MAGNETIC PHASE TRANSITIONS OF T`-PHASE Gd_2CuO_4 SINGLE CRYSTAL

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ABSTRACT

MAGNETIC PHASE TRANSITIONS OF T`-PHASE Gd_2CuO_4 SINGLE CRYSTAL. The magnetic properties of T'-phase Gd_2CuO_4 Single crystal grown by the Traveling Solvent Floating Zone (TSFZ) method have been investigated by means of ac-magnetic susceptibility and dc-magnetization measurements. The result of these studies revealed an anomaly in the temperature dependent magnetization at temperature of T = 6.5, 290 and 20 K, associated respectively with the long-range antiferroniagtietic ordering of the Gd and Cu ions and Cu-spin reorientation transitions. The complex magnetic structure of this compound shown by the weak ferromagnetic behavior below the copper ordering temperature (T_N(Cu) = 290 K) is induced by an effective field due to exchange interactions between the ordered copper moments and the rare-earth ions. These results, together with the previous neutron diffraction measurement, establishes the existence of ferromagnetic Gd layers in the ab-plane which are stacked antiferromagnetically along c-direction, indicating a quasi-two dimensional antiferromagnetic nature of Gd_2CuO_4.

Key words : Gd_2CuO_4 single crystal, Traveling Solvent Floating Zone (TSFZ) method, magnetic properties.

ABSTRAK

TRANSISI FASE MAGNETIK PADA KRISTAL TUNGGAL FASE-T'Gd_2CuO_4. Telah ditelaah dalam studi ini sifat-sifat magnetik dari kristal tunggal fase-T'Gd_2CuO_4 melalui pengukuran suseptibilitas AC-magnetik don magnetisasi DC. Hasil-hasil dari studi ini menunjukkan sebuah anomali pada kebergantungan temperatur dari magnetisasi pada suhu 6,5 , 290 dan 20 K, masing-masing diasosiasikan dengan *ordering* antiferromagnetik berjangkauan panjang dari ion-ion Gd dan Cu dan transisi reorientasi spin dari ion-ion Cu. Struktur magnetik yang kompleks dari sistem ini seperti diperlihatkan oleh kelakuan ferromagnetik lemah pada temperatur di bawah suhu *ordering* Cu(T_N(Cu) = 290 K), disebabkan oleh sebuah medan efektiv dari interaksi tukar antara momen-momen magnetik Cu yang terorder dan ion-ion tanah jarang. Hasil-hasil eksperimen ini bersama-sama dengan hasil pengukuran defraksi neutron terdahulu, menkonfirmasi kehadiran dari lapisan ferromagnetik Gd pada bidang-ab yang tersusun secara antiferromagnetik sepanjang arah-c, mengindikasikan sifat quasi-dua dimensi dari sistem Gd_2CuO_4.

Kata kunci : Kristal tunggal Gd_2CuO_4, metoda *Traveling Solvent Floating Zone (TSFZ), sifat-sifat magnet.*

INTRODUCTION

The magnetic properties of R_2CuO_4 (R = Pr, Nd, Sm, Eu, Gd) crystallize in the tetragonal T'-phase structure of Nd_2CuO_4 [1] with the Cu sublattice presents three-dimensional antiferromagnetic (AF) order below T_N = 250-290 K [2]. Among them, the compound Gd_2CuO_4 occupies a unique place in this family for some reasons: first, although it is easily doped with Ce or Th, it does not become superconducting as the other members; second, the square array of oxygen ions surrounding the Cu atoms is rotated around the c-axis, leading to a reduced orthorhombic symmetry (space group Acam) [3,4]. As the result, the AF order is not perfect in this case and weak ferromagnetism (WF) appears due to canting of the Cu moments in the CuO_2 planes [5-7]. The origin of this canting has been attributed to an antisymmetric exchange interaction of Dzyaloshinskii-Moriya (DM) type [8] allowed in this case by the orthorhombic distortion.

We present in this study the magnetic properties of a Gd_2CuO_4 single crystal grown by means of the

Volume 3 No. 2, Pebruari 2002, hal : 43 - 46
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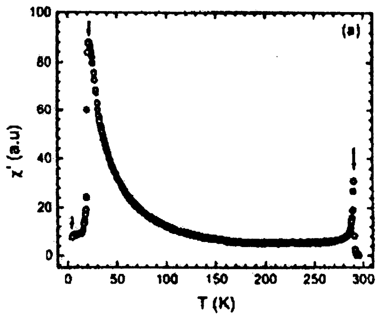
Traveling Solvent Floating Zone (TSFZ) method. The magnetic properties have been explored by ac-magnetic susceptibility and dc-magnetization measurements. In ref. [6] it has been suggested that the in-plane anisotropy Gd_2CuO_4 compound was relatively small. For that reason we ignore the in-plane anisotropy in our measurement. The detailed study about the in-plane magnetization anisotropy has been reported in ref. [9].

