Magnetic phase transitions in Gd_2CuO_4

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We have made neutron-diffraction, magnetization, and antiferromagnetic-resonance investigations of Gd_2CuO_4 single crystals. Neutron-diffraction experiments on ^{158}Gd-enriched Gd_2CuO_4 single crystals show that the gadolinium atoms order antiferromagnetically at $T_N=6.4$ K with the propagation vector $\mathbf{k}=(0,0,0)$. The two Gd atoms of the primitive unit cell related by the center of symmetry are oppositely oriented. The antiferromagnetic-resonance spectra indicate that Gd_2CuO_4 belongs to the class of "easy-plane" antiferromagnets and show the presence of a second-order reorientation phase transition at $H_c=0.88$ T ($\mathbf{H}\|[100]$, $T=1.8$ K). The magnetization measurements confirm the presence of this reorientation transition and also show a complete ferromagnetic alignment of Gd moments at $H=11$ T and $T=1.5$ K.

I. INTRODUCTION

The magnetic properties of the high-temperature superconductors have been investigated in great detail following theoretical suggestions that these might play an important role in the underlying superconducting mechanism.[1,2] Fluctuating two-dimensional antiferromagnetic spin correlations in CuO_2 planes have been reported to exist up to very high temperatures in these compounds and persist even in the samples which are doped to become superconductors and in which the Néel temperature is reduced to zero.[2,3] Until the recent discovery of a series of the form $R_{2-x}Ce_xCuO_4$ (R=Pr, Nd, Sm, or Eu) and also with Ce replaced by Th,[4-7] all the known cuprate superconductors were hole doped. This new series is particularly interesting because electrons, rather than holes, in the CuO_2 planes are suggested as the charge carriers involved in the high-T_c superconductivity. The Cu magnetic moments in R_2CuO_4 order at temperatures in the range 250–275 K.[8-13] At lower temperatures, spin-reorientation transitions have been reported in Nd_2CuO_4 (Refs. 5–10) and magnetic ordering of the rare-earth moments in Nd_2CuO_4 and Pr_2CuO_4.[8,13] Recently, Thompson *et al.*[14] have made a detailed study of the magnetic properties of Gd_2CuO_4 single crystals and have reported three magnetic ordering temperatures (6.5, 20, and 270 K) in a magnetic field of 0.1 T. Out of the R_2CuO_4 family, Gd_2CuO_4 seems to be especially interesting because of the following distinct properties.

(1) Although Gd_2CuO_4 is as easily doped with Ce or Th as the other members of the R_2CuO_4 (R=Pr, Nd, Sm, and Eu) family, it does not, as they do, become superconducting. There have been several suggestions for the absence of the superconductivity in this compound,[15] but there is no clear explanation for this as yet.

(2) The antiferromagnetic ordering of the CuO_2 layers is accompanied by the appearance of a weak ferromagnetic moment, whose magnitude is inversely proportional to the temperature.[14,16,17] The existence of the weak ferromagnetic moment is forbidden in the crystal symmetry $I4/mmm$ of Gd_2CuO_4.[18] The weak ferromagnetism may be due to the presence of the impurity, but this should be proved by new experiments.

(3) In contrast to the La_2CuO_4 and $YBa_2Cu_3O_{6+x}$, the doped and the undoped R_2CuO_4 compounds preserve tetragonal symmetry down to the lowest temperature investigated. Because of the tetragonal symmetry, the possibility of a noncollinear double-**k** structure exists in this

system and this possibility should be investigated by neutron-diffraction studies under magnetic field or uniaxial stress.

We have studied the magnetic properties of the Gd_2CuO_4 single crystals by magnetization and antiferromagnetic resonance. Because of the very large absorption cross section of natural Gd for thermal neutrons, we have grown single crystals of Gd_2CuO_4 enriched with the ^{158}Gd isotope. Using these crystals we have studied the magnetic structure of Gd_2CuO_4 by neutron diffraction. We have shown that the Gd layers in the $a-b$ planes are ferromagnetic and are stacked antiferromagnetically along the c axis.

