ИНСТИТУТ ЯДЕРНОЙ ФИЗИКИ СО АН СССР

O.P. Sushkov

THE POSSIBILITY OF INVESTIGATION OF HIGH T_c-SUPERCONDUCTORS SPIN STRUCTURE USING THE NEUTRONS WITH ENERGY 1—10 eV

PREPRINT 90-128

НОВОСИБИРСК

The Possibility of Investigation of High T_c-Superconductors Spin Structure Using the Neutrons with Energy 1-10 eV

O.P. Sushkov

Institute of Nuclear Physics
630090, Novosibirsk, USSR

ABSTRACT

The suggestion is to use polarized neutrons with the energy 1-10 eV for the investigation of spin structure of copper oxide high T_c-superconductors as well as for investigation of magnetic excitons in these compounds. Corresponding dynamic formfactors are calculated.

Presently the idea of spin liquid (SL) is almost common view on the spin structure of copper oxide high T_c-superconductors. The main point of interest is CuO_2-sheet which is probably responsible for the superconductivity. There are the localized spins s=1/2 at the ions Cu^{2+}. In undoped state (say $La_{2-x}Sr_xCuO_4$ at x=0) the compound is insulator with the Neel ordering of localized spins: $\langle s_z \rangle = \pm 1/2$. More precisely $\langle s_z \rangle$ is smaller due to the quantum fluctuations: $\langle s_z \rangle = \pm 1/2 \cdot 0.55$ [1]. At the doping ($x > 3 \cdot 10^{-2}$) Neel ordering disappears and average value of electron spin at the copper vanishes: $\langle s_z \rangle = 0$. (The qualitative phase diagram for $La_{2-x}Sr_xCuO_4$ is presented at Ref. [2].) Due to the some experimental data on nuclear quadrupole resonance at Cu the Neel ordering does not disappear, but $\langle s_z \rangle$ becomes very small: $\langle s_z \rangle \sim 10^{-2}$ [3]. Picture of the SL corresponds to transition into the quantum state with localized spins $\langle s_i^2 \rangle = 3/4$, but $\langle \vec{s}_i \rangle = 0$. It is not one and only possible scenario. For example $\langle \vec{s}_i \rangle = 0$ if the spins become delocalized and the

system transfers into Fermi liquid (FL) state. The FL picture does not contradict to existing experimental data as well.

To demonstrate the difference between these two scenarios let us consider the model of one double link Cu-O-Cu. Let $a^+_{1\sigma}$ be operator of the hole creation in closed $3d^{10}$-shell of the first copper ion (σ is the spin projection), and $a^+_{2\sigma}$ be the same for second ion. The wave function of Neel state is of the form

$$|\psi_N\rangle = a^+_{1\uparrow} a^+_{2\downarrow} |0\rangle, \qquad \langle s_{iz}\rangle = \pm 1/2. \qquad (1)$$

Spin liquid is the state with spin zero

$$|\psi_{SL}\rangle = 1/\sqrt{2}\ (a^+_{1\uparrow} a^+_{2\downarrow} - a^+_{1\downarrow} a^+_{2\uparrow})|0\rangle, \qquad \langle s_{iz}\rangle = 0. \qquad (2)$$

Fermi liquid corresponds to the hybridization of orbitals

$$|\psi_{FL}\rangle = 1/\sqrt{2}\ (a^+_{1\uparrow} + a^+_{2\uparrow}) \cdot 1/\sqrt{2}\ (a^+_{1\downarrow} + a^+_{2\downarrow})|0\rangle, \qquad \langle s_{iz}\rangle = 0. \qquad (3)$$

In SL the correlator of neighboring spins does not vanishes, and for distant spins the correlator drops down with the distance. Therefore it is evident that to probe SL one needs the magnetic scattering with momentum transfer $q \sim \pi/a$, where a=3.8Å is Cu-Cu distance. The energy transfer ω should be of the order of energy of system excitation. Antiferromagnetic interaction of the localized Cu spins is $J\,\vec{s}_1\vec{s}_2$, $J \approx 0.1$ eV (see e.g. Ref [4]). Therefore $\omega \sim J \approx 0.1$ eV. Thus for investigation of the SL one needs the neutrons with energy $E > 0.1$ eV.

