

Magnetic properties of $\text{La}_2\text{Cu}_{1-x}\text{M}_x\text{O}_4$ with $M = \text{Zn}$ and Ni

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The temperature ($1.8 \leq T \leq 400$ K) and concentration ($0 \leq x \leq 0.10$) dependence of the magnetization and magnetic susceptibility of $\text{La}_2\text{Cu}_{1-x}\text{M}_x\text{O}_4$ with $M = \text{Zn}$ and Ni have been measured. The results are compared with the recent measurements of the concentration ($0 \leq x \leq 0.10$) dependence of the muon spin rotation of $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$. The antiferromagnetic order present in La_2CuO_4 , which is very sensitive to the introduction of impurities which create mobile holes (such as Sr substitution of the La site), is much less sensitive to impurities which do not create mobile holes in the CuO_2 planes. The Néel temperature T_N is depressed rapidly with Zn doping and the width of the peak in the susceptibility increases with x , and thus the peak becomes less well defined. The depression of T_N in Ni-doped La_2CuO_4 and the increase in the width of the susceptibility peak are less rapid compared to those in the Zn-doped material. We also study the influence of doping on the metamagnetic behavior which is concomitant with the antiferromagnetism in these systems.

I. INTRODUCTION

Much of the recent research on the Cu-O based high- T_c superconductors have been directed at studying the common salient features in their magnetic properties. One important feature is that as the mobile charge carrier density is decreased there is a concomitant suppression of the superconducting transition temperature T_c followed by the appearance of a long-range three-dimensional (3D) antiferromagnetic state.¹⁻³ The phase diagram reported for these systems is partially responsible for the suspicion that a pairing mechanism other than phonon mediated is operative in these systems. Most of this attention has been directed at pairing models originating from a variety of approaches applied to the two-dimensional (2D) Hubbard, t - J , and other related models.⁴ Generally these models rely on some type of magnetic interactions to drive the charge carrier pairing responsible for the superconductivity. Thus, it is critical to establish a more complete understanding of the unusual magnetic response displayed by the parent compounds, e.g., La_2CuO_4 and the change of their magnetic response with changes in carrier density and impurities.

The effect of both chemical and magnetic impurities on conventional superconductivity has long been recognized for its contributions to the understanding of conventional pairing and the interplay between magnetism and superconductivity. In high- T_c oxides, substitutional studies have been done for Cu in both La_2CuO_4 and $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ (1:2:3), for Y and Ba sites in 1:2:3 and substitutional studies have been performed where Ca is replaced by several of the rare-earth ions in the Bi-based oxide superconductors. Some of the most interesting effects have been obtained in Sr-doped

$\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$,² in studies where Zn is used to substitute Cu in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (Ref. 5) and 1:2:3 materials,⁶ and Pr-doped 1:2:3 materials.⁷ Small amounts of disorder in the CuO_2 planes, e.g., the substitution of Zn^{2+} for Cu^{2+} in both $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and 1:2:3, causes a rapid depression of T_c along with a rapid increase in the resistivity as $T_c \rightarrow 0$ due to a possible disruption of the Cu 3d-O 2p hybridization.⁶ In Pr-doped 1:2:3 and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, T_c is depressed rapidly due to a combination of p -hole filling and/or magnetic pair breaking.⁷ While there have been numerous studies focusing on the effects of impurities on superconducting properties, little is known about the effects of impurities on many of the normal-state properties and, in particular, the long-range magnetic order phases associated with these materials.⁸

Here, we report results of measurements of the magnetization and magnetic susceptibility of polycrystalline samples of $\text{La}_2\text{Cu}_{1-x}\text{M}_x\text{O}_4$ with $M = \text{Zn}$ and Ni and compare these results to recently reported behavior obtained in muon spin rotation (μSR) studies for Zn-doped La_2CuO_4 .⁹ The La_2CuO_4 was chosen because of the simplicity of the system as compared to 1:2:3, i.e., the latter has nonequivalent Cu sites. The pure La_2CuO_4 system is considered to be¹⁰ an excellent realization of a two-dimensional spin- $\frac{1}{2}$ quantum Heisenberg antiferromagnet with an unusually large in-plane Cu-Cu exchange coupling $J \simeq 1500$ K and with very weak exchange anisotropies. There is a weak antiferromagnetic coupling between CuO_2 planes which is about five orders of magnitude smaller than the in-plane coupling. However, the presence of an antisymmetric spin-spin interaction allowed by the broken inversion symmetry around a planar Cu site (due to the rocking of the CuO_8 octahedra in the material) gives rise to an interesting metamagnetic

