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Title: Magnetic Behavior in Li-Doped La_2CuO_4

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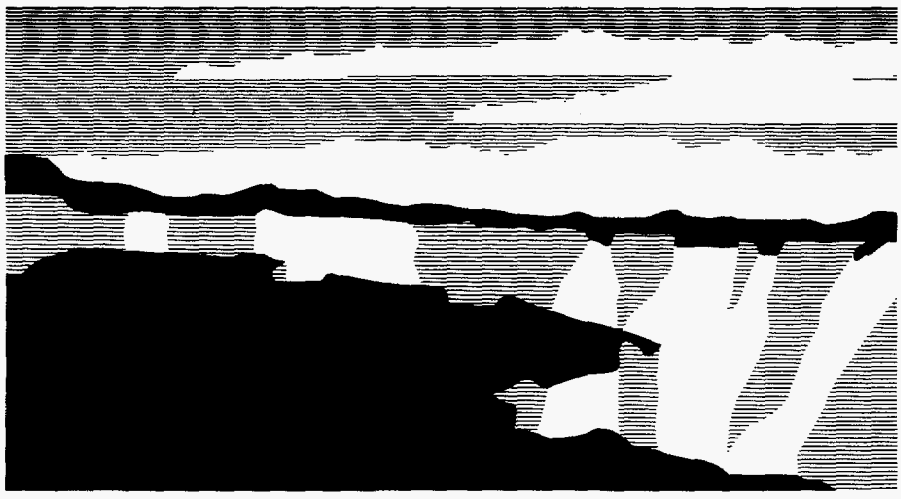

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$\mu\mathcal{SR}$ STUDIES OF Li-DOPED La_2CuO_4

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Magnetic properties of the Li-doped cuprates $La_2Cu_{1-x}Li_xO_4$ (where $x = 0.01$, 0.05, 0.10, 0.45, and 0.50) have been studied by $\mu\mathcal{SR}$. For low Li concentrations ($x \leq 0.10$) we find a rapid suppression of T_N as x increases, but little change in the magnitude and temperature dependence of the AFM order parameter. This indicates that Li doping effectively destroys AFM without strongly affecting the onsite Cu moments and the shape of the spinwave excitation spectrum. For high Li concentrations we find magnetic clusters in about 15% of the sample volume; the remaining volume is non-magnetic, suggesting possible singlet-state formation.

1. Introduction

The interplay between magnetism and superconductivity in the cuprate superconductors continues to be a subject of primary interest. Recently, the effects of substituting Li for Cu have begun to be investigated [1]. Naively, the addition of a Li atom is roughly equivalent to adding both a Sr and a Zn atom, in the sense that Li both removes a Cu moment (as does Zn) and adds a hole (as does Sr). One key difference between Sr- and Li-doping, however, is that the Sr-added holes are mobile, while the Li holes are not [2]. For $x = 0.5$ this compound forms an ordered sublattice of Cu and Li, and the onsite Cu moments disappear [3]. The object of our investigation is to use $\mu\mathcal{SR}$ as a microscopic probe of the Li concentration dependence of both the loss of magnetic order and the onset of the non-magnetic state (perhaps like a Zhang-Rice singlet) [4]. Here we report our $\mu\mathcal{SR}$ measurements on $La_2Cu_{1-x}Li_xO_4$. We find that small amount Li doping will strongly suppress AFM ordering temperature without significantly affecting the onsite Cu moments and the temperature dependence of the sublattice magnetization. Additional Li doping leads to an inhomogeneous magnetic phase with low ordering temperature ($x = 0.05$–0.10), and eventually destroy the Cu moments for x near 0.5.

2. Experiment

The μSR measurements were carried out at TRIUMF, Vancouver, using the M15 and M20 surface muon channels. Polycrystalline samples of $La_2Cu_{1-x}Li_xO_4$ were prepared by standard solid state reaction techniques at Florida State University. The samples were carefully annealed so that they contained no oxygen deficiency, pressed to pellets and then mounted in a He gas-flow cryostat.

