Experimental study of the spin-wave spectrum of a Bi_2CuO_4 single crystal by the inelastic neutron scattering method

G. A. Petrakovskiĭ, K. A. Sablina, V. V. Val'kov, and B. V. Fedoseev
Institute of Physics, Siberian Branch, Russian Academy of Sciences, 660036, Krasnoyarsk

A. Furrer, P. Fischer, and B. Roessli
Laboratory for Neutron Scattering ETH, Zurich, CH-5232, Villigen, Switzerland

(Submitted 1 April 1992; resubmitted 10 July 1992)
(Pis'ma Zh. Eksp. Teor. Fiz. **56**, No. 3, 148–152 (10 August 1992)

The spin-wave spectrum and the critical behavior of the sublattice magnetization of a Bi_2CuO_4 single crystal have been studied by the neutron-scattering method. The structural features of the spectrum have been explained theoretically on the basis of an effective spin Hamiltonian. The basic role of the four-spin interaction has been determined.

The study of Me_2CuO_4 compounds (Me = Nb, Pr, Sm, Bi, La) is important for understanding of the function of the magnetic subsystem in high-T_c superconducting materials. Accordingly, the Bi_2CuO_4 compound, in which the spin moments of Cu^{2+} ions are antiferromagnetically ordered at $T < T_N \sim 45$ K, is presently being studied extensively.[1–6] Experiments on the scattering of neutrons by a single crystal and the results on the measurements of the magnetic susceptibility have shown that magnetization of the sublattice is oriented in the basal plane of the tetragonal Bi_2CuO_4 compound (space group $P4/ncc$), and that the magnetic moment of the Cu^{2+} ion is 0.85 μ_B. The paramagnetic Néel temperature and the g-factor are strongly anisotropic: $\theta^{\parallel} = -32$ K, $\theta^{\perp} = -40$ K; $g^{\parallel} = 2.290$, $g^{\perp} = 2.017$.

In the present article we report the results of an experimental study of the critical behavior of the magnetic excitation of Bi_2CuO_4 and the dispersion relation. Large Bi_2CuO_4 single crystals ($V \sim 1$ cm^3) were grown in the Institute of Physics, Siberian Branch, Russian Academy of Sciences.[5] The temperature dependence of the sublattice magnetization was measured by using the method of elastic scattering of neutrons ($\lambda = 2.345$ Å) on a biaxial P2AX spectrometer. The inelastic scattering of neutrons by spin waves was measured using a triaxial PZAX spectrometer of the Sapphire reactor of the Paul Scherrer Institute. The energy of the inelastic neutron scattering was held constant at 14.96 meV, with a resolution of 1.0 meV. The single-crystal sample was placed in a cylindrical vanadium container built into a closed-cycle helium refrigerator. The inelastic neutron scattering was measured at a temperature of 8 K for spin waves which propagated along the C axis.

The temperature variation of the intensity of the magnetic Bragg [100] reflections is shown in Fig. 1. A computer analysis of the experimental data obtained near the Néel temperature gives $T_N = 45.78 \pm 0.19$ K and a critical index $\beta = 0.36 \pm 0.08$. Such a value of the parameter β confirms the assumption that the magnetic ordering in Bi_2CuO_4 is three dimensional in nature. Figure 2 shows the temperature dependence of the sublattice magnetization in the critical region. As can be seen from the plot, the behavior is described well by a power law.

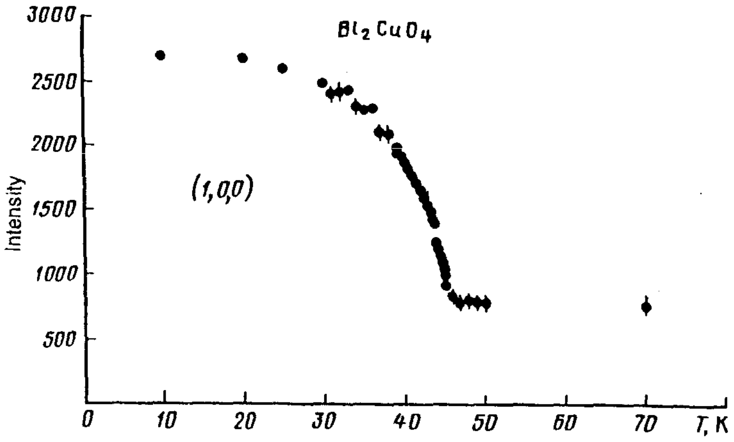

FIG. 1.

Figure 3 is a plot of the experimental dependence of the intensity of inelastic scattering versus the energy transfer at a relatively low momentum transfer. The experimental points clearly indicate two inelastic-scattering peaks, whose locations correlate with the excitation energies of two spin-wave branches. Such a structure of the intensity is retained even when the momentum transfer increases, but the peaks in this case converge. Only one peak has been observed experimentally near the boundary of the Brillouin zone. This experimental observation is interpreted as a matching of excitation frequencies (within experimental error) of the two branches. The results of a computer analysis of the experimental data on the multiple measurements of the inelastic scattering for each value of the wave vector are shown in Fig. 4 (the experimental data points).