behavior.³ In this paper we study the effects of replacing the Cu atoms by Zn or Ni on both the antiferromagnetic order and the metamagnetic behavior of these materials. We find that the effects of these two substitutions are different and this is in part so because of the different local spin environments in the CuO_2 planes created by these two different atoms, while both Zn and Ni have small effect on the charge of the CuO_2 plane. In particular, Zn has a more significant effect on the destruction of the antiferromagnetic order and the metamagnetic behavior as compared to that of Ni.

II. EXPERIMENTAL RESULTS

Samples of average composition $\text{La}_2\text{Cu}_{1-x}\text{M}_x\text{O}_4$ with $M=\text{Zn}$ or Ni and $0 \leq x \leq 0.1$ were prepared using a two-stage dilution method (so as to produce greater homogeneity of the M cation throughout the ceramic), in a manner similar to that described in Ref. 6, except that the calcining temperature was changed to 1100°C . Powder x-ray diffraction studies using $\text{Cu-K}\alpha$ radiation indicated that the samples crystallized in an orthorhombic structure for all x . The magnetization $M(T)$ was measured using a Quantum Design superconducting quantum interference device magnetometer for $1.8 \leq T \leq 400$ K and $0 \leq H \leq 5$ T. The μSR experiments reported in Ref. 9, were done at the stopped muon channel of the Los Alamos Clinton P. Anderson Meson Physics Facility using standard zero-field techniques.

First, we discuss our results on polycrystalline samples of undoped La_2CuO_4 . In Fig. 1, we show M/H as a function of temperature T for various values of the applied magnetic field $H = 0.3, 0.7, 1.2$ and 2.8 T. Notice that M/H is significantly different for different values of H . This is due to the metamagnetic behavior of these materials, which is demonstrated in Fig. 2, where the magnetization M as a function of H is shown for various temperatures below and above the critical temperature for antiferromagnetic ordering $T_N \simeq 250$ K. Notice that for a given value of temperature $T < T_N$, there exists a critical field $H_c(T)$ where there is significant deviation from the straight line.

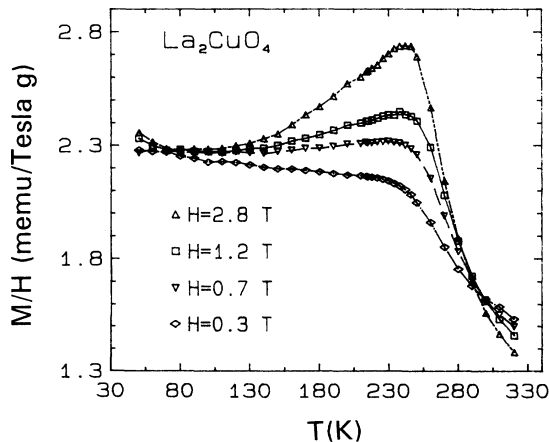


FIG. 1. The M/H vs temperature for polycrystalline samples of La_2CuO_4 at various fields.

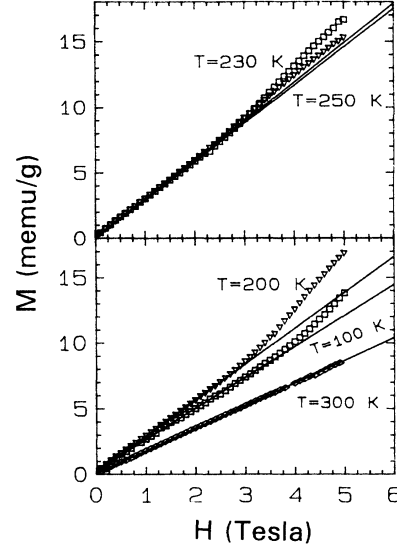


FIG. 2. Magnetization as a function of H for various temperatures for pure La_2CuO_4 .

