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SPIN FREEZING AND RECOVERY OF SUBLATTICE MAGNETIZATION IN LIGHTLY DOPED LANTHANUM CUPRATE

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^{139}La NQR studies in lightly doped $La_2Cu_{1-x}Li_xO_4$ and $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ are reviewed. A strong enhancement of the ^{139}La relaxation rate with a peak accompanied by a sudden increase of the local field at low T has been observed similarly to $La_{2-x}Sr_xCuO_4$. The anomalous magnetic properties are discussed in the light of the microscopic segregation of doped holes into hole-rich domain walls separating undoped AF domains.

1 Introduction

Understanding of the rich phenomenology associated with holes doped into the antiferromagnetic (AF) insulator cuprate continues to be a crucial problem in high temperature superconductors (HTSC). In $La_{2-x}Sr_xCuO_4$ (LSCO) compounds, the AF ordering temperature T_N is suppressed extremely rapidly from ≈ 300 K for $x = 0$ to ~ 0 K by $x \approx 0.02$. Surprisingly, the extrapolated zero-temperature sublattice magnetization M_s^0 is simultaneously depressed with increasing x (decreasing T_N).[1,2] Most interesting phenomenon in lightly doped LSCO is the abrupt recovery of the sublattice magnetization $M_s(T)$, almost to $x = 0$ values, at ≈ 30 K followed by the continuous freezing of the spin degrees of freedom observed from ^{139}La nuclear quadrupole resonance (NQR) studies.[1,2] From the recent ^{139}La NQR measurements in lightly doped $La_2Cu_{1-x}Li_xO_4$ (LCLO)[3] and $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ (LESCO),[4] we strikingly observed the spin freezing and the recovery of $M_s(T)$ nearly identical to the behavior found in LSCO.[1,2] We will start with the brief summary of the ^{139}La NQR results in LCLO and LESCO. The anomalous magnetic properties at low T will then be discussed in association with the coupling across the charged domain walls formed by the microscopic segregation of doped holes into the hole-rich domain walls separating undoped AF domains.

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2 ^{139}La NQR in Lightly Hole-Doped $La_2Cu_{1-x}Li_xO_4$

The effects of in-plane hole-doping due to Li^{1+} substitution for Cu^{2+} on the macroscopic structural and magnetic properties of La_2CuO_4 are very similar to those due to Sr^{2+} substitution for La^{3+}.[5] T_N is suppressed to zero by $x \approx 0.03$ in $La_2Cu_{1-x}Li_xO_4$ (LCLO). However, the charge transport properties of LCLO are different from those of LSCO, as well documented in Ref. [5]. The principle difference of Sr and Li substitution is the absence of the metallic as well as the superconducting phase in the Li case up to the maximum doping level $x = 0.5$.

Recent ^{139}La NQR measurements in lightly hole-doped AF LCLO revealed that the microscopic magnetic properties of LCLO are remarkably similar to those of LSCO even though the origin (dopant) of doped holes is different (out-of-plane vs in-plane).[3] The correspondence between the suppression of M_s^0 and T_N by doping is nearly identical to the Sr case.[6] At low T, a strong enhancement of the ^{139}La spin-lattice relaxation rate $1/T_1$ with a peak (at a temperature $T_f = 11 - 16$ K depending on x) and the abrupt recovery of $M_s(T)$ are observed, which is also identical to the Sr case [Figs. 1(a)and 1(b)]. In addition, the ^{139}La spin-spin relaxation rate $1/T_2$ enhances similarly to $1/T_1$ and saturates below T_f as shown in Fig. 1(b) and the inset. The behavior of $1/T_1$ and $1/T_2$, which is typical for the motional slowing-down, clearly indicates that the sharp peak of $1/T_1$ is associated with the continuous freezing of spin degrees of freedom rather than a cooperative phase transition. Analyzing the data in terms of activated behavior, $T_1^{-1}(T) \propto \exp(E_a/k_BT)$, gives values of $E_a/k_B \cong 120$ K similar to those in LSCO with similar hole concentration and/or T_N.[3] Finally, the temperature dependence of dynamical susceptibility [obtained from the analysis of $T_1^{-1}(x,T)$] exhibits the same finite-size effects[3] as were observed in the static susceptibility $\chi(x,T)$.[7]

