Two-magnon light scattering in Bi_2CuO_4

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We present a calculation of the intensities for two-magnon Raman scattering in Bi_2CuO_4. The calculation is done using spin-wave theory, including magnon-magnon interaction. Obtained results are compared with polarized Raman-scattering spectra. The values of the exchange constants and orientation of the magnetic moments are determined. [S0163-1829(96)07026-9]

I. INTRODUCTION

The interest for the study of light scattering by spin waves in Cu-O based material has been initiated since the discovery of the high-T_c materials. This is based on the fact that almost all isolating phases of Cu-O based high-temperature superconductors exhibit antiferromagnetic ordering. The investigation of light scattering in these materials, besides phonon excitations, has been focused on two-magnon scattering since it is much more intense than the one-magnon scattering process. From these studies the nature of the magnetic interaction was analyzed. For a review, see Ref. 1.

The properties of Bi_2CuO_4 have been highly investigated in the last several years. This oxide attracts attention because of its interesting crystal structure and magnetic properties. The crystal structure of Bi_2CuO_4 is tetragonal with isolated CuO_4 square-planar units of Cu^{2+} ions that are stacked on the top of each other in a staggered manner along the c axis,[2] Fig. 1. The phonon and magnon excitations in this material, studied using IR and Raman spectroscopies, were subjects of our interest.[3,4] The inelastic neutron-scattering experiments,[5,6] show the existence of the three-dimensional antiferromagnetic ordering below $T=45$ K. The magnetic ordering is confirmed with an antiferromagnetic resonance experiment as well.[7] Furthermore, the spin waves were analyzed using Raman[8] and neutron[9]-scattering experiments. However, different experimental techniques gave inconsistent results concerning magnetic moments orientation. Namely, in Refs. 6, 7, and 9 it is stated that the magnetic moments lay in the xy plane, while a recent Raman-scattering magnon selection rules study[10] suggests the z axis orientation of the magnetic moments. Such disagreements provoke the reexamination of the dispersion curves analysis,[9,11] since the dispersion curve is incorporated in the two-magnon scattering calculation.

In this study, the calculation of the intensities for two-magnon Raman scattering at $T=0$ in Bi_2CuO_4 is presented using spin-wave theory that includes the magnon-magnon interaction. The values of the exchange integrals are determined from the best agreement between experimental data and theoretical calculation and the z-axis magnetic moment orientation is confirmed.

II. THEORETICAL MODEL

In order to calculate the two-magnon Raman-scattering intensities we started with the anisotropic exchange antiferromagnetic Hamiltonian:

