Conversion of the Ising Axis in $DyCu_2$ under High Magnetic Field

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The magnetic a and c axes are switched by applying high magnetic field in $DyCu_2$ single crystal. The virgin state has the Ising axis along the a direction with the two-step metamagnetism but the Ising axis is switched to the c direction by applying the field higher than 13 T along the c axis. The recovery of the virgin state is obtained either by increasing temperature above 100 K or applying the field higher than 5 T to the a axis. The switching cycle is quantitatively explained by the first order phase transition model.

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Much work has been reported on the spin-flop or field-induced spin reorientation in magnetic materials but no clear report on the conversion of the magnetic symmetry axis has been published. Although there has been the precursor work concerning cooperative Jahn-Teller distortions in some rare-earth insulators [1] which exhibit field-induced switching of magnetic domain [2], the transformation was not drastic. The first clear example, conversion of the Ising axis, is found in $DyCu_2$ single crystal at liquid helium temperatures when high magnetic field is applied to the c axis and the conversion is retained when the field is removed.

$DyCu_2$ is an intermetallic rare-earth compound with the orthorhombic $CeCu_2$ type crystal structure as is shown in Fig. 1 [3]. The Néel temperature T_N is 31.5 K [4,5] with the spin structure given in Fig. 1 [6] where three crystallographic unit cells are regarded as a magnetic unit cell. The Dy spin has the Ising-like axis along the a direction with two-step metamagnetization as is shown in Fig. 2 [5,7]. The gJ value along the a axis is about $10\mu_B$ in accordance with usual Ising-like Dy^{3+} spin. Magnetizations along the b and c axes are smooth and small up to 6 T [4].

Under high magnetic field, however, this material shows a peculiar magnetization process given in Fig. 3. A pulsed field higher than 30 T with the width of 40 msec is applied to the c axis. The technical details are shown elsewhere [8]. The c-axis magnetization is found as expected below 13 T but a sudden change into the a-axis magnetization occurs around 13 T. The c-axis magnetization does not recover by decreasing field but shows the a-like behavior as is given by the a-like line in Fig. 3 which will be called the a'-axis magnetization hereafter. It should be noted that the virgin state recovers by increasing temperature above about 100 K. Then, it is necessary to keep the sample at low temperatures to investigate the switched state. After the conversion, the low field magnetizations along the a and c axes could be observed by rotating the sample in the liquid helium cryostat and it shows the exchange of the a with the c, namely, the a-axis magnetization gives the c-like (c') one and the c axis shows the a' magnetization, respectively, as are shown in [I] of Fig. 4. A little change with hysteresis is found along the b axis but it does not switch to any axis

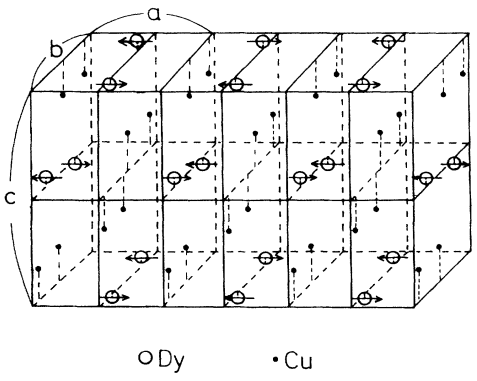

FIG. 1. Crystal and spin structures in $DyCu_2$.

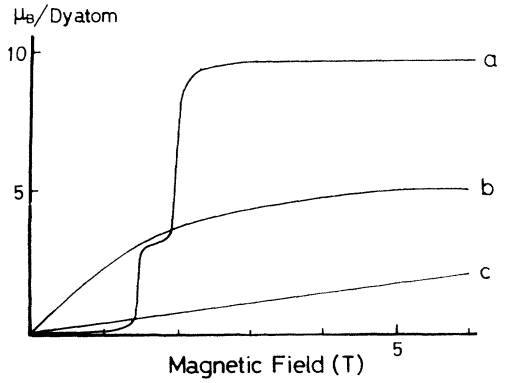

FIG. 2. Magnetization curves of $DyCu_2$ up to 6 T (Ref. [3]).

