Spin Susceptibility in Underdoped Cuprates: Insights from a Stripe-Ordered Crystal

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We report a detailed study of the temperature and magnetic-field dependence of the spin susceptibility for a single crystal of La$_{1.875}$Ba$_{0.125}$CuO$_4$. From a quantitative analysis, we find that the temperature-dependent anisotropy of the susceptibility, observed in both the paramagnetic and stripe-ordered phases, directly indicates that localized Cu moments dominate the magnetic response. A field-induced spin-flop transition provides further corroboration for the role of local moments. Contrary to previous analyses of data from polycrystalline samples, we find that a commonly-assumed isotropic and temperature-independent contribution from free carriers, if present, must be quite small. Our conclusion is strengthened by extending the quantitative analysis to include crystals of La$_{2-x}$Ba$_x$CuO$_4$ with $x = 0.095$ and 0.155. On the basis of our results, we present a revised interpretation of the temperature and doping dependence of the spin susceptibility in La$_{2-x}$(Sr,Ba)$_x$CuO$_4$.

I. INTRODUCTION

A central controversy in the field of high-temperature superconductivity concerns the nature of the magnetic response in the normal state: does it come from local moments or mobile conduction electrons? The bulk spin susceptibility, $\chi^s$, has been central to the discussion, but the analysis so far has been ambiguous. For example, in underdoped cuprates one experimentally observes a decrease in the normal-state $\chi^s$ on cooling below some characteristic temperature $T_{\text{max}}$ \cite{1}. When this behavior was first identified in underdoped YBa$_2$Cu$_{3-y}$O$_{6+y}$ through measurements of the Y Knight shift by Alloul and coworkers\cite{2}, it was interpreted as evidence for the development of a pseudogap in the electronic density of states. Such a perspective attributes the bulk magnetic response to paramagnetism of the conduction electrons (Pauli paramagnetism), and it is often cited\cite{3,4,5,6}. In contrast, Johnston proposed that $\chi^s(T)$ consists of two-components: 1) a temperature-dependent portion that scales with doping and that is associated with antiferromagnetically-correlated Cu moments, and 2) a Pauli contribution that happens to be roughly proportional to the doped hole density. This alternative approach also remains popular\cite{7,8,9}.

Clearly, the temperature dependence of $\chi^s$ is closely tied to the issue of the pseudogap\cite{10}, a continuing conundrum and challenge. The nature of the spin response is also part of the ongoing discussion on the dynamic susceptibility, where the momentum- and energy-dependent structure observed by neutron scattering has generally been interpreted either as the response of conduction electrons scattering across the Fermi surface\cite{11,12,13,14,15,16} or as fluctuations of local Cu moments\cite{17,18,19,20,21,22}. While there is much debate on the dynamic response, we have seen little critical discussion of the alternative interpretations of the static susceptibility, or reconsideration of the assumptions that went into analyses first developed two decades ago, typically applied to measurements on polycrystalline samples\cite{23,24,25}.

In this paper, we present bulk susceptibility measurements performed on a single crystal of La$_{1.875}$Ba$_{0.125}$CuO$_4$. We go through a stepwise analysis of the data, considering the extent to which we can distinguish between contributions from mobile charge carriers and local moments. One criterion for such a distinction is anisotropy: Pauli paramagnetism is expected to be isotropic, whereas the paramagnetic response of local moments depends on the orientation of the applied field due to anisotropic gyromagnetic factors. Thus, from the fact that we observe a temperature-dependent anisotropy of the susceptibility in the paramagnetic regime, we can conclude that there are significant contributions from local moments.

Our most direct evidence for local moments comes from the temperature regime below the spin-ordering transition that has been characterized by neutron diffraction\cite{26,27,28}. There we see an evolution of the anisotropic susceptibility that can be simply interpreted in terms of the classic expectations for a non-collinear antiferromagnet (see Fig. 1). Furthermore, on following the magnetization vs. applied field, we find evidence for a spin-flop transition at $H = 6$ T. Such a transition is consistent with an anisotropy of the superexchange interaction between local moments.

