Neutron scattering and magnetization studies of $Ba_2Cu_{2.95}Co_{0.05}O_4Cl_2$: A decorated two-dimensional antiferromagnet

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$Ba_2Cu_3O_4Cl_2$ has two interpenetrating square Cu sublattices, one with square root 2 times the in-plane spacing of the other. Isotropic magnetic interactions between the two sublattices are completely frustrated. Quantum fluctuations resolve the intrinsic degeneracy in the ordering direction of the more weakly coupled sublattice in favor of collinear ordering. We present neutron scattering and magnetization studies of the magnetic structure when the Cu ions are substituted with Co. The Co spins create new magnetic interactions between the two sublattices. The ordering behavior of both Cu sublattices is retained largely unmodified. Between the phase transitions of the two sublattices spin-glass behavior is observed. Magnetization results show a strong enhancement to the ferromagnetic aspect of the magnetic structure. The combination of glassy behavior and large moments strongly suggest that the Co moments induce the formation of local canted states.

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I. INTRODUCTION

A wealth of experiments have explored the competition between different types of fluctuation which disrupt ordered phases.[1–4] A typical combination is thermal fluctuations and quenched random fluctuations due to disorder. The strength of the thermal fluctuations is controlled via the temperature while the strength of the quenched fluctuations is controlled by dilution of the constituents. Since both forms of fluctuation perturb the order they are never in profound competition. Here, by contrast, we study the interaction between quantum fluctuations, which give rise to an order-from-disorder transition,[5] and quenched random fluctuations. As the quantum fluctuations drive the ordering in the chosen material the two sets of fluctuations are in direct competition and the results can be expected to be fascinating.

$Ba_2Cu_3O_4Cl_2$ is a lamellar cuprate with two Cu sublattices.[6] The Cu_I sublattice has CuO_2 layers and orders antiferromagnetically at $T_{N,I}=332.4\pm0.8$ K. The Cu_{II} sublattice interpenetrates the Cu_I sublattice with one Cu_{II}^{2+} ion at the center of every second plaquette. The position of the Cu_{II} sublattice relative to a Cu_I plaquette is shown in Fig. 1(a). The plaquettes are staggered from one plane to the next. The Cu_{II} moments order antiferromagnetically at $T_{N,II}=31.31\pm0.01$ K. The isotropic couplings between the Cu_I and Cu_{II} sublattices are completely frustrated and this means that more subtle interactions are manifest.[7] Collinear ordering of the second sublattice with respect to the first is preferred due to the contribution of quantum fluctuations. As a result the Cu_{II} system has Ising class critical properties.

Detailed studies of the pure 2342 materials have been carried out in recent years.[6–11] In an applied magnetic field, in the temperature range $T_{N,II}<T<T_{N,I}$, $Sr_2Cu_3O_4Cl_2$ develops a small magnetic moment on the Cu_{II} sites. Chou and co-workers studied the behavior as a function of temperature, field strength, and field orientation.[7] Combined with the known behavior of the Cu_I order parameter it was possible to determine the nature of the coupling between Cu_I and Cu_{II} moments. The isotropic average coupling $J_{av}=-12\pm9$ meV and may originate from superexchange and direct exchange. The anisotropic coupling is $|J_{pd}|=27\pm1$ μeV and is pseudodipolar. Noro and co-workers studied the weak ferromagnetism in $Ba_2Cu_3O_4Cl_2$ in the pure material and with Zn or Ni substitution.[8] The behavior of the pure material has similar magnetization, ferromagnetism, and susceptibility to the Sr material. Neither substitution of the spin system resulted in any new phase transitions or glassy behavior. Adding Zn

