On the "spin-freezing" mechanism in underdoped superconducting cuprates

M. Eremin\textsuperscript{1,2}, A. Rigamonti\textsuperscript{1}

Department of Physics " A. Volta", University of Pavia,
I-27100 Pavia, Italy

\textsuperscript{2}Physics Department, Kazan State University, 420008 Kazan, Russia

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Abstract

The letter deals with the spin-freezing process observed by means of NMR-NQR relaxation or by muon spin rotation in underdoped cuprate superconductors. This phenomenon, sometimes referred as coexistence of antiferromagnetic and superconducting order parameters, is generally thought to result from randomly distributed magnetic moments related to charge inhomogeneities (possibly stripes) which exhibit slowing down of their fluctuations on cooling below $T_c$. Instead, we describe the experimental findings as due to fluctuating, vortex-antivortex, orbital currents state coexisting with $d$-wave superconducting state. A direct explanation of the experimental results, in underdoped $Y_{1-x}Ca_xBa_2Cu_3O_{6.1}$ and $La_{2-x}Sr_xCuO_4$, is thus given in terms of freezing of orbital current fluctuations.

74.20.Mn, 74.25.-q, 74.25.Ha
NMR-NQR and \( \mu \)SR experiments show that in underdoped cuprates, on cooling from about \( T_c \), divergent behaviour of the relaxation rates occur. This phenomenon, generally interpreted in terms of "glassy spin-freezing" or of "coexistence of superconductivity and magnetic ordering", is believed to be related to magnetic moments resulting from charge inhomogeneities, possibly stripes. For \(^{139}\)La NMR-NQR observations in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \), see Julien \textit{et al.} and References therein; a review of early data is given in Ref. \( \text{2} \) the muon longitudinal relaxation rate and spin precessional frequencies in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) and in \( \text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_6 \) have been measured by Niedermayer \textit{et al.}. More recently the low-temperature spin-freezing process in underdoped \( \text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_6.1 \) ( \( T_c \approx 35 \text{ K} \) ) has been studied by \(^{89}\)Y NMR relaxation in a field \( H_0 = 9.4 \text{ T} \)(Ref. \( \text{4} \)). The occurrence of a fast-relaxing component in the \(^{89}\)Y spin-lattice relaxation below \( T_c \) confirmed the \( \mu \)SR findings interpreted in terms of coexistence of superconductivity and spin-freezing process. A new significant aspect was the detection of the stretched character of the relaxation time, while the \( \mu \)SR data had been discussed in terms of a single exponential recovery.

Very recently Mook \textit{et al.} by means of inelastic neutron scattering measurements in underdoped superconducting YBCO detected longitudinal with respect to \( c \)-axis magnetic moments of unknown origin, with fluctuation frequencies well below the energy resolution of 1 meV. Similar observation, but with transverse magnetic moments, was reported by Sidis \textit{et al.}. NMR-NQR and \( \mu \)SR spin-lattice relaxation data have been interpreted in terms of fluctuations of a local \( h(t) \) originating from magnetic moments associated to hole localization, or from staggered moments within locally ordered mesoscopic domains, with non-zero effective spin or from stripes. The microscopic origin of the fluctuating field \( h(t) \) is still an open issue. Here we address this problem within a substantially different picture, namely the one of the extended charge density waves with imaginary order parameter (\textit{id-CDW}). Important features of this new scenario are described here and it is pointed out that the fluctuating field originates from vortex-antivortex orbital currents.

We start from the so-called \( t-J \) model by including the inter-site Coulomb interaction:
\[
H = \sum_{i,j} t_{ij} \psi_i^{pd,\sigma} \psi_j^{\sigma,dp} + \frac{1}{2} \sum_{i,j} j_{ij} \left[ (S_i S_j) - \frac{n_i n_j}{4} \right] + \\
+ \frac{1}{2} \sum_{i,j} G_{ij} \delta_i \delta_j
\]  