tion from the straight line. This metamagnetic property of La_2CuO_4 has been observed³ more clearly in single crystals where it appears as a more pronounced jump in M vs H . The critical field $H_c(T)$ has temperature dependence: it is zero above T_N and it is nonzero for any $T < T_N$. Notice that at $T = 300$ K (Fig. 2) the critical field H_c is zero while it attains a value around 3–4 T very quickly below T_N . In Figs. 3(a)–3(c), we give the derivative $\chi(H) = \frac{\partial M(H)}{\partial H}$, i.e., the susceptibility at a finite field H , as a function of H . Notice that, for example, at $T = 100$ K (Fig. 3(a)), $\chi(H)$ as a function of H is a constant below $H_c \simeq 3.5$ T (within some fluctuations in its value which we believe is due to noise), while there is a systematic rise for fields higher than 3.5 T. We can use this method to define the critical field. At $T = 200$ K [Fig. 3(b)] the value of the critical field has decreased to $H_c \simeq 2.5$ T. As we approach the critical point $T_N \simeq 250$ K the signal to noise ratio decreases, while $H_c(T)$ decreases.

In Fig. 4, we give the temperature dependence of the susceptibility measured at constant field H , namely $\chi(H) = \frac{\partial M(H)}{\partial H}$ and it has been computed by subtracting the measured values of the magnetization at the following two values of the field: $H_{\pm} = H \pm 0.1$ T (i.e., 0.1 T below and above the desired value of H). At lower fields the calculation of the derivative involves larger fluctuations and noise. The solid line of Fig. 4 represents the results of our extrapolation to zero-field susceptibility using the formula that

$$\chi(H \rightarrow 0) = \chi(0) + \alpha H^2. \quad (1)$$

Here, a linear term has been excluded because of the symmetry $\chi(-H) = \chi(H)$. Our data for $H = 1.0, 2.0$, and 3.0 T follow this relationship. Notice that the peak in the susceptibility shifts to higher temperatures by decreasing H to zero.

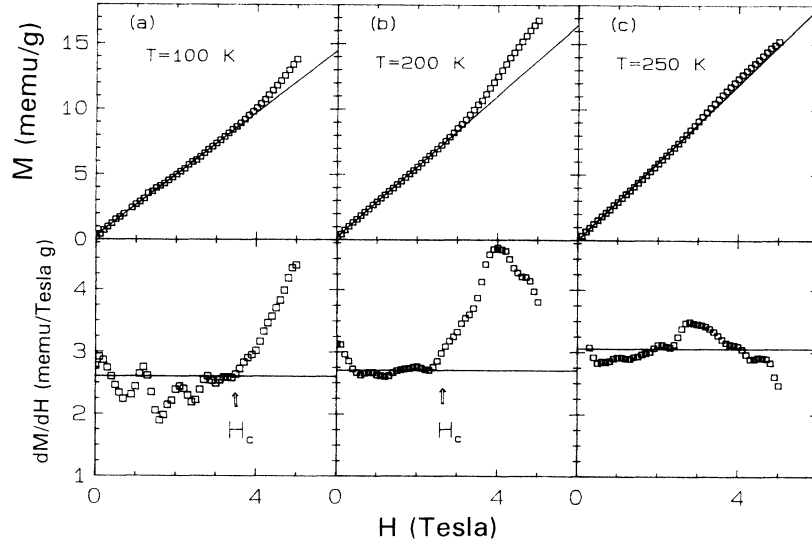


FIG. 3. Magnetization and its derivative with respect to H as functions of H for (a) $T = 100$ K, (b) $T = 200$ K, and (c) $T = 250$ K for pure La_2CuO_4 .

First, we need to determine T_N from the susceptibility. We have used the minimum of the second derivative of $\chi'' = \partial^2 \chi(T)/\partial T^2$ as a function of T to determine the critical point. This assumes that the other contributions to the susceptibility (i.e., from spins which do not participate in the antiferromagnetic ordering), around the critical temperature have a rather smooth (nonsingular) temperature dependence. Thus, by taking the second derivative, these contributions are eliminated and the singularity due to the antiferromagnetic ordering becomes more pronounced giving rise to a minimum in χ'' . In Fig. 5(a), we give $\chi''(T)$ measured for fields $H = 1, 2$, and 3 T. Notice that the presence of the metamagnetic behavior can underestimate the Néel temperature by about 20 K when a field of 3 T is used. We need to extract the Néel temperature for various values of the field and tem-

