Comment on ”Strong dependence of the interlayer coupling on the hole mobility in antiferromagnetic La$_{2-x}$Sr$_x$CuO$_4$ ($x < 0.02$)”

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Abstract

Using the experimental data given in the above paper, we show that – unlike the effective coupling discussed in this paper – the net average antiferromagnetic interlayer coupling in doped lanthanum cuprates depends only weakly on the doping or on the temperature. We argue that the effective coupling is proportional to the square of the staggered magnetization, and does not supply new information about the origin of the suppression of the magnetic order in doped samples. Our analysis is based on a modified version of the equation describing the spin-flip transition, which takes into account the decrease of the staggered moment with temperature and doping.

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The prototype high-temperature superconductor is La$_{2-x}$Sr$_x$CuO$_4$, which is antiferromagnetic, with the Néel temperature $T_N$ strongly decreasing with increasing doping for $x \lesssim 0.02$, and superconducting at $x > 0.05$. Stoichiometric La$_2$CuO$_4$ has alternate weak ferromagnetic moments in the c-direction (perpendicular to each CuO$_2$ plane), due to the Dzyaloshinsky-Moriya (DM) interaction, allowed by its orthorhombic structure. These moments flip into the same direction (together with a flip of the in-plane staggered moments), generating a net ferromagnetic moment, at a temperature-dependent spin-flip (SF) magnetic field $H_c(T)$ along the c direction.$^1$

In a recent paper,$^2$ Hücker et al. studied the temperature and field dependent spin-flip transition in La$_{2-x}$Sr$_x$Cu$_{1-z}$Zn$_z$O$_4$. In a separate paper$^3$ they also studied this transition in La$_{2-x-y}$Eu$_y$Sr$_x$CuO$_4$. For each sample, they also measured the ferromagnetic moment at the SF transition, $M_F(T)$. The aim of the study was, according to the authors, “to find out the primary controlling parameter for the suppression of the 3D AF order in La$_{2-x}$Sr$_x$CuO$_4$”. To this end, they “have studied the magnetic interlayer coupling $J_\perp$ as a function of Sr and/or Zn doping” (see p. 1 of their paper). Actually, however, Hücker et al. calculated the product

$$\Delta E(T) = M_F(T)H_c(T),$$

and defined an “effective interplanar coupling”, $J_\perp(T) = \Delta E(T)/S^2$. It is from the doping and temperature dependences of this quantity that they tried to find out the reason of the suppression of the AF order in doped lanthanum cuprates.

In this Comment we show that for doped samples the “effective interlayer coupling” is not related directly to $J_\perp$ even at $T = 0$. Then we use the experimental data by Hücker et al. to conclude that in fact $J_\perp$ depends only weakly on $T$ and $x$. It is probably also independent of $z$. Therefore, $J_\perp$ is not the primary controlling parameter for the suppression of the 3D order. The decrease in $J_\perp(T)$ results from the decrease in the staggered moment, which is probably caused by doping dependent changes in the intraplanar correlation length, due to changes in the intraplanar interactions.
The energy $\Delta E(T)$ should balance the net interplanar antiferromagnetic exchange coupling energy at the (first order) flip transition. Therefore, at $T = 0$ and $x = z = 0$, Thio et al.$^1$ equated the energy $\Delta E(0)$ to $J_\perp S^2$, where $S$ represents the ground state staggered moment per Cu ion, to deduce the interlayer exchange energy $J_\perp$. Since $J_\perp$ results from a delicate balance between four bonds which couple each Cu ion to its neighbors in the next plane, one might expect this energy to depend on doping even at $T = 0$.

Our main argument relates to the value of $\Delta E(T)$ at non-zero temperature and/or doping. As $T$ increases from 0 to $T_N$, the staggered moment per Cu ion, $M^\dagger$, decreases from $S$ to zero. A similar decrease may result from the doping, even at $T = 0$. Therefore, the relation $\Delta E(0) = J_\perp S^2$ of Ref. 1, which was also used in Ref. 2, should be generalized into the effective mean-field interlayer energy given by

$$\Delta E(T) = J_\perp [M^\dagger]^2. \quad (2)$$

Thus, $J_\perp(T) = J_\perp[M^\dagger/S]^2$, and the decrease in $J_\perp$ results mainly from the decrease in $M^\dagger$. H"ucker et al. claimed that $J_\perp(0) = J_\perp$ (see p. 3 of the paper), and derived from this relation $J_\perp$ for doped samples. However, as argued above, this relation holds only for pure samples, when $M^\dagger = S$. Therefore, the values of $J_\perp$ reported in Table 1 of Ref. 2 do not represent the interlayer coupling $J_\perp$.