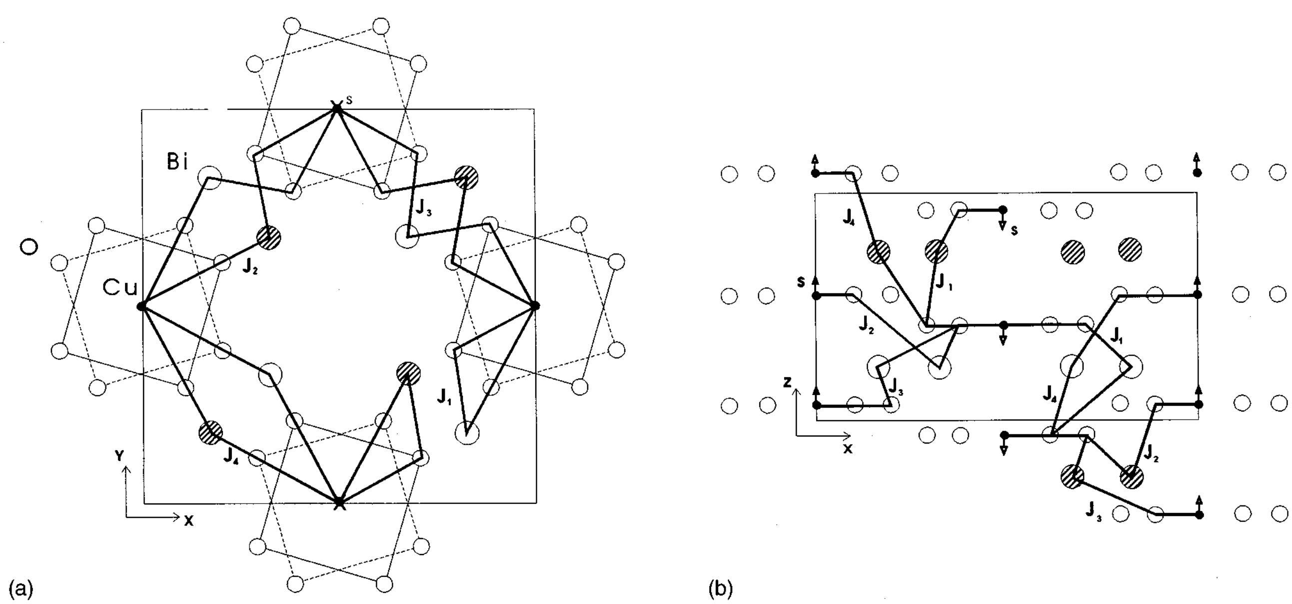

FIG. 1. The crystal structure of Bi_2CuO_4 shown in (a) (001) and (b) (010) projections. The thick lines represent the four antiferromagnetic superexchange pathways.

$$H=-2\sum_{(i,j)\text{pair}}(J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j+D_{ij}S_i^zS_j^z), \quad (1)$$

where the summation goes over two sublattices and J_{ij}, D_{ij} represents the isotropic and anisotropic part of the exchange interaction between spins $\mathbf{S}_i$ and $\mathbf{S}_j$. This Hamiltonian is formally identical with one presented in Ref. 11 but with the axis orientations adapted to a crystal structure given in Fig. 1.

The first step was the calculation of the spin-wave dispersion. After applying the Holstein-Primakoff transformation[12] (we adopt the z-axis magnetic moment orientation) and performing the standard diagonalization procedure,[11] we obtain the eigenvalues with the form

$$\hbar\omega(\mathbf{k})=2S\sqrt{[\mu+J_{11}(\mathbf{k})]^2-[J_{12}(\mathbf{k})]^2}, \quad (2)$$

where

$$\mu=[J_{12}(0)+D_{12}(0)-J_{11}(0)-D_{11}(0)],$$

$$J_{11}(\mathbf{k})=2J_1\cos(\mathbf{k}\cdot\mathbf{c}),$$

$$J_{12}(\mathbf{k})$$

$$=4\cos\left(\frac{\mathbf{k}\cdot\mathbf{a}}{2}\right)\cos\left(\frac{\mathbf{k}\cdot\mathbf{b}}{2}\right)$$

$$\times\sqrt{[J_2+(J_3+J_4)\cos(\mathbf{k}\cdot\mathbf{c})]^2+[(J_4-J_3)\sin(\mathbf{k}\cdot\mathbf{c})]^2},$$

$$D_{11}(0)=2D,\quad D_{12}(0)=4(D_2+D_3+D_4). \quad (3)$$

We underline that the case of spin orientation along the z axis brings the spin-wave dispersion degeneracy at the symmetry points at the edge of the Brillouin zone, and that will be discussed in Sec. III. This fact is also confirmed simply using the symmetry consideration.[10]

The second step for evaluation of the two-magnon Raman intensities at $T=0$ involves the calculation of the imaginary part of the retarded Green function:

$$G(\delta,\delta')=\langle\langle P(\delta);P(\delta')\rangle\rangle_\omega, \quad (4)$$

where δ and δ' are vectors connecting nearest neighbors. It was shown[13,14] that the dominant contribution to the two-magnon scattering comes from the part of $P(\delta)$ defined as

$$P_{|0+\rangle}(\delta)=\sum_{\mathbf{r}}(S_{\mathbf{r}}^xS_{\mathbf{r}+\delta}^x+S_{\mathbf{r}}^yS_{\mathbf{r}+\delta}^y). \quad (5)$$

Following the standard procedure,[14] the two-magnon cross section is proportional to

$$\mathrm{Im}G_0=\mathrm{Im}\left[\frac{1}{N}\sum_k\frac{\Phi(\mathbf{k})}{\omega^2-4\omega^2(\mathbf{k})}\right]$$

$$=\frac{\pi}{4N}\sum_k\frac{\Phi(\mathbf{k})}{\omega(\mathbf{k})}\delta(\omega-2\omega(\mathbf{k})), \quad (6)$$

where $\Phi(\mathbf{k})$ are weighting functions for different polarization configurations. Assuming the existence of the magnon-magnon interaction, the two-magnon cross section becomes proportional to