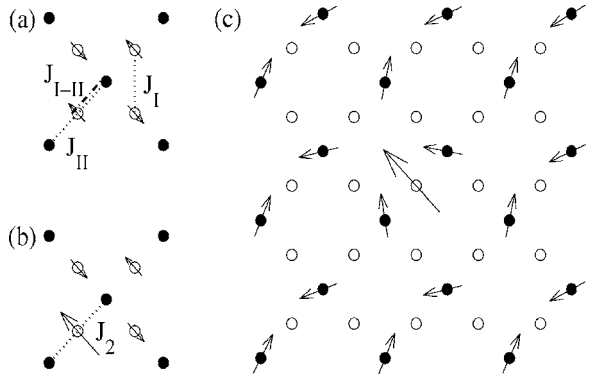

FIG. 1. Schematic illustration of the magnetic couplings. Open (closed) circles indicate $Cu_I(Cu_{II})$ sites. (a) J_I couples the Cu_I moments and J_{II} couples the Cu_{II} moments. The isotropic coupling J_{I-II} between sublattices is completely frustrated. (b) When Co is added (large arrow) there is a new effective coupling J_2 between the Cu_{II} moments. (c) Schematic illustration of the canted structure. The Co moment in the center induces a ferromagnetic ordering in nearby Cu_{II} moments. Antiferromagnetic order can form independently in the orthogonal direction.

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reduced the ordered moment by a small amount. Adding Ni reduced the ordered moment by almost a factor of 2 without altering the transition temperatures. Apart from very close to the two ordering transitions the ferromagnetic component is entirely suppressed. Furthermore, the anisotropy in the in-plane susceptibility disappears. These effects suggest that Ni substitution weakens the coupling between the two sublattices.

Kim and co-workers[9,10] made a comprehensive elastic, quasielastic, and inelastic neutron scattering study of $Sr_2Cu_3O_4Cl_2$. The role of quantum fluctuations in coupling Cu_I and Cu_{II} subsystems were demonstrated via the dramatic variation in the energies of the spin-wave modes as the Cu_{II} subsystem orders. The behavior is in quantitative agreement with theoretical predictions.[12] The dispersion of a Cu_{II} mode was measured out to the zone boundary and this provided precise verification of theoretical calculations. The major interactions were found and are indicated in Fig. 1(a). For the Sr material $J_I \sim 130$ meV while $J_{II} \sim |J_{I-II}| \sim 10$ meV (J_I and J_{II} had not been determined by Chou and co-workers[7]). The similar transitions and transition temperatures suggest that the coupling strengths will be very similar for the Ba material.

Here we are studying this intriguing two sublattice antiferromagnet with the Cu moments mixed with ~2% Co. This is seen to modify some of the magnetic behavior substantially. Similar magnetic systems have been considered theoretically and were called a *decorated* model. Villain considered whether spin-glass behavior could occur for an insulator with only antiferromagnetic interactions.[13] He first demonstrated that competing ferromagnetic and antiferromagnetic interactions emerge for a two sublattice Ising antiferromagnet with spin dilution of one of the sublattices. Subsequently a number of promising scenarios were considered including local canted states: An antiferromagnetic ground state is perturbed when an interstitial magnetic impurity is introduced. The neighboring moments cant due to their interaction with the interstitial moment and this produces a local canted state. The local canted state is characterized by an $(n-1)$ component magnetic moment which is perpendicular to the ordering direction of the antiferromagnetic ground state. If there are many local canted states in a system they will interact and undergo a spin-glass transition. Hence different components of the magnetic moment order separately via antiferromagnetic and spin-glass transitions.

Using an alternative approach, Henley considered the role of thermal, quantum, and concentration fluctuations in a frustrated antiferromagnet.[14,15] He considered a two-sublattice antiferromagnet, in contrast to $Ba_2Cu_3O_4Cl_2$, both sublattices were on the same sized lattice. He, following others,[5] argued that thermal and quantum fluctuations of one sublattice give rise to collinear ordering in the other. This is because the largest fluctuations are transverse to the spin direction for both sublattices. The coupling between these fluctuations reduces the energy. The fluctuations in composition change the magnitude of the magnetic moment at random and this is equivalent to a longitudinal fluctuation. In order to couple transverse fluctuations to these longitudinal fluctuations the neighboring spins on the other sublattice align perpendicular to them. If the perturbation due to variations in composition is too small the thermal and quantum fluctuations will maintain the Ising-type symmetry. The composition fluctuations will then give random exchange fields which will pin the ordering direction in various locations and may prevent the formation of a long-range-ordered state.

