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LETTER TO THE EDITOR

Anisotropy of the antiferromagnetic spin correlations in the superconducting state of $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$

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Abstract
We present evidence that the antiferromagnetic spin correlations in optimally doped $YBa_2Cu_3O_7$ and underdoped $YBa_2Cu_4O_8$ develop a surprisingly strong anisotropy in the superconducting state. Comparing the ratio of the nuclear spin–lattice relaxation rates of the planar copper and oxygen, measured at the lowest and highest temperatures as well as at T_c, we conclude that the antiferromagnetic in-plane correlations vanish as the temperature goes to zero. This observation is corroborated by the measurement of the copper linewidth in $YBa_2Cu_4O_8$. In contrast, the out-of-plane correlations do not change appreciably between $T = T_c$ and $T = 0$. Within a model of fluctuating fields this extreme anisotropy of the antiferromagnetic correlations also explains the observed temperature dependence of the anisotropy of the copper relaxation measured in a low external magnetic field.

(Some figures in this article are in colour only in the electronic version)

Nuclear magnetic/quadrupole resonance (NMR/NQR) experiments have shed light on the antiferromagnetic (AFM) spin correlations in the normal state of high-temperature superconductors. It was found that the correlations become progressively stronger as the temperature is reduced towards the superconducting transition temperature T_c. The question is to what extent these correlations persist below T_c. As the temperature is lowered in the superconducting state, the spin–lattice relaxation rates ${}^kT_{1\alpha}^{-1}$ of the planar copper ($k = 63$) and planar oxygen ($k = 17$) nuclei decrease rapidly, with the applied field either parallel to the CuO_2 plane ($\alpha = ab$) or perpendicular to it ($\alpha = c$). This decrease goes approximately like T^3, which is the temperature dependence expected for d-wave orbital pairing [1]. Great emphasis has been put on the different temperature behaviours of the copper and oxygen relaxation rates in the normal state. However, less attention has been paid to the ratios of these rates [2–9] in the superconducting state. A difficulty arises, since in the normal state the spin–lattice relaxation is largely insensitive to the strength of the applied magnetic field [10, 11], while the relaxation rate becomes field dependent in the superconducting state at low temperature, with ${}^{63}T_{1c}^{-1}$ showing a stronger dependence than ${}^{63}T_{1ab}^{-1}$ and ${}^{17}T_{1c}^{-1}$ [5–7, 12]. In order to draw any conclusions about magnetism in the superconducting state it is therefore very important to look for intrinsic effects which can only be obtained from experiments done in weak magnetic fields so as to minimize the flux line influence. We will consider four NMR/NQR experimental results: the ratio ${}^{63}T_{1c}^{-1}/{}^{17}T_{1c}^{-1}$, the NQR copper linewidth in $YBa_2Cu_4O_8$, the nuclear spin–spin relaxation rate T_{2G}^{-1} and the ratio ${}^{63}T_{1ab}^{-1}/{}^{63}T_{1c}^{-1}$. From these experiments we conclude that in the superconducting state the AFM in-plane correlations vanish as the temperature goes to zero, whereas the out-of-plane correlations do not change much between $T = T_c$ and $T = 0$. In the case of the first three experiments the discussion is model independent. For the fourth the argumentation is based on a model of fluctuating fields developed recently [13]. For convenience and clarity, we have however adopted throughout the paper the formalism and notation of this model, which we outline below.