3 ^{139}La NQR in Lightly Hole-Doped $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$

The structural phase transition (SPT) from the low temperature orthorhombic (LTO) to the low temperature tetragonal (LTT) structure in $La_{2-x}Ba_xCuO_4$ and in rare-earth-doped $La_{2-x-y}M_ySr_xCuO_4$ (M = Nd, Eu) has attracted much attention due to its association with the anomalous suppression of the superconducting transition at a certain range of hole concentration[8,9] and the occurrance of static charge order into stripes.[10]

We have performed ^{139}La NQR and relaxation measurements in lightly hole-doped $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ (LESCO) which undergo the SPT at $T_{LT} = 134 \pm 2$.[4] Note that the values of T_N are the same as those of LSCO at the same hole (Sr) concentration; the rare-earth co-doping does not alter the doping dependence of T_N. At T_{LT}, a sharp asymmetric peak in the ^{139}La NQR relaxation

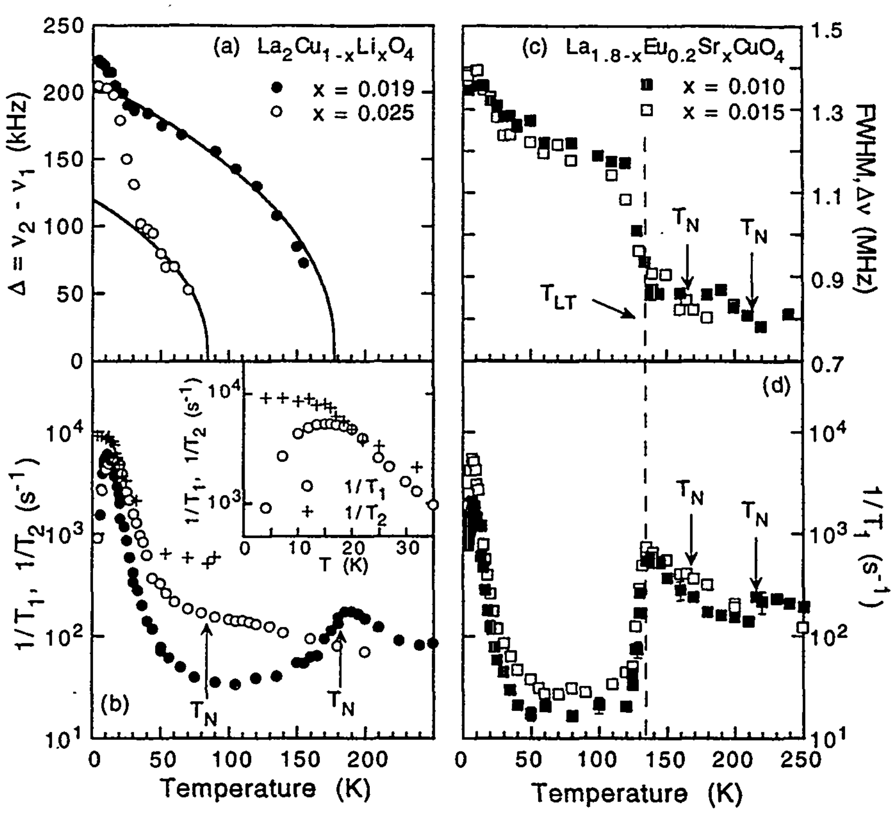

Figure 1: Summary of ^{139}La NQR in lightly hole-doped $La_2Cu_{1-x}Li_xO_4$ [(a), (b)] and $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ [(c), (d)]: $\Delta = \nu_2 - \nu_1$ in (a) is the splitting of the NQR line below T_N which is proportional to the component of the internal magnetic field **H** at the La site along the axis of the electric field gradient (EFG). Solid curves are fits to a power law $\Delta(T) = \Delta_0(1 - T/T_N)^\beta$, with $\beta = 0.44 \pm 0.01$ and T_N obtained from dc magnetization measurements. In (c), the linewidth $\Delta\nu$ is plotted instead of the splitting Δ since the splitting below T_N is not visible due to the broad line of quadrupolar origin arising from local inhomogeneity of the EFG due to the high density of Eu ions in LESCO. However, the additional small broadening below T_N and the substantial jump of $\Delta\nu$ at T_{LT} are found to be of magnetic origin from the ratio $\Delta\nu_{\rm ratio} \equiv \Delta\nu(\text{at } 3\nu_Q)/\Delta\nu(\text{at } 2\nu_Q)$. $1/T_1$ in both (b) and (d) was obtained by fitting the recovery data for the first decade to single exponential function. Although below ≈ 30 K, revcovery law deviates to stretched exponential behavior similarly to LSCO case, The same fitting procedure was applied for the entire temperature range investigated; while this increases the uncertainty in $1/T_1$ at low T, we find that varying the fitting procedure has essentially no effect on the position of the peak at T_f and at T_{LT}, and the value of the activation energy E_a. Decay of the spin-echo amplitude $S(t)$ cannot be determined by either single exponential or gaussian function. Thus, an effective $1/T_2$ in (b) was determined by fitting $S(t)$ to the expression: $S(t) = S(0)\exp[-(t/T_2)^\alpha]$.

