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Interplay between the magnetic fluctuations and superconductivity in the lanthanum cuprates

G. B. Teitel'baum[1)], *V. E. Kataev*, *E. L. Vavilova*, *P. L. Kuhns*[+], *A. P. Reyes*[+], *W. G. Moulton*[+]

E. K. Zavoiskii Institute for Technical Physics RAS, 420029 Kazan, Russia

[+]*NHMFL, Tallahassee FL 32310, USA*

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We report the analysis of the magnetic fluctuations in the superconducing $La_{2-x}Sr_xCuO_4$ and the related lanthanum cuprates having the different symmetry of the low temperature structure. The NMR and ESR investigations revealed the dynamical coexistence of the superconductivity and the antiferromagnetic correlations in the large part of superconductivity region of the phase diagram. We show that for all compounds, independent on their low temperature symmetry and on their superconducting properties, the enhancement of the spin stiffness near 1/8 doping takes place.

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The interest to the microscopic phase separation in the high-T_c superconducting materials has received a strong impetus after the discovery of stripe correlations [1]. They were observed only in the compounds specially doped with the rare earth ions whose role is to induce the low temperature tetragonal (LTT) phase favorable for the pinning of the stripe fluctuations. Recent neutron scattering experiments [2] in the low temperature orthorhombic (LTO) phase of $La_{2-x}Sr_xCuO_4$ with $x = 0.12$ reveal the presence of modulated antiferromagnetic order very similar to that found in LTT compound $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. But on the larger time scale the magnetic fluctuations in $La_{2-x}Sr_xCuO_4$ are dynamical especially for the superconducting state and their relevance to the stripe structure is a matter of debate. In particular, the dynamical character of the microscopic phase separation hinders the investigation of its properties by means of low frequency local methods such as conventional NMR [3, 4].

The main aim of the present work is to analyze the phase diagram and the properties of magnetic fluctuations for superconducting $La_{2-x}Sr_xCuO_4$ and related compounds with a help of experiments whose characteristic frequency is shifted to larger values in comparison with the conventional NMR. We consider ESR ($\nu \backsim 10\,\text{GHz}$) and high field NMR ($\nu \backsim 0.1\,\text{GHz}$) measurements which are focused on a comparative analysis of the magnetic fluctuations for the different metalloxides. With this purpose we discuss the ESR data obtained for such compounds as $La_{2-x}Sr_xCuO_4$ (LSCO) [5], $La_{2-x}Ba_xCuO_4$ (LBCO) [6], $La_{2-x-y}Eu_ySr_xCuO_4$ (LESCO) [7] together with the conventional NMR data for $La_{2-x-y}Nd_ySr_xCuO_4$ (LNSCO) [8] and new high field NMR data for superconducting LSCO. All the measurements were carried out on powder samples with various hole doping. For LSCO the doping level covers the entire superconducting region of the phase diagram, for LBCO we studied the doping region in the vicinity of the well known T_c dip, whereas the LESCO and LNSCO series correspond to the nonsuperconducting LTT phase. The samples which were used for the ESR measurements were doped with 1 at. % of Gd, used as ESR probe [5]. Such tiny concentration of Gd ensured only the small suppression of T_c via pair breaking.

We analyzed the temperature and concentration dependence of the width of the most intense component of multiline Gd^{3+} ESR spectrum, corresponding to the fine splitting of the spin states $S = 7/2$ in the crystalline electric field [5]. The typical temperature dependence of the linewidth δH is shown in Fig.1.

The temperature behaviour for $T > T_c$ is qualitatively very similar for all samples under study: a linear dependence of δH on temperature which is followed by the rapid growth of the linewidth at low T. But after cooling below 40 K the behaviour of superconducting and nonsuperconducting samples becomes different: the linewidth of superconducting LSCO exhibits the downturn starting at a temperature T_m dependent on x whereas for other samples which are not bulk superconductors the linewidth continues to grow upon further lowering temperature (see Fig.1).