3. Results and Discussions

For the lightly-doped systems a spontaneous muon spin precession is observed in zero applied field, indicating the onset of magnetic ordering. (The quantity of precession frequency ν is proportional to the sublattice magnetization M.) Figure 1(a) shows the temperature dependence of ν for $x = 0$, 0.01, 0.05 and 0.10. The rapid suppression of T_N is evident. However, one sees that despite the reduction of T_N by a factor of 60 (from 300 K to 5 K for $x = 0$ and $x = 0.10$, respectively), the zero-temperature frequency declines only by 1/3 (from 5.8 MHz to about 4 MHz). This indicates that the Li doping efficiently breaks down the strength of the magnetic correlations, but only weakly reduces the onsite Cu moments. As seen in other μSR experiments, this also occurs with different dopants, such as with excess oxygen [5], doping Sr on the La site [6] and Zn on the Cu site [8].

In Fig. 1(b) we show the normalized linewidth $\delta\nu/\nu$ vs. temperature for various Li concentrations. We see that the spread in local fields becomes larger as the Li concentration is increased, indicating increasing microscopic inhomogeneity. The observed linewidth and change in zero-temperature frequency could be due either to a dilution of the Cu moments or to a spread in the magnitude of the Cu moments. The differentiate between these two scenarios can be identified, however. For a dilute spin system with small amount of non-magnetic sites a small peak appears in the field distribution, which corresponds to a missing spin at the origin. The dominant peak is unshifted with respect to the undoped system. For the 10%-diluted system the large peak has roughly a Lorentzian lineshape with a normalized linewidth of 0.06. This number is significantly small than 0.15 and 0.3 for the 5% and 10% Li-doped systems, respectively. Thus, Li doping must decrease the average onsite moments and broaden the width, though these effects are much weaker than the reduction of $T_N(x)$ with x.

Figure 2 shows a comparison of the magnetic phase diagrams in $La_2Cu_{1-x}Li_xO_4$, $La_2Cu_{1-x}Zn_xO_4$ and $La_{2-x}Sr_xCuO_4$. We see that Li doping

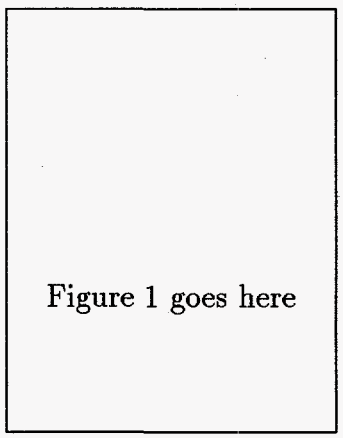

Fig. 1. Temperature dependence of ZF-μSR (a) precession frequency and (b) normalized linewidth observed in $La_2Cu_{1-x}Li_xO_4$, with $x = 0$ (taken from [5]), 0.01, 0.05 and 0.10. Note the different temperature scales for $x \leq 0.01$ and $x \geq 0.05$.

strongly depresses the antiferromagnetic order, similar to Sr doping. If we fit the small-x Li data phenomenologically to $T_N(x)/T_N(0) = 1 - (x/x_c)^2$, we obtain a critical value $x_c = 0.03$, slightly larger than 0.02 for Sr doping, but much smaller than 0.12 for Zn doping [10]. The difference between Sr, Li and Zn doping is that Zn doping only removes spins from the Cu sites, so that the remaining spins are still well connected if the doping concentration is not large. On the other hand, both Sr and Li create holes in the CuO_2 planes which frustrate the AFM coupling between the neighboring spins. Thus the presence of holes in the CuO_2 planes is much more effective in destroying the AFM order than simple dilution of the Cu moments.

There are differences between Sr and Li doping, however. First, Li doping removes a Cu spin while Sr doping does not. As shown above, this has only a weak effect on $T_N(x)$. Second, Li doping creates a more localized hole than Sr doping. This conclusion is mainly derived from the behavior in the heavily doped compounds; namely, Sr doping leads to superconductivity, while Li doping leads to insulating diamagnetism at $x = 0.50$. However, in the low Li-doping range the high-temperature resistivity actually becomes smaller and reaches a minimum near $x \approx 0.10$ [2]. Similarly, the residual susceptibility χ_0 initially increases with increasing x and reaches a maximum also near $x \approx 0.10$ [7]. These results indicate that as long as x is small, Li doping introduces somewhat delocalized holes in the CuO_2 plane, similar to Sr doping. This is consistent with our observation that Sr doping is only marginally more effective than Li doping in destroying the AFM coupling.