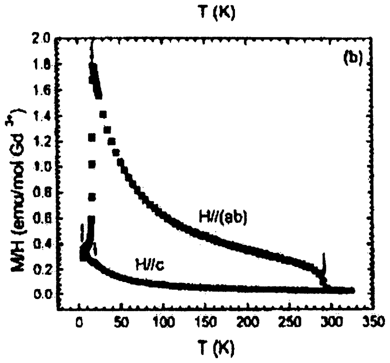
EXPERIMENTS

The T'-phase Gd_2CuO_4 single crystal was grown by the Traveling Solvent Floating Zone (TSFZ) method using a four-mirror furnace from Crystal system, Inc. The quality of the as-grown crystal was examined by X-ray LAUE diffraction pattern using DIFFRACTIS 582 of ENPAF NONIUS, and Electron Probe Micro Analysis (EPMA) using JEOL JXA-8621. The AC-susceptibility measurement has been carried out in a homemade system using a mutual inductance technique. With this system, the measurements can be performed in the temperature range between 4.2 and 300 K at an appropriate cooling or heating rate by manipulating the vacuum in the heat-exchange space of the measurement insert. The dc-magnetization measurements were measured by a commercial Quantum Design (SQUID) MPMS 5 magnetometer. Both of these measurements were performed at the Van der Waals-Zeeman Institute, University of Amsterdam.

RESULTS AND DISCUSSION

The results of the temperature dependent AC-susceptibility and dc-magnetization measurements are given in Fig. 1. The development of anisotropy is readily apparent from this figure. The AC-susceptibility





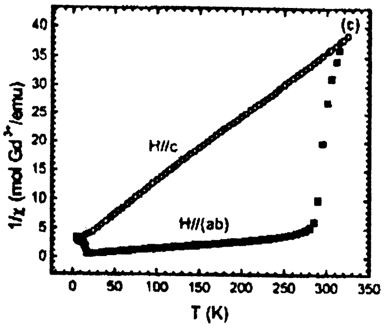
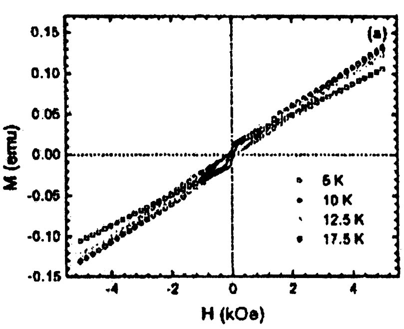


Fig. 1(a). Temperature dependent AC-magnetic susceptibility, *(b).* dc-magnetization measurements and *(c).* inverse molar susceptibility for field direction in the plane (χ_{ab}^{-1}) and along the c-axis (χ_c^{-1}).

and dc-magnetization measurement for field parallel to the plane (H//ab; χ_{ab}) show an anomaly at T = 6.5, 20 and 290 K, in good agreement with the previous report [5,10-13]. The dc-magnetization measurement for field parallel perpendicular to the plane *(H//c;* χ_c)on the other hand, shows the anomaly at 6.5 and 20 K only. The Currie-Weiss bellavior is shown by $1/\chi_c$, curve for $T > 20$ K as depicted in Fig. 1(c). The effective magnetic moment determined from this plot is μ_{eff} = 8.37 μ_B/Gd^{3+}-atom and paramagnetic Curie temperature θ = 15.51 K. These values are to be compared with previous reports [2,5] and close to its free ion value of μ_{eff} = 7.94 μ_B/Gd^{3+}[14]. We note that for H parallel to the planes and temperature less than ~ 200 K, μ_{eff} = 25.3 μ_B/Cd^{3+}-atom and θ = 27.37 K; these values are bigger than the previous report by Thompson ct.al. [5]. The reasons for the lower and higher anomalies are associated with the antiferromagnetic ordering of Gd and Cu ions. At T_N(Cu) = 290 K, the Cu magnetic moments order to a La_2NiO_4-like antiferromagnetic (AF) structure with spin direction of Cu ions parallel to [110]. At T_N(Gd) = 6.5 K the Gadolinium ions order antiferromagnetically with special configuration, namely the two centrosymmetrically related Gd atoms in the primitive unit cell are oppositely oriented. The explanation for the mid temperature anomaly T_c= 20 K will be presented in the following paragraph.