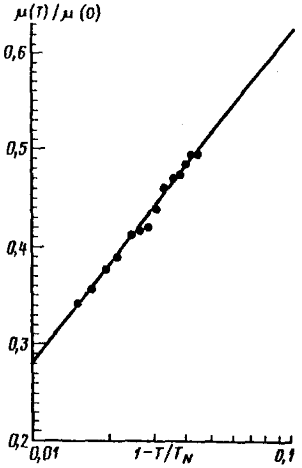

FIG. 2.

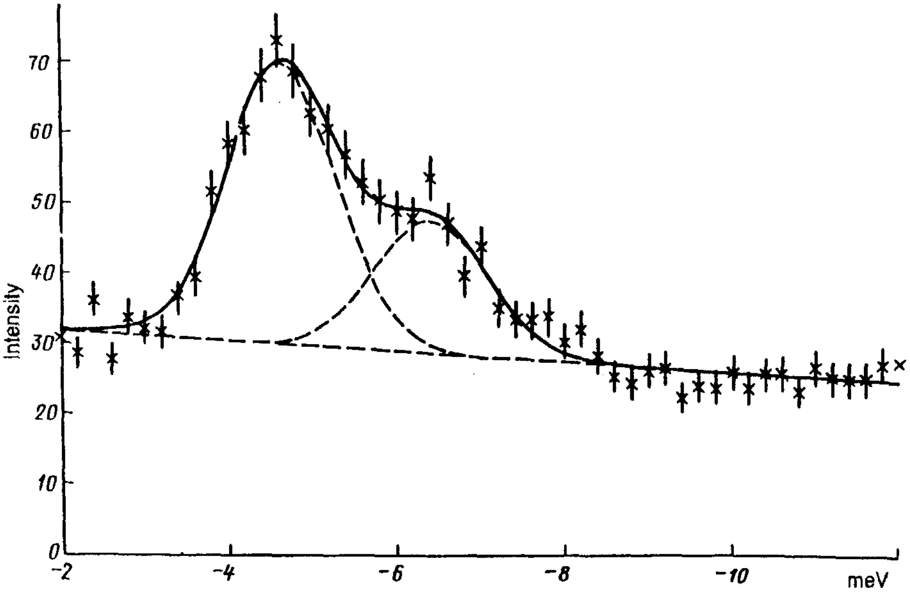

FIG. 3.

To obtain information about the magnetic interaction in Bi_2CuO_4, we calculated the collective excitation spectrum on the basis the effective spin Hamiltonian H_{eff}. In writing it for Bi_2CuO_4 we took into account two important factors. This first factor has to do with the fact that its spin is $S = 1/2$, and hence H_{eff} cannot contain terms corresponding to a single-ion anisotropy. Consequently, the experimentally observed anisotropy of the magnetic properties is attributable to the anisotropy of the two-site and the multiple-site spin–spin interaction (four-site spin–spin interaction in our case, since three-site interaction is absent because $S = 1/2$). The second factor is that Bi_2CuO_4 in the magnetic phase has a tetragonal symmetry and the sublattice magnetization is in the basal plane. Given such a geometry, allowance in H_{eff} of only the invariants quadratic in the spin operators would result in that the lower branch of the spin-wave vector would clearly have no gaps. The experimental data, on the other hand, can be described better if the lower branch exhibits activation behavior.[1] We have therefore included a four-site volume in H_{eff}:[8]

$$H_{eff} = -\frac{1}{2}\sum_{\substack{\alpha\beta\\ fm}} I^{\alpha\beta}_{fm} S^{\alpha}_{f} S^{\beta}_{m} - \frac{1}{4}\sum_{\substack{\alpha\beta\gamma\delta\\ fmgl}} K^{\alpha\beta\gamma\delta}_{fmgl} S^{\alpha}_{f} S^{\beta}_{m} S^{\gamma}_{g} S^{\delta}_{l} \equiv H(2) + H(4), \qquad (1)$$

where the four-spin interaction is taken into account only for the four-site plaquettes which have the smallest perimeter and which are oriented in the basal plane and in the plane parallel to the tetragonal axis. Additionally, for simplification we have used in $H(4)$ only the invariants of the cubic symmetry. This procedure gives us three constants for each type of plaquette, from which we can determine whether the lower

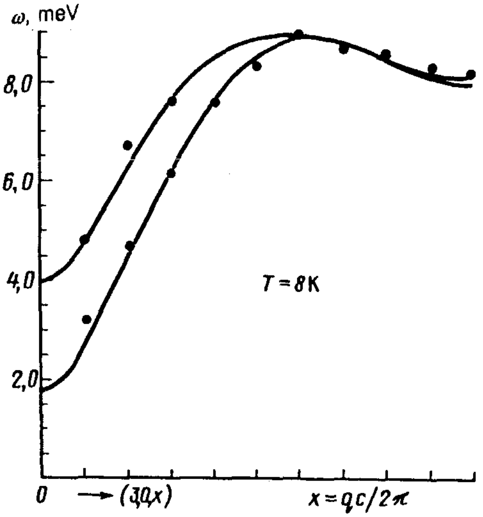

FIG. 4.

branch has a gap. The choice of cubic symmetry over tetragonal symmetry of the four-site interaction is justified by the simplification of the final equations, without losing any fundamental features from a physics point of view.