Some of us have recently proposed a new phenomenological model for the analysis of nuclear spin–lattice relaxation rate experiments in the normal state of the cuprates [13]. Special attention was paid to the question of to what degree the hyperfine fields originating from magnetic moments on copper ions should be added coherently. That the question is of importance is demonstrated by the anisotropy ratio ${}^{63}T_{1ab}^{-1}/{}^{63}T_{1c}^{-1}$.[1] The in-plane and out-of-plane AFM correlations could be determined in [13] for the optimally doped and underdoped compound of the $YBa_2Cu_3O_y$ family. The temperature dependences of the in-plane and out-of-plane correlations were found to be similar in the normal state. In the present report we extend the application of the model to the analysis of data obtained in the superconducting state, in particular to the question of the various measured ratios of the relaxation rates. In the model of fluctuating fields [14] the NMR spin–lattice relaxation rate of a nuclear species k is determined by fluctuating fields in the direction β and γ perpendicular to the applied static field direction α and is thus expressed as

$$ {}^{k}T_{1\alpha}^{-1}(T) = [{}^{k}V_{\beta}(T) + {}^{k}V_{\gamma}(T)]\tau_{\text{eff}}(T). \tag{1} $$

The term $\tau_{\text{eff}}(T)$ is an effective electronic spin–spin correlation time, and ${}^{k}V_{\beta}(T)$ and ${}^{k}V_{\gamma}(T)$ correspond to the square of the components β and γ of the effective hyperfine fields at the nucleus. In the cuprates the hyperfine fields are produced by more or less localized electronic moments on the copper ions. In particular, for a planar oxygen one gets [13]

$$ {}^{17}V_{\beta}(T) = \frac{1}{4\hbar^2}2C_{\beta}^2[1 + K_{01}^{\beta}(T)], \tag{2} $$

where C_β is the hyperfine field which is transferred from the two Cu moments (at site 0 and 1) adjacent to the O. K_{01}^{β} is the β-component of the normalized nearest-neighbour electron spin–spin correlation defined as $K_{01}^{\beta}(T) = 4\langle S_0^{\beta} S_1^{\beta}\rangle$ and can take values between -1 (fully antiferromagnetically correlated and yielding ${}^{17}V_{\beta} = 0$) and 0 (no correlation). The antiferromagnetism observed in the parent compounds is well described by a nearly isotropic two-dimensional Heisenberg model ($J^c \approx J^{ab}$). Upon doping, the long range order is destroyed but a short range order persists, characterized by spin–spin correlation lengths λ^c and λ^{ab} of the order of a lattice constant. In [13] the spin–spin correlations were parametrized according to $K_{01}^{\beta}(T) = -\exp[-1/\lambda^{\beta}(T)]$. Typical values for $YBa_2Cu_3O_7$ at T_c (as determined

[1] The experimental value for the anisotropy ${}^{63}T_{1ab}^{-1}/{}^{63}T_{1c}^{-1}$ in the normal state of $YBa_2Cu_3O_7$ is significantly higher than what is calculated assuming either a fully coherent or a fully incoherent addition of the hyperfine fields. Therefore, the experimental result can only be explained if the model provides an interpolation scheme which exhibits a maximum between these two extremes.

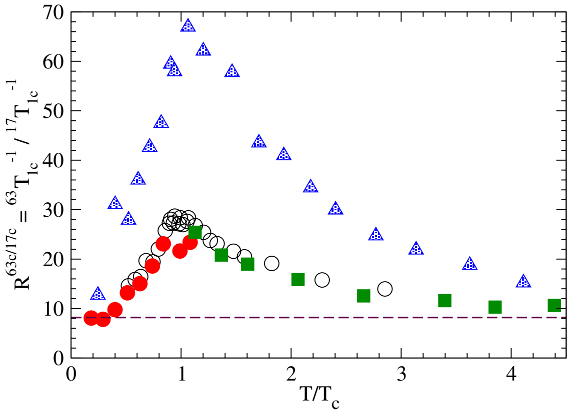

Figure 1. $R^{63c/17c}(T) = {}^{63}T_{1c}^{-1}/{}^{17}T_{1c}^{-1}$ versus T/T_c for $YBa_2Cu_3O_{6.96}$ in high field (empty circles, data from Yoshinari *et al* [4]) and for $YBa_2Cu_3O_7$ in low field (filled circles, data from Martindale *et al* [7]). The squares denote values calculated from combining ${}^{63}T_{1c}^{-1}$ data from Barrett *et al* [15] and ${}^{16}T_{1c}^{-1}$ data from Nandor *et al* [16]. The ratio for $YBa_2Cu_4O_8$ (half-filled triangles) was combined in high field for the oxygen measurement only, from the data from Bankay *et al* [9]. The dashed line is the model prediction for vanishing AFM correlations ($K_{01}^{ab} = 0$).

in [13]) are $K_{01}^{ab}(T = T_c) = -0.4$ and $K_{01}^{c}(T = T_c) = -0.5$. It should be emphasized that they are static correlations with respect to typical NMR times.

