Journal of Magnetism and Magnetic Materials 63 & 64 (1987) 114–116
North-Holland, Amsterdam

NEUTRON DIFFRACTION AND MAGNETIC STUDIES OF CeZn AND NdZn SINGLE CRYSTALS

H FUJII, Y UWATOKO, K MOTOYA*, Y ITO** and T OKAMOTO

Faculty of Integrated Arts and Sciences, Hiroshima University, Hiroshima 730, Japan
**Faculty of Science, Saitama University, Urawa 338, Japan*
***Institute of Solid State Physics, University of Tokyo, Tokyo 106, Japan*

Neutron diffraction studies on CeZn indicate that the Ce moment is 1 91μ_B and Γ_8 is the ground state The observed Ce moment suggests that the Kondo effect should be considered, while magnetic properties of NdZn single crystal are qualitatively understood by taking into account the CEF effect

1. Introduction

The equiatomic compounds of the rare earth with zinc crystallize in the CsCl-type structure The heavy-rare earth RZn compounds exhibit ferromagnetism, while the light-rare earth ones are antiferromagnetic Powder neutron diffraction studies of the latter compounds [1,2] indicate that the magnetic structure is described by the propagation vector $(0, 0, \frac{1}{2})$ with a quadratic magnetic cell $(a, a, 2a)$ We consider only three different structures shown in fig 1 for cubic lattice as the $(00\frac{1}{2})$ magnetic reflection is absent, which are indiscernible on powder neutron patterns However, as a tetragonal distortion is detected below T_N for CeZn and PrZn, we can deduce that the structure is the collinear-type structure (fig 1a) On the other hand, since NdZn does not show any distortion, it is impossible to determine the magnetic structure of NdZn from the neutron diffraction studies, assuming multi-domains

Among them, CeZn exhibits some anomalous behavior owing to possibly the extent of the 4f shell and the consequent strong mixing with the conduction electron band states The especially interesting phenomena are (1) the appearance of pressure-induced ferromagnetism with a pronounced Kondo anomaly at high pressures above 8 kbar [3] and (2) the damping of the crystal electric field excitations (CEF) which lead to the collapse of the inelastic line into the quasi-elastic peak [4]

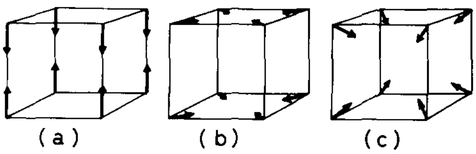

Fig 1 Possible three equivalent magnetic structures in the cubic structure with the $(00\frac{1}{2})$ propagation vectors

2. Results

Fig 2 shows the product of the effective magnetic form factor by magnetic moment $\mu f(\boldsymbol{k})$ as a function of the scattering vector $\sin\theta/\lambda$ ($\mathrm{\AA}^{-1}$) for CeZn at 8 K and NdZn at 4 2 K The form factor seems to be weakly anisotropic in CeZn, whereas that of NdZn is more anisotropic within our experimental errors The magnetic moments of CeZn and NdZn are estimated to be 1 91 and 2 52μ_B/R-atom by extrapolating $\mu f(\boldsymbol{k})$ to $\sin\theta/\lambda = 0$, respectively These values are in good agreement with those obtained by Schmitt et al [1] and Morin and Pierre [2] The temperature dependence of the magnetization is shown in fig 3 for both CeZn and NdZn, which were deduced from the $(10\frac{1}{2})$ magnetic reflections CeZn reveals a first-order transition at $T_N = 30$ K, below which a tetragonal distortion appeared and the $(c/a - 1)$ value reached 1 7% at 8 0 K from analysis of the (200) nuclear reflection The magnetic moment of NdZn decreases sensibly near $T_R = 18$ K and vanishes at $T_N = 70$ K

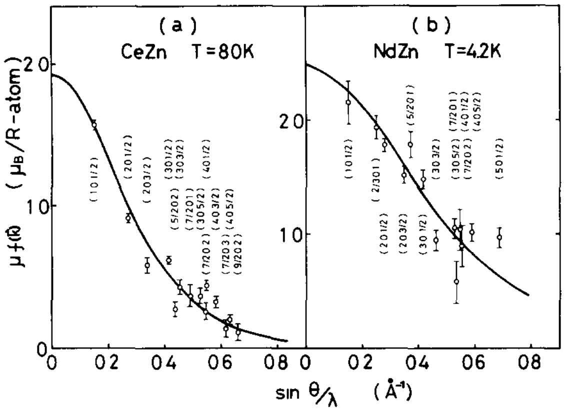

Fig 2 Effective magnetic amplitude $\mu f(\boldsymbol{k})$ as a function of scattering vector $\sin\theta/\lambda$ The solid lines show the calculated form factors according to the formula [6] $f = \langle J_0\rangle + \sum_{i=2}^{4} C_i\langle J_i\rangle$ ($C_2 = 0$ and $C_4 = 0\ 71$ for CeZn, and $C_2 = 0\ 56$ and $C_4 = 0$ for NdZn, respectively) The neutron diffraction experiments were performed for CeZn using PANSI of ISSP installed at the JRR-2 reactor, Tokai, JAERI and for NdZn using ND at the Research Reactor Institute, Kyoto University in Japan