To describe the doping dependence of the susceptibility of polycrystalline La$_{2-x}$Sr$_x$CuO$_4$, Johnston\cite{6} proposed that there should be a component $\chi^\text{Cu}$ due to antiferromagnetically-coupled local moments. Furthermore, he assumed that it had the form $\chi^\text{Cu}(x) f(T/T_{\text{max}}(x))$, where $f$ is a normalized “universal” function. He found that he could get a good fit to the data if he included a temperature-independent component with a magnitude roughly proportional to $x$. The same scaling analysis was later applied to a more extensive set of measurements by Nakano et al\cite{2}. The temperature-independent component is generally assumed to correspond to the Pauli susceptibility, $\chi^\text{P}$, associated with the doped carriers.

This successful, but ad hoc, decomposition has been widely applied, and we initially attempted to use it on our own results to separate out the contribution from
local moments. We found, however, that a $\chi^P$ component of the commonly accepted magnitude is not compatible with the large anisotropy observed in the single crystal data. We have strengthened our case by extending the analysis to include data for La$_{2-x}$Ba$_x$CuO$_4$ with $x = 0.095$ and $0.155$. We conclude that any temperature-independent $\chi^P$ component must be an order of magnitude smaller than that reported in Refs. 17,18,19.

As a result of the much reduced $\chi^P$, the extracted $\chi^{\text{Cu}}$ is larger at low temperature than previously assumed. Based on our new insights, we discuss the implications for the evolution of magnetic correlations in La$_{2-x}$Sr$_x$CuO$_4$ and La$_{2-x}$Ba$_x$CuO$_4$ vs. doping and temperature. In particular, we show that the maximum of $\chi^{\text{Cu}}(T)$, which occurs at $T_{\text{max}}(x)$, is inversely proportional to the effective superexchange energy $J$ determined by Raman and neutron scattering.

The rest of this paper is organized as follows. The next section describes the experimental methods. In Sec. III, we present the data and work step-by-step through the analysis. The implications are discussed in Sec. IV, and a brief conclusion is given in Sec. V.

II. EXPERIMENTAL METHODS

The La$_{1.875}$Ba$_{0.125}$CuO$_4$ crystal studied here, with a mass of 0.6 g, was cut from the same single-crystal rod grown by the travelling-solvent floating-zone method that was the source of samples for previous work.

For the complementary measurements on $x = 0.095$ and $x = 0.155$, similarly sized crystals were used. The magnetization $M$ was measured with a SQUID (superconducting quantum interference device) magnetometer at Brookhaven ($H_{\text{max}} = 7$ T), and a vibrating sample magnetometer at IFW-Dresden ($H_{\text{max}} = 15$ T). In the case of La$_{1.875}$Ba$_{0.125}$CuO$_4$, $\chi = M/H$ was recorded for $H || [001] (c$-axis$)$ as well as $H || [100], [010]$ and $[110] (ab$-plane) in zero-field-cooled (ZFC) and field-cooled (FC) modes. [See the high-temperature-tetragonal (HTT) unit cell in Fig. 2(b) for definition of directions.] Measurements of $\chi_{[100]}$ and $\chi_{[010]}$ produce identical results, while $\chi_{[110]}$ shows clear distinctions. Bulk superconductivity (SC) in this sample appears below $\sim 2.3$ K in $H = 20$ G. SC fluctuations are detectable up to $\sim 40$ K for $H || c$ in 18,19,20 but are suppressed down to $\sim 20$ K ($\sim 5$ K) by a field of 2 T (7 T).

III. DATA AND ANALYSIS

Figure 2(a) shows the raw FC data for $\chi_c$ and $\chi_{[100]}$ in La$_{1.875}$Ba$_{0.125}$CuO$_4$. While there is no signature of the 235-K structural transition ($T_{\text{HT}}$) from the HTT to the low-temperature-orthorhombic (LTO) phase, there are clear changes at $T_{\text{LT}} = 54.5$ K, where the crystal transforms into the low-temperature tetragonal (LTT) phase 18,19,20,21,22. We know that charge-stripe order sets in at $T_{\text{LT}}$, followed by a significant decrease below 42 K. This decrease is not due to diamagnetism from SC, but is connected to the onset of spin stripe order at $T_{\text{SO}}$, as detected by $\mu$SR 23 and neutron diffraction 18,19,20. Below $H \sim 0.5$ T, $T_{\text{SO}}$ is marked by a clear kink in $\chi_{[100]}$ and $\chi_{[110]}$ as is shown in the insets of Fig. 3 as well as in Ref. 14.