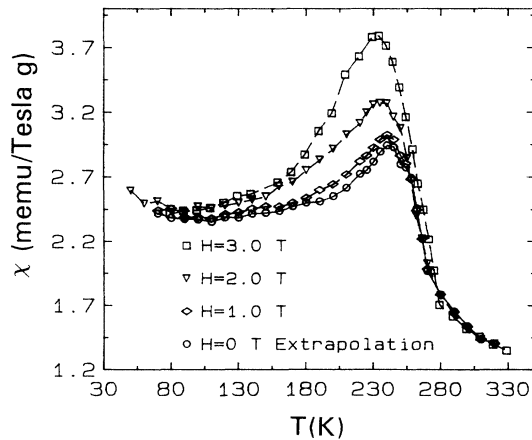


FIG. 4. Susceptibility $\chi(H) = \partial M/\partial H$ as a function of T measured for fields $H = 1.0, 2.0$, and 3.0 T. The solid line is the extrapolation to $H \rightarrow 0$ using the form given by Eq. (1).

perature and for several doped samples. This requires a procedure which involves several measurements of M versus H . We decided to use the following simpler but somewhat less accurate method, which provides a good

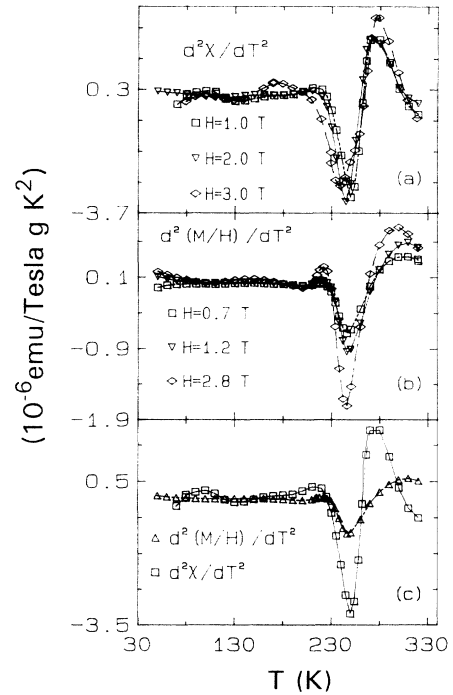


FIG. 5. (a) The second derivative of χ with respect to T as a function of T for $H = 1.0, 2.0$, and 3.0 T. (b) The second derivative of M/H with respect to T as a function of T for $H = 0.7, 1.2$, and 2.8 T. (c) Comparison of the minimum in the second derivatives of χ and of M/H with respect to T for $H \sim 1.0$ T.

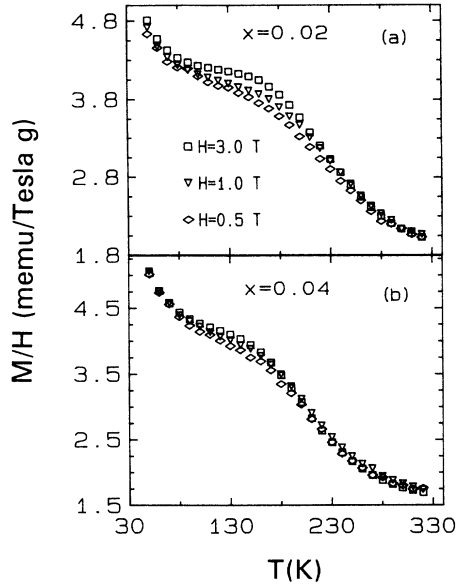


FIG. 6. M/H vs T for $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$ with $x = 0.02$ and 0.04 and for $H = 0.5, 1.0$, and 3.0 T.

estimate of T_N with a small (about 2%) systematic error. In Fig. 5(b), we present the second derivative of M/H with respect to T and in Fig. 5(c) we compare the minima obtained from $\partial^2\chi/\partial T^2$ and from $\partial^2(M/H)/\partial T^2$ at relatively low $H \sim 1$ T. Notice that the minimum occurs at approximately the same temperature: We find that $T_N \simeq 250$ K from χ while M/H gives $T_N \simeq 246$ K. Thus, it appears that estimating T_N from the second temperature derivative of M/H only underestimates T_N by $\sim 2\%$. We shall use this simpler method to determine the Néel temperature for the doped samples.