We have organized this paper in the following way: In Sec. II we describe the experimental procedures. In Sec. III results of the magnetization and antiferromagnetic resonance investigations are described. Section IV gives the results of neutron-diffraction investigations. Section V is devoted to the discussion of the experimental results. Finally, in Sec. VI, we summarize the conclusions obtained from the present investigations.

II. EXPERIMENT

Single crystals of Gd_2CuO_4 were grown by the flux method using CuO as the flux material. Magnetic measurements were made using a conventional ac susceptometer in the frequency range 20–1000 Hz and in magnetic fields up to 8 T. The value of the ac field in the excitation coil was in the range 0.001–0.01 T. High-field magnetization measurements were made using the extraction method in a superconducting cryomagnet of the Laboratoire Louis Néel[19] which is capable of producing a maximum magnetic field of 16 T.

Antiferromagnetic resonance (AFMR) experiments were performed in the frequency range 15–37 GHz. The sample was in a cavity of the appropriate frequency range and a reflecting videospectrometer circuit was used to register the absorption signal. The sample could be rotated in the vertical plane inside the cavity. The cavity together with the sample was placed in a superconducting cryomagnet capable of generating a maximum magnetic field of 8 T.

Neutron-diffraction investigations were made using the diffractometer D15 of the Institut Laue-Langevin. The plate-shaped crystal ($4\times4\times0.4$ mm^3) was fixed to the sample stick of a conventional helium "orange" cryostat with the crystallographic [110] axis parallel to the ω axis of the diffractometer. The measurements were made with a calibrated wavelength of 1.176(1) Å and a small, two-dimensional, position-sensitive detector. The lattice constants were determined from the angles of 15 centered reflections and were found to be $a=3.892(2)$ Å and $c=11.863(8)$ Å at 2.5 K.

III. MAGNETIZATION AND RESONANCE INVESTIGATIONS

The magnetic field variation of the real part of the differential magnetic susceptibility, dM/dH was measured in the temperature range 1.8–6.5 K with magnetic field applied parallel to [100]. At $T=1.8$ K and $H_c=0.88$ T, an anomaly is observed in the field variation of dM/dH which is shown in Fig. 1(a). This anomaly is observed at all temperatures below T_N but the magnitude of the field at which it occurs shifts to lower field at higher temperatures. This result indicates the existence of a second- order spin-orientation transition in Gd_2CuO_4 for magnetic fields applied parallel to [100].

Figure 1(b) shows the field dependence of the resonance frequency for magnetic fields parallel to [100] and [110]. A strong softening of the antiferromagnetic resonance frequency is observed when the magnetic field is applied parallel to [100], again suggesting a second-order phase transition. For $\mathbf{H}\|$ [110], the magnetic field dependence of the resonance frequency follows the simple quadratic law: $\omega_1^2=\omega_{10}^2=\gamma^2H^2$, which is typical of the oscillation of a spin system without degeneracy and with no ferromagnetic moment.