In the simple model of one link Cu-Cu it is easy to calculate the cross sections of magnetic scattering on the states (2), (3) which correspond to SL and FL. If interaction of the neutron with electron equals $g\,\vec{S}_n\vec{s}_e$, then

$$\sigma_{SL} \sim 3/8 g^2 (1-\cos\vec{q}\vec{a})\delta(\omega - J), \quad \sigma_{FL} \sim 3/16\ g^2 (1-\cos\vec{q}\vec{a})\ \delta(\omega - t). \qquad (4)$$

For the FL ω equals to hybridization energy t~2-3 eV. Surely for infinite system the ω-distribution is not δ-function but has the width of the order of J or t correspondingly. Thus in magnetic scattering we can easily differ the spin liquid from the Fermi liquid: the ω-distributions are completely different.

Let $E = p^2/2M$ be initial energy of the neutron. Then $\omega = p^2/2M - (\vec{p} - \vec{q})^2/2M \approx \vec{p}\vec{q}/2M = pq_\parallel/M$. It is easy to verify that if a=3.8Å and ω and E are taken in eV then $q_\parallel a \approx 42\omega/\sqrt{E}$. At ω=0.1 eV and E=10 eV we get $q_\parallel a \approx 1.33$. This is exactly suitable magnitude of longitudinal momentum transfer. At transverse momentum transfer $q_\perp \sim \pi/a$ the scattering angle equals $\alpha \approx q_\perp/p \approx 4 \cdot 10^{-2}/\sqrt{E}$. For the E=10 eV we have $\alpha \sim 10^{-2}$ rad.

Let us do more accurate calculation. Matrix element of the Hamiltonian of neutron magnetic interaction with the Cu $3d$-electron equals

$$H_M = 8\pi\ \mu_n\ \mu_B\ S_\alpha\ (\delta_{\alpha\beta} - n_\alpha n_\beta)\ (l_\beta + 2s_\beta). \qquad (5)$$

Here $\vec{n} = \vec{q}/q$, $\vec{S}$ is spin of the neutron, $\vec{l}$ and $\vec{s}$ are orbital angular momentum and spin of the electron, μ_B is Bohr magneton, and $\mu_n = -1.91\mu_N$ is the neutron magnetic moment. Spin ex-

system transfers into Fermi liquid (FL) state. The FL pic-citations are due to the term in eq.(5) with spin of electron. Cross section averaged over the neutron polarizations and reduced to the one cell of CuO_2-sheet equals

$$\frac{d\sigma}{d\omega\, d\Omega} = 2W(\omega, \vec{q})\, \sigma_0,$$

$$W(\omega, \vec{q}) = 1/N \sum_n |\langle n| \sum_{i=1}^{N} s_z^{(i)} e^{i\vec{q}\vec{r}} |0\rangle|^2\, \delta(\omega - \omega_n), \qquad (6)$$

$$\sigma = (4 m_n\, \mu_n\, \mu_B)^2 = 2.9 \cdot 10^{-25}\ \mathrm{cm}^2.$$

Here m_n is the neutron mass and $W(\omega, \vec{q})$ is dynamic formfactor of SL. For the liquid axis z can be directed in arbitrary direction. Spin liquid was simulated on the clusters consist of N=8, 9, 10 Cu ions on square lattice. The wave functions was found by direct diagonalization of Hamiltonian matrix $\Sigma\, J\, \vec{s}_i \vec{s}_j$ (sum over nearest neighbors). The formfactor depends on the $\vec{q}_p = \vec{q}_x + \vec{q}_y$. The axes x, y lie in CuO_2-plane and are directed along the CuO-links. For simplicity let us consider formfactor averaged over the directions of $\vec{q}_p$ in the plane. This corresponds to the experiment with polycrystalline sample but with aligned CuO_2-sheets. At Fig.1 the plot of static formfactor is presented: $W(q_p) = \int_0^{\omega} d\omega\, W(\omega, q_p)$.