In Figs. 6(a) and 6(b) we present the measurements of M/H for $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$ and in Figs. 7(a) and 7(b)

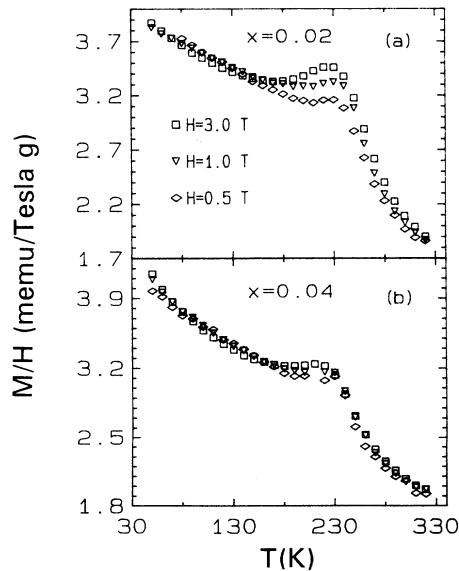


FIG. 7. M/H vs T for $\text{La}_2\text{Cu}_{1-x}\text{Ni}_x\text{O}_4$ with $x = 0.02$ and 0.04 and for $H = 0.5, 1.0$, and 3.0 T.

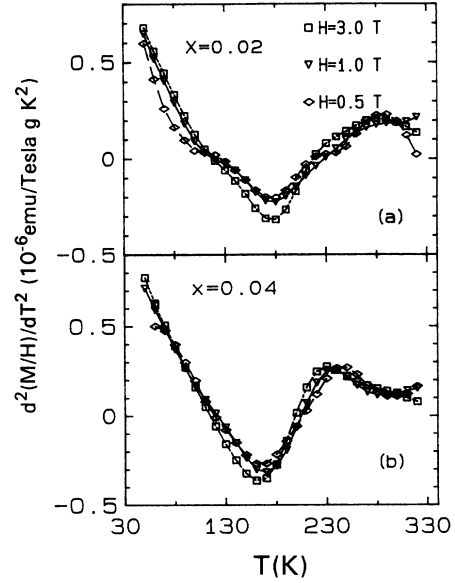


FIG. 8. (a) The second derivative of M/H with respect to T for $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$ for $x = 0.02$ and $H = 0.5, 1.0$, and 3.0 T. (b) the same as (a) for $x = 0.04$.

for $\text{La}_2\text{Cu}_{1-x}\text{Ni}_x\text{O}_4$ for $x = 0.2$ and $x = 0.4$ and for fields $H = 0.5, 1.0$, and 3.0 T. The main observation is that the peak associated with the antiferromagnetic order becomes less well defined upon doping. In addition, doping suppresses the value of the Néel temperature. Another interesting effect is that Zn suppresses the antiferromagnetic peak more than the same amount of Ni. In Figs. 8(a), 8(b), 9(a), and 9(b) we give the second temperature derivative of M/H versus T for these two compounds for the above two doping concentrations to determine T_N . Clearly the Néel temperature is sup-

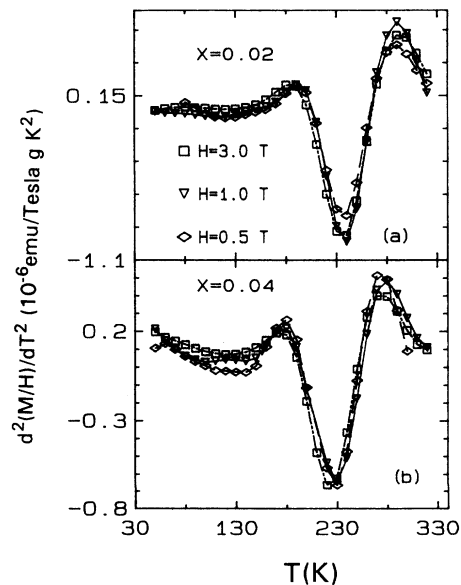


FIG. 9. (a) The second derivative of M/H with respect to T for $\text{La}_2\text{Cu}_{1-x}\text{Ni}_x\text{O}_4$ for $x = 0.02$ and $H = 0.5, 1.0$, and 3.0 T. (b) the same as (a) for $x = 0.04$.