Figure 2 shows the magnetization corrected for demag-

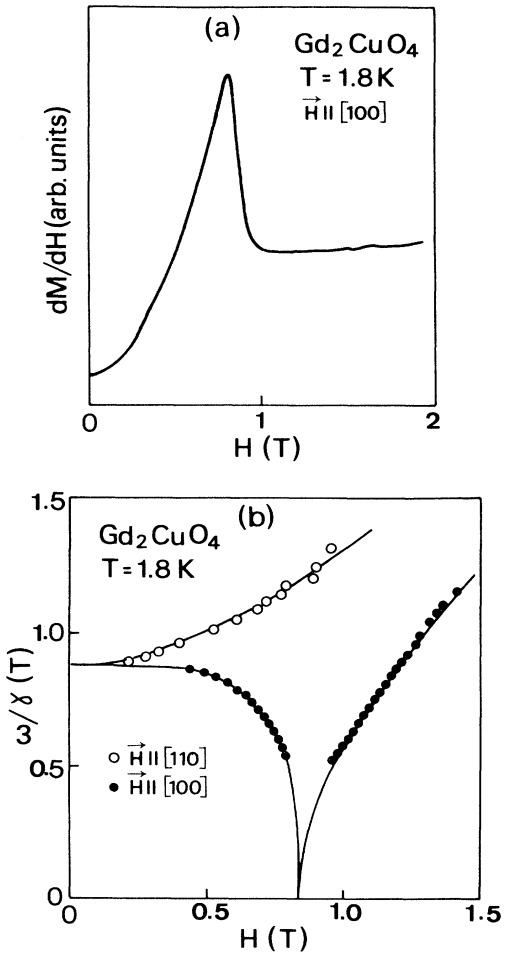

FIG. 1. (a) Magnetic field dependence of the real part of the differential magnetic susceptibility dM/dH of Gd_2CuO_4 with magnetic field applied parallel to [100]. (b) Magnetic field dependence of the antiferromagnetic resonance frequency of Gd_2CuO_4 with magnetic fields applied parallel to [100] and [110].

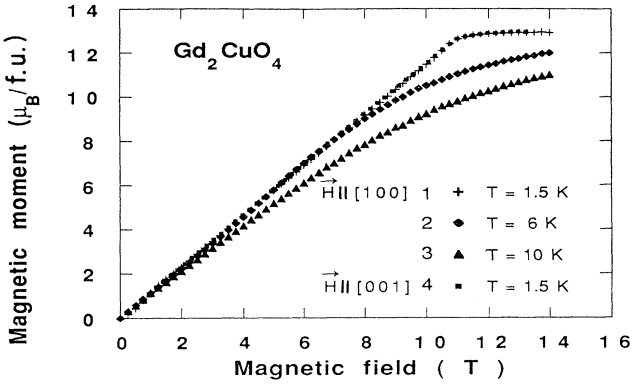

FIG. 2. Magnetization of Gd_2CuO_4 as a function of magnetic field applied parallel to [100] at 1.5, 6, and 10 K (curves 1–3). Also shown is the magnetization (curve 4) with magnetic field applied parallel to [001] at $T=1.5$ K. The magnetization has been corrected for the demagnetization effects.

netization effects, at several temperatures in the temperature range 1.5–20 K as a function of the magnetic field up to 14 T. The measurements were made on a single crystal of rectangular shape ($5\times1.5\times0.075$ mm^3) with the longest dimension parallel to [100]. Curves 1–3 are for the magnetic field $\mathbf{H}\|$ [100]. At $T=1.5$ K (curve 1) a sharp change of the slope of $M(H)$ is observed at $H_s=11$ T, showing saturation of the aligned Gd moments at $\mu=6.5\mu_B$. The saturation field shifts to lower field at higher temperatures. Figure 3 shows the temperature variation of the saturation field. Figure 2 also shows the magnetization with magnetic field parallel to [001] at $T=1.5$ K (curve 4) which coincides almost exactly with that for the magnetic field parallel to [100] at the same temperature.

IV. NEUTRON-DIFFRACTION INVESTIGATIONS

A search for magnetic reflections at superlattice points corresponding to the propagation vector $\mathbf{k}=(\frac{1}{2},\frac{1}{2},0)$ (in analogy with Nd_2CuO_4) and other simple antiferromagnetic wave vectors was unsuccessful. However, measurements of 24 symmetry-independent nuclear reflections at 1.5 and 13.4 K showed that there were magnetic contributions to the nuclear reflections at 1.5 K indicating a propagation vector $\mathbf{k}=(0,0,0)$. This is well demonstrated by the strong temperature dependence of the intensity of the 112 reflection whose nuclear structure factor is very small and whose intensity is therefore almost entirely of magnetic origin. In Fig. 4 we show the variation of the square root F of the magnetic intensity of the 112 reflection which is proportional to the sublattice magnetization, as a function of the reduced temperature $t=1-T/T_N$, where T_N is the temperature at which the gadolinium sublattice orders. The magnetic contribution to the intensity of the 112 reflection was obtained by subtracting the nuclear contribution measured just above T_N from the total intensity. The sublattice magnetization decreases continuously with increasing temperature and becomes very small at $T_N=6.4$ K. The temperature variation of the sublattice magnetization does not follow the Brillouin function $B_{J=7/2}$. The solid curve is the least-squares fit of the expression