Experimental studies of $Ba_2Cu_{2.95}Co_{0.05}O_4Cl_2$ are described and the results presented (Sec. II) and discussed (Sec. III) below.

II. RESULTS

A single crystal of $Ba_2Cu_{3-x}Co_xO_4Cl_2$ with $x=0.05$ was prepared by a direct melt method from powder samples.[8] The laminar single crystal had dimensions $5\times4\times0.8$ mm^3 and a well developed (0 0 1) plane. The crystal is tetragonal[16] (space group $I4/mmm$) with lattice constants $a=5.514$ Å and $c=13.711$ Å at $T=30$ K. The Cu_I ordering was studied using neutron scattering with the crystal mounted in the $(H\ 0\ L)$ plane while the Cu_{II} magnetism was observed in the $(H\ H\ L)$ scattering plane. The location of reflections in these planes is shown inset to Fig. 2(a). The neutron scattering measurements were performed on the BT7 and BT9 instruments at the National Institute for Standards and Technology, Center for Neutron Research (NCNR). The incident neutrons of energy 14.7 meV were selected with a (0 0 2) graphite monochromator and the collimation was generally [60′-40′-40′-80′] on BT7 and [40′-40′-40′-80′] on BT9. A pyrolytic graphite filter was used to suppress higher-order contamination. An (0 0 2) graphite analyzer was used to improve the instrumental resolution and to suppress background counts. For the quasielastic study of the diffuse scattering a two-axis configuration was employed. The magnetization was measured using a superconducting quantum interference device magnetometer over a temperature range 4–400 K.

The temperature dependence of the neutron scattering intensity of the (1 0 1) reflection is shown in Fig. 2(a). The additional component to the scattering below $T=324.9\pm0.4$ K is due to the Cu_I antiferromagnetic structure. The solid line shows the order-parameter behavior for pure $Ba_2Cu_3O_4Cl_2$. For this line the exponent $\beta=0.29\pm0.02$ and this is close to, but somewhat less than, the value $\beta=0.35$ expected for a 3D XY phase transition. The addition of Co is seen to have very little effect on this transition.

Different behavior occurs at lower temperatures involving the Cu_{II} spins. Figure 2(b) shows the neutron scattering observed at the $(\frac{1}{2}\ \frac{1}{2}\ 4)$ position for temperatures above 30 K. The choice of a large L component to the wave vector enhances the scattering from magnetic moments lying within the planes. The scattering was weak with no clear peak and the results presented are integrated counts. The measurements were made in two-axis configuration in order to reduce the energy resolution. The intensity is observed to decrease slowly with decreasing temperature with a more substantial drop beginning around $T\sim100$ K. This diffuse scattering is different in character to that observed in the pure material.[10,11] In the pure case rods of scattering are observed in the vicinity of $T_{N,II}$ that reach a peak intensity at the transition and diminish in the ordered phase. The diffuse scatter-