A. Contributions to the measured susceptibility

The measured susceptibility $\chi_i(T)$ ($i = c, ab$) can be written as

$$\chi_i(T) = \chi_i^c(T) + \chi_i^{\text{core}} + \chi_i^{\text{VV}}.$$  (1)

The core diamagnetism $\chi_i^{\text{core}}$ amounts to $-1.01 \times 10^{-4}$ emu/mol, as determined from standard tables 23. The Van
Vleck susceptibility of the Cu$^{2+}$ ions, $\chi_i^{VV}$, depends on the direction of the field, but is independent of temperature; we will come back to it shortly. We can write the spin contribution as

$$\chi_i^s(T) = \chi_i^{Cu}(T) + \chi^P.$$  \hspace{1cm} (2)\

Here $\chi_i^{Cu}$ is the contribution from local moments, and

$$\chi_i^{Cu} = \frac{N g_i^2 \mu_B^2}{kT} \sum_r \langle S_r^S S_r^S \rangle,$$  \hspace{1cm} (3)\

where $N$ is the density of Cu atoms, $S_r^S$ is the spin component in direction $i$ for the Cu atom at site $r$; in the paramagnetic phase, $\chi_i^{Cu}$ remains anisotropic, as $g_{ab}$ and $g_c$ typically differ by 15% for Cu$^{2+}$ compounds. The isotropic Pauli susceptibility can be expressed as

$$\chi^P = (g \mu_B/2)^2 \rho(\epsilon_F),$$  \hspace{1cm} (4)\

where $g \approx 2$ and $\rho(\epsilon_F)$ is the electronic density of states at the Fermi energy, $\epsilon_F$. (In principle, the paramagnetic response of conduction electrons due to their spins is partially offset by their orbital response, in the form of Landau diamagnetism.) For a Fermi liquid, the density of states is independent of temperature, so that $\chi^P$ is a constant. As the cuprates do not follow conventional Fermi-liquid behavior, $\chi^P$ might be temperature dependent.

In simple metals, such as Na or Al, all of the spin response comes from delocalized conduction electrons. The doped cuprates are unusual metals, but one can nevertheless consider the extent to which the charge carriers also determine the magnetic response. Thus, we start our analysis with the question: Can we explain all of the temperature dependence in our single-crystal $\chi_i(T)$ simply in terms of $\chi^P(T)$?

To answer this, we consider the anisotropy between $\chi_{ab}(T)$ and $\chi_c(T)$. From Eqs. (1) and (2), we see that there are two sources of anisotropy, $\chi_i^{VV}$ and $\chi_i^{Cu}$. Looking at Fig. 2(a), we see that $\chi_c - \chi_{[100]}$ varies with temperature over the entire measurement range, and especially below $T_{SO}$. As noted above, the Van Vleck component is temperature independent, and hence the temperature-dependent anisotropy must come from the local-moment contribution. Thus, we immediately conclude that the temperature dependence of the spin susceptibility cannot come uniquely from a temperature dependence of the electronic density of states.

### B. Initial analysis of anisotropy

In principle, we can determine $\chi_i^{Cu}$ from the anisotropy of the measured susceptibility; however, to do this, we need to know the values of $\chi_i^{VV}$ and the $g_i$ factors. For isolated Cu$^{2+}$ ions, one has

$$\chi_i^{VV} = \frac{8 \mu_B^2}{\epsilon_0},$$ \hspace{1cm} (5)\
$$\chi_i^{ab} = \frac{2 \mu_B^2}{\epsilon_1},$$ \hspace{1cm} (6)\
$$g_c = 2 + 8 \lambda/\epsilon_0,$$ \hspace{1cm} (7)\
$$g_{ab} = 2 + 2 \lambda/\epsilon_1,$$ \hspace{1cm} (8)\

where $\lambda$ is the spin-orbit coupling, and the energies $\epsilon_0$ and $\epsilon_1$ are the crystal-field splittings between the 3$d_{x^2−y^2}$ and the tetragonally-split $t_{2g}$ orbitals. In the CuO$_2$ planes, the 3$d$ states turn into bands, and one must average over the Brillouin zone to properly evaluate these quantities. The only calculation of such factors for cuprates that we know of was performed for hypothetical Sc$_2$CuO$_4$. While we cannot apply those results directly to our case, we can use them as guidance.