At simulation on cluster the dynamic formfactor consists of separate lines. To obtain the smooth curve each line was spread by the factor

$$f = \frac{\Gamma}{2\pi} \frac{1}{(\omega - \omega_0)^2 + \Gamma^2/4}\, \phi(\omega),$$

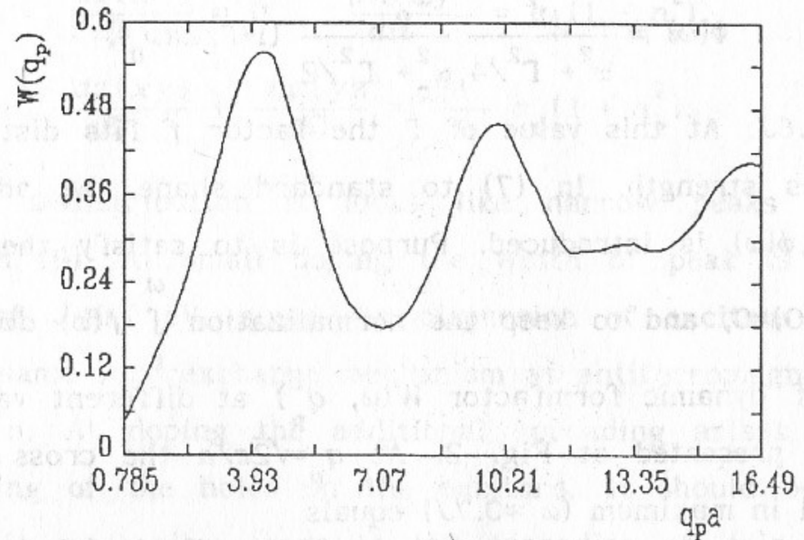

Fig. 1. Static formfactor of spin liquid $\pi/4 \le qa \le 21\pi/4$. First maximum is in the point $q = \pi\sqrt{2}/a$.

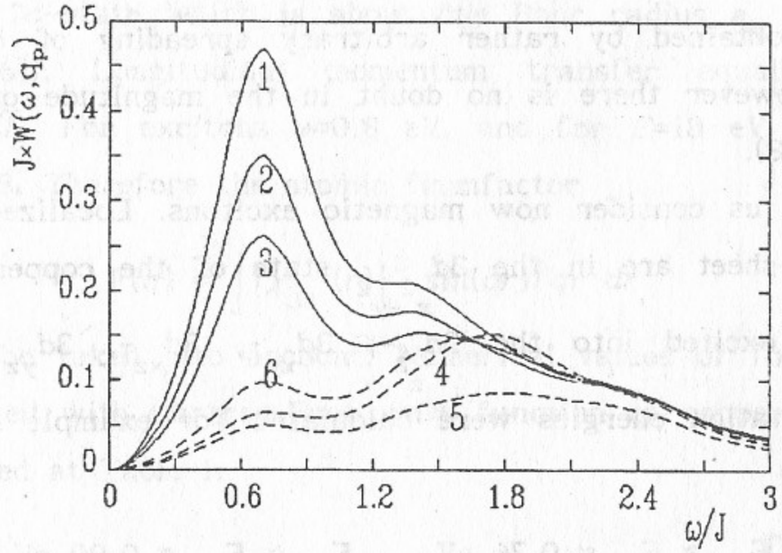

Fig. 2. Dynamic formfactor of spin liquid. Solid curves correspond to the maximums of static formfactor: $q_1 a = \sqrt{2}\,\pi$, $q_2 a = 13\pi/4$, $q_3 a = 5\pi$ (see Fig. 1). Dashed curves correspond to the minimums: $q_4\, a = 3\pi/4$, $q_5\, a = 9\pi/4$, $q_6\, a = 15\pi/4$.

$$\Phi(\omega) = \frac{\omega^2}{\omega^2 + \Gamma^2/4} \frac{\omega_0^2 + \Gamma^2}{\omega_0^2 + \Gamma^2/2} (1+\Gamma/2\pi\omega_0), \qquad (7)$$

at $\Gamma=0.6J$. At this value of Γ the factor f fits distribution of lines strength. In (7) to standard shape the additional factor $\phi(\omega)$ is introduced. Purpose is to satisfy the condition $f(0)=0$, and to keep the normalization $\int_0^\omega f(\omega)\, d\omega \approx 1$. The plots of dynamic formfactor $W(\omega, q_p)$ at different values of q_p are presented at Fig. 2. At $q_p = \sqrt{2}\pi/a$ the cross section per cell in maximum ($\omega \approx 0.7J$) equals

$$\frac{d\sigma}{d\omega\, d\Omega} \approx 2.7\cdot 10^{-25}\,\mathrm{cm}^2/J \approx 2.7\cdot 10^{-24}\,\mathrm{cm}^2/\mathrm{eV}. \qquad (8)$$

Surely this calculation is not very reliable one since $W(\omega, q_p)$ is obtained by rather arbitrary spreading of discrete lines. However there is no doubt in the magnitude of cross section (8).