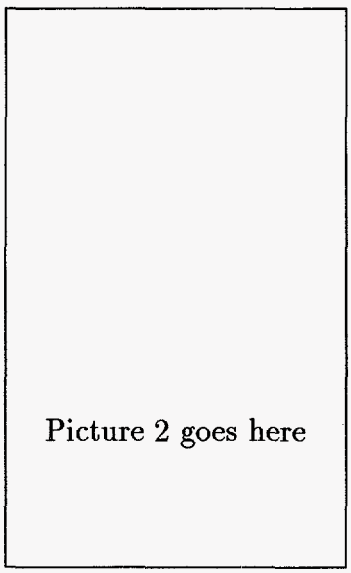

Fig. 2. Magnetic ordering temperature as a function of doping concentration in $La_2Cu_{1-x}Li_xO_4$ (open circles from [7]), $La_2Cu_{1-x}Zn_xO_4$ [8], and $La_{2-x}Sr_xCuO_4$ [9].

As shown in Fig. 2, T_N changes behavior for $x \geq 0.03$ in both the Sr- and Li-doped systems, namely, T_N is small but persists to a rather high concentration, of the order of 10%. To investigate the nature of this magnetic state, we conducted the longitudinal field measurements. We find that the oscillation is decoupled completely in 1 kG (or about 4 times of the internal field), indicating a static magnetic correlations. We note that the magnetic correlations for $x \geq 0.03$ Li doping have not been seen in neutron scattering measurements [11]. The absence of magnetic Bragg peaks means the formation of either a short-range magnetic correlation or possibly an incommensurate magnetic structure. The relatively narrow linewidth (about 0.15 for the 5%-Li system) rules out the spin-glass state.

One of the interesting observations in our experiments is the universal scaling of the temperature dependence of frequency for $0 \leq x \leq 0.10$, as shown in Fig. 3. This scaling behavior is remarkable because it apparently holds over a wide range of T_N, including a possible change in the magnetic structure, as discussed above. A possible explanation for this scaling behavior is that both the intraplane and interplane exchange coupling between the Cu moments ($J_{||}$ and $J_{\perp}$, respectively) decrease as a result of Li doping. To see this, we note that $J_{\perp}$ is primarily due to the orthorhombic distortion of the CuO_2 lattice, which lifts the frustration of the body-centered Cu spin. In

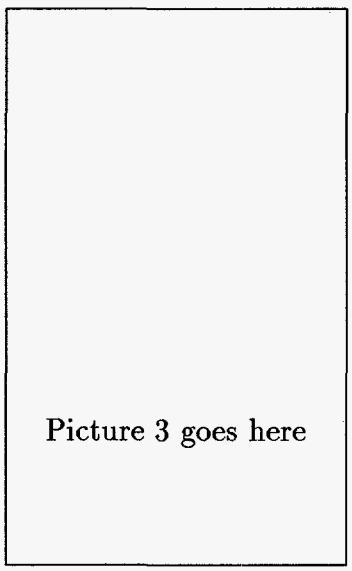

Fig. 3. Reduced frequency vs. reduced temperature in $La_2Cu_{1-x}Li_xO_4$. A universal temperature dependence is seen for $x = 0$, 0.01, 0.05 and 0.10.

the Li-doped system the orthorhombic strain does decreases with increasing x and disappears above $x = 0.10$ [1,7]. Consequently, the shape of magnon spectrum remains largely unchanged, with its characteristic energy scaled by T_N. This leads to the universal scaling of $M(T)$.