In contrast to oxygen, a copper nucleus is affected by an on-site anisotropic field (A_β) and by transferred isotropic hyperfine fields (B) originating from its four nearest neighbour copper ions. This leads to an expression for ${}^{63}V_\beta(T)$ that contains further distant spin correlations as well. For simplicity however, these further distant correlations have been assumed to depend on K_{01}^{β} and to decrease exponentially with the distance between spins. As a result ${}^{63}V_\beta(T)$ can be expressed as

$$ {}^{63}V_\beta(T) = \frac{1}{4\hbar^2}[A_\beta^2 + 4B^2 + 8A_\beta B K_{01}^{\beta}(T) + 8B^2|K_{01}^{\beta}(T)|^{\sqrt{2}} + 4B^2|K_{01}^{\beta}(T)|^2]. \quad (3) $$

Valuable information on AFM spin correlations is gained from ratios of relaxation rates, since then τ_{eff} (equation (1)) cancels out. We gather in figure 1 some experimental data for $R^{63c/17c} := {}^{63}T_{1c}^{-1}/{}^{17}T_{1c}^{-1}$ in $YBa_2Cu_3O_7$ (circles [4, 7], squares [15, 16]) and $YBa_2Cu_4O_8$ (triangles [9]). Note that the ratio for $YBa_2Cu_3O_7$ at elevated temperature has been obtained by combining copper data from Barrett *et al* [15] and oxygen measurements from Nandor *et al* [16]. Both optimally and underdoped compounds see an increase of $R^{63c/17c}$ from high temperature down to T_c, followed by a sharp decrease as the temperature is further reduced down to the very same level that is observed at the highest temperature. It is astonishing that this striking similarity of $R^{63c/17c}$ at very low and very high temperatures has not been much interpreted so far. One reason for this may be that in the standard analysis of ${}^{k}T_{1\alpha}^{-1}$ data (the so-called MMP theory [17]) the copper and oxygen are treated very differently. In the model outlined in equations (1)–(3), however, the relaxation rate of both nuclei is determined by the same τ_{eff}. It is only ${}^{63}V_\beta$ and ${}^{17}V_\beta$ that differ due to the different contribution of hyperfine field values and spin correlations.

The normal state temperature dependence of ${}^{63}T_{1c}^{-1}$ and ${}^{17}T_{1c}^{-1}$ could be fitted very well with the model (1)–(3), whereby their ratio is given by

$$ R^{63c/17c}(T) = \frac{2\,{}^{63}V_{ab}(T)}{{}^{17}V_a(T) + {}^{17}V_b(T)}. \quad (4) $$

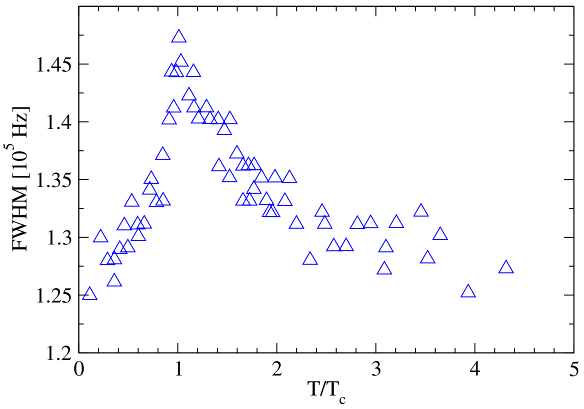

Figure 2. Linewidth versus T/T_c for $YBa_2Cu_4O_8$. Data from Mali *et al* [18].

