Observation of magnetic resonance in Pr_2CuO_4 and Nd_2CuO_4

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Absorption lines which are intense and broad ($\Delta H \approx 0.5T$) have been observed in the wavelength region 5–1.5 mm in tetragonal Pr_2CuO_4 and Nd_2CuO_4 single crystals at 2–10 K in a magnetic field directed along the tetragonal axis. These lines appear to be associated with resonance excitations of copper ions, whose state is strongly affected by the rare-earth ions.

Previous attempts to observe a magnetic resonance in single crystals of the La_2CuO_4 type have not met with success. Certain reports[1] have not been verified by subsequent studies. According to the theoretical understanding, a spin ordering in the tetragonal phase of such crystals should be antiferromagnetic, with the main antiferromagnetism vector directed along the *a* axis in the plane of a layer. It follows that there should be an energy gap in the spectrum of the magnetic resonance and that the resonant frequencies should be most sensitive to an external magnetic field when this field is oriented along the *a* axis (Ref. 2, for example). We have now established that in the temperature interval 2–10 K magnetic resonance lines are observed over a wide frequency range in an external field directed along the tetragonal axis, the *c* axis.

The spectrometer used in these experiments had a test cell similar to that described in Ref. 3 and was part of a computer-controlled data acquisition and processing system with a video display. The errors in the measurement of the wavelength and the magnetic field strength were 0.2% and 0.5%, respectively, and were much smaller than the experimental error set by the width of the absorption lines being measured.

The samples were platelets with dimensions of $3 \times 3 \times 0.5$ mm, made of single crystals grown by controlled crystallization from the surface of a molten solution.[4] Their quality and tetragonal structure were monitored at room temperature by x-ray diffraction. A linear dependence (linear within the experimental error) of the resonant frequencies was observed over the region 2–6 cm^{-1} in a pulsed external magnetic field directed parallel to the *c* axis in the Pr_2CuO_4 crystals; these results are shown in Fig. 1. Figure 2 shows a spectrogram of the absorption at the frequency 3.55 cm^{-1}. The width of the lines is 0.5–0.6 *T* over the entire region, and the integral intensity is comparable to the intensity of the magnetic resonances in other magnetically ordered crystals.

A similar absorption, with completely identical parameters, was observed in the Nd_2CuO_4 crystals. In addition, some fainter and extremely narrow absorption lines

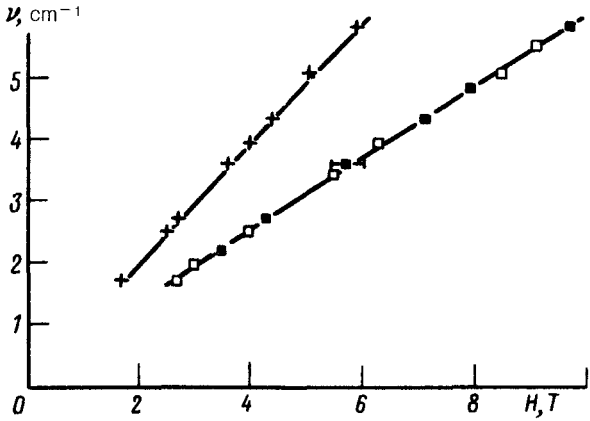

FIG. 1. Field dependence of the frequency of the magnetic resonance ■—Cu excitations in Pr_2CuO_4; □—Cu excitations in Nd_2CuO_4; + —Nd excitations in Nd_2CuO_4.

were observed in weaker magnetic fields. The field dependence of the frequency of these lines is also shown in Fig. 1.

In an external magnetic field up to 15 T applied in the *ab* plane (in two mutually perpendicular directions), we observed only faint lines in the frequency region 2–15 cm^{-1}. The field dependence of the frequency of these lines did not depend on the orientation of the external field. There were no intense absorption lines in this case.

Since identical absorption lines are present in Pr_2CuO_4 and Nd_2CuO_4, these lines can be identified as a resonance associated with copper ions. The faint lines additionally observed in Nd_2CuO_4 probably correspond to the excitation of paramagnetic Nd ions.

The observed behavior of the frequencies of the magnetic resonance of the copper ions is exceedingly unusual and does not correspond to the case in which the antiferromagnetism vector is directed along the *a* axis.

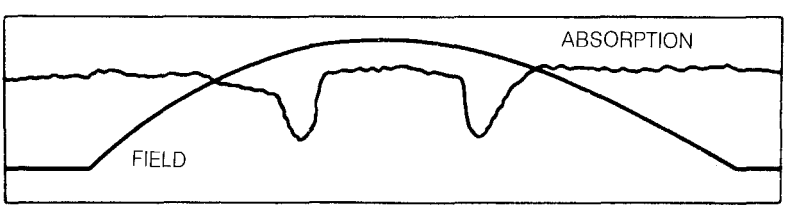

FIG. 2. Absorption spectrogram in Pd_2CuO_4 at a frequency of 3.55 cm^{-1}. This line was recorded at the leading edge and trailing edge of the magnetic field pulse.

Measurements of the temperature dependence of the absorption spectra in these crystals revealed that heating them from 2 K to 8–10 K has no substantial effect on the spectra. As the temperature is raised further, the frequencies and widths of all the lines remain the same, while the absolute intensity decreases. At 20–25 K, the lines disappear. This temperature dependence may mean that the rare-earth subsystem is participating in the formation of magnetic excitations of copper ions.

In our opinion, the least contradictory explanation of these results comes from the assumption that a magnetically ordered state of the rare-earth ions and of the copper ions exists in these crystals in the temperature region 2–10 K. According to this interpretation, these two ion species have strong effect on each other. The nature of this state and the reasons for its appearance require further research.

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