedral tilts are coupled, the magnetic broadening might indicate a distribution of the angle \( \alpha \) of the azimuthal rotation of the tilt axis. This interpretation is consistent with our conclusion from magnetization data that, in the LTT phase, the spin-flip field \( H_c \), and therefore the effective interlayer coupling \( J_1^* \), are not well defined.

**IX. CONCLUSION**

In summary, we have studied the magnetism of the CuO\(_2\) planes in the different structural phases of
La$_{2-x-y}$Eu$_x$Sr$_y$CuO$_4$ over a broad range of Eu and Sr doping, with focus put on the antiferromagnetic regime. To separate the Cu spin magnetism, we have carefully subtracted the much larger Van Vleck magnetism of the Eu$^{3+}$ ions. Our results show that Dzyaloshinsky–Moriya superexchange stabilizes a canted Cu spin structure in the LTT phase, which is in sharp contradiction to theoretical predictions. Although our result agrees with neutron diffraction data, there is disagreement with the latter on other questions. Most intriguing is the decrease of the spin-flop field, which suggests that the in-plane spin-wave gap decreases in the LTLO phase while neutron scattering data indicate an increase. Next, according to our data, the size of the canting moment of La$_{1.8}$Eu$_{0.2}$CuO$_4$, compared to that of pure La$_2$CuO$_4$, is about 50% larger in the LTLO and the LTO phases, which we attribute to the larger octahedral tilt angle. Moreover, no significant change of the canting moment was detected at the LTO$\leftrightarrow$LTT transition itself. The major difference of the LT phase compared to the LTO phase is the loss of a well-defined interlayer coupling, which macroscopically results in the disappearance of the spin-flip transition. Though the interlayer coupling still puzzles us, it seems that it is not uniform, but has an average much weaker than in the LTO phase. In the LTT phase, magnetization curves become weak ferromagnetic, and exhibit a small remanent moment perpendicular to the CuO$_2$ planes. Spontaneous weak ferromagnetism is not observed. The remanent moment, as well as the weak ferromagnetism, exists only in the antiferromagnetic LTT phase and disappears for $x > 0.02$ within the resolution of our experiment.

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