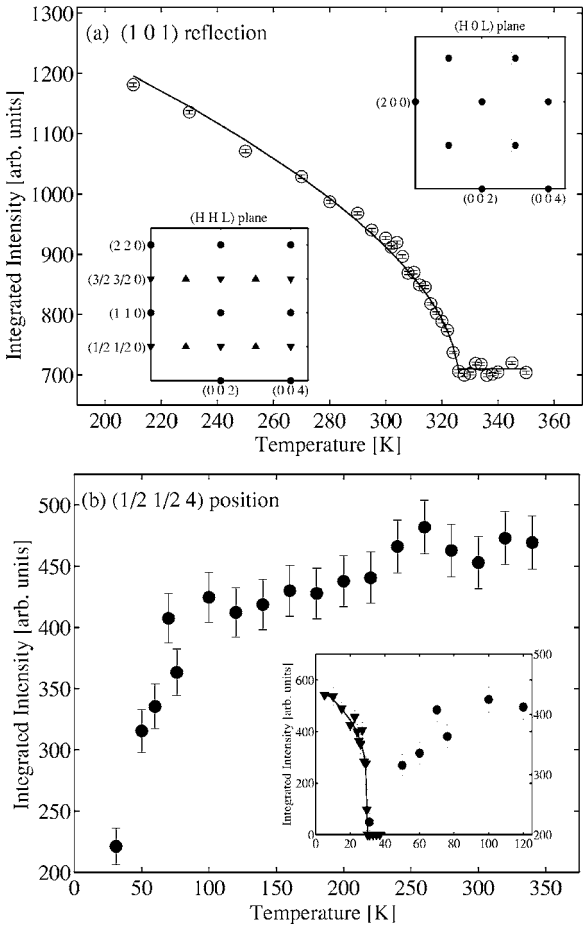

FIG. 2. (a) The temperature dependence of the scattered intensity at the (1 0 1) position. The growth in intensity corresponds to the ordering of the moments on the Cu_I sublattice. The solid line is the known behavior of the pure material (Ref. 6). Inset: reciprocal-lattice diagrams of the (H 0 L) and (H H L) planes explored in these experiments. The full circles are the nuclear Bragg reflections; the open circles are the Cu_I magnetic Bragg reflections and the full (up and down) triangles are the magnetic Bragg reflections from the two Cu_{II} domains. (b) The temperature dependence of the short-range correlations at the $(\frac{1}{2}\ \frac{1}{2}\ 4)$ position. The intensity drops for $T \leqslant 100$ K. Inset: the triangles and the circles show the temperature dependence of the $(\frac{1}{2}\ \frac{1}{2}\ 2)$ and $(\frac{1}{2}\ \frac{1}{2}\ 4)$ positions, respectively. The growth of intensity at the $(\frac{1}{2}\ \frac{1}{2}\ 2)$ position is due to antiferromagnetic order involving the Cu_{II} sites.

ing observed here corresponds to very short-range correlations which appear to freeze out well above $T_{N,II}$. The results were confirmed to be similar via more sparse measurements at the $(\frac{1}{2}\ \frac{1}{2}\ 2)$ position. The addition of 2% of Co increases the effective interactionstrength between the Cu_{II} spins. One quarter of the Cu_{II} moments have a Co as a nearest neighbor on one of the two sublattices. For these moments the *transition temperature* effectively triples—in line with the diffuse scattering. Inset to Fig. 2(b) is the scattering measured at the $(\frac{1}{2}\ \frac{1}{2}\ 2)$ position below $T=30$ K superimposed on the temperature dependence of the diffuse scattering. The enhanced intensity of scattering results from the ordering of the Cu_{II} moments. In this case $\beta=0.102\pm0.011$. As seen here the temperature dependence of the order parameter is very similar to that of the pure material[6] where $\beta=0.117\pm0.007$. Both of these are close to the 2D Ising value of $\beta=0.125$. Figure 3 demonstrates the short-range nature of the low-temperature magnetic structure perpendicular to the planes. Within the Cu_3O_4 planes the $(\frac{1}{2}\ \frac{1}{2}\ 3)$ peak width is comparable to the instrumental resolution. By contrast the magnetic reflection is seen to be broader than the instrumental resolution out of the plane. The magnetism is correlated over 157 ± 14 Å in the [0 0 L] direction which is a little over ten unit cells. This result suggests that two-dimensional Ising behavior is retained.