$$F=F_0(1-T/T_N)^{\beta} \tag{1}$$

to the data. The fit gave $F_0=134(2)$, $T_N=6.370(5)$, and $\beta=0.234(9)$. The goodness of the fit $\chi^2=13.7$. The inset of Fig. 3 gives the corresponding log-log plot. The critical exponent β obtained is considerably less than the three-dimensional Heisenberg critical exponent $\beta=0.38$ and also less than the three-dimensional Ising value $\beta=0.312$. Because of the layer structure of Gd_2CuO_4, one expects quasi-two-dimensional behavior in this sys-

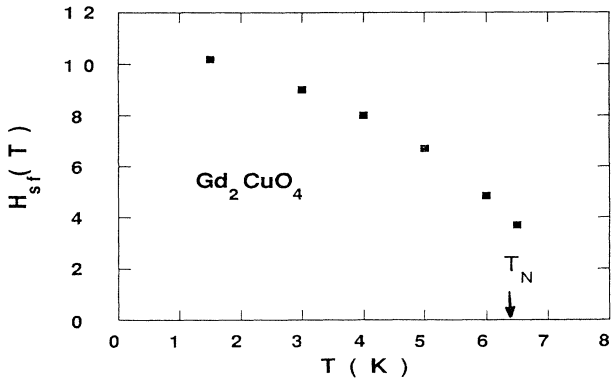

FIG. 3. Temperature variation of the saturation field of Gd_2CuO_4 obtained from the peaks of dM/dH vs H curves.

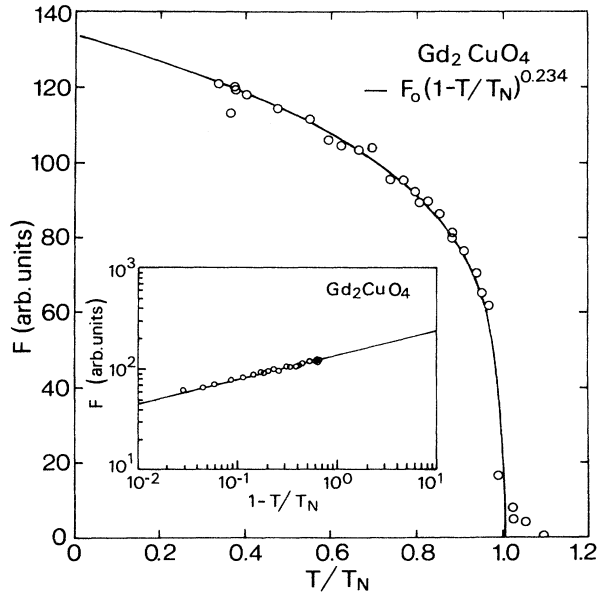

FIG. 4. Temperature variation of the square root of the intensity of the 112 reflection as a function of the reduced temperature T/T_N. The solid curve is the result of the least-squares fit of the data points with the expression $F=F_0(1-T/T_N)^{\beta}$ ($\beta=0.234$). The inset shows the corresponding log-log plot.