Finally, we discuss our results obtained in the high Li concentration regime ($x = 0.45$ and 0.50). We find that the ZF-$\mu\mathcal{SR}$ spectra at high temperatures are well described by a Kubo-Toyabe relaxation function due to nuclear dipoles. Below about 200 K, however, a fast relaxing component is observed which grows both in amplitude and relaxation rate as the temperature is reduced. At the lowest measured temperature, this component corresponds to about 15% of the sample volume and a static magnetic field of 260 G. Both the magnitude of internal field and the onset temperature are comparable to those obtained in the lightly-doped systems. Thus, the formation of the fast-relaxing signal is most likely due to magnetic clusters with partially frozen Cu moments. Such uncompensated moments could also give rise to the Curie tails seen in the susceptibility measurements [1,7]. The remaining slow component (about 85% of the sample volume) are essentially the same as those found at high temperatures. We estimate the upper limit of the magnetic field contributed by the electron moments is about 1 G, which corresponds to a frozen moment of $10^{-3}\mu_B$ per Cu ion.

This is consistent with a spin-singlet state in the 50% doped system.

4. Conclusions

We have performed μSR measurements on the Li-doped La_2CuO_4. We obtained the magnetic phase diagram for doping concentrations varying from $x = 0$ to 0.5. In the low doping range ($x \leq 0.03$) a rapid suppression of T_N is seen, similar to Sr doping, but different from Zn doping, indicating that the presence of holes in the CuO_2 planes is much more effective in destroying antiferromagnetic correlations than simple dilution of the Cu moments. In the intermediate doping range ($x = 0.05$–0.10), we find that the magnetic correlations persist, but with increasing inhomogeneity. Up to $x = 0.10$ we found little change in the onsite Cu moments and the temperature dependence of the magnetic order parameter, despite the fact that T_N declines by a factor of 60. As the doping concentration increases further, we find no evidence of magnetic relaxation in the majority of the sample volume, consistent with the formation of the singlet ground state for $x = 0.45$–0.50.