Let us consider now magnetic excitons. Localized holes of CuO_2-sheet are in the $3d_{x^2-y^2}$ state of the copper. They can be excited into the $3d_{z^2}$-, $3d_{xy}$-, $3d_{xz}$-, $3d_{yz}$-states. The excitation energies were calculated for example in Ref. [5]:

$$E_{xy} \approx E_{z^2} \approx 0.76 \text{ eV}, \qquad E_{xz} \approx E_{yz} \approx 0.90 \text{ eV}. \qquad (9)$$

For these excitations the term with 1 in matrix element (5) is responsible. Averaged over the neutron polarizations cross section per Cu ion equals

$$\frac{d\sigma(z^2)}{d\Omega} = 0, \qquad \frac{d\sigma(xy)}{d\Omega} = \sigma_0(1 - n_z^2),$$
$$\frac{d\sigma(xy)}{d\Omega} + \frac{d\sigma(yz)}{d\Omega} = \frac{1}{4}\sigma_0(1 + n_z^2). \qquad (10)$$

In the ω-distribution it looks like narrow peaks at the energies (9). At small doping the width of peak is of the order of $J\approx 0.1$ eV, since the dispersion of excitons is due to the same superexchange mechanism as antiferromagnetic interaction. At doping the additional spreading arises due to scattering of the holes on the excitons. It should be noted that this mechanism leads to the spreading of spin excitation dynamic formfactor (eq. (6) and Fig. 2) as well.

The formulae (10) do not take into account the size of atomic $3d$-state which is about one Bohr radius a_B (see e.g. Ref. [6]). Longitudinal momentum transfer equals $q_\parallel a_B \approx 5.8\omega/\sqrt{E}$. For excitons $\omega\approx 0.8$ eV, and for $E=10$ eV we have $q_\parallel a_B \approx 1.8$. Therefore the atomic formfactor

$$F(q) = \int |\chi_{3d}(r)|^2 \sin(qr)/qr\, dr \qquad (11)$$

should be taken into account. Numerical values of formfactor calculated with Hartree-Fock wave function of copper ion are presented at Table 1.

Table 1

Formfactor of Cu 3d-state

qa_B	0.	0.5	1.	1.5	2.	2.5	3.	3.5
$F(q)$	1.	0.95	0.82	0.66	0.50	0.37	0.27	0.18

If we neglect the higher angular harmonics in neutron wave function the cross sections (10) should be simply multiplied by factor F^2. At E=10 eV this reduce exciton cross sections by a factor 3 - 3.5.

The observation of excitons is important by itself. However I would like to stress that it could be very useful for interpretation of spin excitation data. Actually calculation of the cross sections (10) is rather reliable. Therefore using them for normalization one can find from experimental data the absolute normalization of dynamic formfactor of spin liquid.

Let us discuss now independent of neutron spin mecha-nisms of forward scattering with energy transfer ω~0.1 eV. They contribute to the background and can hamper the obser-vation of magnetic scattering. First is the scattering on the optical phonons. Most hard are the oscillations of the in plane oxygen ions. They have frequency $\omega_0 \approx 0.1$ eV (see e.g. Ref. [7]). Simple calculation gives (per cell of CuO_2-plane).

$$\frac{d\sigma(ph)}{d\Omega} \approx l^2 (q/\sqrt{M\omega_0})^2 \approx 6\cdot 10^{-28}\text{cm}^2 \ll \sigma_0. \qquad (12)$$