Magnetization measurements have been carried out for $Ba_2Cu_3O_4Cl_2$ and for two levels of Co substitution ($x=0.05, 0.1$). One of these is similar to the sample described above. Figure 4(a) shows the magnetization along the [1 1 0] direction. The two traces for each sample are for zero-field cooling (ZFC) and cooling in a 0.1 T field (FC). All samples exhibit a transition at $T\sim330$ K corresponding to ordering on the Cu_I sites. The transition shifts up in temperature with Co substitution due to the increase in the average moment. For the pure $Ba_2Cu_3O_4Cl_2$ sample the ZFC curve shows a sharp discontinuity at $T\sim30$ K corresponding to the onset of antiferromagnetic order. The key feature of these data is the evidence of a transition at intermediate temperature induced by the Co substitution. For $x=0.05$ the first transition occurs at $T\sim150$ K and appears rounded; a second discontinuity is observed at $T\sim30$ K. These results suggest that order is occurring on the Cu_{II} sites in two steps. The magnetization observed is greatly enhanced as a result of substitution of a small amount of Co.

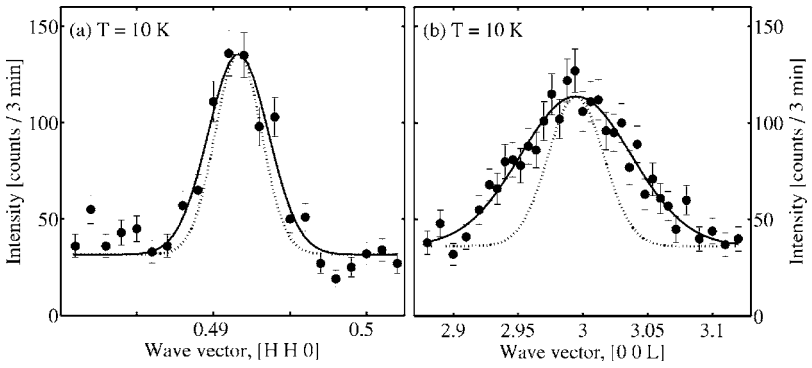

FIG. 3. Scans of neutron scattering intensity versus wave-vector transfer for (a) $\mathbf{q}=(HH3)$ and (b) $\mathbf{q}=(\frac{1}{2}\ \frac{1}{2}\ \mathrm{L})$. The reflection corresponds to the antiferromagnetic order of the Cu_{II} sublattice. The dotted line is the instrumental resolution. Correlation between the Cu_{II} ordering on different planes is evidently short range.