Note that because of the in-plane and out-of-plane spin exchange anisotropy, the staggered moment in lanthanum cuprates is finite at finite $T$ even at vanishing interlayer coupling, and hence it need not be very sensitive to the interlayer coupling. Its decrease with the increase of $T$ is due to thermal fluctuations. At relatively high doping, this decrease may also be due to stripe formation.$^5$ However, the possibility of stripe formation in the (low doping) AF ordered region of lanthanum cuprates is still controversial: The recent experimental results by Gozar et al.$^6$ exclude the phase separation scenario suggested in Ref. 5 for Sr doping in the relevant range $x \leq 0.02$. At these concentrations, the strong decrease of $M^\dagger$ with localized hole doping is most probably due to frustration in the planes.$^7$

Given the mean field DM free energy per site, $-4DM^\dagger M_F$, where $D$ is the DM interaction
energy, the ferromagnetic moment is given by

\[ M_F(T) = \chi_\perp 4DM^\dagger, \]

where \( \chi_\perp \) is the transverse ferromagnetic susceptibility. For the undoped system below \( T_N \), \( \chi_\perp \approx 1/(8J) \), where \( J \) is the intralayer exchange energy. Therefore, equating the two expressions for \( \Delta E(T) \) yields

\[ \alpha \equiv \frac{H_c(T)}{M_F(T)} = \frac{J_\perp}{(4D\chi_\perp)^2}. \]

Using the data from Fig. 3 and Table 1 of Ref. 2, we plot in Fig. 1 the ratio \( \alpha \) versus \( T/T_N \) for different samples, doped with Sr and Zn. We also calculated \( \alpha \) for Eu doped samples, using the data from Fig. 24 of Ref. 3. Roughly, \( \alpha \) is seen to be essentially the same for all temperatures and Sr (holes) or single crystal Eu doping (data for Eu doping are shown only at temperatures where the crystal remains orthorhombic). The overall slow increase of \( \alpha \) with \( T \) can be explained by the decrease of \( D \), see Eq. (4), because of the decrease of the orthorhombic distortion with increasing temperature. This latter decrease may also cause a small decrease in \( J_\perp \), which is apparently compensated by the larger decrease in \( D \). In any case, these changes are all small. In contrast, substitution of Cu by Zn apparently yields somewhat smaller values of \( \alpha \). However, this could still be consistent with no change in \( J_\perp \). It is known that in vacancy doped planar isotropic AF systems the susceptibility \( \chi_\perp \) diverges at any doping concentration. Magnetic anisotropy removes the divergency, but the susceptibility may still be large, since the anisotropy in lanthanum cuprates is small. This increase may also account for the increase of \( M_F \) observed in Ref. 2, see our Eq. (3), and the decrease of \( \alpha \) in Fig. 1. An alternative source for the decrease of \( \alpha \) would involve a doping-dependent octahedral tilt angle, which would lead to the increase of \( D \).\textsuperscript{2} The same mechanism can explain the decrease of \( \alpha \) in polycrystalline Eu doped samples.

Thus, the values which we deduce for \( \alpha \) are consistent with a scenario in which \( J_\perp \) essentially does not depend on \( T \) or on doping. Since \( J_\perp \) represents a net superexchange energy, which is an average local quantity, this result implies that fluctuations due to doping
average out and have no strong effect on the measured $J_\perp$; the local $J_\perp$ increases or decreases depending on which sub-lattice is doped. Given that the average $J_\perp$ is constant, the “effective interlayer coupling” $J_\perp(T)$ does not give more information than the staggered magnetization $M$. The approach of $J_\perp(T)$ to zero when $T$ approaches $T_N$ does not imply that the interlayer coupling diminishes. Thus, the statement in the abstract of Ref. 2, that the “interlayer coupling plays a key role in the suppression of the AF phase”, is unjustified.

The title of Ref. 2, which states that the interlayer coupling (i.e., $J_\perp$ rather than $J_\perp$) depends strongly on the hole mobility, is also misleading. First, as shown above, the change, if any, of $J_\perp$ due to doping is small. Secondly, the paper presents no direct evidence that the hole mobility has any direct effect on the magnetic properties of lanthanum cuprates. In contrast, it was shown that the strong suppression of the AF order by Sr (hole) doping – in variance with Zn (vacancy) doping – can be explained by the long-range dipole-type magnetic distortion introduced by localized holes. The theory based on this model describes quantitatively the phase diagrams of Sr doped as well as of Sr and Zn doped lanthanum cuprates. Hence the attempt of the authors to explain this difference by the effect of hole mobility is only a suggestion, which has no quantitative support. The idea of dynamic magnetic antiphase boundaries evoked by the authors to support their statements is, as noted above, still controversial.

In conclusion, we have shown that all the available data are consistent with a constant $J_\perp$, essentially independent of $T$, $x$, $y$ and possibly also $z$. Therefore, it is not necessarily the interlayer coupling which controls the AF order in Sr doped lanthanum cuprates. In fact, the suppression of the AF order can be fully explained by the reduction of the in-plane correlation length with Sr doping, due to frustration.

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FIG. 1. The ratio $\alpha = H_c/M_F$ versus temperature for lanthanum cuprate doped with Sr (concentration $x$), Zn ($z$) and Eu ($y$). The experimental data are taken from Refs. [2] and [3]. The typical error bars are from Table 1 in Ref. [2]. “s” and “p” stand for single and polycrystal samples.