(1)

with \(\delta_i\) being the number of extra holes per unit cell of bilayer, \(\psi_i^{pd,\sigma}, (\psi_j^{\sigma,dp})\) creation (annihilation) operator constructed on the basis of the singlet combination of copper (d) and oxygen states (p) (see Ref. 3 for details). It should be noted that the temperature dependence of Cu(2) Knight shift in YBa\(_2\)Cu\(_4\)O\(_8\), the evolution of the Fermi surface in Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_8+y\) and the \(k\)-dependence of the pseudogap in the normal state have been successfully explained\(^{13}\) in the framework of that model, including the d-wave orbital symmetry of the superconducting state (SC)\(^{14}\). The d-wave SC can coexist with extended charge density waves (for short \(id\)-CDW)\(^{15,16}\). Chakravarty et al.\(^{17}\) have stressed that the corresponding ordered phase is a staggered pattern of orbital currents. The region in which d-wave SC coexists with \(id\)-CDW strongly depends on doping\(^{14}\). For \(\delta_{opt}\) the SC phase forces out \(id\)-CDW state (see Fig. 1 in Ref. 13), but when \(T^*/T_c \leq 2.5\) then SC and \(id\)-CDW state can coexist even at low temperatures. Non-monotonic temperature dependence of energy gap parameter, due to the competition of SC and \(id\)-CDW, was predicted in Ref. 13 and confirmed by Ekino et al.\(^{18}\) by means of break-junction tunneling spectroscopy in Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_8+y\).

Starting from another form of Hamiltonian Murakami\(^{19}\) has predicted (for \(G < 0\)) the coexistence \(id\)-CDW, antiferromagnetic (AF) and \(\eta\)-triplet states near half filling. Coexistence of d-CDW and spin density waves (SDW) was also derived by Bouis et al.\(^{20}\).

The main issue related to the scenario recalled above is whether it is possible to have a coexistence of \(d\)-SC and AF or SDW in the frame of the singlet - correlated band Hamiltonian (4). Using the Roth - type decoupling scheme\(^{21}\), in a mean field approximation, from Eq.(1) one has

\[
H^{MF} = \sum_k \Psi_k^+ M_k \Psi_k
\]  

(2)

where the matrix \(M_k\) is
Let us focus now at the order parameters:

\[ \eta_{k,Q}^\uparrow = G^{ph} + \left( t_{k+Q} - \frac{(s_i s_j) t_{ij}}{(P_{pd})^2} \right) \left\langle \frac{1}{2} \epsilon_Q + s_Q^z \rightangle - \]

\[ - \frac{1}{2NP_{pd}} \sum_{k'} \left\{ J_{k'-k} \left\langle \psi_{k'+Q}^{pd,\uparrow \downarrow} \psi_{k'}^{\downarrow \uparrow} \right\rangle + 2G_{k'-k} \left\langle \psi_{k'+Q}^{pd,\uparrow \downarrow} \psi_{k'}^{\downarrow \uparrow} \right\rangle \right\} \]

and

\[ U_k^\uparrow = - \frac{1}{NP_{pd}} \sum_{k'} (t_{k'+Q} + t_{k'}) \left\langle \psi_{k'}^{pd,\uparrow \downarrow} \psi_{k'-Q}^{\downarrow \uparrow} \right\rangle + \]

\[ + \frac{1}{2NP_{pd}} \sum_{k'} [J_{k'-k} + J_{k'+k+Q} - 2G_{k'-k}] \left\langle \psi_{k'}^{pd,\uparrow \downarrow} \psi_{k'-Q}^{\downarrow \uparrow} \right\rangle \]

where \( t_k, J_k, G_k \) are the Fourier transforms of transfer, superexchange and Coulomb coupling parameters, respectively. \( \epsilon_Q \) and \( s_Q^z \) are the Fourier amplitudes of the conventional charge density waves and spin density waves. For the commensurate instability wave vector \( Q = (\pi, \pi) \) as it was observed in neutron scattering, one derives the following relations

\[ \eta_{k,Q}^\uparrow = S_{1k}^\uparrow + S_{2k}^\uparrow + iD_k^\uparrow \]

\[ \eta_{k+Q,-Q}^\uparrow = -(S_{1k}^\uparrow)^* + (S_{2k}^\downarrow + iD_k^\uparrow)^* \]

\[ (\eta_{k,-k}^\downarrow)^* = S_{1k}^\downarrow + S_{2k}^\downarrow - iD_k^\downarrow \]

\[ (\eta_{k-Q,Q}^\downarrow)^* = -(S_{1k}^\downarrow)^* - (S_{2k}^\downarrow - iD_k^\downarrow)^* \]