The electrical resistivities ρ of LaZn, CeZn and NdZn along the ⟨100⟩-axis are shown in fig 4 A remarkable discontinuity is observed for CeZn at T_N, which is characteristic of a first-order transition The magnetic resistivity ρ_m for Ce was determined by subtracting the resistivity

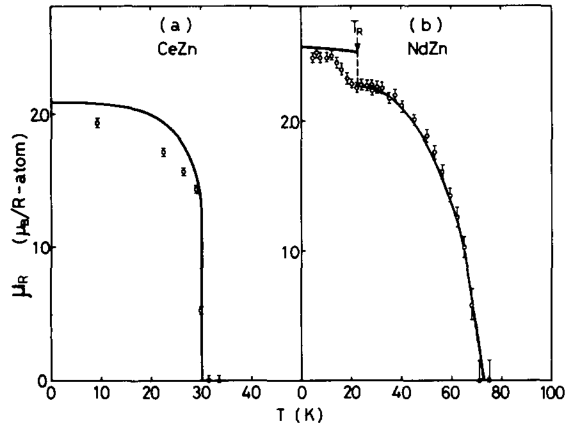

Fig 3 Temperature dependence of magnetization deduced from the $(10\frac{1}{2})$ magnetic reflection for (a) CeZn and (b) NdZn The solid lines are the calculated ones using the Hamiltonian of eq (1)

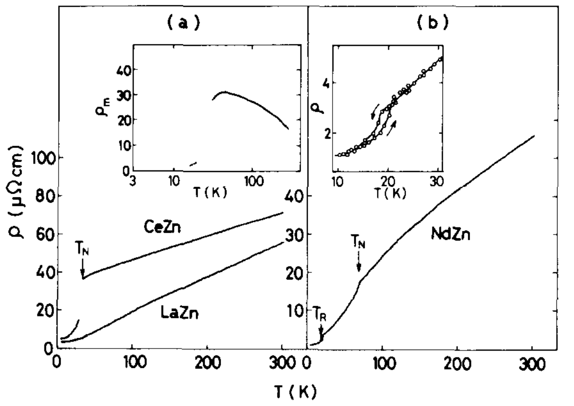

Fig 4 Electrical resistivity ρ as a function of temperature T for (a) LaZn and CeZn, and (b) NdZn along the [100]-axis

of LaZn The temperature dependence of ρ_m is given in the insertion of fig 4a The occurrence of a maximum in ρ_m can be ascribed to an incipient Kondo effect on the CEF split multiplet of Ce This result is in agreement with that obtained by Pierre et al [4] The ρ vs T curve of NdZn reveals (1) an anomaly at T_N corresponding to a second-order transition and (2) a well defined first-order transition at T_R with a thermal hysteresis of ≈2 K as shown in the insert of fig 4b, suggesting a magnetic order–order transition

In order to obtain direct information on the order–order transition, we measured the magnetization curves along the ⟨100⟩, ⟨110⟩ and ⟨111⟩-axes above and below T_R (see fig 5) The

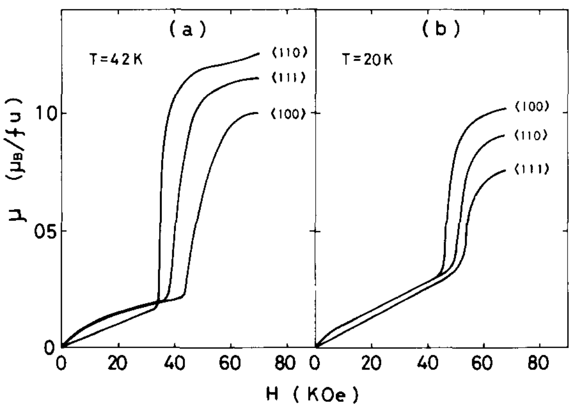

Fig 5 Magnetization vs magnetic field curves of NdZn at (a) 4 2 K < T_R and (b) 20 K > T_R, where T_R is the reorientation temperature

metamagnetism below T_R can be interpreted by a non-collinear model with the Nd moments of $2.52\mu_B$ directed to the ⟨110⟩-axes (fig 1b), assuming that the Nd moments hold in the ⟨110⟩ directions even though the field is applied to any directions because of strong magnetic anisotropy The magnetic structure above T_R can be also understood by a non-collinear one with the Nd moments along the ⟨111⟩-axes (fig 1c) as discussed by Kitai et al [5] as well These results give further confidence in the triple q to double q transition proposed by Morin and de Combarieu from the specific heat measurements [7]