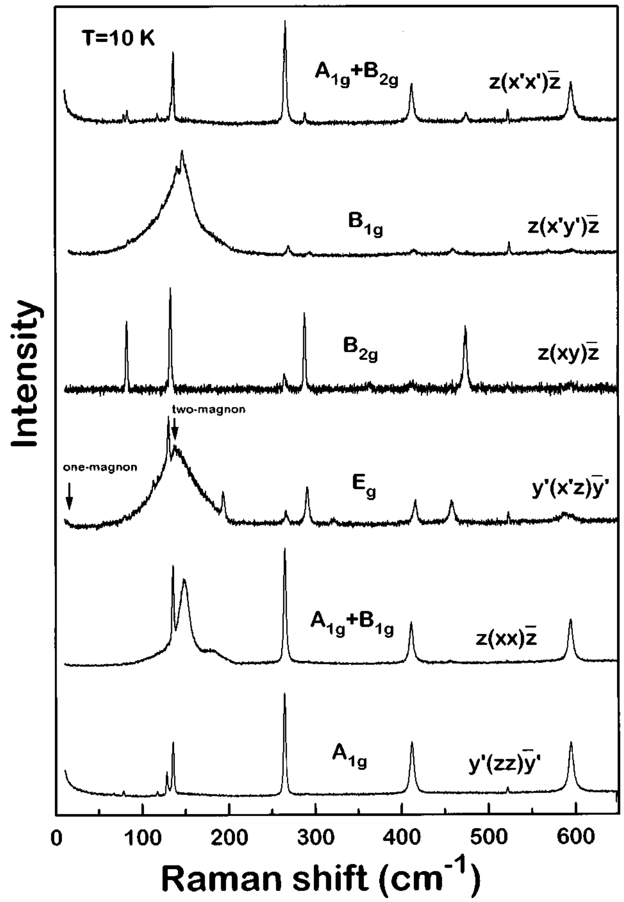

FIG. 2. The Raman-scattering spectra of the Bi_2CuO_4 single crystal at $T=10$ K in the spectral region from 10 to 650 cm^{-1}, for six different polarization configurations.

$$I_{\text{two-magnon}}\sim\mathrm{Im}\left[\frac{G_0(\omega)}{1+bG_0}\right], \quad (7)$$

where parameter b describes the strength of magnon-magnon interaction. Both $\Phi(\mathbf{k})$ and b are symmetry-dependent quantities and their evaluation requires knowledge of the magnetic structure in the antiferromagnetically ordered phase. According to the conclusion given in Ref. 10, we use the unitary subgroup D_4^2 of the magnetic space group $P4/n'c'c'$ to find the projectors of 12×12 matrices that are formed considering 12 nearest neighbors. From such a study[14] the three representations and corresponding symmetry factors are picked out:

$$\Phi(\mathbf{k})=\begin{cases}4\cos^2(k_xa)\cos^2(k_ya) & \text{for } \Gamma_1\\ 4\sin^2(k_xa)\sin^2(k_ya) & \text{for } \Gamma_3\\ 4\sin^2(k_xa)\cos^2(k_ya) & \text{for } \Gamma_5\end{cases} \quad (8)$$

with b approximately ($|J_4|\gg|J_2|,|J_3|,|J_1|$) given by

$$b=J_4(4\mu-J_4)\quad \text{for } \Gamma_3 \text{ and } \Gamma_5. \quad (9)$$

III. RESULTS AND DISCUSSION

The Raman-scattering spectra of the Bi_2CuO_4 single crystal at a temperature of $T=10$ K, for six different polarized