This behaviour may be explained if to take into account that in addition to the important but temperature independent residual inhomogeneous broadening

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[1)] e-mail: grteit@dionis.kfti.knc.ru

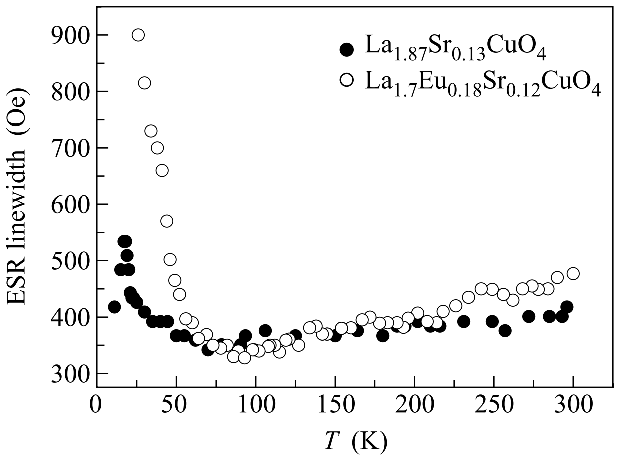

Fig.1. The typical ESR linewidth temperature dependences for LTO superconducting LSCO and LTT nonsuperconducting LESCO

the linewidth is given by different homogeneous contributions linked to the magnetic properties of CuO_2 planes:

i) the interaction of Gd^{3+} spins with the charge carriers, i.e. the Korringa relaxation channel. The simplest Korringa term in the linewidth is $\delta H = a + bT$ with $b = 4\pi(JN_F)^2 P_M$, Ref. [9], where $P_M = [S(S+1) - M(M+1)]$ – is the squared matrix element of the Gd spin transitions between the M and $M+1$ states, N_F is the density of states at the Fermi level, J is the coupling constant between the Gd and charge carriers spins [5]. The factor P_M describes the Barnes-Plefka enhancement [9] of the relaxation with respect to the standard Korringa rate. Such an enhancement occurs in exchange-coupled crystal field split systems where the g-factors of localized and itinerant electrons are approximately equal but the relaxation of conduction electrons towards the "lattice" is strong enough to inhibit bottleneck effects. For the system under study it was discussed in Ref. [5]. Note, that the enhancement of the linear slope for LESCO compound relative to that for LSCO seen in Fig.1 is due to the influence of the depopulation of the first excited magnetic Eu level [7].

ii) the interaction of Gd with copper spins, giving rise to homogeneous broadening of Gd ESR line (a close analogue of nuclear spin-lattice relaxation):

$$\delta H = \frac{1}{2}(\gamma H)^2 P_M \left[(\tau/3) + (2\tau/3)/(1+(\omega\tau)^2)\right] \quad (1)$$

where τ is the magnetic fluctuations life-time, H is the internal magnetic field at Gd site. Following Ref. [8] we assume the activation law for the fluctuation lifetime temperature dependence $\tau = \tau_\infty \exp(E_a/kT)$ with τ_∞ being the lifetime at the infinite temperature and E_a – the activation energy, proportional to the spin stiffness ρ_s ($E_a = 2\pi\rho_s$).

The second contribution describes the standard Bloembergen-Purcell-Pound (BPP) behaviour: the broadening of the ESR line upon cooling with the downturn at certain freezing temperature T_m corresponding to $\omega\tau = 1$. Here ω is the resonant frequency. This expression is written for the case when the fluctuating magnetic fields responsible for Gd spin relaxation are induced by local Cu moments. In the polycrystalline samples the averaging over the random orientation of the local Cu moments with respect to the external magnetic field yields by a factor of 2 larger probability of their perpendicular orientation as compared to the collinear one.

We observed that depending on the Sr content the linewidth behaviour transforms from the BPP-like (with the maximum at T_m) to the pure Korringa (linear) temperature dependence. Basing on the observation that the relative weight of the BPP-contribution, compared with the Korringa one, decreases with increasing Sr doping we conclude that at low x the Gd spin probes almost magnetically correlated state and at the high x end – almost nonmagnetic metal. Such a transformation may be explained it terms of the microscopical phase separation to the metallic and AF correlated phases. It is worth to remind that very soon after the discovery of the high T_c superconductivity in cuprates it was suggested [10], that the microscopical phase coexistence is the inherent feature of these materials. Note that according to the phase diagram shown in Fig.2 the obtained T_m values are lower than the respective T_c, although for certain hole doping they are lying close to each other. The relative amount of the AF phase falls abruptly in the vicinity of $x = 0.20$ so that for $x = 0.24$ any traces of it are absent. One cannot exclude that this boundary is connected with the existence of the widely discussed quantum critical point [11] at this doping values.