tem. Note that the fit to the critical exponent is very good over the whole range of temperature investigated. This is typical for a two-dimensional system. The critical exponent obtained is larger than the two-dimensional Ising value $\beta=0.125$. This is not very unusual and many quasi-two-dimensional magnetic systems have critical exponents of about this magnitude. Similar two-dimensional behavior has been observed in $GdBa_2Cu_3O_{7-\delta}$ and $ErBa_2Cu_3O_7$,[22-24] although the critical exponents obtained in these systems are closer to the two-dimensional Ising value.[25,26] No magnetic intensity could be detected for reflections hkl with $h+k+l=2n+1$ showing that the body centering is retained in the magnetic structure. The magnetic unit cell is therefore identical with the chemical unit cell. The magnetic moments of the two Gd atoms of the primitive unit cell related by the center of symmetry are oppositely oriented and the magnetic moments of the Gd atoms related by the translation $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ are parallel. The existence of magnetic intensity in the 004 and 006 reflections excludes the possibility that the magnetic moments are parallel to [001]. Figure 5 shows the proposed magnetic structure of Gd_2CuO_4. The orientations of the spins within the a-b plane cannot be determined because, since the propagation vector is $\mathbf{k}=(0,0,0)$, the intensity of the magnetic reflections is the sum of contributions from separate domains with tetragonally related spin directions. The remaining question is whether the magnetic moments are tilted out of the a-b plane. The neutron-diffraction intensity data are severely affected by the presence of absorption and extinction. The isotopic substitution is not 100%. The linear absorption coefficient measured at $\lambda=1.176$ Å, the wavelength of the experiment, was 2.2 mm^{-1}, giving a ratio of about 8 between the maximum and minimum transmission coefficients for the measured reflections from the platelike crystal. Since the [001] axis is perpendicular to the plate, the absorption factors for the 001 reflections are particularly high and the crystal shape could not be determined with sufficient accuracy to calculate them reliably. The neutron-diffraction data can therefore not be used to provide evidence for such tilting.

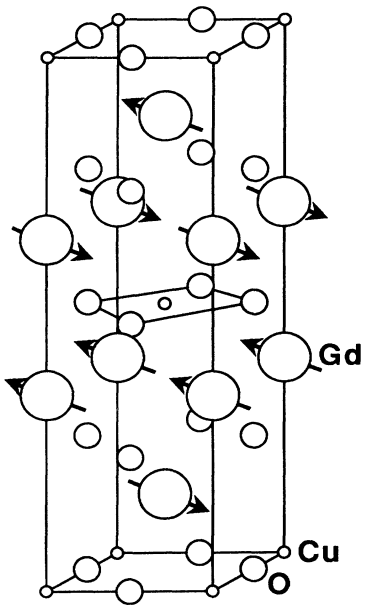

FIG. 5. The proposed magnetic structure of Gd_2CuO_4. The directions of the Gd moments are indicated by the arrows. The exact spin orientation cannot be determined from the neutron diffraction. The magnetization and the antiferromagnetic resonance investigations suggest that the spins of the Gd atoms are oriented parallel to [110].

The limited beam time and high absorption did not permit us to investigate the magnetic structure of the Cu sublattice which is presumably ordered antiferromagnetically as in the case of Nd_2CuO_4 and Pr_2CuO_4. For this we plan to perform a further investigation on a larger single crystal at higher temperature with hot neutrons ($\lambda=0.48$ Å).

V. DISCUSSION

The present magnetization, antiferromagnetic-resonance, and neutron-diffraction investigations provide partial answers to some of the questions raised in Sec. II. We have established by neutron diffraction that Gd_2CuO_4 undergoes an antiferromagnetic phase transition in zero applied magnetic field at $T_N=6.4$ K. Antiferromagnetic resonance and low-field ac susceptibility investigations have established a second-order spin-reorientation transition at 0.88 T at $T=1.8$ K when the magnetic field is applied parallel to [100] and high-field magnetization investigations show that a complete ferromagnetic alignment of Gd moments occurs with fields greater than 11 T at $T=1.5$ K.