We find that we must resort to fitting the data in order to determine the anisotropy factors. To do so, we will first consider the case $\chi^P = 0$; we will later come back to consider the impact of a finite $\chi^P$. From Eqs. (1)–(3), we find that, in the paramagnetic regime,

$$\chi_{ab} = \chi_{ab}^2 = \chi_c^2 = \chi_{ab}^2,$$ \hspace{1cm} (9)\

or, equivalently,

$$(\chi_{ab} - \chi_{ab}^{core})/\chi_{ab}^2 = (\chi_c - \chi_{ab}^{core})/\chi_{ab}^2 = \gamma.$$ \hspace{1cm} (10)\\

Thus, we must vary the anisotropic factors in order to get a unique $\chi_i^{Cu}/g_i^2$ from the experimentally measured $\chi_{ab}$ and $\chi_c$. We note from Eqs. (9) – (10) that

$$(g_c - 2)/(g_{ab} - 2) = \chi_c^{VV}/\chi_{ab}^{VV} = \gamma,$$ \hspace{1cm} (11)\

where one expects $\gamma \leq 4$. The analysis is relatively insensitive to the precise value of $\gamma$, so we reduce the number of adjustable parameters to two by setting $\gamma = 3$, approximately equal to the ratio calculated for Sc$_2$CuO$_4$. Applying the analysis to the temperature range 100 K $\leq T \leq 300$ K, we obtain $g_{ab} = 2.19, g_c = 2.58, \chi_{ab}^{VV} = 0.09 \times 10^{-4}$ emu/mole and $\chi_c^{VV} = 0.28 \times 10^{-4}$ emu/mole (see top row of Table 1), as well as the polycrystalline averages $g_{av} = 2.32$ and $\chi_{av}^{VV} = 0.15 \times 10^{-4}$ emu/mole. Finally, the results obtained for $\chi_i^{Cu} g_i^2/\chi_{ab}$ and $\chi_{[100]} g_{[100]}^2/\chi_{ab}$ are plotted in Figs. (c) and (d).
quite comparable to the theoretical results. In fact, we will reproduce this effect in Sec. IV.F.

![Graph showing field dependence of $\chi_{[100]}$ and $\chi_{[110]}$](image)

FIG. 3: (color online) $\chi$ of La$_{1.875}$Ba$_{0.125}$CuO$_4$ obtained in FC mode. (a) for $H \parallel [100]$. (b) for $H \parallel [110]$. Small deviations between curves (within error bar at bottom) for different fields and magnetometers due to experimental error were corrected by shifts in $\chi$ so that curves match for $T > T_{LT}$ where no field dependence was observed. Insets: FC and ZFC data for $H = 0.1$ T showing clear kinks at $T_{SO}$ and $T_{LT}$. (c) Comparison of the field dependence of $\chi^\text{Cu}_{av}/g^2_i$.

Our fitted values for the Van Vleck susceptibilities are quite comparable to the theoretical results of $\chi^{VV} = 0.15 \times 10^{-4}$ emu/mol and $\chi^{VV} = 0.4 \times 10^{-4}$ emu/mol. We note that considerably larger values for Van Vleck susceptibilities have been reported; however, those studies did not include the possibility of anisotropy in $\chi^\text{Cu}$. In fact, we will reproduce this effect in Sec. IV.F.

![Graph showing field dependence of $M$](image)

FIG. 4: (color online) $M(H)$ of La$_{1.875}$Ba$_{0.125}$CuO$_4$ obtained in ZFC mode. (a) for $H \parallel [100]$. (b) for $H \parallel [110]$. Small experimental errors were corrected so that at 15 T $M/H$ is consistent with $\chi$ in Fig. 3 (see error bar). For the 10 K curve, the data is limited to $H > 3$ T to avoid effects of superconductivity at lower fields. Inset: derivative $dM/dH$ calculated after 3-point averaging of $M(H)$ (except for $T = 20$ K with $H \parallel [100]$, where 2-point averaging was used due to the larger point spacing).