rate R_1, a clear evidence for phonon softening associated with the SPT, is observed [Fig. 1(d)]. The width of the ^{139}La NQR line, $\Delta\nu$, increases suddenly at T_{LT} [Fig. 1(c)], indicating the distribution of ordered moment orientations in *ab*-plane. This is attributed to faults in the spin-stacking pattern due to the reduction of the AF interlayer coupling in LTT or *Pccn* phase.[4] Regarding the anomalous magnetic properties at low T, a strong enhancement of $1/T_1$ with a peak at $T_f \cong 6$ K [Fig. 1(d)] and an increase in the local magnetic field at the La site below ≈ 30 K [Fig. 1(c)] are observed; all these are reminiscent of very similar features found in LSCO [1,2] and LCLO.[3] This behavior now seems to be universal in lightly hole-doped AF La214. The values obtained in LESCO, in particular for $x = 0.015$: $T_f \cong 6$ and $E_a/k_B \cong 62 \pm 5$ K obtained from fitting to activated behavior,[4] are considerably smaller than those obtained in LSCO: $T_f \cong 12$ K and $E_a/k_B \cong 120$ K at similar hole concentration [1], even though the origin (dopant) of the doped holes is the same. This is in contrast to the similar doping dependence of T_N mentioned earlier. This is also in contrast to the observation that T_f and E_a/k_B found in LSCO and LCLO are essentially identical even though they have different dopants. The significant difference between LSCO and LESCO is the local structure. Clearly, the local structure plays a crucial role in determining the low-T magnetic properties in lightly hole-doped La214.

4 Phase Separation and Magnetism in Lightly Hole-Doped La214

It is well recognized that holes added to AF La214 segregate into hole-rich domain walls separating undoped AF domains. Evidence for such an effect in lightly doped LSCO was obtained from a scaling relation of the magnetic susceptibility: $\chi(x,T) = \chi\{f(x)[T - T_N(x)]\}$, with the scaling function $f(x) = 0.02/x^2$.[7] This, so-called, finite-size effect indicates that the magnetic correlation length is limited to the linear dimension $L \propto 1/x$ of the AF domains. We obtained the same finite-size scaling of dynamic susceptibility from $1/T_1T(x,T)$ in the vicinity of T_N in LCLO with the same scaling function $f(x)$.[3] Therefore, phase separation or microscopic segregation is believed to be responsible for the rich magnetic phase diagram of lightly doped La214 although the mobility of the doped holes is still imporant factor to determine the magnetic properties.[11]