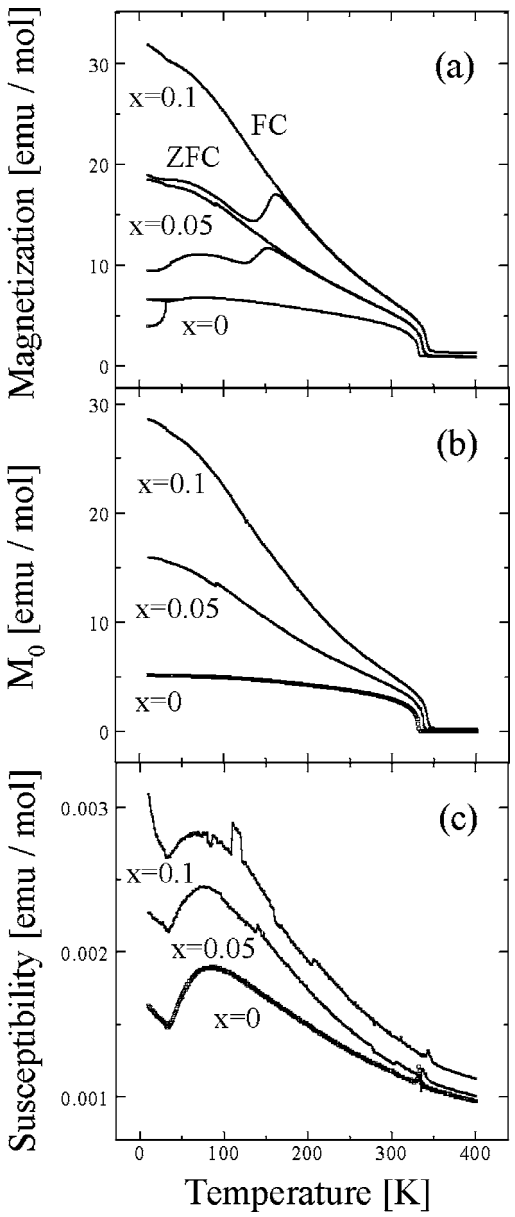

FIG. 4. Temperature dependence of (a) the magnetization along [1 1 0] in $Ba_2Cu_{3-x}Co_xO_4Cl_2$ single crystals, measured after field cooling (0.1 T) and zero-field cooling. The magnetic hysteresis shows the presence of the short-range order. (b) the weak ferromagnetic moment M_0 along [1 1 0] in $Ba_2Cu_{3-x}Co_xO_4Cl_2$ single crystals. The canting angle of Cu_{II} and Cu_I moments increases steeply due to the doping of Co ions. (c) the magnetic susceptibility along [1 1 0] in $Ba_2Cu_{3-x}Co_xO_4Cl_2$ single crystals. The increase in the paramagnetic susceptibility by Co doping is associated with the formation of local canted states.

The dependence of magnetization on the applied field strength can be fitted by $M(H)=M_0+\chi H$, indicating theexistence of a weak ferromagnetic component M_0. The temperature dependence of M_0 is shown in Fig. 4(b) and the susceptibility in Fig. 4(c). The results for pure $Ba_2Cu_3O_4Cl_2$ along [1 1 0] shown here and also along other directions have been published previously by Noro and co-workers.[8] In contrast to Zn and Ni substitution, Co greatly strengthens the weak ferromagnetism. Figure 4(b) shows $M_0(T)$ along [1 1 0]. The pure $Ba_2Cu_3O_4Cl_2$ trace indicates that a ferromagnetic component is developed at $T\sim 330$ K and quickly saturates. Co substitution evidently has two effects: first, the transition temperature for the appearance of the Cu_I structure rises. Second, the ferromagnetic moment never saturates but continues to rise steadily down to low temperatures. A 2% substitution triples the ferromagnetic moment with an increase up to 4% giving a sixfold enhancement! In $Sr_2Cu_3O_4Cl_2$ it was shown that the ferromagnetic moment tracked the behavior of the order parameter for the Cu_I sites.[7] Here the order-parameter behavior is shown in Fig. 2(a). Evidently the ferromagnetic contribution departs from this. The ferromagnetic contribution to the magnetic structure of pure $Ba_2Cu_3O_4Cl_2$ is attributed to a canting of the magnetic moments. This canting of the moments is augmented, both by Co substitution and then by decreasing temperature.

Figure 4(c) shows the magnetic susceptibility as a function of temperature and Co substitution along the [1 1 0] direction. The overall shape changes little. The magnitude of the susceptibility is observed to increase with Co substitution and this is the first indication of the presence of the Co moments on the Cu_{II} sites. On top of this the susceptibility climbs strongly at the lowest temperatures with increasing Co substitution.

III. DISCUSSION

We have made a study of the magnetic properties of $Ba_2Cu_{2.95}Co_{0.05}O_4Cl_2$ using neutron scattering and magnetization measurements. For the pure material the behavior for $T<T_{N,I}$ is marked by a single phase transition. Here we observe evidence of a different transition for $T\sim 100$ K and a transition very similar to that of the pure material at around $T_{N,II}$. The Co substituted material also exhibits a much larger ferromagnetic moment than the pure material.

The Cu_I spins with Co substitution order in the same way as they do in the pure material, albeit with an increased average moment. Unlike in the pure material case there is now an uncompensated moment component that is experienced by the Cu_{II} sites. There are two useful ways to think about the effect on the Cu_{II} sites. First, the Co substitution can be thought of as creating new, effective, interactions between the Cu_{II} spins. Second, the Co substitution can be thought of as longitudinal fluctuations which interact with the thermal and quantum fluctuations of the Cu_{II} spins.