C. Spin anisotropy in the ordered state

Let us consider how the anisotropy is connected with the spin ordering. In the classic case of collinear AF order, Fig. (b), one measures $\chi^\text{Cu}$ for $H \parallel S$ and $\chi^\text{Cu}$ for the two orthogonal orientations; $\chi^\text{Cu}_i \to 0$ as $T \to 0$ while $\chi^\text{Cu}_i$ grows relative to the paramagnetic state. In contrast, for La$_{1.875}$Ba$_{0.125}$CuO$_4$ $\chi^\text{Cu}$ behaves like $\chi^\text{Cu}_i$, but $\chi^\text{Cu}_i$ and $\chi^\text{Cu}_i$ each seem to correspond to $\chi^\text{Cu} = \frac{1}{2}(\chi^\text{Cu}_i + \chi^\text{Cu}_i)$, as indicated in Fig. (a). This anisotropy is consistent with the stripe model. The stripes rotate by $\pi/2$ from one layer to the next, following the crystalline anisotropy and the effective spin anisotropy should rotate as well, with the Cu spins ordering within the ab-plane. (Note that twinning could also yield the same average non-collinear response for a completely collinear AF.)

D. Spin-flop transition

Next, consider the field dependence of $\chi_{[100]}$ shown in Fig. (a). Increasing the field above 4 T causes $\chi_{[100]}$ to rapidly increase for $T \lesssim 40$ K; when $H = 15$ T, $\chi_{[100]} = (g^2_{av}/g^2_i)$ matches $\chi^\text{Cu}_i(g^2_{av}/g^2_i)$. This behavior is
consistent with a spin-flop in the spin stripes. The spin-flop transition is clearly seen in the $M_{[100]}(H)$ curves of Fig. 5(a). $M_{[100]}$ becomes nonlinear for $T < 50$ K, with the transition marked by a peak in $dM_{[100]}/dH$ at $H_{SF} \sim 6$ T, as shown in the inset. Similar data for $\chi_{[110]}$ in Fig. 5(b) and $M_{[110]}$ in Fig. 5(b) exhibit a much broader transition with no peak in $dM_{[110]}/dH$ (see inset of Fig. 5(a)). The anisotropy in $dM/dH$ implies that the spins that flop at the transition initially have $S \parallel [100]$. Figure 5 illustrates the situation. At zero field, we presume that $S \parallel [010]$ in plane $z = 0$, and $S \parallel [100]$ in plane $z = 0.5$. For $H > H_{SF}$ along [100], spins in plane $z = 0.5$ flop so that the staggered moments are approximately perpendicular to the field (Fig. 5(b)). It is the gain in Zeeman energy, by slightly polarizing the spins in the field direction, that stabilizes the flopped state. When applying $H \parallel [110]$, spins in all planes continuously rotate until the staggered moment axis is perpendicular to $H$ (Fig. 5(c)), so there is no sharp transition.

Figure 6 shows the resulting phase diagram for $H \parallel [100]$. The LTO+LTT transition at $T_{LT}$ is field independent, while the onset of spin stripe order shifts from $T_{SO} = 42(3)$ K at 0.1 T to 49(3) K at 15 T. X-ray diffraction data indicate that charge stripes order at $T_{CO} = T^{LT}$ \cite{18,21,22}. Hence, for $T_{SO} < T < T_{CO}$ there are static charge stripes but no static spin stripes, which is consistent with recent inelastic neutron scattering results indicating the opening of a spin wave gap only for $T \lesssim T_{SO}$ \cite{18}. On the other hand, $\chi_e$ increases right at $T_{CO}$, which suggests that spins parallel to the CuO$_2$ planes; see Fig. 5(a). This means that below $T_{CO}$ spin dimensionality is effectively reduced from 2D-H to 2D-XY. (Exchange anisotropy becomes relevant even in the absence of static order.) This is extremely interesting because a 2D-XY system allows for topological order without additional anisotropies or interlayer coupling, with 2D SC being one possibility. Finally, Fig. 4 indicates the spin-flop transition for $T < T_{SO}$. $H_{SF}$ is of the same magnitude as in the LTT phase of antiferromagnetic

![Figure 5](image)

**Figure 5:** (color online) Model for spin structure of site-centered stripes as a function of field. (●, ○) Half-filled charge stripes. Stripes in adjacent planes at $z = 0$ and 0.5 are perpendicular. (a) spin structure for $H = 0$, (b) $H \parallel [100]$, (c) $H \parallel [110]$.