Neutron-diffraction and antiferromagnetic-resonance experiments at low magnetic fields enable us to determine the ground-state magnetic structure of Gd_2CuO_4. According to these, Gd_2CuO_4 belongs to the class of "easy-plane" collinear antiferromagnets with a small anisotropy in the basal plane. We can describe this system phenomenologically by a two-sublattice antiferromagnet whose normalized energy contains four terms:

$$\varepsilon=E/M_0=H_e l^2-mH+H_{a1}l_x^2l_y^2/4+H_{a2}l_z^2/2\ , \quad (2)$$

where H_e is the exchange field, $l=(M_1-M_2)/2M_0$, $m=(M_1+M_2)/2M_0$, M_0 is the saturation magnetization, mH is the Zeeman term, and the other two terms are the anisotropy terms. This simple phenomenological description permits us to understand the magnetization and antiferromagnetic resonance data of Gd_2CuO_4 such as the existence of two gaps in the AFMR spectra, frequency-field dependence of the AFMR mode, spin-reorientation phase transition of the second order, etc. The spin-reorientation phase transition can be understood in the following way.

At temperatures below T_N, Gd_2CuO_4 is a collinear antiferromagnet made up of ferromagnetic (001) planes with spins parallel to $\langle 110\rangle$ directions stacked antiferromagnetically along the c axis. When the magnetic field is applied parallel to [100] or [010], the moment direction of the ferromagnetic planes starts to rotate in the (001)

plane from its zero-field direction [110] to the direction perpendicular to the magnetic field. The rotation is completed when the magnetic field reaches the critical field H_c. This picture of the phase transition is inconsistent with a noncollinear double-**k** structure in which the angle between the magnetic moments along [100] and [010] is $\pi/2$ because, in this case, as can be shown by simple calculations, a first-order phase transition rather than a second-order one should be observed. The resonance spectrum also rules out the existence of two mutually perpendicular orthorhombic domains, for, in that case, both the curves of Fig. 1(b) would be observed simultaneously. These two conclusions also follow from the neutron-diffraction data since the propagation vector $\mathbf{k}=(0,0,0)$ is incompatible with a multiple-**k** structure, but consistent with the tetragonal symmetry. The antiferromagnetic resonance and magnetization data allow the added conclusion that the moments are parallel to $\langle 110 \rangle$ directions.

We have observed saturation of the ferromagnetic alignment of Gd moments at $H=11$ T at $T=1.5$ K. This enables one to determine the exchange and anisotropy terms in Gd_2CuO_4. Using the saturation field of 11 T at $T=1.5$ K as $2H_e$ (where H_e is the exchange field), it is possible to extract from the gaps in the AFMR spectra $\omega_i/\gamma=(2H_eH_{ai})^{1/2}$ two anisotropy fields H_{a1} and H_{a2}. The values of these gaps are $\omega_1/2\pi=25$ GHz and $\omega_2/2\pi=86$ GHz.[20] Using these values, we get values, for the two anisotropy fields, $H_{a1}=0.075$ T and $H_{a2}=0.9$ T. H_{a1} is the small interplanar field due to fourth-order anisotropy whereas H_{a2} is the easy-plane anisotropy field. Calculations of the magnetic dipole-dipole interaction in the Gd_2CuO_4 lattice with ferromagnetically ordered Gd layers (see Sec. IV) give $H_{a2}=1.17$ T, which is close to the value of H_{a2} obtained from AFMR data. We can expect a small reduction of H_{a2} because, in the EPR experiments on Gd^{3+} ions in Eu_2CuO_4 (Ref. 21), a single-ion anisotropy of the opposite sign $H_{a2}=-0.1$ T is detected. In magnetization experiments with magnetic field $\mathbf{H}\|$ [001], a shift of about 1 T due to H_{a2} in the value of H_s is expected. In fact, the magnetization curves corrected for the demagnetization effects, corresponding to the magnetic fields parallel to [100] and [001], coincide at all temperatures investigated. The possible reason for this very surprising result is discussed below.