Let us consider now the quantities

\[ S_{1k}^\uparrow = \left[ t_{k+Q}^{(1)} - \frac{(s_i s_j) t_{ij}^{(1)}}{(P_{pd})^2} \right] \left\langle \frac{1}{2} \epsilon_Q + s_Q^z \right\rangle \]

\[ S_{2k}^\uparrow = G^{ph} + \left( t_{k+Q}^{(2)} - \frac{(s_i s_j) t_{ij}^{(2)}}{(P_{pd})^2} \right) \left\langle \frac{1}{2} \epsilon_Q + s_Q^z \right\rangle \]
where the indexes 1 and 2 refer to the first and the second nearest neighbour hopping parameters. $S_{1k}^\uparrow$ changes the sign under the transformation $k \rightarrow k + Q$ and as one can see from Eq. (3) this yields a damping factor, since the energy dispersion becomes complex. The value $S_{2k}^\uparrow$ is invariant under such transformation and can generate $s$-wave CDW and SDW. Therefore, for open Fermi surface one finds that at the hopping parameter $t_1$ larger enough the expectation values $<e_q>$ and $<s_q>$ must be zero, and conventional SDW/CDW do not exist. This conclusion is in agreement with the one derived in Ref. 22 on the basis of neutron scattering data. However, we stress that the above conclusion is valid only for the energy dispersion of the form $(\cos k_x + \cos k_y)$, but not for the case $\epsilon_k = (\cos k_x + \cos k_y)^2$ when pockets are formed around the points $(\pm \pi/2, \pm \pi/2)$ in the Brillouin zone. In our picture this happens when antiferromagnetic correlation (parameter $J$) hampers the nearest neighbour hopping i.e. $t_1 \left(P_{pd} + \frac{\langle s_i s_j \rangle}{t_{pd}} \right) \approx 0$. It is noted that the order parameter $U_k$ resembles the so-called $\eta$-singlet pairing introduced by Yang23. Formally the $U_k$-order parameter construction in Eq.(5) also resembles the Larkin-Ovchinnikov, Fulde-Ferrell parameter24,25 with the important difference that the instability wave vector $q$ is completely different. The leading term of $U_k$

$$U_0^\uparrow = -\frac{1}{NP_{pd}} \sum_{kk'} \left( t_{k'Q} + t_{k'} \right) \left\langle \psi_{k'}^{\uparrow,dp} \psi_{-k'Q} \right\rangle$$

is determined by the second- and third- neighbours hopping integrals. Numerical solution of Eq. (9) in correspondence to the parameters ($t_1=80$ meV, $t_2 = 0$ and $t_3 = 12$ meV) used23 for the Fermi surface near optimal doping, yields a critical temperature of the onset $U$-phase below 1 K. One should remark that the onset of this inhomogeneous superconducting state is very sensitive to magnetic impurities and to the details of the Abrikosov’s vortex lattice. In addition we note that the $U$-ordering is particle-particle pairing at opposite spin orientations and must be sensitive to the external magnetic field. Since there is no evidence of such an effect of the magnetic field (see the data later on) we conclude that the peak in relaxation rate is not related to phase transition involving this $U$ order parameter. In the matrix given by Eq. (3) there are two imaginary components $iD^\uparrow$ and $iD^\downarrow$. It is useful to introduce their
combinations

\[ i(D_{k,Q}^1 + D_{k,Q}^\dagger) = -\frac{1}{2NP_{pd}} \sum_{k'} (J_{k'-k} + 2G_{k'-k}) \left\{ \langle \psi_{k'+Q}^\dagger \psi_{k}^\dagger dp \rangle + \langle \psi_{k'}^d \psi_{k'+Q} dp \rangle \right\} \] (10)

\[ i(D_{k,Q}^\dagger - D_{k,Q}^1) = -\frac{1}{2NP_{pd}} \sum_{k'} (J_{k'-k} - 2G_{k'-k}) \left\{ \langle \psi_{k'+Q}^\dagger \psi_{k}^\dagger dp \rangle - \langle \psi_{k'}^d \psi_{k'+Q} dp \rangle \right\} \] (11)

the first corresponding to charge current and the second one to spin current \cite{26,27} or, in other terminology, to the spin-nematic state \cite{28,29}. It is clear from Eqs. (10)-(11) that \( T_{cr}^{id-CDW} >> T_{cr}^{id-SDW} \), and when \( 2G \approx J \) Eq. (11) has no solution at all. Concluding this theoretical analysis, we see that the real situation is strongly dependent on the Fermi surface. Because there are no indication about pockets-like Fermi-surface in superconducting underdoped cuprates, we do expect coexistence of the \textit{d}-SC, \textit{id}-CDW and \textit{id}-SDW or \textit{U}-state at low temperatures. Let us see now what the experimental findings tell us.