![Figure 6](image)

**Figure 6:** (color online) $H$-$T$ diagram of La$_{2-x}$Ba$_x$CuO$_4$ as determined for $H \parallel [100]$. Only for $H \lesssim 0.5$ T and $H \geq 7$ T an anomaly at $T_{SO}$ is observed. For SC properties see Ref. \cite{13}. La$_{1.8}$Eu$_{0.2}$CuO$_4$ \cite{30} suggesting that the in-plane gap has changed only moderately.

### E. Diamagnetism in paramagnetic state

The reader may already have noted in Fig. 7(c) the apparent anisotropy at $T \gtrsim 54$ K, where $\chi^c_{Cu}/g^2 < \chi^c_{Eu}/g^2$. This behavior does not make sense in terms of exchange anisotropy. Instead, we believe that $\chi^c$ bears a weak two-dimensional diamagnetic contribution associated with the vortex liquid state proposed by Li et al. \cite{36} based on magnetization measurements on La$_{2-x}$Sr$_x$CuO$_4$ for $0.03 \leq x \leq 0.07$. Similar behavior appears above $T_c$ at other compositions of La$_{2-x}$Ba$_x$CuO$_4$. Examples for $x = 0.095$ and 0.155 can be seen in Fig. 7.

### F. Anisotropy and doping-dependence of susceptibility

Now we will reconsider our initial assumption that $\chi^P = 0$. Obviously, that assumption is in conflict with the scaling analysis \cite{17,31}, which gives approximately $\chi^P \sim x$. To properly evaluate the latter form, we need to jointly analyze anisotropic measurements for samples with different hole contents. In Fig. 8(a) we show the raw data of $\chi_e$ on single crystals of La$_{2-x}$Ba$_x$CuO$_4$ with compositions from $x = 0.095$ to 0.155. Keeping in mind that $\chi_{Cu}^V$ and $g_i$ should be independent of doping, we now repeat our fitting of these parameters for all three samples simultaneously and with various choices for $\chi^P$. These choices and the fitted parameter values are listed in Table I, and the best-fit results for $\chi_{Cu}^P$ are plotted in Fig. 7(b-d). The quality of fit parameter $R$ is equal to the sum of the squared differences between the two versions of $\chi_{Cu}^P(g_{av}^2/g_i^2)$, with $H \parallel ab$ and $H \parallel c$, evaluated over the fit interval of 100 K $\leq T \leq 300$ K.

Case I corresponds to $\chi^P = 0$, and clearly it gives an excellent match of $\chi_{Cu}^P(g_{av}^2/g_i^2)$ for all three compositions. The fitted parameters are quite close to what we obtained for $x = 0.125$ alone (case 0). For cases III and IV, we fix...
TABLE I: Parameters from analysis of $\chi^P_i$ for different cases, as discussed in the text. Bold numbers indicate parameters that were varied during fit. Numbers in the first row result from individual fit for crystal with $x = 0.125$; see Fig. 1(c). In contrast, the data in rows I-IV were obtained by simultaneous fits for all $x$, see Fig. 2(b-d). Susceptibilities are given in units of $10^{-4}$ emu/mol. $R$ is the sum of squared differences.

| Case | $\chi^P_i$ at $x = 0.125$ | $\chi^P_i$ at $x = 0.155$ | $\chi^{VV}_i$ | $\chi^{VV}_i$ | $\chi^{ab}_i$ | $\chi^{c}_i$ | $R$ |
|------|-------------------------|-------------------------|---------------|---------------|---------------|-------------|-----|
| 0    | -                       | -                       | 0.09          | 0.28          | 2.19          | 2.58        | 0.005|
| I    | 0                       | 0                       | 0.11          | 0.33          | 2.18          | 2.56        | 0.16 |
| II   | 0.3                     | 0.3                     | 0.18          | 0.54          | 2.18          | 2.56        | 0.16 |
| III  | 0.66                    | 0.89                    | 1.13          | 0.33          | 2.58          | 3.74        | 36  |
| IV   | 0.66                    | 0.89                    | 1.13          | 1.04          | 2.03          | 2.10        | 9.4  |

$\chi^P(x)$ to values interpolated from results in Refs. [refs]. In case III, we use the $\chi^{VV}_i$ of case I and only vary the $g_i$, while all parameters are varied for case IV. In both, the matching is very poor. In fact, for case IV the resulting $\chi^{Cu}(g_{av}^2/g_i^2)$ becomes negative as a result of the large anisotropy in $\chi^{VV}_i$ that is required to compensate for the reduced magnitude of $\chi^{Cu}(g_{av}^2/g_i^2)$. With reduced values of $\chi^{VV}_i$, in case III, the $g_i$ factors become unreasonably large.