The different temperature dependences of the linewidths for the superconducting and nonsuperconducting compounds may be consistently explained assuming that for the superconducting samples the linewidth below T_c is governed by fluctuating fields which are transversal to the constant field responsible for the Zeeman splitting of the Gd spin states (the second term in Eq.(1) for δH). Since these fluctuations are induced by Cu moments lying in the CuO_2 planes, it means that Gd ions are subjected to the constant magnetic field normal to these planes. This may indicate that the magnetic flux lines penetrating in the layered superconduting sample tend to orient

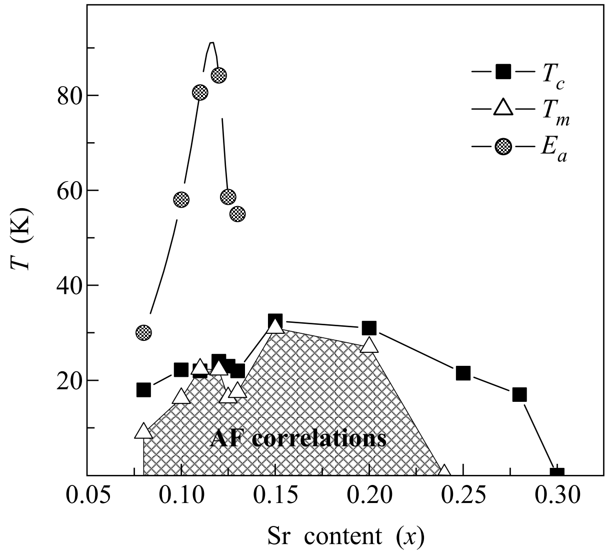

Fig.2. The phase diagram of the magnetic fluctuations of superconducting LSCO. The triangles correspond to the magnetic transition temperature $T_m(x)$, squares – to the superconducting transition temperature $T_c(x)$ and the circles to the magnetic fluctuations activation energy $E_a(x)$. The coexistence region is shown with a grey color

normally to the basal planes where the circulating superconducting currents flow (it is also possible, that Gd ions pin the magnetic fluctuations connected with the normal vortex cores). The important argument in favor of the magnetic fluctuations contribution to the Gd ESR linewidth is given by the fact that the BPP peak at $T = T_m$ ($x = 0.10$) $\approx$ 16 K is in a reasonable agreement with that observed near 4 K in the ^{139}La nuclear spin relaxation rate temperature dependence for LSCO compound with $x = 0.10$ at the frequency of 140 MHz [12].

In principle there might be also a second possibility of the different low temperature behaviour of the linewidth for superconducting samples in comparison with that for nonsuperconducting ones. The nonresonant field dependent microwave absorption in the superconducting state may distort the shape of the ESR spectrum. But these distortions should be especially pronounced for the broad lines, typically for the samples with the small amount of holes, whereas the temperature T_m, characteristic for small x, is considerably lower than T_c. Thus the possible distortion of ESR lineshape owing to the nonresonant microwave absorption as the main reason for the apparent narrowing of the ESR line below T_c seems to be improbable.

Since the measurements were carried out at nonzero external field it is very important to consider the flux lattice effects. At typical ESR fields of approximately 0.3 T, oriented normally to the CuO$_2$ layers, the period of lattice is 860 Å, whereas the vortex cores sizes for LSCO are approximately 20 Å. As the upper critical field amounts to 62 T, it is clear that in the case of ESR the vortex cores occupy only 0.5% of the CuO$_2$ planes. According [13, 14] the Cu spins in the vortex cores may be AF ordered. Therefore the phase diagram in Fig.2 indicates that not only the spins in the normal vortex cores are AF correlated, but the AF correlations are spread over the distances of the order of magnetic correlation length which at low doping reaches 600–700 Å [14].

Numerical simulations of the Gd ESR linewidths for the compounds with the different Sr content enable us to estimate the values of the parameters in the expression for the linewidth. For example the maximal effective internal field H in the rare earth positions is about 200 Oe; the life time τ_∞, which was found to be material dependent, for LSCO is equal to $\tau_\infty = 0.3 \cdot 10^{-12}$ sec, and the activation energies E_a for all investigated compounds are shown in Figs.2, 3. Note that since the influence of the Nd magnetic moments for the LNSCO compound hinders the ESR measurements the activation energy for this compound was estimated from the measurements of the nuclear spin relaxation on Cu and La nuclei.

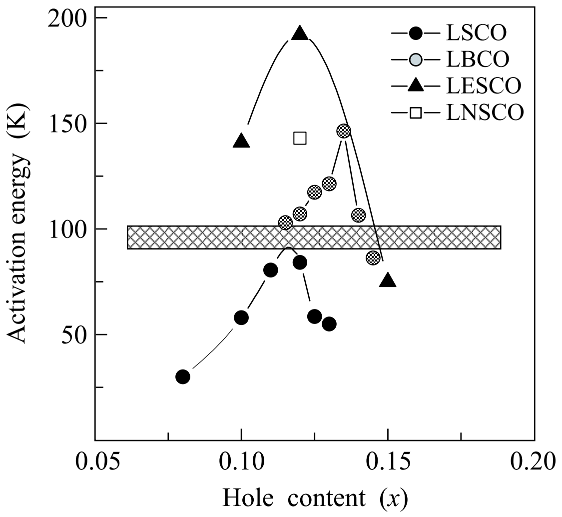

Fig.3. The activation energies $E_a(x)$ for the magnetic fluctuations in the different cuprates versus the hole doping x. Shadowed is the boundary separating the $E_a(x)$ values corresponding to nonsuperconducting and bulk superconducting phases