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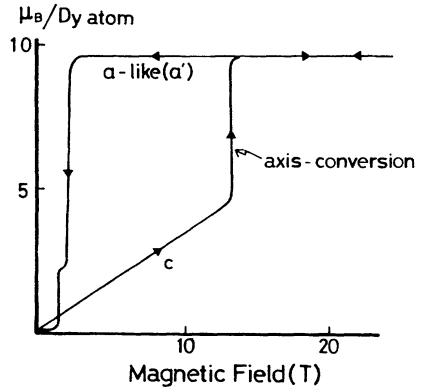

FIG. 3. Conversion of the Ising axis from the a axis to the c axis in $DyCu_2$ at 4.2 K. An external magnetic field is applied along the c axis.

up to 40 T. The recovery of the virgin state is obtained as well by applying the field higher than 5 T along the c' axis as is shown in [II] of Fig. 4. The c' axis, which is the original a axis, shows a small magnetization below 5 T where a sharp step magnetization appears and the decreasing field clearly shows the typical a-axis magnetization reflecting the recovery of the virgin state.

The observed phenomenon may be called the conversion of the Ising axis or more generally the conversion of the magnetic axis. The results are explained by using the first order phase transition model similar with the martensitic transition. A typical martensite (bcc)–austenite (fcc) transition in iron and nickel alloys has been discussed by Patel and Cohen [9] with the free energy diagram in Fig. 5. Let us start from the temperature A_s on the A line which gives the free energy of the austenite state. The thermodynamical equilibrium with the martensite state given by the B line is achieved at T_0 but no

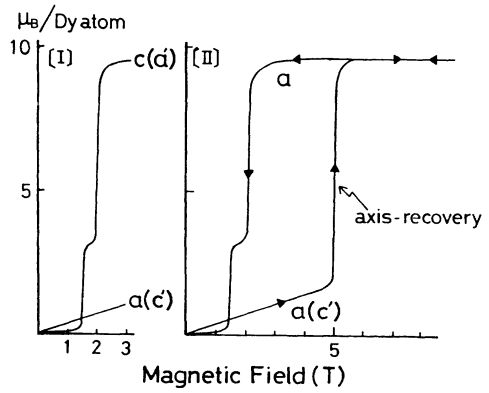

FIG. 4. Magnetization process after the conversion. [I] shows the low field magnetization which shows the a and c axes exchange. [II] shows the inverse conversion, that is the recovery of the virgin state induced by a magnetic field along the original a axis.

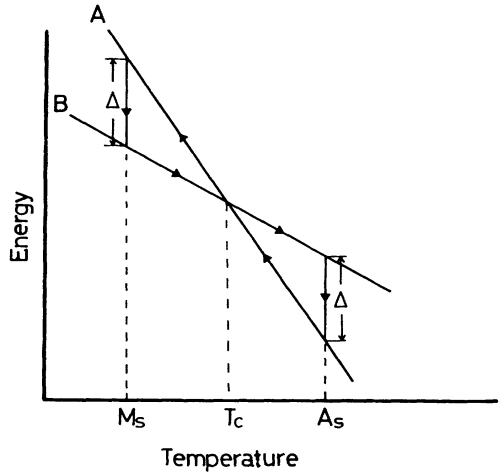

FIG. 5. Schematic picture of the first order martensitic transition with thermal hysteresis.

transition occurs here because the driving energy Δ is necessary to induce the transition so that the transition is found at M_s. The same energy is needed at A_s. The field-induced martensitic transition of various alloys are systematically investigated by our group [10] and the Patel-Cohen model has been satisfactorily confirmed. A similar transition model is introduced to the present system as is shown in Fig. 6. Two electronic states ψ_a and ψ_c are considered. ψ_a is the virgin state, that is the electronic state with the Ising axis along the a direction and ψ_c represents an excited state with the Ising axis along the c direction. The corresponding energies E_a and E_c in the external magnetic field H_c along the c axis are given as

$$E_a = -(\chi_c/2)H_c^2, \tag{1}$$

$$E_c = E_0 - gJ\mu_B H_c, \tag{2}$$

where E_0 is the energy gap at $H_c = 0$ and χ_c is the mag-

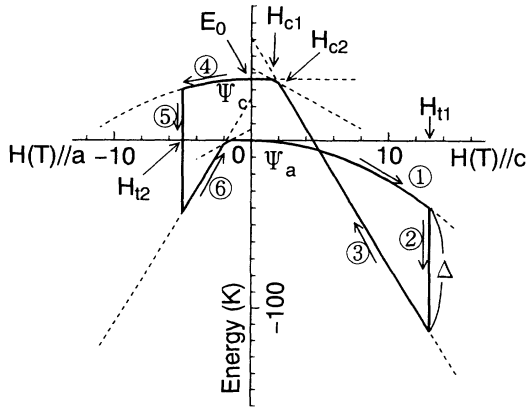

FIG. 6. A cycle of the conversion in $DyCu_2$ at 4.2 K.