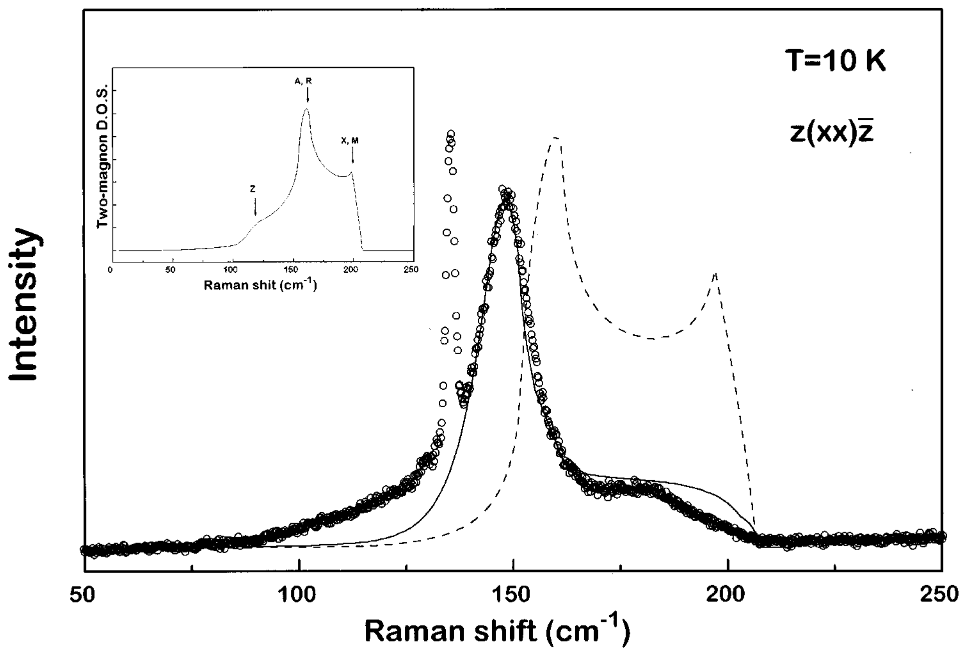

FIG. 3. The intensity of the two-magnon light scattering of Bi_2CuO_4 in the $z(xx)\overline{z}$ polarization configuration. The open circles represent the experiment, the solid line is the calculated spectrum obtained using Eq. (7), and dashed line represents the noninteracting spin-wave theory. Inset: The two-magnon density of states.

configurations, are presented in Fig. 2. The experimental details and the assignations of the observed modes are already done.[4,8] We remind the reader here that the wide dominant feature observed in $z(x'y')\overline{z}$,$y'(x'z)\overline{y'}$ and $z(xx)\overline{z}$ belongs to the two-magnon scattering process.

The two-magnon intensities in Bi_2CuO_4, obtained using the numerical calculation of Eq. (7), including numerical integration in order to evaluate the **k** summation over the Brillouin zone, are given in Figs. 3 and 4 for both Γ_3 and Γ_5 polarized configurations, respectively. According to the experiment the Γ_1 polarized configuration, although allowed by selection rules, gives no noticeable two-magnon scattering since the dominant contribution of this type comes from the center of the Brillouin zone (low two-magnon density of states, see inset of Fig. 4).

The values of the exchange integrals are given in Table I. These values are obtained from a fit of two-magnon Raman-scattering intensities[8] and the neutron dispersion measurements[9] at the same time. The value of $b=98.3$ $(\mathrm{meV})^2$ is obtained for the b parameter which represents the strength of the magnon-magnon interaction.

(xx) polarization: In the case of the (xx) polarization configuration, according to Raman-scattering tensors,[10] the symmetry of the two-magnon excitation is described with a Γ_3 representation of the D_4^2 group. The calculation of the two-magnon intensities is done following Eq. (7) with the corresponding $\mathbf{\Phi}(\mathbf{k})$ function and b parameter as in Eqs. (8) and (9). The results are presented in Fig. 3, where the solid line represents the calculated intensity and the circles are experimental data. The agreement between theory and ex-