We note that the temperature dependence of $R^{63c/17c}$ comes solely from $K_{01}^{ab}(T)$, the in-plane AFM correlations. At very high temperature we expect all AFM correlations to go to zero. In such a case (4) reduces to

$$R_0^{63c/17c} = \frac{A_{ab}^2 + 4B^2}{C_a^2 + C_b^2}. \tag{5}$$

Using the hyperfine constants calculated in [13] (in units of 10^{-6} eV: $A_{ab} = 0.168$, $B = 0.438$, $C_a = 0.259$, $C_b = 0.173$), we find that $R_0^{63c/17c} = 8.2$, a value marked by the dashed line in figure 1. It is obvious from figure 1 that this is also, to a very good agreement, the experimental $T \to 0$ limit of this ratio. Therefore, we conclude that the in-plane correlations reduce to zero in the superconducting state. It has of course been recognized in earlier works that the significant decrease in ${}^{63}T_{1c}^{-1}/{}^{17}T_{1c}^{-1}$ in the superconducting state suggests a loss of AFM fluctuations [8]. However, the connection with the measurements at elevated temperature had not been made. We would like to emphasize that equation (5) is a widely accepted result in the case of no correlations and is quite independent of our phenomenological model. In particular, equation (5) will result from any model that explains spin–lattice relaxation rates in terms of fluctuating hyperfine fields added incoherently.

Further indication that the in-plane AFM correlations decrease below T_c is given by the temperature dependence of the copper NQR linewidth of $YBa_2Cu_4O_8$. This underdoped compound has the advantage of being stoichiometric with a well ordered and stable structure, without the otherwise inevitable disorder effects created by extrinsic dopants. The temperature dependence of the planar copper linewidth from [18] is reproduced in figure 2. The measurements were made on a loose polycrystalline powder sample. The observed linewidth results from two contributions: a large temperature independent quadrupolar contribution and a smaller temperature dependent magnetic contribution. As seen in figure 2, the linewidth increases with decreasing temperature down to T_c, very much like $R^{63c/17c}$, and upon entering the superconducting state also decreases sharply. The exact origin of the temperature dependent magnetic contribution is at the moment not known precisely. However, the two main sources of the magnetic line broadening are the static local magnetic fields stemming from the material-imperfection induced staggered magnetization, and the indirect nuclear spin–spin interactions mediated by the electron spin system in the plane. In contrast to NMR, where the static magnetic line broadening is predominantly caused by the local magnetic field

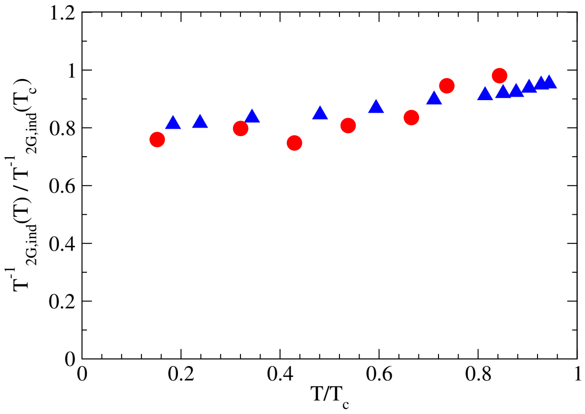

Figure 3. $T^{-1}_{2G,\mathrm{ind}}(T)/T^{-1}_{2G,\mathrm{ind}}(T_c)$ versus T/T_c for $YBa_2Cu_3O_7$ (circles) and $YBa_2Cu_4O_8$ (triangles). Data from Stern *et al* [20].

components *parallel* to the large applied magnetic field, the NQR lines are broadened mainly by local magnetic fields that are *perpendicular* to the NQR quantization axis [19]. This axis is material specific and is fixed by the largest principal axis of the electric field gradient (EFG) tensor. At the plane-copper site in $YBa_2Cu_4O_8$ the largest principal axis of the EFG tensor coincides with the crystallographic *c*-axis. Therefore, any decrease of the in-plane magnetic fields due to the loss of in-plane AFM correlations will result, as observed in the experiment, in a decrease of the magnetic contribution to the linewidth of the plane-copper NQR line.