The plots of field dependent magnetization at certain temperature, $M(H)_T$ for H//(a,b) is shown in Fig. 2 and 3 associated respectively for temperature range below and above T_c = 20 K. Two important features



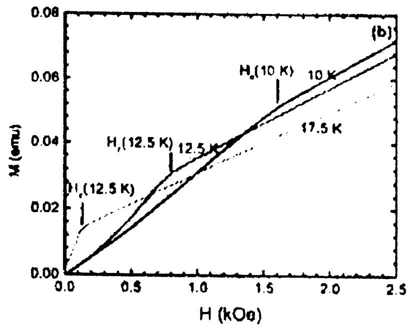


Fig. 2(a). Isothermal magnetic hysteresis loop of a Gd_2CuO_4 single crystal measured at various temperature below T_c=20 K. ***(b).*** The *M*(*H*) curves in the temperature range of T_N(Gd) < *T*< T_c, showing two linear parts of the curves with different slope, intersect each other at H_c See text for discussion.

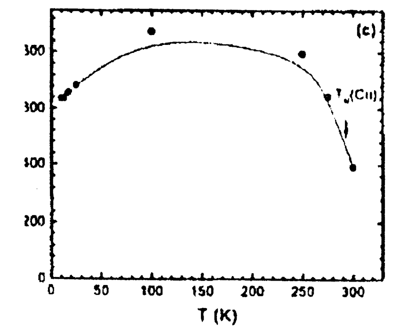
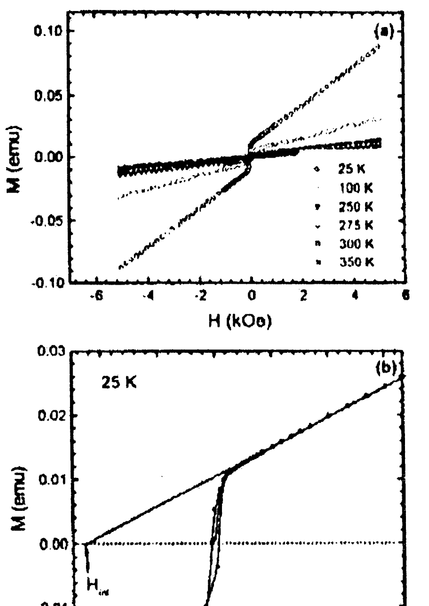


Fig. 3(a) Isothermal magnetic hysteresis loop of a Gd_2CuO_4 single crystal measured at various temperature above T_c ***(b).*** The schematic picture showing the determination of internal field H_{int}, for *T*= 25 K. ***(c).*** Temperature dependent internal field H_{int} (*T*). See text for discussions.

observed which indicate the presence of a phase transition: (1) The small but significant hysteresis at $H\approx0$, which is present at all temperatures below T_N(Cu) and gradually disappear below T_c = 20 K. This behavior is clearly seen in Fig. 3(b) for *T*= 25 K. The form of the *M(H)* curves in the temperature range of T_N(Gd) < *T* < T_c have two almost linear parts with different slopes, intersect each other at H_c as depicted by arrowheads in Fig. 2(b). From this figure one can see that the value of H_c decreases with increasing temperature. Further, the lower part of the *M(H)* curves (*H*< H_c) passes through the origin when extrapolated to *H*= 0, indicate a pure AF phase is stable when *H* < H_c. These results clearly show that the WF phase only exists in Gd_2CuO_4 in zero-field for *T* >T_c, although below T_c it can be induced by a magnetic field applied in the ab-plane. We concluded that a WF-AF phase transition does occur in zero-field at *T*= 20 K. An important characteristic of the WF phase is the value of the internal field H_{int}, which induces the weak



ferromagnetic moment in the ab-plane. We determined the H_{int} values by extrapolating the linear high-field part of the curves (according to linearity criterion of *R* =1) to *M*=0 as illustrated in Fig. 3(b) for *T*= 25 K. The temperature dependent internal field H_{int} (*T*) is shown in Fig. 3(c).

Having gone through the above analysis, we are attempting now to discuss the complex magnetic structure of this Gd_2CuO_4 compound. In the temperature region of T_c<*T*<T_N(Cu), the Cu-spins have long-range antiferromagnetic order with a significant weak ferromagnetic moment parallel to the ab-planes, arises from polarization of the paramagnetic Gd sublattice by exchange interaction with the ordered Cu subsystem. The weak ferromagnetism results from a strong intraplanar spin-spin interaction. At *T* = T_c a phase transition to a purely antiferromagnetic state with weak ferromagnetism behavior takes place. These weak ferromagnetic moments in the temperature region of T_N(Gd) < *T* < T_c are easily induced by relatively weak magnetic fields applied perpendicular to the c-axis. Such behavior suggests that a weak ferromagnetic moment exists independently in each CuO_2 plane and that the phase transition at T_c is associated with the appearance of substantial antiferromagnetic correlation between weak ferromagnetic moments of adjacent CuO_2 planes. The role of the