Ignoring the magnon–magnon interaction and diagonalizing the quadratic form of the Bose analog of Hamiltonian (1), we find two branches of the excitation spectrum

$$\Omega_1 = S\{(J_0^{\perp} - J_q^{\perp} + I_0^{\perp} - I_q^{\perp} + 2S^2 A_q^{-})(J_0^{\perp} + J_q^{\parallel} + I_0^{\perp} - I_q^{\parallel} + 2S^2 A_q^{+})\}^{1/2},$$

$$\Omega_2 = S\{(J_0^{\perp} - J_q^{\parallel} + I_0^{\perp} - I_q^{\parallel} + 2S^2 A_q^{-})(J_0^{\perp} + J_q^{\perp} + I_0^{\perp} - I_q^{\perp} + 2S^2 A_q^{+})\}^{1/2}, \tag{2}$$

where S is the spin, and $J_q^{\parallel}(I_q^{\parallel})$ and $J_q^{\perp}(I_q^{\perp})$ are the Fourier components of the exchange parameters which describe the interaction of a spin pair from different (same) superlattices when the moments are oriented along the tetragonal axis and at right angles to it, respectively. The functions A_q describe the effect of a four-spin exchange:

$$A_q^{\pm} = 2K^1 + 4K^1 - 4K^2 \cos(q_z \frac{c}{2}) - K^3\{\cos(q_x a) + \cos(q_y a)\} \pm \delta_q,$$

$$\delta_q = 4\{K^2 + K^2 + K^3 \cos(q_z \frac{c}{2})\} \cos(q_x a) \cos(q_y a). \tag{3}$$

Here $K^i_\perp$ and $K^i_\parallel$ ($i = 1, 2, 3$) represent the aforementioned four-spin exchange constants for the plaquettes, which are oriented in the basal plane (the subscript $\perp$), and for the vertical plaquettes (the subscript $\parallel$). The constants were normalized in such a way that each $K^i_{\parallel,\perp}$ is equal to the spin interaction energy in one plaquette with a corresponding spin configuration.

The dispersion relations (2) can be used to describe fairly well the experimental points (Fig. 4) for several reasonable sets of constants. Because of the limitations of the experimental data, an unambiguous choice of the parameters for the Hamiltonian cannot be made. A numerical analysis of the results nonetheless leads us to conclude that the excitation spectrum of the lower branch has a gap, Δ_1, since the best agreement between theory and experiment occurs in this case.

The presence of a gap, Δ_1, in the lower branch suggests that the multiple-spin exchange is important in the formation of the magnetic properties of Bi_2CuO_4. Since the magnetic subsystem of Bi_2CuO_4 is similar to that of high-T_c superconducting materials, we can assume that the multiple-spin exchange also plays an important role in high-T_c superconducting systems. Such an assumption, for example, renders irrelevant the question concerning the stability mechanism of long-range order at $T = 0$, since a four-site exchange in a square lattice with $S = 1/2$ leads to excitations with a gap, which stabilizes the magnetic structure.

The excitation energy was observed to decrease in the region of large wave vectors. The decrease occurred either because of the presence of frustrated bonds or because of the large renormalization due to the interaction of quasiparticles.[9] The latter possibility appears to be unlikely, since the gap-like behavior of the spectrum does not allow appreciable development of quantum fluctuations, which justifies the use of the noninteracting-magnon approximation in our analysis.

[1)] In preparing this paper for publication we have learned of the results of NMR studies[7] of Bi_2CuO_4, in which a gap in the lower branch, $\Delta_I = 1.7$ meV, was detected.

---

[1] J. L. Gasia-Muncz, J. Rodrigues-Carvajal, E. Sapina *et al.*, J. Phys.: Cond. Matt. **2**, 2205 (1990).

[2] R. Troc, J. Janicki, I. Filatov *et al.*, J. Phys.: Cond. Matt. **2**, 6989 (1990).

[3] E. Ong, G. Kwei, R. Robinson *et al.*, B **42**, 4255 (1990).

[4] J. Konstantinovic, G. Stanisic, M. Ain, and G. Parette, J. Phys.: Cond. Matt. **3**, 381 (1991).

[5] G. A. Petrakovskii, K. A. Sablina, A. M. Vorotynov *et al.*, Solid State Commun. **79**, 317 (1991).

[6] K. Yamada, K. Takada, S. Hosoya *et al.*, J. Phys. Soc. Jpn. **60**, 2406 (1991).

[7] M. Motokava, Abstract of Intern. Conf. on Magnetics Electr., Krasnoyarsk, 1992.

[8] E. L. Nagaev, *Magnets with complex exchange interaction*, Nauka, Moscow, 1988.

[9] V. V. Val'kov and T. A. Val'kova, Pis'ma Zh. Eksp. Teor. Fiz. **52**, 1179 (1990) [JETP Lett. **52**, 589 (1990)].

Translated by S. J. Amoretty