We turn now to an experiment that provides information on the component of the correlations along the *c*-axis. It has been known from T^{-1}_{2G} measurements that AFM correlations do subsist in the superconducting state almost as strongly as in the normal state. Figure 3 reproduces a plot from Stern *et al* [20] showing normalized NQR measurements of $T^{-1}_{2G,\mathrm{ind}}$, the Gaussian contribution to the nuclear spin–spin relaxation T^{-1}_{2G} caused by the indirect nuclear spin–spin coupling mediated by the non-local static spin susceptibility. However, in the $YBa_2Cu_3O_y$ compounds, $T^{-1}_{2G,\mathrm{ind}}(T)$ depends only on the component of the real part of the electronic spin susceptibility along the *c*-axis [21], and hence it depends only on the correlations along this axis. In our notation therefore, $T^{-1}_{2G,\mathrm{ind}}(T)$ depends on K^{c}_{01} and not on K^{ab}_{01}. The measurements on $YBa_2Cu_3O_7$ (circles [22]) and $YBa_2Cu_4O_8$ (triangles [6]) reported in figure 3 indicate that the out-of-plane AFM correlations vary little from their value at T_c when the temperature is lowered. This suggests that whereas the in-plane AFM correlations vanish in the superconducting state, the out-of-plane correlations remain more or less frozen in.

Finally, we check these drastically different temperature dependences for the in- and out-of-plane components of the correlation on experiments where both components are involved. The development of such an extreme anisotropy of the AFM correlations in the superconducting state has visible consequences for the planar copper relaxation rate anisotropy $R^{63ab/63c} := {}^{63}T^{-1}_{1ab}/{}^{63}T^{-1}_{1c}$. Figure 4, reproduced from Bankay *et al* [6], shows the temperature dependence of $R^{63ab/63c}$ in $YBa_2Cu_3O_7$ (filled circles [22]) and $YBa_2Cu_4O_8$ (filled triangles [6]). Both ratios were measured in a weak magnetic field. In the normal state, the anisotropy is temperature independent and larger for $YBa_2Cu_3O_7$ (dotted line) than for $YBa_2Cu_4O_8$ (dashed line) [23]. After entering the superconducting state an upturn occurs, so that at low temperature the anisotropy of $YBa_2Cu_4O_8$ exceeds that of $YBa_2Cu_3O_7$.

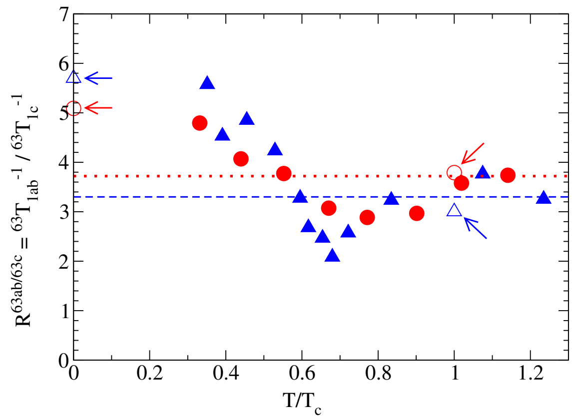

Figure 4. ${}^{63}T_{1ab}^{-1}/{}^{63}T_{1c}^{-1}$ versus T/T_c for $YBa_2Cu_3O_7$ (filled circles, data from Takigawa *et al* [22]) and $YBa_2Cu_4O_8$ (filled triangles, data from Bankay *et al* [6]) in low field. The dotted ($YBa_2Cu_3O_7$) and dashed lines ($YBa_2Cu_4O_8$) are the normal state measurements. The empty symbols are model predictions.