Acknowledgment

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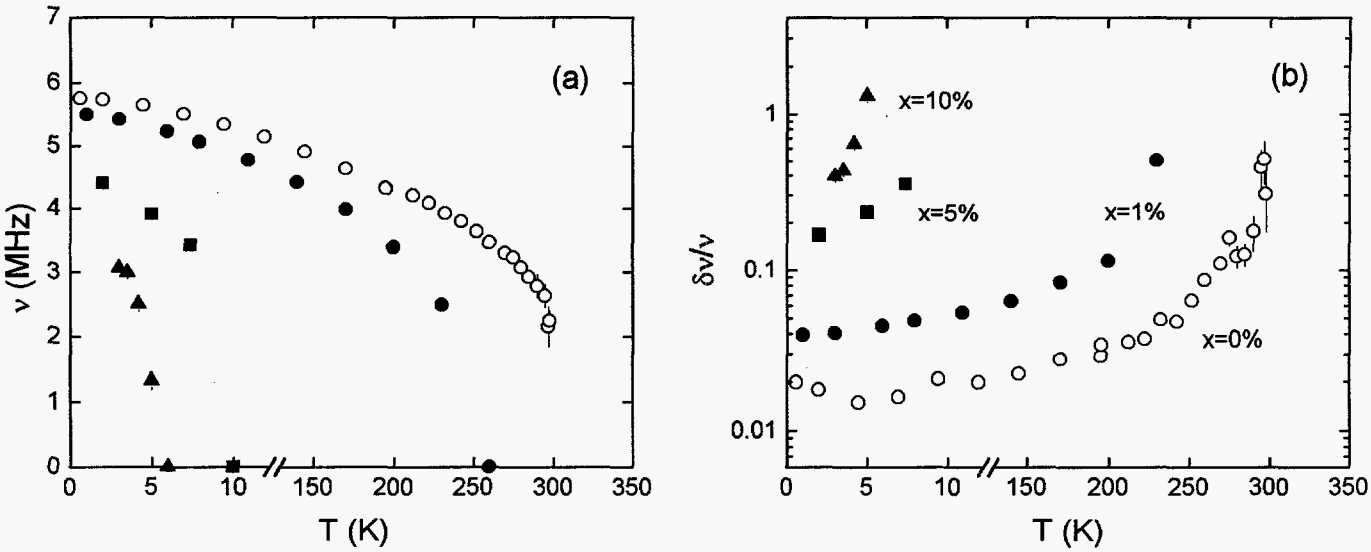
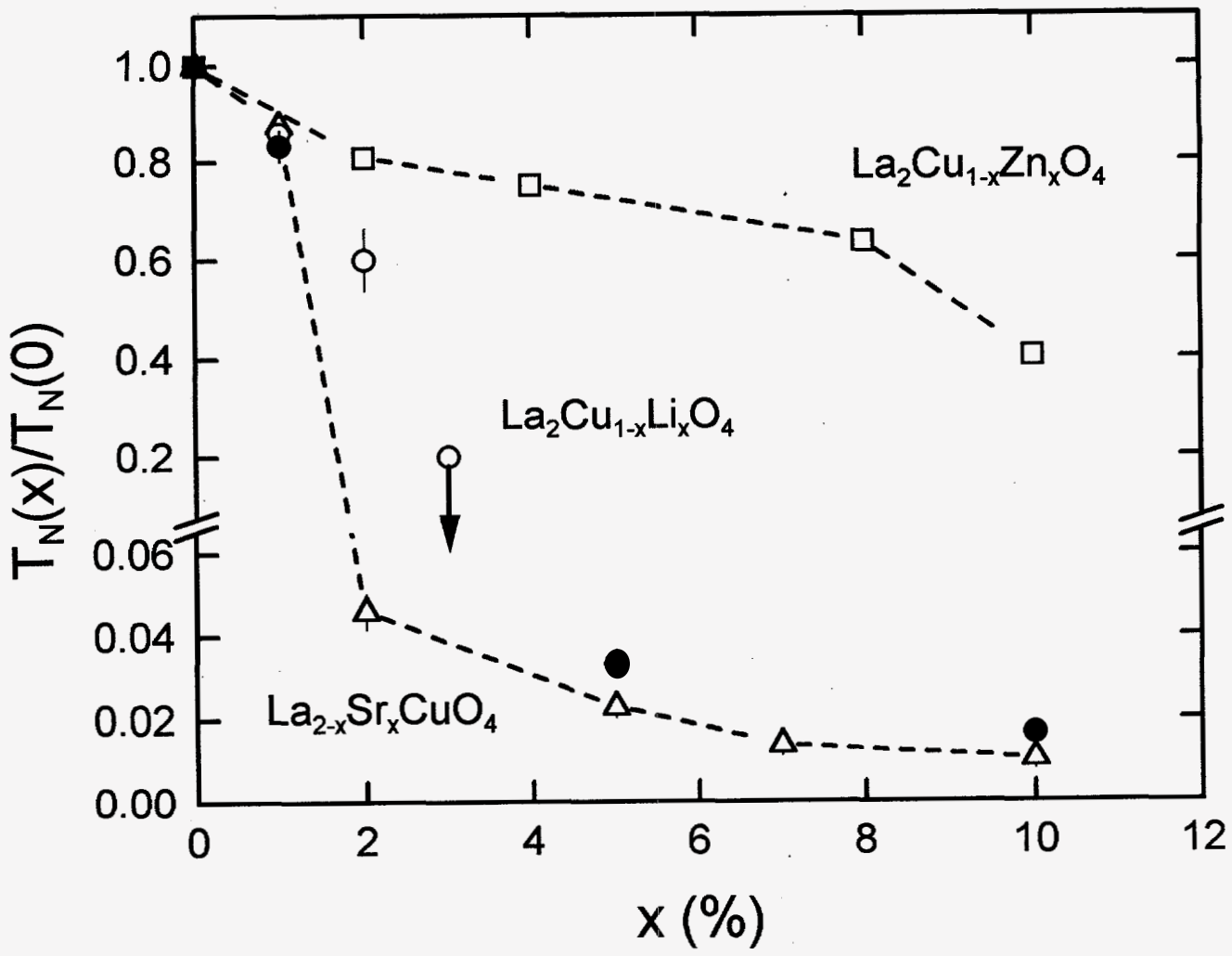
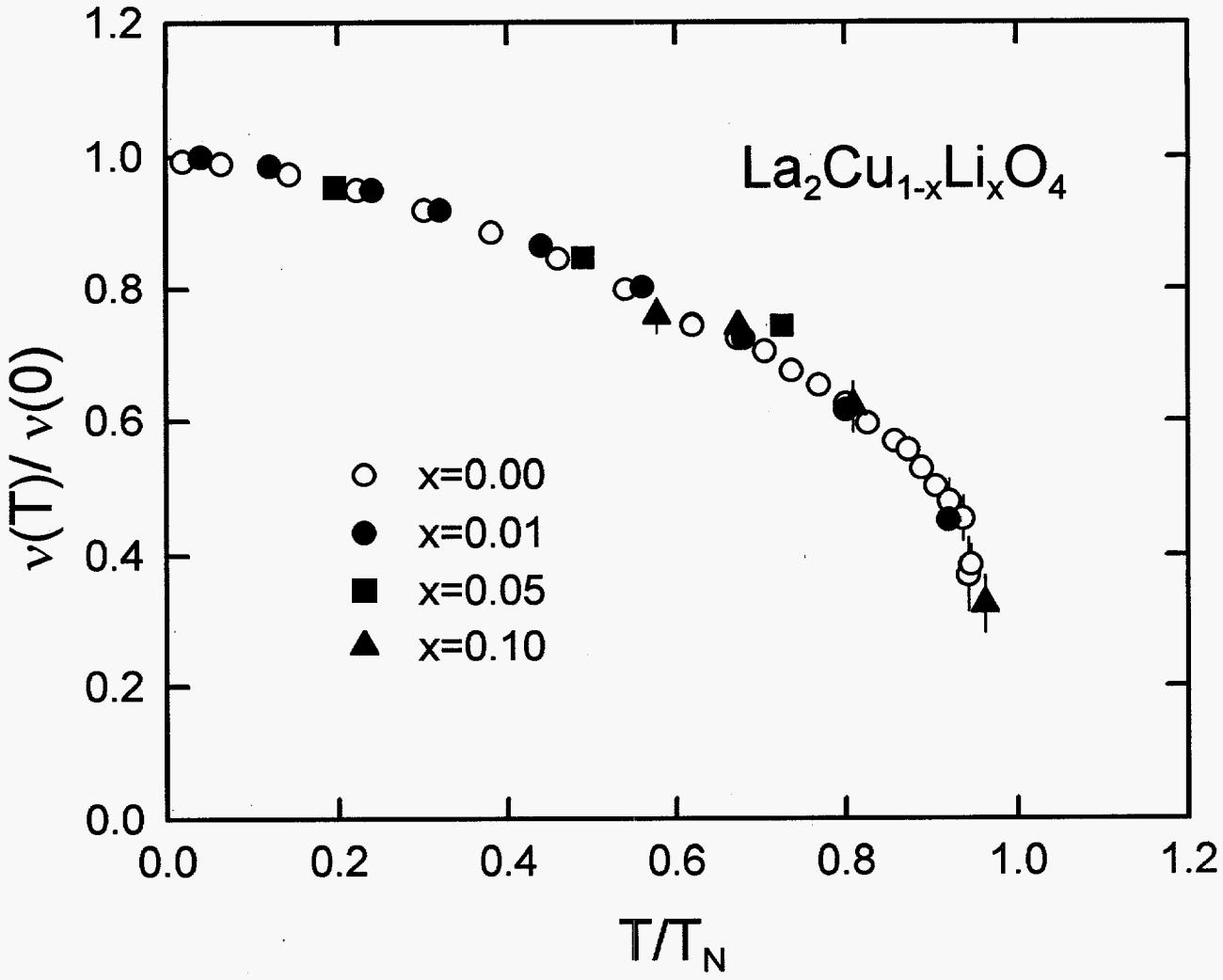

(a)
ν (MHz)
T (K)
(b)
δν/ν
x=10%
x=5%
x=1%
x=0%
T (K)


1.0
0.8
0.6
0.4
0.2
0.06
0.04
0.02
0.00
$T_N(x)/T_N(0)$
$La_2Cu_{1-x}Zn_xO_4$
$La_2Cu_{1-x}Li_xO_4$
$La_{2-x}Sr_xCuO_4$
0
2
4
6
8
10
12
x (%)


La2Cu1-xLixO4
x=0.00
x=0.01
x=0.05
x=0.10
ν(T)/ ν(0)
T/TN
0.0
0.2
0.4
0.6
0.8
1.0
1.2