It is important to note that the analysis is insensitive to a doping-independent but finite $\chi^P$. Although unphysical, we present this situation in case II, which demonstrates a match of equal quality to Case I. We conclude this case simply to show a limitation in terms of uniqueness of our analysis.

While the values of $\chi^P(x)$ inferred from Refs. [refs] clearly seem to be unreasonable when anisotropy is taken into account, it is still possible that there is a $\chi^P(x)$ contribution of smaller magnitude. Hence, we have performed fits allowing $\chi^P(x) \sim x$ with a variable scale factor. Figure 7(e) shows $R$ as a function of the corresponding $\chi^P(x = 0.155)$. We see that $R$ strongly increases for $\chi^P(x = 0.155) \gtrsim 0.25 \times 10^{-4}$ emu/mol, which is a limit nearly five times smaller than the value given by the scaling analysis (see case IV in Table I). We conclude that if a temperature-independent contribution from mobile carriers is present, it must be an order of magnitude smaller than the estimates based on the scaling analysis of the static magnetic susceptibility.

IV. DISCUSSION

Our analysis indicates that the bulk spin susceptibility $\chi^b$ in La$_{1.875}$Ba$_{0.125}$CuO$_4$ comes dominantly from local Cu moments and that $\chi^b$ does not make a substantial contribution. In Fig. 1(c), we compare our results for $\chi^{Cu}_i$ with the prediction for the 2DHAF with $J = 100$ meV, where the estimate for $J$ is taken from a neutron scattering study of the magnetic excitation spectrum [22]. A clear implication of our analysis is that $\chi^b$ of the doped cuprate is greater than that of the undoped antiferromagnet. A comparison of the anisotropy below $T_{SO}$ suggests a substantial ordered moment, as found by muon spin-rotation studies [23,38]. If we estimate $\chi^{Cu}_i$ as

$$\chi^{Cu}_i = \frac{2\chi^{Cu}_i}{g_{av}^2/g_i^2} - \chi^{Cu}_i(g_{av}^2/g_i^2),$$

FIG. 7: (color online) (a) $\chi_i(T) \times 10^{-4}$ emu/mol for different dopings $x = 0.095$, 0.125, and 0.155. Note that the crystals with $x = 0.095$ and 0.155 are bulk superconductors with $T_c = 32$ K and 30 K, respectively. (b-d) Spin susceptibility $\chi_i(T)/(g_{av}^2/g_i^2)$ extracted from data in (a) under different conditions, as discussed in the text. Roman numbers I – IV refer to corresponding parameter sets in Table I. (e) Least squared deviation $R$ vs $\chi^P$ at $x = 0.155$ doping for fits in which a linear $x$ dependence of $\chi^P(x)$ is assumed. Open circles indicate $R$ values for cases I and IV from Table I.
then it appears that $\chi_{\parallel}^{Cu}$ remains finite at low temperature, in contrast to the expectation $\chi_{\parallel}^{Cu} \to 0$ for an undoped antiferromagnet. The lack of perfect spin ordering was already indicated by a Cu nuclear quadrupole resonance (NQR) study, where attempts to measure the antiferromagnetic resonance at temperatures down to 0.35 K provided evidence for a broad distribution of time scales.

For another comparison, Fig. 8 shows $\chi_{av}^{Cu}$ data for polycrystalline La$_{2-x}$Sr$_x$CuO$_4$ with $x = 0.08$ and 0.15, extracted from the bulk susceptibility using the same values for the core diamagnetism and Van Vleck susceptibility as for the La$_{1.875}$Ba$_{0.125}$CuO$_4$ result, which is shown as a polycrystalline average. We note that the La$_{2-x}$Ba$_x$CuO$_4$ result is consistent with the La$_{2-x}$Sr$_x$CuO$_4$ data both in magnitude and in terms of the trend of increasing $\chi_{av}^{Cu}$ with doping. We note that the latter observation is contrary to what has been inferred based on the scaling analysis.

