Effect of high hydrostatic pressure on the exchange interaction in Eu_2CuO_4 single crystals

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The pressure dependence of the exchange integral J in the CuO_2 plane has been determined over the pressure range 0–410 kbar. The value of J was found from the spectra of two-magnon Raman scattering in the antiferromagnet Eu_2CuO_4. The relative change in the exchange interaction with decreasing lattice constant is considerably smaller than in classical antiferromagnets.

Most of the high-T_c superconducting materials share the structural feature of CuO_2 planes. It has now been established that the insulating phases of these superconductors are magnetically ordered. It has been found by neutron scattering[1] that these phases are antiferromagnetic with a spin $S = 1/2$, which is localized at the copper atoms. These phases have comparatively high Néel temperatures T_N, which may be above room temperature. In the cuprate planes, on the other hand, there are strong antiferromagnetic correlations at temperatures well above the 3D ordering temperature T_N.

Interest in research on the magnetic properties of the high-T_c materials derives from the importance from determining the role played by magnetic interactions in the mechanism for high-T_c superconductivity.[2,3] Raman scattering is an effective method for studying antiferromagnetic ordering in cuprate planes. Numerous studies have demonstrated that the Raman spectra of insulating crystals of La_2CuO_4 and their analogs[4–6] exhibit a broad band of B_{1g} symmetry, with a peak near 3000 cm^{-1}. This band corresponds to a scattering of light accompanied by the creation of two magnons. A corresponding band is observed in the insulating phase of $YBa_2Cu_3O_{6+x}$ crystals.[7,8] The two-magnon band is not observed in the Raman spectrum of the superconducting phases of the same crystals or in the bismuth- and thallium-based high-T_c superconductors. On the other hand, one does observe a broad, essentially structureless background of electron origin up to frequencies > 4000 cm^{-1}.

According to the theory[9,10] describing the position and shape of the two-magnon band in the Raman spectrum of a 2D antiferromagnet with a coordination number z and a spin S, the position of the maximum, ω_{max}, is related to the exchange integral[1)] J by $\omega_{max} \approx 1.37$ JSz. For the case of the cuprate plane of a high-T_c compound, this relation becomes ($S = 1/2$, $z = 4$)

$$\omega_{max} \approx 2.74J. \quad (1)$$

The value of J is determined by the extent to which the wave functions of neighboring

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Cu and O atoms overlap (*p–d* overlap) in the CuO_2 plane. This overlap of wave functions in turn depends on the distance between atoms. There is accordingly considerable interest in experiments at a high hydrostatic pressure. Such experiments would make it possible to directly determine the dependence of the exchange integral on the distance between neighboring atoms in the CuO_2 plane.

In this letter we are reporting a study of the Raman-scattering spectra of Eu_2CuO_4 single crystals, which are structural analogs of La_2CuO_4, over the pressure range from 0 to 410 kbar. A diamond-anvil chamber was used to produce the high pressure.[11] The test samples were single-crystal wafers with a large *ab* plane, with dimensions $\approx 40 \times 40 \times 10$ μm. The samples were placed in an aperture $\simeq 100$ μm in diameter in a rhenium gasket. Helium was used as a pressure-transmission medium. The use of helium ensured that the compression would remain hydrostatic over the pressure range studied.[12] The pressure in the chamber was determined from the shift of the R_1 fluorescence line of ruby.[12] The measurements were carried out with the help of an Ar^+ laser with an excitation wavelength $\lambda = 4880$ Å on a DILOR XY triple Raman spectrometer with a microscope attachment.

Figure 1 shows experimental Raman spectra of Eu_2CuO_4 at various pressures. As the pressure is raised, the peak of the two-magnon band undergoes a noticeable shift toward a higher energy. At $P = 410$ kbar, the shift amounts to $\simeq 1000$ cm^{-1}. Figure 2