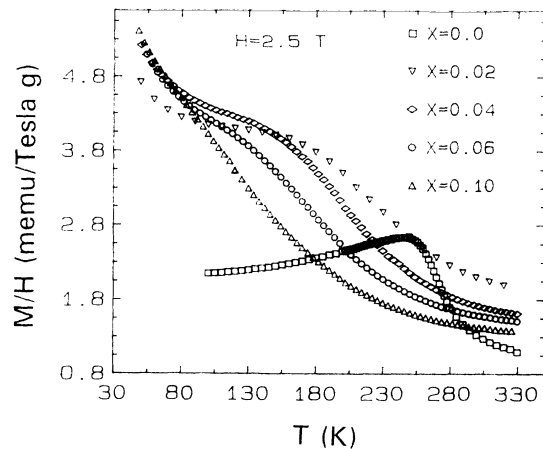


FIG. 10. Comparison of the various curves of M/H for different doping fractions x for $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$.

pressed by doping. In Figs. 10 and 11 we compare the results for M/H for $\text{La}_2\text{Cu}_{1-x}\text{M}_x\text{O}_4$ with $M = \text{Zn}, \text{Ni}$, and $x = 0.0, 0.02, 0.04, 0.06, 0.10$. In Fig. 12 we present the overall x dependence of T_N determined from M/H as discussed above. The open squares in Fig. 12 present results for T_N for the Zn-doped materials as found by the muon spin-rotation experiments (μSR).⁹ Notice that within error bars the results for the Zn-doped materials from μSR agree with those obtained from M/H . The results obtained from M/H are systematically somewhat lower than those determined by μSR ; we believe that this is due to the small systematic error introduced by the fact that we use M/H to determine T_N and not the zero-field susceptibility χ . Again, notice that the effect of Ni on T_N is much less significant as compared to that of Zn.

Another interesting question is the effect of doping on the metamagnetism of these compounds. In Fig. 13(a), we present M as a function of H for $x = 0.04$ Zn-doped compound for various temperatures below $T_N \sim 200$ K. The straight lines are fits at low fields and they are drawn

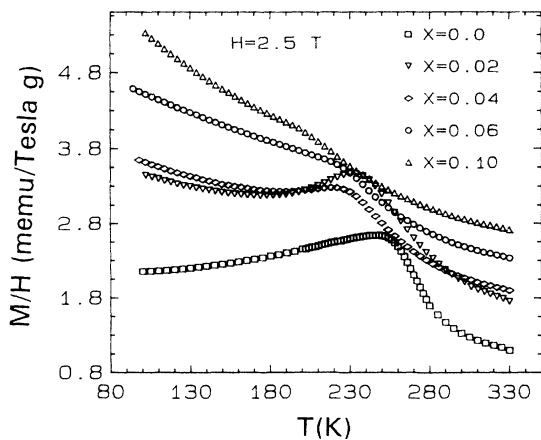


FIG. 11. Comparison of the various curves of M/H for different doping fractions x for $\text{La}_2\text{Cu}_{1-x}\text{Ni}_x\text{O}_4$.

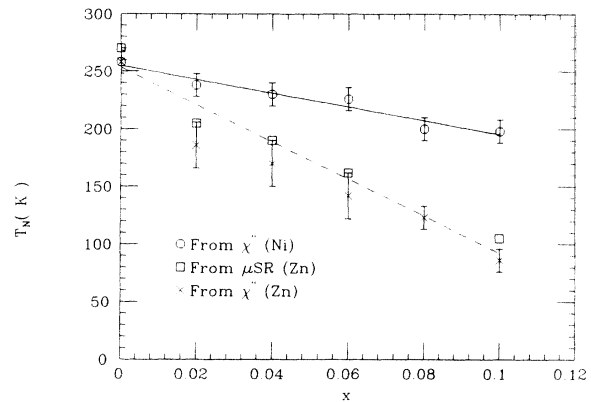


FIG. 12. The dependence of the Néel temperature T_N on x for Ni-doped (open circles) and Zn-doped (crosses) La_2CuO_4 as extracted from the second derivative of M/H . The open squares are the μSR results for Zn-doped samples (Ref. 9).

as guides to the eye. Notice that M vs H at $T = 110$ K has a clear metamagnetic transition with a critical field $H_c \sim 2 - 3$ T. The critical field increases by lowering the temperature while at higher temperatures the critical point cannot be determined in these samples. In Fig. 13(b) we give the derivative $\partial M/\partial H$ as a function of H in order to determine the critical field $H_c(T)$ as discussed above. We notice the presence of a well-defined $H_c \sim 2 - 3$ T at $T = 110$ K and for $T = 10$ K $H_c \sim 3$ T; however, the signal is much weaker as we approach T_N . In the Ni-doped material [Figs. 14(a) and 14(b)] the metamagnetic behavior is weaker and the departure for the straight line much less clear.