The magnetic behavior of Gd_2CuO_4 in high fields cannot be understood in the framework of just a two-Gd-sublattice model. There are three important magnetic interactions: $J_{\text{Cu-Cu}}$, $J_{\text{Gd-Gd}}$, and $J_{\text{Cu-Gd}}$, of which $J_{\text{Cu-Cu}}$ is the strongest intraplaner Cu-Cu interaction and is of the order of 1000 K estimated from the high Neél temperature of Gd_2CuO_4 (270 K) and also from the spin-wave velocity[2,3] measured in similar compounds, La_2CuO_4 and $YBaCu_3O_6$. The intermediate interaction is $J_{\text{Gd-Gd}}$, which is the interaction between two Gd atoms belonging to neighboring Gd ferromagnetic layers antiferromagnetically stacked. The corresponding exchange field is 11 T as determined from the saturation magnetic field H_s. The smallest interaction is $J_{\text{Cu-Gd}}$, which is the interaction between the Cu and Gd atoms belonging to the neighboring Cu and Gd layers and is of the order of 1 T as determined from the saturation fields for $\mathbf{H}\|$ [100] and $\mathbf{H}\|$ [001]. In spite of the very large Cu-O-Cu exchange interaction ($J_{\text{Cu-Cu}}=1000$ K) and the very small magnetic susceptibility suggesting that the Cu subsystems contribute little to the magnetization value, the Cu subsystem must have a significant influence on the Gd subsystem leading to considerable modification of the high-field behavior. In particular, we suggest that a polarization field of about 1 T, originating from the Cu ions, acts on the Gd ions to counteract their anisotropy field.

The most surprising result obtained during the present investigations is the discovery of magnetic scattering in Gd_2CuO_4 corresponding to the propagation vector $\mathbf{k}=(0,0,0)$ rather than $\mathbf{k}=(\frac{1}{2},\frac{1}{2},0)$, establishing the existence of ferromagnetic Gd layers in the *a-b* plane which are stacked antiferromagnetically along [001]. Such ferromagnetic rare-earth layers have not been observed in any other cuprate materials. The temperature variation of the sublattice magnetization obtained from the neutron diffraction indicates that Gd_2CuO_4 is a quasi-two-dimensional antiferromagnet.

VI. CONCLUSIONS

In conclusion, we have established the three-dimensional magnetic ordering of Gd moments below $T_N=6.4$ K by neutron diffraction and a second-order spin-orientation transition at $H_c=0.88$ T and $T=1.8$ K by magnetization and antiferromagnetic resonance. Magnetization measurements also show that complete alignment of Gd moments parallel to $\langle 100 \rangle$ or $\langle 001 \rangle$ is obtained in fields of 11 T or more at 1.5 K. The most important result of the neutron-diffraction investigations is the evidence for the existence of the ferromagnetic Gd layers parallel to the *a-b* plane. This ferromagnetic rare-earth layer has not been observed in any other cuprate material. It is important to investigate whether the absence of superconductivity in Gd_2CuO_4 upon Ce doping is related to the presence of such ferromagnetic Gd layers.

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[1] P. W. Anderson, Science **235**, 1196 (1987).

[2] R. J. Birgeneau and G. Shirane, in *Physical Properties of High Temperature Superconductors,* edited by D. M. Ginsberg (World-Scientific, Singapore, 1989), and references therein.

[3] H. Chou, J. M. Tranquada, G. Shirane, T. E. Mason, W. J. L.

Buyers, S. Shamoto, and M. Sato, Phys. Rev. B **43**, 5554 (1991); J. Rossat-Mignod, L. P. Regnault, M. J. Jurgens, C. Vettier, P. Burlet, J. Y. Henry, and G. Lapertot, Physica B **163**, 4 (1990), and references therein.

[4]Y. Tokura, H. Takagi, and S. Uchida, Nature **337**, 345 (1989).