The enhancement of E_a (that is of a spin stiffness ρ_s) near $x = 0.12$ shown in Fig.3 gives evidence of the developed antiferromagnetic correlations for all investigated compounds and explains both the anomalously narrow peak in inelastic neutron scattering [15] and the elastic incommensurate peak with a narrow q-width [2] reported for the superconducting $La_{2-x}Sr_xCuO_4$ for this Sr doping. This indicates the important role of the commensurability and gives evidence of the plane character of the inhomogeneous spin and charge distributions. The maximal activation energies are 80 K for LSCO, 144 K for LBCO, 160 K for LESCO and 143 K for LNSCO. Note that for LBCO and LESCO the signatures of the bulk superconductivity [6, 7] become visible upon the suppression (in course of the Ba or Sr doping) of the activation energy down to 80–85 K. Therefore it is plausible to assume that these values of the activation energy are probably the critical ones for the realization of the bulk superconducting state. The corresponding boundary is shown in Fig.3. Fluctuations with the higher activation energies (spin stiffness) are effectively pinned and suppress the superconductivity.

To obtain the information about the ordered magnetic moments for the compounds with the enhanced spin stiffness the NMR measurements at 20–25 T were carried out in a high homogeneity resistive magnet of the NHMFL in Tallahassee FL. The temperature and doping dependencies of $^{63,65}Cu$ and ^{139}La NMR field sweep spectra of the oriented powders $La_{2-x}Sr_xCuO_4$ were studied. According to the previous La NQR results [3, 12] the measurements of oriented powder samples in a magnetic field perpendicular to **c** axis revealed that for Sr content near 1/8 the central lines of the observed spectra both for Cu and La exhibit the broadening upon cooling below 40–50 K (Fig.4).

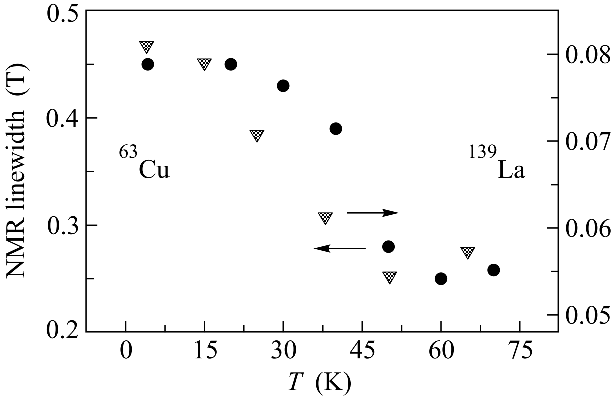

Fig.4. The temperature dependence of the ^{63}Cu and ^{139}La NMR linewidths for the superconducting LSCO with $x = 0.12$

Such a behaviour is connected with the slowing down of the magnetic fluctuations, which are gradually slowing down upon ordering. The broadening of the La NMR line allows us to estimate that the additional magnetic field at La nucleus is 0.015 T. If we consider that for the antiferromagnet La_2CuO_4 the copper moment of $0.64\mu_B$ induces at the La site the field of 0.1 T [16], then the effective magnetic moment in the present case is $\sim 0.09\mu_B$. Note that the manifestation of the magnetic order only in the vicinity of $x = 1/8$, when the AF structure is commensurate with the lattice, indicates that the magnetic inhomogeneities are of a plane character.

In conclusion our investigation reveals that for all studied compounds independent on the symmetry type (LTO or LTT) in the neighbourhood of 1/8 doping the enhancement of the spin stiffness takes place. The compounds with the spin stiffness larger than the certain critical value (See Fig.3) reveal no bulk superconductivity.

According to the phase diagram the inherent feature of the superconducting state in cuprates is the presence of frozen antiferromagnetic correlations. Such a coexistence seems to be a result of phase separation at the microscopic scale as it was discussed in pioneering paper of Gor'kov and Sokol [10].

In the neighbourhood of 1/8 doping this coexistence may be realized in a form of dynamic stripes, since the corresponding enhancement of the spin-stiffness reveals the plane character of the spin (and charge) inhomogeneities.

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