On general physical grounds NMR-NQR or muon relaxation rates can be written

\[ \frac{1}{T_1} \approx \gamma^2_n \left\langle h_{eff}^2 \right\rangle J(\omega_m, \tau_e) \] (12)

where \( \left\langle h_{eff}^2 \right\rangle \) is a mean square amplitude of the transverse effective field at the nuclear or muon site and \((\omega_m, \tau_e)\) is the spectral density of the fluctuations at the measuring frequency \( \omega_m \) and an effective correlation time \( \tau_e \) is assumed. The stretched exponential recovery, of the form \( \exp \left[ -t/T_1^{e} \right]^{1/2} \) observed experimentally \cite{30} is naturally explained by charge density waves scenario, as it was pointed out by Philips\cite{30}. Usually this process is described as a superposition of Debye relaxation or in an ideal limit as a Laplace transform

\[ \exp \left[ -t/T_1^{e} \right]^{1/2} = \int_0^\infty \rho(1/T_1) \exp \left[ -t/T_1 \right] d(1/T_1) \] (13)

where \( \rho(1/T_1) = \frac{T_1}{2\sqrt{\pi}} \left( \frac{T_1^e}{T_1^e} \right)^{1/2} \exp \left[ -T_1/4T_1^e \right] \) is the distribution function. It is conceivable to assume that the distribution of \( (1/T_1) \) is related to a distribution of correlation time \( \tau_e \) and in turn to a distribution of the energy barrier \( E \) pinning the fluctuating - sliding current motions. Then we write
\[ \langle \tau_e \rangle = \tau_0 \exp(\langle E \rangle / k_B T) \] (14)

and

\[ J(\omega_m, \tau_e) = \frac{2 \langle \tau_e \rangle}{1 + \omega_m^2 \langle \tau_e \rangle^2} \] (15)

When the progressive slowing-down, on cooling, of the orbital current excitations causes \( \langle \tau_e \rangle \approx \frac{1}{\omega_m^{-1}} \), one has the maximum in the relaxation rates (frequently used to define \( T_g \), commonly called "spin-glass" freezing temperature). From this maximum one can extract the effective magnetic field induced by the currents at the nuclear or muon site:

\[ \sqrt{\langle h_{\text{eff}}^2 \rangle} = \frac{1}{\gamma_n} \left[ \frac{\omega_m}{(T_1^*)_{\text{max}}} \right]^{1/2} \] (16)

One could remark that on the basis of the above equation a slight underestimate of \( \sqrt{\langle h_{\text{eff}}^2 \rangle} \) is obtained, since the distribution of correlation times tends to level the maximum of the spectral function. The distribution also accounts for the deviation of the experimental data from the simple law (Eq. (14)) in the low temperature region (see Fig. 1). The values of \( \sqrt{\langle h_{\text{eff}}^2 \rangle} \) derived from the experimental data on the basis of Eq. (16) are collected in Table 1.

In Fig. one sees that Eq. (12), with Eqs. (14)-(15) rather well fits the observed temperature dependence of relaxation time in spite of the roughness of the assumptions. The energy barrier \( \langle E \rangle \) is roughly the same, for different kind of experiments and systems (see also Table 1). It is naturally related to periodic lattice potential. It should be stressed that the energy barrier is almost independent on the applied magnetic field. This fact would be hard to explain in the spin-freezing scenario. On the contrary, in our picture the correlation functions entering Eq. (10) are of particle-hole type and they are proportional to differences of Fermi functions with the same spin orientation\(^1\). Therefore, Zeeman energy comes out from these functions and they become independent of the external magnetic field. As a result the pseudo-gap temperature \( T^* \) and the pinning process caused by the lattice periodic potential are insensitive to the magnetic field.
As one can see from Table that the order of magnitude $\sqrt{\langle h_{\text{eff}}^2 \rangle}$ of the magnetic field agrees with theoretical estimation made on the basis of similar bond currents patterns (flux-phase) Ref.\textsuperscript{[2]}