[5]H. Takagi, S. Uchida, and Y. Tokura, Phys. Rev. Lett. **62**, 1197 (1989).

[6]J. T. Market and M. B. Maple, Solid State Commun. **70**, 145 (1989).

[7]J. T. Market, E. A. Early, T. Bjornholm, G. Ghamaty, W. B. Lee, J. Neumier, R. D. Price, C. L. Seeman, and M. B. Maple, Physica C **158**, 178 (1989).

[8]Y. Endo, M. Matsuda, Y. Yamada, K. Kakurai, Y. Hidaka, G. Shirane, and R. J. Birgeneau, Phys. Rev. B **40**, 7023 (1989).

[9]S. Skanthakumar, H. Zhang, T. W. Clinton, W.-H. Li, J. W. Lynn, Z. Fisk, and S.-W. Cheong, Physica C **160**, 124 (1989).

[10]D. E. Cox, A. I. Goldman, M. A. Subramanian, J. Gopalakrishnan, and A. W. Sleight, Phys. Rev. B **40**, 6998 (1989).

[11]M. Matsuda, K. Yamada, K. Kakurai, H. Kadowaki, T. R. Thurston, Y. Endoh, Y. Hidaka, R. J. Birgeneau, M. A. Kastner, P. M. Gehring, A. H. Moudden, and G. Shirane, Phys. Rev. B **42**, 10 098 (1990), and references therein.

[12]M. J. Rosseinsky, K. Prassides, and P. Day, J. Chem. Soc. Commun. **1989**, 1734 (1989).

[13]T. Chattopadhyay, P. J. Brown, U. Köbler, and V. L. Sobolev, Physica C **177**, 294 (1991).

[14]J. D. Thompson, S.-W. Cheong, S. E. Brown, Z. Fisk, S. B. Oseroff, M. Tovar, D. C. Vier, and S. Schultz, Phys. Rev. B **39**, 6660 (1989).

[15]S. B. Oseroff, D. Rao, F. Wright, D. C. Vier, S. Schultz, J. D. Thompson, Z. Fisk, S.-W. Cheong, M. F. Hundley, and M. Tovar, Phys. Rev. B **41**, 1934 (1990).

[16]C. L. Seaman *et al.*, Physica C **159**, 391 (1989).

[17]G. Xiao, M. Z. Cieplak, and C. L. Chien, Phys. Rev. B **40**, 4538 (1989).

[18]K. A. Kubat-Martin, Z. Fisk, and R. Ryan, Acta Crystallogr. C **44**, 1518 (1988).

[19]P. Lethuiller, J. F. Guilland, and J. Voiron (unpublished).

[20]A. I. Zvyagin, M. I. Kobets, V. A. Pashchenko, A. A. Stepanov, and E. N. Khatsko, Physica B **165 & 166**, 1321 (1990).

[21]P. Rao, M. Tovar, S. B. Oseroff, D. C. Vier, S. Schultz, J. D. Thompson, S.-W. Cheong, and Z. Fisk, Phys. Rev. B **38**, 8920 (1988).

[22]T. Chattopadhyay, H. Maletta, W. Wirges, K. Fischer, and P. J. Brown, Phys. Rev. B **38**, 838 (1988).

[23]T. Chattopadhyay, P. J. Brown, B. C. Sales, L. A. Boatner, H. A. Mook, and H. Maletta, Phys. Rev. B **40**, 2624 (1989).

[24]H. Maletta, E. Pörschke, T. Chattopadhyay, and P. J. Brown, Physica C **166**, 9 (1990).

[25]C. Meyer, H.-J. Borneman, H. Schmidt, R. Ahrens, D. Ewart, B. Renker, and G. Czjzk, J. Phys. F **17**, 1345 (1987).

[26]J. W. Lynn, T. W. Clinton, W.-H. Li, R. W. Erwin, J. Z. Liu, K. Vandervoort, and R. N. Shelton, Phys. Rev. Lett. **63**, 2606 (1989).