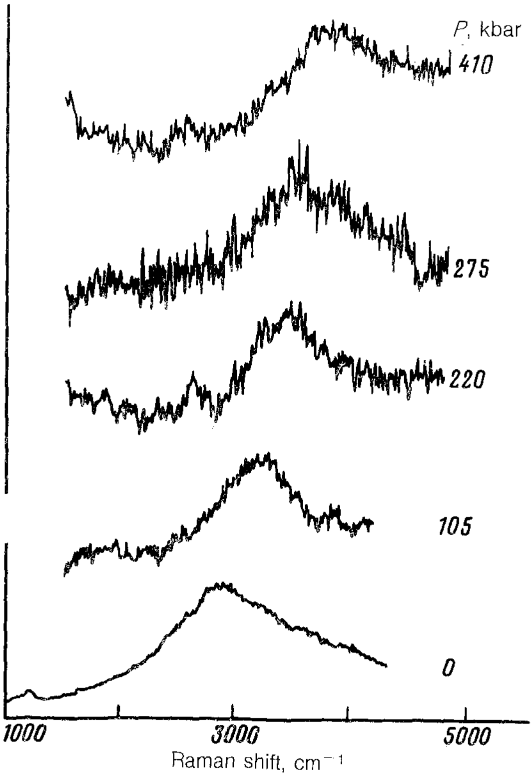

FIG. 1. Raman scattering spectra of a Eu_2CuO_4 single crystal in the region of two-magnon scattering at various pressures *P*.

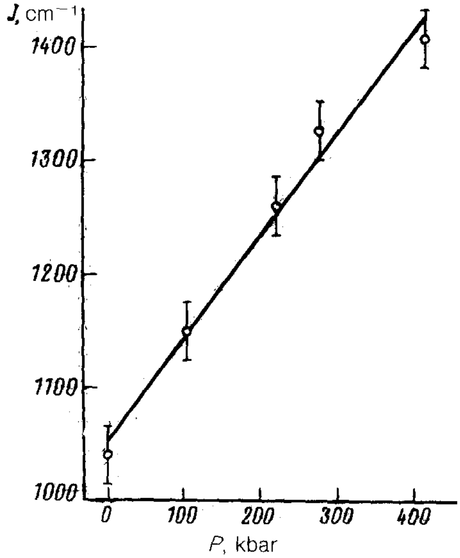

FIG. 2. Exchange integral in Eu_2CuO_4 crystals versus the pressure P.

shows the exchange integral J as a function of the pressure according to (1). This plot is essentially linear over the pressure range from 0 to 410 kbar.

As the pressure is raised from 0 to 400 kbar, the exchange integral changes by ~30%. To estimate the change in the lattice constant, we used data from a study[13] of $(La_{0.9}Sr_{0.1})_2CuO_{4-y}$ crystals over the pressure range $P = 0$–70 kbar. The compressibility according to those measurements is $(2.5 \pm 0.4) \times 10^{-4}$ kbar^{-1}. In addition, measurements[4] of the lattice constant of $YBa_2Cu_3O_{6+x}$ crystals at pressures up to 200 kbar show that the values of the compressibility are approximately the same in the *ab* plane. The change in the distance between the Cu and O atoms in the *ab* plane of Eu_2CuO_4 at a pressure of 400 kbar is thus ~10%.

The dependence of the exchange integral on the distance between atoms, r, is determined by the overlap of the corresponding wave functions. In general, this dependence may be quite complex. Understandably, it is thus not possible to reconstruct this dependence accurately from experimental data when the change in r is small. However, to describe the rate of change of the exchange integral upon a change in the distance between atoms, one can follow the customary approach of approximating $J(r)$ by a power law.[6,15] For example, a detailed analysis[15] of numerous experimental data and numerical calculations has revealed $J \propto r^{-12}$ for the case of a 180° superexchange interaction in classical antiferromagnets (e.g., K_2NiF_4 and K_2MnF_4).

In the case of Eu_2CuO_4, this dependence is considerably weaker, $J \propto r^{-n}$, with $n = 3 \pm 0.5$, according to the experimental data. This behavior of the exchange integral as a function of the distance may stem from an initially pronounced overlap of the wave functions, with the result that the interaction is not greatly intensified when the Cu and O atoms move closely together. Indeed, evidence for a pronounced overlap of the wave functions comes from the values of the exchange interaction in the high-T_c materials ($J \simeq 1000$ cm^{-1}).[4-8] These values are considerably higher than the typical values in the 2D antiferromagnets which have been studied previously.[5] Still, such arguments are only qualitative. Reaching a more comprehensive understanding of the nature of the exchange interaction in the Cu–O–Cu system and finding a quantitative description of this interaction will require a corresponding theoretical analysis and corresponding calculations based on models.

[1)] In determining J we ignored the renormalization of this quantity for quantum fluctuations in a 2D antiferromagnet with a spin $S = 1/2$ (Ref. 10).

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