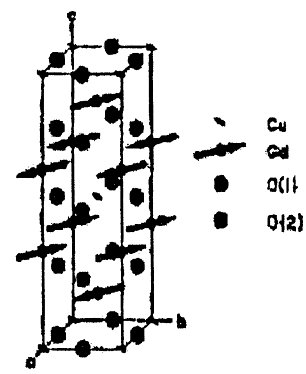


Fig. 4 The proposed magnetic structure of Gd_2CuO_4 in the antiferromagnetic phase at *T*<T_N(*Gd*)= 6.5 K

Volume 3 No. 2, Pebruari 2002, hal. 43 - 46
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relatively weak interplanar interaction is to produce some correlation between the orientation of the weak ferromagnetic moments in neighboring CuO_2 planes. The crystal structure of Gd_2CuO_4 deduced from neutron diffraction measurement [11] and in conformity with the above analysis is shown in Fig. 4. This figure establishing the existence of ferromagnetic Gd layers in theab-plane which are stacked antiferromagnetically along c-direction, thus indicating a quasi two dimensional antiferromagnetic nature of Gd_2CuO_4 system.

SUMMARY

We have presented in this study the magnetic properties Gd_2CuO_4 single crystal grown by the TSFZ method. Temperature dependent magnetization shows a clear anomaly at T = 6.5, 290, and 20 K, associated respectively with the long-range antiferromagnetic ordering of the Cd and Cu ions and Cu-spin reorientation transitions. In addition, a weak ferromagnetic behavior induced by an effective field due to exchange interactions between the ordered copper moments and the rare-earth ions was developed below the copper ordering temperature (T_N(Cu) = 290 K). These results establishes the existence of ferromagnetic Gd layers in the ab-plane which are stacked antiferromagnetically along c-direction, indicating a quasi-two dimensional antiferromagnetic nature of Gd_2CuO_4 in concurrent with neutron diffraction results.

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REFERENCES

[1]. HK. MÛLLER~BUSCHBAUM and W. WOLLSCHLÄGER, Z. *Anorg. Allg. Chem.* **414** (1975) 76.

[2]. R. SAEZ PUCHE, M. NORTON, and W.S. GLAUSINGER, *Mater. Res. Bull.* **17** (1982) 1523; R. SAEZ PUCLIC, M. NORTON, T.R. WHITE and W.S. GLAUSINGER, *J. Solid State Chem.* **50** (1983) 281.

[3]. M. BRADEN, W. PAULUS, A. CAUSSON, P. VIGOUREUX, G. HEGER, A. GAUKASSOV, P. BOURGES and D. PETITGRAND, *Europhys. Lett.* **25** (1994) 625

[4]. H.M. LUO, Y.Y. HSU, B.N. LIN, Y.P. CHI, T.J. LEE and H.C. KU, *Phys. Rev. B* **60** (13) (1999) 119.

[5]. 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** (1989) 6660

[6]. 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** (1990) 1934.

[7]. M. TOVAR, X. OBRADORS, F. PEREZ, S.B. OSEROFF, R.J. DURO, J. RIVAS, D. CHATEIGNER, P. BORDET, and J.CHENAVAS, *Phys. Rev. B* **45** (1992) 4729.

[8]. I.E. DZYALOSHINSKII, *J. Chem. Phys. Solids* **4** (1958) 241 ; T. MORIYA, *in Magnetism*, Edited by G.T. RADO and H. SUHL, Academic Press, New York, **Vol. 1** (1966) 85.

[9]. A. BUTERA, M. TOVAR, S.B. OSEROFF, Z. FISK, *Phys. Rev. B* **52** (13) (1995) 444.

[10]. CHATTOPADHYAY, P.J. BROWN, A.A. STEPANOV, P. WYDER, J. VOIRON, A.I. ZVYAGIN, S.N. BARILO, D.J. ZHIGUNOV and I. ZOBKALO, *Phys. Rev. B* **44** (1991) 9486.

[11]. A.A. STEPANOV, P. WYDER, T. CHATTOPADBYAY, P.J. BROWN, G. FILLION, I.M. VITEBSKY, A. DEVILLE, B. GAILLARD, S.N. BARILO and D.I. ZHIGUNOV, *Phys. Rev. B* **48** (12) (1993) 979.

[12]. P.W. KLAMUT, *Phys. Rev. B* **50** (13) (1994).

[13]. P.W. KLAMUT, K. ROGACKI, A. SIKORA and B. DABROWSKI, *J. Appl. Phys.* **84** (1998) 5129.

[14]. R.S. TEBBLE, D.J. CRAIK, *Magnetic Materials*, John Wiley and Sons, (1969) 184.