In Fig. 8 we also compare calculations for the 2-dimensional Heisenberg antiferromagnet (2DHAF) with different values of $J$. As one can see, $\chi_{2DHAF}^{Cu}$ provides a reasonable estimate of $\chi_{max}^{Cu} = \chi_{av}^{Cu}(T_{max})$ for an appropriate $J$, but gives poor agreement for the temperature $T_{max}$ at which $\chi_{av}^{Cu}$ reaches its maximum. Now, we know from neutron scattering studies that doping significantly modifies the dynamic spin correlations from those of the undoped insulator, so it should not be surprising that $T_{max}$ would be reduced from that of a spin-only model system. On the other hand, $\chi_{Cu}^{max}$ might be less affected, as it corresponds to the response as antiferromagnetic correlations just begin to overcome disturbances from thermal or electron-scattering excitations. Indeed, Imai et al. concluded quite some time ago, on the basis of NQR measurements of the $^{63}Cu$ nuclear spin-lattice relaxation rate, that the Cu spin correlations in

![FIG. 8: (color online) $\chi_{Cu}^{av}$ vs $T$ for the La$_{1.875}$Ba$_{0.125}$CuO$_4$ single crystal and for polycrystalline La$_{2-x}$Sr$_x$CuO$_4$. Solid lines correspond to $\chi_{2DHAF}$ for different $J$.](image)

![FIG. 9: (color online) Comparison of experimental $\chi_{Cu}^{av}(T_{max})$ (evaluated with our $\chi_{av}^{Cu}$) and values calculated from experimental $J$ values and $\chi_{av}^{Cu}$ for La$_{2-x}$Sr$_x$CuO$_4$ and for polycrystalline La$_{2-x}$Sr$_x$CuO$_4$.](image)
The developing picture in underdoped cuprates is one of considerable incoherence of the electronic correlations at high temperatures. Optical conductivity studies of La$_{2-x}$Sr$_x$CuO$_4$, La$_{2-y}$Ba$_y$CuO$_4$, and YBa$_2$Cu$_3$O$_{y+4+x}$ indicate that the Drude peak, often taken as an indication of coherent behavior, disappears as the temperature is increased towards the pseudogap crossover temperature, $T^*$. A recent analysis of temperature- and doping-dependent Hall-effect measurements suggests that not only the doped holes but also thermally-activated carriers participate in the transport at high temperature. It has been noted that $T^*$ identified from transport properties is quite close to $T_{\text{max}}$ from the bulk susceptibility. Thus, it appears that the coherent nodal metallic states and the antiferromagnetic spin correlations develop together on cooling below $T^*$. This co-evolution is compatible with the dynamic susceptibility in underdoped cuprates as suggested by neutron scattering studies.

There have been suggestions that the pseudogap might be the consequence of spin-density-wave (SDW) correlations. In that case, the magnetic response would come dominantly from the charge carriers. This possibility now seems unlikely, as we have argued that the static susceptibility is dominated by the response of local moments, and there is no evidence of coherent electronic states at $T \gtrsim T^*$ from which a SDW might develop.

Finally, our interpretation of the static susceptibility is compatible with the distinct dynamic responses detected by nuclear magnetic resonance studies at Cu and O in-plane sites in La$_{2-x}$Sr$_x$CuO$_4$. The site-dependent responses can be understood in terms of the spatial inhomogeneity associated with stripe correlations. By symmetry, the O site does not see the spins on the two nearest-neighbor Cu sites in the uniform antiferromagnetic state; however, stripe correlations can break this symmetry, allowing O to detect the spin susceptibility. In fact, it is the existence of the stripe correlations that allows for the coexistence of regions of antiferromagnetically-coupled Cu moments and mobile charge carriers.

V. CONCLUSION

To conclude, we have presented evidence that the bulk susceptibility in underdoped and optimally-doped La$_{2-x}$(Sr,Ba)$_x$CuO$_4$ is dominated by the response of antiferromagnetically-coupled Cu moments, with little contribution from free carriers. This conclusion has implications for the interpretation of the dynamic magnetic susceptibility. If electronic quasiparticles were to contribute significantly to the dynamic susceptibility through Fermi-surface-nesting effects, then one would expect an associated contribution to the static susceptibility. The absence of a substantial isotropic component of electronic origin in $\chi^s$ raises questions about the relative importance of quasiparticle contributions to the dynamic susceptibility in underdoped cuprates.

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