Here $M=16m_n$ is O^{2-} mass and $l\approx 5.8$ fm is the neutron scattering length on oxygen nucleus [8]. Numerical value of cross section is taken at $q=\pi/a$. Thus the scattering on phonons is negligible in comparison with magnetic scattering. More important is double scattering of a neutron in sample with momentum transfers $q_1, q_2 \gg q$, $\vec{q}_1+\vec{q}_2=\vec{q}$. In this case we can consider the atoms as unbounded and energy transfer equels $\omega=q_1^2/M$. Let L be the length of a sample and L_0 be the mean free path of neutron. If $L \le L_0$ the probability of double scattering equals

$$\frac{dW}{d\omega\ d\Omega} \approx \frac{\pi M}{4m_n}\frac{l^4}{E}(nL)^2. \qquad (13)$$

Here l is the length of nuclear scattering, E is the neutron energy, and n is density of the atoms which contribute to rescattering. Let us consider for example La_2CuO_4. Let N be the density of elementary cells. Due to Eq.(13) the most effective are heavy atoms. Therefore $n \approx n_{La}=2N$. Scattering length $l\approx 5$ fm is roughly the same for all atoms, and therefore $L_0 \approx (7N\sigma)^{-1} \approx (28\pi l^2 N)$. To compare the probability of double scattering (13) with the cross sections presented above we should divide it by NL.

$$\frac{1}{NL}\frac{dW}{d\omega\ d\Omega} \approx 2.5\ l^2/E\ (L/L_0). \qquad (14)$$

At E=10 eV and L/L_0=1 it equals to $0.6\cdot 10^{-25}$cm/eV. Thus even at L/L_0=1 the probability of magnetic scattering (8) is by a factor ~50 larger then the probability of rescattering. To avoid the misunderstanding we remind that this is scattering at small angles.

What kind of experiment is discussed?

1. This is forward neutron scattering: $\alpha \sim 4\cdot 10^{-2}/\sqrt{E}_{eV} \sim 10^{-2}$rad.

Resolution inside this angle is desirable to measure the $\vec{q}$-dependence of formfactor.

2. One needs to separate the events with neutron spin flip to measure the magnetic scattering. Due to the estimations presented above nonmagnetic background is small. However these estimations are based on the suppositions on the spin liquid structure, and those should be checked by themselves.
3. The resolution in energy losses should be better then 0.1 eV.
4. The dependence of formfactor on the orientation of $\vec{q}$ with respect to CuO_2-sheet is important. To measure this dependence the sheets in a sample should be aligned.
5. Control of the value of CuO_2-sheet doping by the holes is desirable. Probably the most effective way to do it is to measure the shift of frequency of Cu nuclear quadrupole resonance [9].

First of all I am grateful to V.V. Flambaum. In discussions with him the importance of experiment with energetic neutrons was realized. I am grateful as well to V.P. Alfimenkov, V.I. Lushikov, S.V. Maleyev, A.I. Okorokov, R. Osborn, L.B. Pikelner, A.P. Serebrov for stimulating discussions.

REFERENCES

1. *D. Vakhin et al.* Phys. Rev. Lett., 58 (1987) 2802.
2. *Amnon Aharony et al.* Phys. Rev. Lett., 60 (1988) 1330.
3. *K. Kumagai and Y. Nakamura.* Physica **C157** (1989) 307.
4. *Hong-Qiang Ding and M.S. Makivic.* Phys. Rev. Lett., **64** (1990) 1449.
5. *V.V. Flambaum and O.P. Sushkov.* Physica **C168** (1990) 565.
6. *V.V. Flambaum and O.P. Sushkov.* Physica **C159** (1989) 586.
7. *R. Feile.* Physica C159 (1989) 1.
8. *S.F. Mughabghab.* Neutron Cross Sections. Vol. 1. Academic Press 1981.
9. *V.V. Flambaum and O.P. Sushkov.* Preprint INP 90-68. Submitted to Physica C.

O.P. Sushkov

The Possibility of Investigation of High T_c-Superconductors Spin Structure Using the Neutrons with Energy 1-10 eV

О.П. Сушков

Возможность исследования спиновой структуры высокотемпературных сверхпроводников с помощью нейтронов с энергией 1-10 эВ

Ответственный за выпуск: С.Г.Попов

Работа поступила - 5 октября 1990 г.
Подписано к печати 14.11 1990 г.
Формат бумаги 60×90 1/16
Объем 0,9 п.л., 0,8 уч.-изд.л.
Тираж 180 экз. Бесплатно. Заказ 128.

Ротапринт ИЯФ СО АН СССР,
г. Новосибирск, 90.