Effective interactions between Cu_{II} spins give rise to the interesting behavior we observe in $Ba_2Cu_{2.95}Co_{0.05}O_4Cl_2$. Figure 1(a) shows the cancellation of the Cu_I contributions at the Cu_{II} sites. A Co moment on the central site in this figure would strengthen the coupling between Cu_{II} moments but would not create anything new. The effective interactions arise from the Co moments on the Cu_I sites pictured in Fig. 1(b). The existence of the uncompensated moment introduces an effective ferromagnetic interaction J_2 between Cu_{II} moments. As pointed out by Villain, ferromagnetic interactions at random locations in an antiferromagnetic material would give rise to spin-glass behavior in an Ising system.[13] In the case of *XY* or Heisenberg symmetry the frustration between ferromagnetic and antiferromagnetic interactions

can be resolved by canting of the Cu_{II} moments close to the Co site. This situation is pictured in Fig. 1(c) and exhibits simultaneous ferromagnetic and antiferromagnetic order. Villain considered a similar case with interstitial magnetic impurities: a *decorated* model.[13] In that case antiferromagnetic coupling between the host and interstitial were chosen. For $Ba_2Cu_{2.95}Co_{0.05}O_4Cl_2$ the local canted states have a nonzero moment as is evident in our magnetization results. In Villains model the local canting compensates the impurity moment and a quadrupolar field results.[13]

The substituted Co moments on the Cu_I sites are longitudinal fluctuations in the field experienced by the Cu_{II} moments. There is a coupling between Cu_I fluctuations and the fluctuations of the Cu_{II} moments. In the pure material the coupling gives rise to collinear ordering.[5,17] This is because the fluctuations are predominantly transverse to the moment direction for both Cu_I and Cu_{II} spins. Here the Co substituted Cu_I spins have large static longitudinal fluctuations. In order for the Cu_{II} thermal and quantum fluctuations to couple to the longitudinal fluctuations it is necessary for the Cu_{II} spins to align perpendicular to the Cu_I spins. This model was studied by Henley.[15] Canting similar to Fig. 1(c) is the prediction of considerations of the effective interaction and the fluctuations. Evidently longitudinal fluctuations also exist due to the Co substitution on the Cu_{II} sublattice. These are at random fixed locations and are dilute. These fluctuations will couple to the thermal and quantum fluctuations of the Cu_I moments and hence these Co moments will also favor perpendicular alignment.

Our elastic neutron diffraction results indicate that the Cu_I and Cu_{II} sublattices of $Ba_2Cu_{2.95}Co_{0.05}O_4Cl_2$ order in a very similar manner to the pure material (Fig. 2). We assume that the low-temperature ordering of the Cu_{II} spins is collinear to the ordering of the Cu_I spins as in the pure material. This assumption is supported by the observation that substitution with Co has hardly changed the ordering temperature and critical properties. If there were competition between collinear and perpendicular ordering, due to the Co substitution, it might have been expected that the low-temperature transition would have moved away from Ising critical properties. The quasielastic measurements [Fig. 2(b)] revealed diffuse scattering at intermediate temperatures which is associated with very short-range correlations. This scattering is not the same as that observed in the pure material.[10] Here the correlations are due to the local ordering close to the Co moments on both sublattices. The Co moments on the Cu_I sites create a preferred orientation for the antiferromagnetic fluctuations which is perpendicular to the Co moment. The local canted states give rise to the short-range order. It appears likely that the drop in quasielastic intensity is associated with a freezing of the perpendicular spin components while the collinear component retains the pure material behavior. It is the interactions between canted regions that lead to spin-glass freezing.