netic susceptibility along the c axis. The two-step metamagnetism is not necessary to consider here because the conversion field is much higher than the metamagnetism critical field H_{c1} and H_{c2}. Assuming that the driving energy gap Δ is obtained at the field H_{t1}, then the relation

$$\Delta = -E_0 + gJ\mu_B H_{t1} - (\chi_c/2)H_{t1}^2 \quad (3)$$

is obtained. The first stage of the conversion cycle starting from ψ_a is illustrated by the path, $1 \rightarrow 2 \rightarrow 3$ in Fig. 6 and the ψ_c state is obtained at zero magnetic field. Then the field H_a along the $a(c')$ axis is applied. The energies E_a and E_c are given by

$$E_a = -gJ\mu_B H_a , \quad (4)$$

$$E_c = E_0 - (\chi_c/2)H_a^2 . \quad (5)$$

The recovery transition to the ψ_a state is assumed to occur at H_{t2} with the same energy gap Δ. Then the relation

$$\Delta = E_0 + gJ\mu_B H_{t2} - (\chi_c/2)H_{t2}^2 \quad (6)$$

is obtained. E_0 and Δ are obtained from Eqs. (3) and (6) as

$$E_0 = gJ\mu_B(H_{t1} - H_{t2})/2 - \chi_c(H_{t1}^2 - H_{t2}^2)/4 , \quad (7)$$

$$\Delta = gJ\mu_B(H_{t1} + H_{t2})/2 - \chi_c(H_{t1}^2 + H_{t2}^2)/4 . \quad (8)$$

Numerical values of these parameters are estimated by using values of $gJ = 10$, $\chi_c = 0.35\mu_B$, $H_{t1} = 13$ T, and $H_{t2} = 5$ T as $E_0 = 27\mu_B = 37$ K and $\Delta = 55\mu_B = 74$ K with the temperature unit. The result of $\Delta \approx 100$ K is consistent with the fact that the recovery process occurs by increasing temperature above 100 K.

To confirm the energy level scheme in Fig. 6, the magnetization has been measured after field cooling process of 8 T along the c axis. According to the energy level scheme, the ψ_c state should be lower than the ψ_a state at 8 T along the c axis, then the ψ_c ground state could be obtained even if the field is removed. After this procedure, the a'-magnetization curve, as is shown in Fig. 4, has been observed along the c axis. This result implies that the ψ_c ground state is obtained by field-cooling procedure and supports the energy level scheme.

It is rather surprising that two metamagnetic critical fields H_{c1} and H_{c2} in the ψ_a and ψ_c states are quite close from each other. This means that the Ising characters and various exchange interactions are very similar in both states. The observed similarity strongly suggests that the orthorhombic nature of the Dy spin may not be explained by the simple point charge crystalline field model but rather by considering the interaction with conduction electrons. Since there is no x-ray data for the ψ_c-state structure, it is difficult to infer that the same point charge configuration exists in both states. The cubic field may come from the surrounding atoms but the lower symmetry may mainly come from the band Jahn-Teller effect which optimizes the conduction electron and localized Dy-level energies. The observed similarity between the ψ_a and ψ_c states is understandable if the latter mechanism is dominant in $DyCu_2$. It is noted that the band Jahn-Teller effect is also necessary to understand the high field magnetization of DyAg [11] where the Ising axes along four equivalent [111] directions are converted to one [111] direction parallel to the magnetic field without large change in the magnetic interactions. In this case, however, the hysteresis is very small and the converted state is not obtained at zero magnetic field.

There are some difficulties to obtain the perfect conversion cycle in $DyCu_2$. The used crystals sometimes show cracks after a conversion cycle because the conversion might accompany considerable magnetostriction. X-ray analysis and magnetostriction experiments are desirable.

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