Borsa *et al.* modeled the system assuming that the mobile holes formed 1D stripes in the CuO_2 planes which effectively decoupled adjacent undoped domains from each other.[2] The suppression of M_s^0 can be understood in the context of the restricted set of spin wave modes accessible in the confined AF domains. We previously proposed that holes form antiphase domain walls which

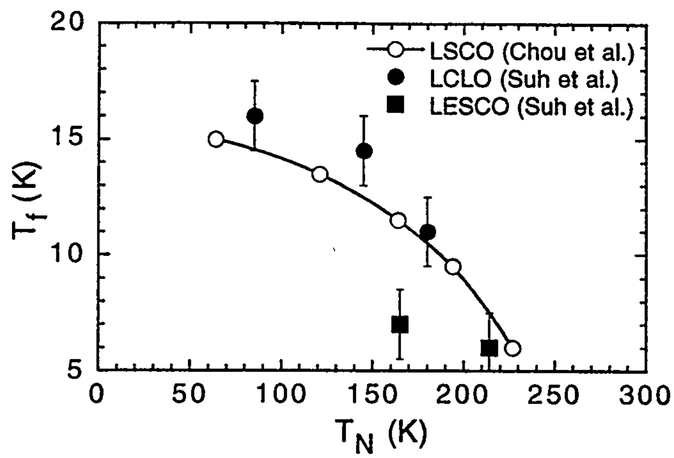

Figure 2: Spin freezing temperature T_f vs T_N in lightly hole-doped La_2CuO_4. The distinct behavior between $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ and $La_{2-x}Sr_xCuO_4$ in spite of the same dopant (Sr) is in contrast to the similar behavior found in $La_{2-x}Sr_xCuO_4$ and $La_2Cu_{1-x}Li_xO_4$ even though they have different origin of doped holes (out-of-plane vs in-plane).

surround mobile domains in which the phase of the AF order is reversed.[3,6] Passage of such anti-phase domains over a given site will reverse the orientation of particular ordered Cu moment. Suppression of M_s^0 as well as T_N can be understood as long as the motion of domains is rapid compared to the NQR measurement time. Both models explain semi-quantitatively the suppression of M_s^0 and the finite-size scaling of $\chi(x,T)$ and $1/T_1T(x,T)$. According to these models, the recovery of the sublattice magnetization $M_s(T)$ and the spin freezing at low T are interpreted as arising from freezing of the domain motion [3,6] and/or disappearance of the domain walls as the constituent holes become pinned to lattice sites.[2]

On the other hand, static charge-stripe order has been observed in the LTT phase of more heavily doped $La_{2-x-y}Nd_ySr_xCuO_4$,[10] indicating that domain walls are more strongly pinned in LTT phase. Thus, the detail nature of our data (Fig. 2), in particular, the smaller values of T_f and E_a/k_B in LESCO,[4] cannot be simply explained by either the freezing of domain motion or the localization of holes at low T.

We propose a likely scenario that the recovery of $M_s(T)$ and the following spin freezing are triggered by the coupling across domain boundries. The finite-size effects suggest that the coupling between the different spin regions separated by domain walls is very weak at high T due to a small coupling constant (J') across domain walls. However, it is universially observed that the domain walls (stripes) serve as anti-phase domain walls between the hole-free domains,[6,10] indicating that the interaction between domains are not totally cut off by the charged domain walls, i.e., J' is not negligiblly small. Coupling strength is proportional to $J'\xi^z$ where ξ is the correlation length within the domains (spin regions) which is a function of the exchange coupling constant J, and the exponent z is close to unity depending upon the dimensionality of domains. Thus, the coupling is not negligible for a small but finite J' when ξ is sufficiently large. As lowering T, the ξ increases due to the large J (≈ 1500 K) if the domain is sufficiently large or long. Then, even a small J' can be effective and trigger the coupling between domains resulting in the suppression of the finite-size effects and hence, the sublattice magnetization recovers.

Acknowledgments

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