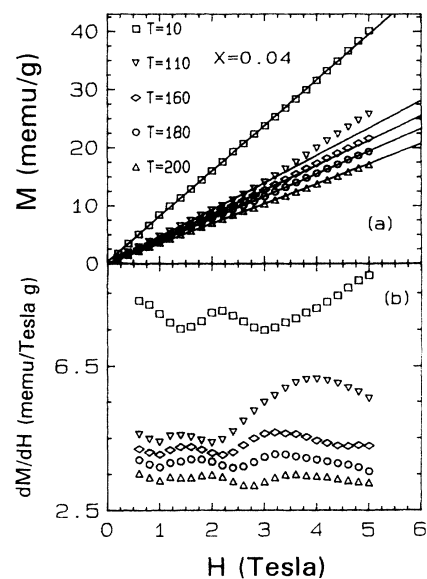


FIG. 13. Magnetization and its derivative with respect to H as functions of H for various temperatures for $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$.

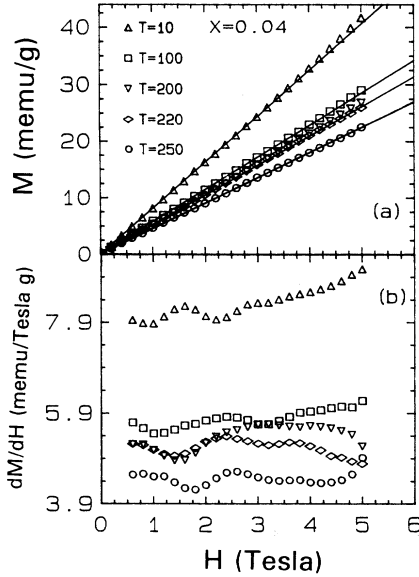


FIG. 14. Magnetization and its derivative with respect to H as functions of H for various temperatures for $\text{La}_2\text{Cu}_{1-x}\text{Ni}_x\text{O}_4$.

III. DISCUSSION

Several of the static and dynamical properties of the undoped and stoichiometric material La_2CuO_4 have been recently revealed. The picture emerging from these studies^{3,10-12} is such that one needs a Hamiltonian which includes a nearly isotropic 2D spin- $\frac{1}{2}$ quantum Heisenberg antiferromagnet (AF) with interaction between Cu spins emerging from the superexchange, mediated by the intervening oxygen ions via virtual hopping processes involving doubly occupied Cu sites. The strength of the AF exchange interaction coupling J between spins on the same CuO_2 plane is large, $J \simeq 1500$ K, while the plane-plane coupling is about five orders of magnitude smaller. Because of the magnitude of the 2D spin correlation length (hundreds of Å) at room temperature (which is low temperature compared to J), a very small inter-layer coupling is large enough to establish AF long-range order at around room temperature and below. Looking at the gaps in the spin-excitation spectrum with neutron scattering, one is led to believe that the magnitude of the anisotropies between the coupling of the components of the spins is very small also. The most important perturbation which needs to be included is a Dzyaloshinskii-Moriya¹³ (DM) antisymmetric term to explain the hidden ferromagnetic behavior and the metamagnetic behavior of the susceptibility. The Hamiltonian of the undoped system is given by

$$H = \sum_{\langle ij \rangle} J_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j) + \sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j), \quad (2)$$

where the summation is over nearest-neighbor Cu spins, $J_{ij} = J$ for spins in a given CuO_2 plane, and $J_{ij} = J'$ for spins in different planes. The last term is

the DM antisymmetric term which is allowed due to the fact that certain crystal symmetries are broken because of the slight rotation of the CuO_6 octahedra around the Cu site. This can produce a canting of the spins away from the plane of staggered magnetization by a small angle $\theta \sim |\mathbf{D}_{ij}|/J$; thus a ferromagnetic moment pointing in the direction perpendicular to the copper-oxygen planes appears, and this can explain the behavior of the uniform susceptibility and the magnetoresistance data of Thio *et al.*³ The coupling $J_{\text{DM}} = |\mathbf{D}_{ij}|$, estimated from the angle of the canted spins, was found to be small compared to the antiferromagnetic coupling J ($J_{\text{DM}}/J \sim 10^{-3}$). However, it does play an important role in determining the ferromagnetic like behavior of the susceptibility close to the 3D Néel temperature. Because the planes are antiferromagnetically coupled the ferromagnetic moment of the planes due to spin canting is canceled by the nearest-neighbor plane. However, the plane-plane coupling is very weak and at a finite value of an external field the ferromagnetic moments of the planes line up, which is the cause of the jump in the magnetization versus temperature behavior of our data.