Other magnetic effects also occur in the same temperature range where we observe changes in the diffuse scattering. The development of antiferromagnetic Cu_I domains in the vicinity of 100 K has been studied in detail by Parks and co-workers.[18] This is a rare magnetization study of antiferromagnetic domains and was possible because of the ferromagnetic moment developed on the Cu_{II} sites due to the pseudodipolar interaction. Two kinds of Cu_I domains are possible and the orientation of the associated ferromagnetic moments are mutually orthogonal and so the domain populations can be studied via the magnetization signal. The application of a high magnetic field leads to a single domain dominating the sample. As the field is reduced domains of the other type begin to nucleate. At 100 K the susceptibility for domain wall formation is maximum. Additional studies were made using neutron scattering showing the decrease in intensity of the (3 2 1) reflection at around 100 K. We do not believe that the decrease in scattering intensity at the $(\frac{1}{2}\ \frac{1}{2}\ 4)$ position at around 100 K is associated with the proliferation of domain walls in the Cu_I spin system. This is true in principle because we did not use an applied magnetic field to create a monodomain sample. It is also true in practice as can be observed from our measurements: At this temperature the Cu_{II} antiferromagnetic fluctuations at $(\frac{1}{2}\ \frac{1}{2}\ 4)$ are very short ranged; there is no peak in reciprocal space. By contrast, the Cu_I antiferromagnetic structure has long-range order. Evidently there is no correspondence between the size of the Cu_I and Cu_{II} antiferromagnetic domains at these temperatures.

Figure 4(a) shows that the three regimes we have observed via neutron scattering are also observed in the magnetization studies presented here. The first regime is the high-temperature Cu_I ordering. Then the spin-glass freezing transition emerges at intermediate temperatures on diluting with Co. Importantly, the behavior in the middle regime is seen to be different: The divergence between field-cooled and zero-field-cooled magnetization measurements for the $x = 0.05, 0.1$ measurements has no parallel in the pure material [Fig. 4(a)]. At low temperatures the ordering of the Cu_{II} sublattice persists (presumably for those moments not pinned by a Co moment). Figure 4(b) shows that in a magnetic field applied along the [1 1 0] direction the ferromagnetic moment M_0 is substantially larger than that in the pure material and has different temperature dependence. In the pure material M_0 tracks the Cu_I order parameter and is induced by pseudodipolar coupling between sublattices. In the Co substituted system the applied field aligns the Co moments on the Cu_I sites and the canted contributions on the Cu_{II} sites as pictured in Fig. 1(c). Some canting of the Cu_I moments will also occur.[7] The combination leads to a large ferromagnetic contribution. Figure 4(c) shows that these local large moments also give an increase in the susceptibility. Magnetization measurements have been made for $Ba_2Cu_3O_4Cl_2$ with the Cu diluted with Zn, Ni, and Co. In the case of Zn and Ni no new transitions appear[8] whereas with Co spin-glass behavior results. These differences stem from the magnitude of the effective interactions induced. The magnetic interaction between the spin of a Cu^{2+} ion on a Cu_I site and the Cu_{II} spins is canceled by the other spins on the Cu_I sublattice. Ni^{2+} and Zn^{2+} have moments that differ from Cu^{2+} by $\frac{1}{2}$. Hence Zn and Ni on Cu_I sites result in an uncompensated spin one-half moment which is apparent to the Cu_{II} moments. Co^{2+} has a moment that differs from Cu^{2+} by 1 hence for Co the effect is doubled. Since J_{II} and J_{I-II} are equal and opposite only Co is likely to give rise to new and dominant interactions between the Cu_{II} spins.

Our neutron scattering and magnetization results are consistent with the Co substitution leading to a new transition in

the Cu_{II} spin system. Spin-glass freezing at ~100 K precedes the antiferromagnetic ordering at ~30 K. We interpret our observations in terms of the involvement of both the parallel and perpendicular spin components on the Cu_{II} sites [Fig. 1(c)]. The change in behavior is due to the influence of the Co moments located on the Cu_I sites; this can be explained in terms of new effective interactions or alternatively coupling between fluctuations. Either way the Cu_{II} moments are caused to form local canted states around the Co moments. The local canted states frustrate each other and lead to spin-glass freezing. The local cluster has a substantial moment and this is reflected in the magnetization measurements. At lower temperatures the portion of the Cu_{II} moment that is collinear with the Cu_I ordering direction (and not pinned by a Co moment) orders antiferromagnetically as in the pure material. This system exhibits a delicate balance of quenched random and quantum fluctuations. The former controls the magnetic behavior at intermediate temperatures while the latter yields order at lower temperatures.

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