3. Discussion

As is evident from fig 2, the form factor is weakly anisotropic in CeZn and the Ce moment is deduced as $1.91\mu_B$ This indicates that Γ_8 is the ground state However, it is to be noted that the form factor decreases more sharply with increasing $\sin\theta/\lambda$ than that expected from the ground state $|\frac{5}{2}, \frac{5}{2}\rangle$, which could be realized in the ordered state at 8 K On the other hand, the form factor of Nd is more anisotropic and it is possible to see that when the scattering vector is near the ⟨100⟩ direction, the observed magnetic form factors are larger than those near the ⟨110⟩ direction This simply suggests that the 4f electron density along the ⟨110⟩ direction is wider than along the ⟨100⟩ direction Then, the domain distribution was confirmed to be uniform from the intensity measurements of some equivalent $\{\frac{1}{2}03\}$ magnetic peaks in the $(h0l)$ plane, within our experimental errors

As an attempt to explain the temperature dependence of magnetizations, we introduce the effective single-ion Hamiltonian as follows

$$\mathcal{H} = B_4(O_4^0 + 5O_4^4) + B_6(O_6^0 - 21O_6^4) - g\mu_B J H_{ex} - G_1(\langle O_2^0\rangle O_2^0 + 3\langle O_2^2\rangle O_2^2) \quad (1)$$

Here, B_n is the CEF parameter, H_{ex} is the bilinear exchange field which is given by $H_{ex} = ng\mu_B\langle J\rangle$ and $n = \theta^*/C$, and the last term expresses the magnetoelastic and/or biquadratic exchange interactions The Ce moment is estimated as $2.05\mu_B$ at 8 K in CeZn using $\theta^* = 32$ K, $G_1 = 0.22$ K and $B_4 = 0.18$ K ($\Delta = 65$ K) according to Pierre et al [4] The calculated value is considerably larger than $1.91\mu_B$ obtained in this work This difference might be attributed to the Kondo moment compensation, that is the Kondo-type antiparallel coupling between the 4f moments and conduction electron spins Furthermore, we cannot satisfactorily understand the M vs T curve of CeZn shown in fig 3a, in which the solid line is the calculated value using the Hamiltonian of eq (1)

On the other hand, the Nd moment is calculated to be $2.60\mu_B$ at 4.2 K using $B_4 = 2.4\times10^{-2}$ K and $B_6 = 1.4\times10^{-3}$ K, $\theta^* = 90$ K and $G_1 = 0$, which is in fairly good agreement with the observed value of $2.52\mu_B$ The moment direction at 4.2 K is deduced to be along the ⟨110⟩-axes and above $T_R = 24$ K, the Nd moments are directed to the ⟨111⟩-axes under the condition which minimizes the free energy The Nd moment calculated is shown by the solid line in fig 3b The agreement between the calculated curve and experimental points is fairly good, but the calculated T_R is a little higher than the observed $T_R \approx 18$ K This suggests that not only G_1 but also G_2 terms must be taken into account, where the G_2 term is expressed by $G_2(\langle 0xy\rangle 0xy + \langle 0yz\rangle 0yz + \langle 0zx\rangle 0zx)$

On the other hand, for better understanding magnetism in CeZn, the Kondo effect should be considered since T_K is not negligible compared to the CEF splitting

References

[1] D Schmitt, P Morin and J Pierre, J Magn Magn Mat 8 (1978) 249
[2] P Morin and J Pierre, Phys Stat Sol (a)30 (1975) 549
[3] H Kadomatsu, H Tanaka, M Kurisu and H Fujiwara, Phys Rev B33 (1986) 4799
[4] J Pierre, A P Murani and R M Galera, J Phys F11 (1981) 679
[5] T Kitai, H Fujii, T Okamoto and Y Hashimoto, Solid State Commun 52 (1984) 407
[6] M Blume, A J Freeman and R E Watson, J Chem Phys 37 (1962) 1245
[7] P Morin and A de Combarieu, Solid State Commun 17 (1975) 975