Within the framework of the phenomenological model equations (1)–(3), the anisotropy is expressed as follows:

$$R^{63ab/63c}(T) = \frac{1}{2}\left(1 + \frac{{}^{63}V_c(T)}{{}^{63}V_{ab}(T)}\right). \tag{6}$$

According to the suggestion that the out-of-plane correlations are frozen at their T_c value in the superconducting state and the in-plane ones have dropped to zero at $T = 0$, $R^{63ab/63c}$ at $T = 0$ becomes

$$R_0^{63ab/63c} = \frac{1}{2}\left(1 + \frac{{}^{63}V_c[K_{01}^c(T = T_c)]}{{}^{63}V_{ab}[K_{01}^{ab} = 0]}\right). \tag{7}$$

In order to compute $R^{63ab/63c}(T_c)$ and $R_0^{63ab/63c}$ we need to know, besides the hyperfine field constants, the values at T_c of the in-plane correlation $K_{01}^{ab}(T = T_c)$ and of the out-of-plane correlations $K_{01}^c(T = T_c)$. For $YBa_2Cu_3O_7$ we take the values determined in [13], $K_{01}^c(T = T_c) = -0.50$ and $K_{01}^{ab}(T = T_c) = -0.40$, and get $R^{63ab/63c}(T_c) = 3.79$ and $R_0^{63ab/63c} = 5.09$. These results are marked by empty circles at $T = T_c$ and $T = 0$ in figure 4. In the case of $YBa_2Cu_4O_8$ we do not have values for $K_{01}^{\alpha}(T = T_c)$, but we can use the results found in [13] for the underdoped compound $YBa_2Cu_3O_{6.63}$, whose planar charge carrier concentration comes close to that of $YBa_2Cu_4O_8$. Therefore, taking $[K_{01}^c(T = T_c)] = -0.61$ and $[K_{01}^{ab}(T = T_c)] = -0.69$, we get for $YBa_2Cu_4O_8$ $R^{63ab/63c}(T_c) = 3.00$ and $R_0^{63ab/63c} = 5.70$. These results are marked by empty triangles at $T = T_c$ and $T = 0$ in figure 4[2]. For both compounds the model predictions reproduce well the limits of the temperature behaviour of this ratio.

In conclusion, we have shown that a range of NMR/NQR experiments indicates that below T_c the AFM spin correlations develop a different behaviour in the in-plane direction and in the out-of-plane direction. From the analysis of $R^{63c/17c}$ and the planar copper NQR

[2] In equation (7), the value of ${}^{63}V_{ab}[K_{01}^{ab} = 0]$ is the same for any compound with the same hyperfine field constants. However, ${}^{63}V_c[K_{01}^c(T = T_c)]$ is higher for $YBa_2Cu_4O_8$ than for $YBa_2Cu_3O_7$, since the AFM correlations at T_c are higher in the underdoped than in the optimally doped compounds. Hence $R_0^{63ab/63c}$ is higher in the former than in the latter.

linewidth in $YBa_2Cu_4O_8$ we deduced that the AFM in-plane correlations disappear gradually in the superconducting state, in contrast to the out-of-plane correlations, which remain almost unchanged, as demonstrated by the measurements of the planar copper spin–spin relaxation T_{2G}^{-1}. Additional evidence that an extreme anisotropy of the AFM correlation develops in the superconducting state is provided by the analysis of $R^{63ab/63c}$ within the model of fluctuating fields. The predicted values at $T = 0$ come astonishingly close to the experiment. Expressed in terms of the AFM correlation lengths λ^c and λ^{ab}, we find that λ^{ab} vanishes as $T \rightarrow 0$. This means that the short range correlations of the spin system, which above T_c retained the nearly isotropic Heisenberg-like character of the parent antiferromagnet, acquire below T_c with decreasing temperature a more and more Ising-like character. We finally point out that these conclusions are drawn from NMR data, which are sensitive to the quasiparticle spectrum at very low energies. It is possible that neutron scattering data taken at higher energy might give a different picture.

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