In mean field theory (MFT), the 3D Néel temperature is determined from the equation $J' \chi_{2D}(T_N) = 1$ where χ_{2D} is the staggered susceptibility of the CuO_2 planes. At low temperatures χ_{2D} can be approximated by $\chi_{2D} \sim \xi_{2D}^2/k_B T$, where ξ_{2D} is the 2D spin-spin correlation length. Accurate studies¹⁰⁻¹² of the pure 2D Heisenberg antiferromagnet suggest that $\xi_{2D}(T) \simeq C_\xi e^{2\pi\rho_s/T}$ where $C_\xi \simeq 1.0$ Å and $\rho_s \simeq 0.20J \simeq 300$ K. There are also theoretical studies of the 2D Heisenberg antiferromagnet doped with static randomly distributed holes.¹⁴ In Ref. 14, it was found that the correlation length increases less rapidly with decreasing temperature with essentially the same functional form but with a prefactor and spin-stiffness constant which depend on the doping fraction x . In the case where the La site is doped with Sr it has been experimentally found that the correlation length and the Néel temperature are much more dramatically affected. In both cases, the Néel temperature is reduced because the 2D susceptibility is reduced and the MFT equation $\chi_{2D}(T_N) = 1/J'$ is satisfied at lower values of T_N . In addition, within MFT, the sharpness of the peak in the total 3D susceptibility is entirely controlled by the rapid variation with T of $\chi_{2D}(T)$. The latter variation depends on the rapid or slow variation of ξ_{2D} with T . The peak in our undoped samples is also sharp while in the doped samples it becomes broadened and the Néel temperature decreases with doping as explained above. Thus, substitution of Cu by a different metal introduces impurities and a different spin value on the Cu site. This reduces the correlation length and makes its temperature dependence weaker than that of the pure material and this broadens the peak in the susceptibility very rapidly with x (Figs. 10-11).

Our studies also indicate that purely magnetic or other static impurities have much weaker effect on the depression of the Néel temperature with impurity concentration x than that of mobile holes. In the latter case it is known that at $x = x_c \sim 0.02$ the AF long-range order is destroyed; here, substitution of the Cu by another metal,

with the same oxidation state as Cu, does not create additional holes or electrons on the CuO_2 plane and it simply acts as a foreign spin or as an impurity that produces less damage in the AF order. It does, however, limit the correlation length on the copper-oxygen plane from thousands of Å close to T_N to much smaller values. This effect broadens the (hidden) ferromagneticlike peak in the susceptibility. The μSR and susceptibility measurements,⁹ however, clearly show that AF long-range order exists up to significantly large values of x .

As discussed earlier when the Zn or Ni atoms are introduced in these compounds they both are in the same oxidation state as that of Cu (i.e., $2+$). Thus, doping with such atoms does not alter the charge excitations in the CuO_2 planes unlike the case of Sr doping. The fact that the effect of Ni doping is less harmful to antiferromagnetism than that of Zn can be understood as follows. Hund's rules indicate that the total spin of Cu^{2+} is $\frac{1}{2}$ while that of Ni^{2+} and Zn^{2+} are $S = 1$ and $S = 0$, respectively. Thus, Ni doping introduces magnetic impurities with different spin from that of the Cu^{2+} ion,

while Zn doping introduces nonmagnetic impurities. The spinless Zn^{2+} ion does not interact antiferromagnetically with the spin of nearest-neighbor (NN) Cu^{2+} ions in the doped system. On the other hand the Ni^{2+} ion has spin 1 and thus it is antiferromagnetically coupled with the NN Cu^{2+} ions via the superexchange interaction. The disruption of the antiferromagnetic order by a nonmagnetic ions is stronger than that produced by a magnetic impurity such as Ni that couples to the other spins in a similar way. This explains our findings, namely, that the effect of the Zn doping is more dramatic (as compared to that of Ni) on both the antiferromagnetic order and the metamagnetic behavior. Therefore the present experiments allow a cleaner study of the effect of pure magnetic impurities since the effect of charge motion, which couples to the spin degrees of freedom strongly, is to a good degree of approximation absent in these systems.

We are in the process of creating single crystals where effects such as the metamagnetic behavior of these systems which depend strongly on the orientation of the sample can be studied in a more quantitative basis.

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