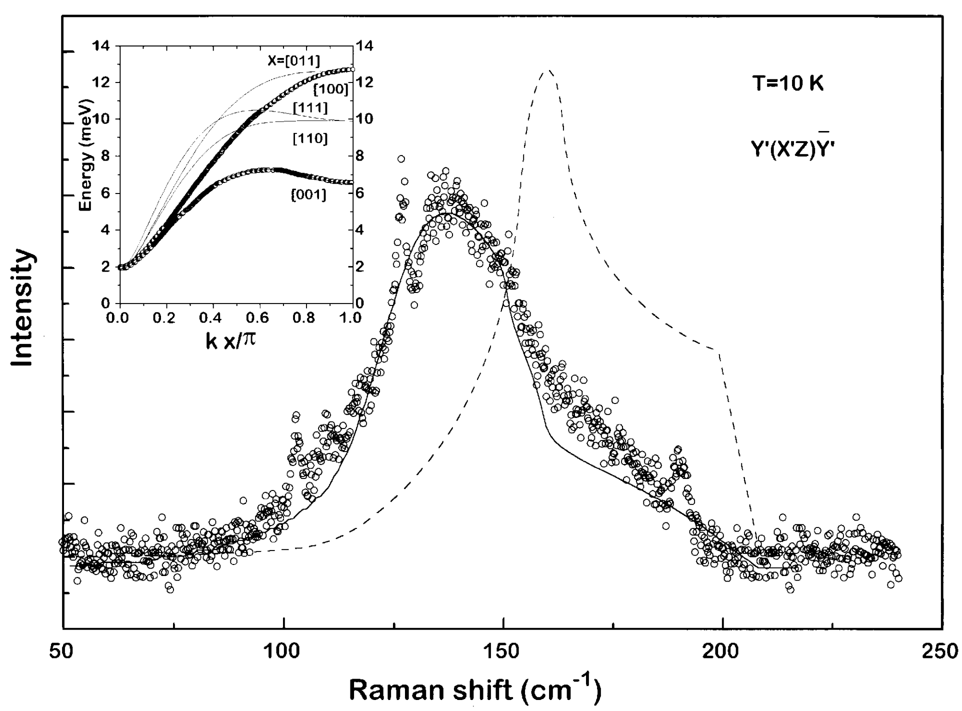

FIG. 4. The intensity of the two-magnon light-scattering spectrum of Bi_2CuO_4 in the $y'(x'z)\overline{y'}$ polarization configuration. Inset: The magnon dispersion curves. The circles represent the neutron-scattering experiments (Ref. 8) and the solid lines are obtained using Eq. (2). **x** denotes the crystallographic axes a, b, and c.

TABLE I. The values of the exchange integrals.

$J_{i,j}$ (meV)	$D_{i,j}$ (meV)
$J_1 = -0.68$	$D_1 = 0$
$J_2 = -0.63$	$D_2 = 0$
$J_3 = -0.19$	$D_3 = 0$
$J_4 = -2.3$	$D_4 = -0.035$

periment is both qualitative and quantitative. The small disagreement between theory and experiment in the spectral range between 80 and 130 cm^{-1} is probably due to existence of a strong phonon mode at 135 cm^{-1} and the fact that our calculation is done for $T=0$ K, while the experimental results are obtained at $T=0$ K. Besides that, an interaction between next-nearest neighbors is not included in the calculation.

In the inset of Fig. 3 we show the calculation of the two-magnon density of states. As mentioned above the z-axis orientation of the magnetic moments bring the spin-wave energy degeneration at the edge of the Brillouin zone. In the case of x-axis orientation of the magnetic moments this degeneration is removed.[11]

In order to show the effect of the magnon-magnon interaction we also present the results obtained using noninteractive spin-wave theory (dashed line in Fig. 3). The magnon-magnon interaction shifts the two-magnon scattering peak position for about 12 cm^{-1}.

$(x'z)$ polarization: The comparison between theory and experiment is done for a $(x'z)$ polarization configuration in Fig. 4. In this case the symmetry of the two-magnon scattering process belongs to a Γ_5 representation. Again we obtained very good agreement between experimental and calculated data. For this polarization the phonon intensities are much smaller then in the (xx) polarization and the agreement between theory and experiment is better. At the inset of Fig. 4 we reproduced the neutron dispersion curve measurement from Ref. 9 together with the dispersion curve calculation based on Eq. (2). As can be seen from this inset our calculation fit the experimental curves very well.

IV. CONCLUSION

In conclusion, the two-magnon scattering spectra of Bi_2CuO_4 can be completely described using interacting spin-wave theory, although the linear approximation is not rigorously applicable for the systems with spin = 1/2. This study gave the values of the exchange integrals by simultaneous fit of the neutron dispersion and Raman-scattering data. This procedure was necessary since the fit of the neutron-scattering measurements of dispersion branches along [100] and [001] does not give a unique solution for the exchange integrals. The values of the exchange integrals we obtained are close to the values obtained by the other authors[9,11] but the essential degeneracy of the magnon branches is not reproduced by their results. Finally, this analysis of the magnetic interaction in Bi_2CuO_4 confirmed the z-axis orientation of the magnetic moments.

ACKNOWLEDGMENT

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