The $z-$axis of the electric field gradient at $^{139}\text{La}$ site is almost parallel to $c-$axis (see for Ref. \textsuperscript{[2]}). Therefore we can interpret $\sqrt{\langle h_{\text{eff}}^2 \rangle} = 255\text{Gs}$ as a field transverse with respect to $c$-axis. The effective field at $Y$ site (9Gs) is relatively small. This means that transverse components within $Cu-O$ bilayer are almost compensated. Two magnetic field components at nuclear spin occur: a direct component - $h(1)$ as dicussed above and so-called hyperfine and supertransferred fields from adjacent $Cu(2)$ states - $h(2)$. The $id$-CDW state contributes to both. The modulation of the hyperfine coupling yields the effective magnetic field which is proportional to charge-spin correlation function

$$h(2) \propto F(q-q')e_qS_{q'}$$

where $F(q)$ is the form factor involved in the Mila-Rice Hamiltonian (see Ref.\textsuperscript{[3]}). This factor is different for $La$ and $Y$ nuclei sites and for electronic shell of ions $Yb^3+ \text{ and Er}^3+$ as well.

In summary, we have described the so-called spin-freezing process in superconducting state of underdoped cuprates with non-zero first neighbours hopping in a frame of a regime of coexistence of $d-$SC and $id-$CDW states.

We have shown:

i) that this scenario is naturally derived in a frame of singlet - correlated band model (or on the basis of the t-J model including inter-site Coulomb repulsion) and it is consistent with recent neutron scattering data and tunneling spectroscopy measurements. ii) the progressive slowing down of the sliding current motions, due to pinning barrier related to periodic lattice potential explains the temperature behavior of the relaxation rates detected in NMR-NQR, $\mu$SR and EPR experiments; in particular it directly justifies the stretched exponential character of the recovery curves; the fluctuating frequency lying in 10-1000 MHz range explain why the resolution limit ($\approx \text{meV}$) makes them invisible in neutron scattering measurements iii) the fact that practically no difference is observed in the relaxation rates
upon increasing the magnetic field from zero \((NQR)\) up to 23 Tesla and when the field is applied parallel or perpendicular to the c-axis (that is not explained in a spin-freezing scenario) is explained by the insensitivity of the current correlation functions to the external magnetic field.

Thus we believe that our picture brings a new and suggestive perspective, which is fully supported by the experimental findings and reveals new insights on a much debated issue.

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FIG. 1. Temperature behavior of the relaxation rates in La$_{2-x}$Sr$_x$CuO$_4$ and in Y$_{1.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.1}$, normalized to the values at T>>T_g ≈ T_{max}, as a function of T$^{-1}$. The data are taken from Ref. 1-4: circles are for La$_{1.94}$Sr$_{0.06}$CuO$_4$ in zero field and resonance frequency 18.6 MHz (NQR), stars - La$_{1.9}$Sr$_{0.1}$CuO$_4$ in H=23.2 Tesla, parallel to the c-axis, uptriangles - La$_{1.9}$Sr$_{0.1}$CuO$_4$ with H=9.4 Tesla, perpendicular to the c-axis, downtriangles - La$_{1.9}$Sr$_{0.1}$CuO$_4$ at H=9.4 Tesla, parallel to the c-axis, squares - Y$_{1.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.1}$ in H=9.4 Tesla.
### TABLES

**Table 1** Maximum values of the relaxation rates, estimated values for the effective field and for the average energy barrier for pinning of the sliding motions of the orbital currents.

| sample                  | Method (Ref.) | $\nu_0$(MHz) | $H_0$(Tesla) | $1/T_1$(s$^{-1}$) | $<E>$ (K) | $\sqrt{<h_0^2>}$ (Gs) |
|-------------------------|---------------|--------------|--------------|-------------------|-----------|------------------------|
| La$\text{1.9} \text{Sr}_{0.1}\text{CuO}_4$ | $^{139}$La NMR$^\dagger$ | 139.528      | 23.2         | $\approx$700      | 19        | 210                    |
| La$\text{1.94} \text{Sr}_{0.06}\text{CuO}_4$ | $^{139}$La NQR$^\dagger$ | 18.6         | 0            | $\approx$8000     | 30        | 255                    |
| Y$_{0.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_{6.1}$ | $^{89}$Y NMR$^\dagger$ | 19.61        | 9.4          | $\approx$1        | 26        | 9                      |
| Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{6.02}$ | $^{\mu}$SR$^\dagger$ | 0.51         | 0            | $\approx$60000    | 0         | 150                    |
| YBa$_2$Cu$_3$O$_{6.85}$ | Er,Yb EPR$^\dagger$ | 9460         | 0.2          | -                 | 25        | 160                    |