Antiferromagnetic structure of CuO

JETP Lett 91, 54 (11) 650-654

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(Submitted 23 April 1991; resubmitted 8 October 1991)
Pis'ma Zh. Eksp. Teor. Fiz. **54**, No. 11, 643–646 (10 December 1991)

An error is pointed out in the interpretation of the neutron-diffraction pattern and a model for the magnetic structure of copper oxide based on it, which was discussed in previous papers. A new model which agrees with the indicated neutron diffraction pattern, is proposed.

In recent years the properties of copper oxide (CuO) have been studied extensively, since it is believed that CuO can be used as a model system in research on copper-based high-T_c superconducting compounds. In copper oxide (as in the high-T_c compounds under certain condition) an antiferromagnetic order is observed in the low-temperature region at $T<213$ K (Refs. 1–4).

We believe that the model for antiferromagnetic order of CuO proposed in Ref. 2 (and the corresponding model from Ref. 4) cannot be deduced from the neutron-diffraction pattern of Ref. 2, since the wave vector of the sought magnetic structures was determined incorrectly in Ref. 2. Since the model proposed in Refs. 2 and 4 is widely used in describing the properties of copper oxide, we feel it necessary to report this error and to propose a different model, which agrees with this pattern.[2]

The crystal structure of CuO has a bcc lattice of monoclinic symmetry and is described by the space group $C2/c$ (C^6_{2h}). The lattice constants are $a = 4.6837$ Å, $b = 3.4226$ Å, $c = 5.1288$Å, and $\beta = 99.54°$, at 20 °C (Ref. 2). (The second space group is C^6_{2h} with a centered C face[5]). The copper ions occupy the $4(c)$ site (Ref. 5) with the coordinates $1(\frac{1}{4}, \frac{1}{4}, 0)$, $2(\frac{3}{4}, \frac{1}{4}, \frac{1}{2})$, $3(\frac{3}{4}, \frac{3}{4}, 0)$, $4(\frac{1}{4}, \frac{3}{4}, \frac{1}{2})$. The primitive chemical cell, in which there are two copper ions is constructed on the vectors of the shortest translations:

$$\vec{a}_1 = (0, 0, c), \quad \vec{a}_2 = (\frac{a}{2}, -\frac{b}{2}, 0), \quad \vec{a}_3 = (\frac{a}{2}, \frac{b}{2}, 0). \tag{1}$$

The translations $\vec{a}_2$ and $\vec{a}_3$ center the face C. The reciprocal lattice is also base-centered with the shortest vectors

$$\vec{b}_1 = (0, 0, \frac{2\pi}{c}), \quad \vec{b}_2 = (\frac{2\pi}{a}, -\frac{2\pi}{b}, 0), \quad \vec{b}_3 = (\frac{2\pi}{a}, \frac{2\pi}{b}, 0), \tag{2}$$

so we have $\vec{a}_i \vec{b}_j = 2\pi\delta_{ij}$.

The family of six peaks of the magnetic scattering of neutrons on the diffraction pattern[2] has an ancestral reflection $(\frac{1}{2}, 0, \frac{1}{2})$. We would like to point out that in Ref. 2 the indexing of the peaks is not done in accordance with the generally accepted standard, but in a reference frame of sides of inverse centering (rather than the ordinary centering) Bravais cell. Yang *et al.*[2] discussed this point directly in their paper, and it is easy to verify this point independently. If we follow the reference frame in Ref. 2 and use the edges of a centered reciprocal lattice,

$$\vec{B}_x = \vec{b}_2 + \vec{b}_3, \quad \vec{B}_y = \vec{b}_3 - \vec{b}_2, \quad \vec{B}_z = \vec{b}_1, \tag{3}$$

then the wave vector of the magnetic structure we are seeking should be of the form

$$\vec{k}_{new} = (\frac{1}{2}, \ 0, \ \frac{\bar{1}}{2}) = \frac{1}{2}\vec{B}_x - \frac{1}{2}\vec{B}_z. \tag{4}$$

If we add an entire reciprocal-lattice vector $\vec{b}_1$, then the vector $\vec{k}_{\text{new}}$ corresponds, in terms of the vectors $\vec{b}_i$ in (2) (in the terminology of Kovalev's handbook[6]) to a single-prong Lifshitz star $\{\vec{k}_9\}$ of the wave vector

$$\vec{k}_9 = \frac{1}{2}(\vec{b}_1 + \vec{b}_2 + \vec{b}_3). \tag{5}$$

Knowing the vector of the magnetic structure, we can determine its magnetic periods.[7] The wave vector $\vec{k}_9$ in (5) corresponds to a magnetic lattice with Bravais edges, a, b, $2c$. All the shortest translations become antitranslations in the magnetic case.

We should now determine the relative orientation of spins 1 and 2 from the primitive chemical cell and their orientation with respect to the crystallographic axes. Once we have done this, it is a simple matter to extend the spin configuration which arises within a primitive cell to the entire structure, making use of the translation

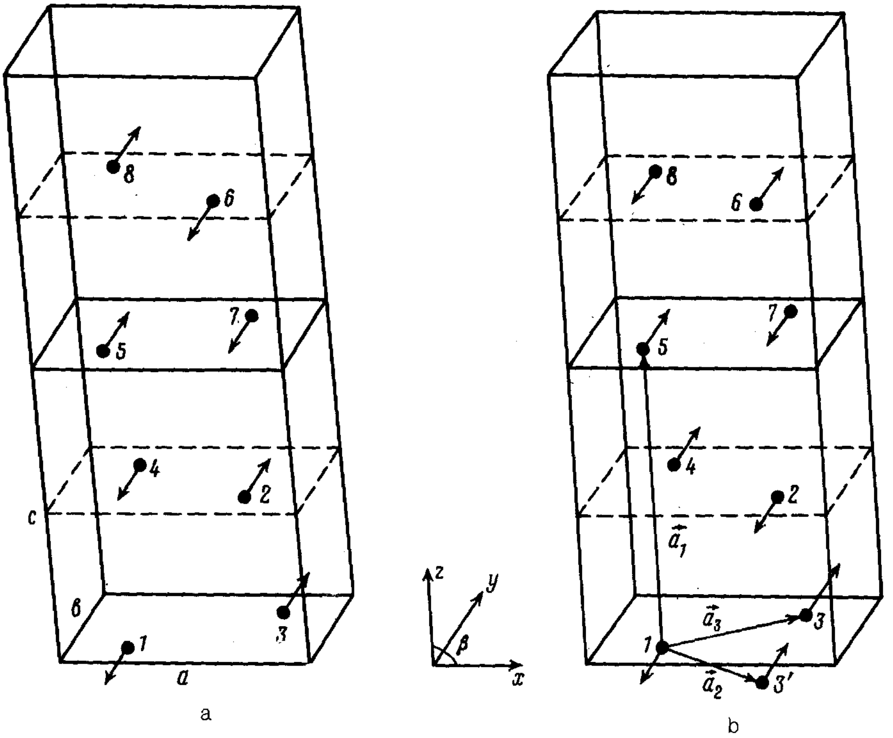

FIG. 1. Magnetic Bravais cells. a—For structure *A* ($\vec S_1\uparrow\downarrow\vec S_2\|\vec y$; b—for structure B ($\vec S_1\uparrow\uparrow\vec S_2\|\vec y$).

properties which we have already found. The necessary spin configuration is found by fitting the calculated intensities of the magnetic-scattering peaks to the intensities of the observed peaks.[7] The best agreement with a neutron diffraction pattern from Ref. 2 is found in the case $\vec S_1\|\vec S_2\|\vec y$. In this case the calculated intensities with $\vec S_1\uparrow\downarrow\vec S_2$ (structure *A*) and $\vec S_1\uparrow\uparrow\vec S_2$ (structure *B*) are completely the same.

We can thus propose two (equally valid) collinear antiferromagnetic structures for copper oxide (Fig. 1), which have identical translation properties.

It is not difficult to verify that the following wave vector corresponds to the antiferromagnetic structure proposed in Refs. 2–4, with the edges of a magnetic Bravais cell $2a$, b, $2c$:

$$\vec k_0 = -\frac{1}{2}\vec b_1 + \frac{1}{4}(\vec b_2 + \vec b_3) = \frac{1}{4}\vec B_x - \frac{1}{2}\vec B_z. \qquad (6)$$

Corresponding to a vector $\vec k_0$ of this sort on the diffraction pattern there should be a

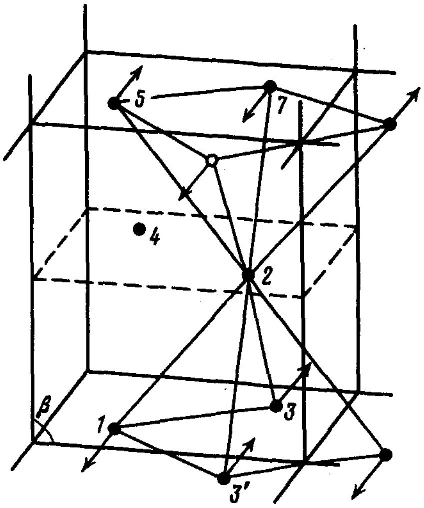

FIG. 2. Origin of the two-dimensional (2D) nature of the spin correlations in copper oxide.

family of peaks with an "ancestral" reflection ($\frac{1}{4}$, 0, $\frac{\bar{1}}{2}$). However, there is no such family on the diffraction pattern. We believe that the error which we have discovered results from neglecting the difference between the recording of the wave vector of the structure in the shortest translations $\vec{b}_1$ in (2) and in the edges of a Bravais cell, (3). This difference is important for centered cells.

From Fig. 2 we draw the conclusion that the spin correlations are of a two-dimensional (2D) nature (in the mean-field approximation). The molecular field induced at ion 2 by the spins of the neighboring layers is zero. Each of the magnetic bonds (indirect exchange through oxygen ions, which are not shown in this figure) is cancelled by its antipode. This aspect of the interlayer interaction is characteristic of both structure *A* and structure *B*. The direction of the spin $\vec{S}_2$ (along or opposite the $\vec{y}$ axis) is not regulated by the spins of the neighboring layers and is not shown in this figure.

Analysis of the exchange energy of copper oxide shows that structures *A* and *B*, having the same energy, can form a domain structure. Yet another conclusion, which emerges from an analysis of the anisotropic part of the energy, is that a slightly noncollinear "cross-shaped" state can occur in copper oxide.[8] In this case we would expect a structure of type *A* with an admixture of structure *B* (or a structure of type *B* with an admixture of *A*) to arise. A cross-shaped structure can arise by virtue of Dzyaloshinskiĭ terms in the energy. A detailed analysis of the magnetic energy will be reported in a separate paper.

We wish to thank T. I. Arbuzov and A. S. Moskvin for useful discussions.

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Translated by D. Parsons