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CRYSTAL-FIELD SPLITTING IN Pr DIDEUTERIDE

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CRYSTAL-FIELD SPLITTING IN Pr DIDEUTERIDE

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Abstract

From inelastic neutron scattering experiments, it is concluded that the crystal-field splitting in $\text{PrD}_{1.95}$ is 41 meV. Because of this high value, the antiferromagnetic ordering below $T_N = 2.3$ K is ascribed to a magnetic ground state, probably Γ_5 , of the Pr^{3+} ions.

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The heavier f.c.c. rare-earth dideuterides, RED_{2+x} ($x > 1.95$), are well known to order antiferromagnetically (AF) /1/. Transitions from commensurate into incommensurate structures occur for concentrations $\text{D}/\text{RE} > 1.95$, due to the occupation of the octahedral (O) sites for D/RE above 1.95 /1,2/. Recently it was shown by us that also $\text{CeD}_{1.95}$ and $\text{PrD}_{1.95}$, where the O sites are still vacant, order AF below T_N equal to 6.2 and 2.3 K, respectively /3,4/.

Particularly the magnetic order of $\text{PrD}_{1.95}$ is of interest in view of the possible nonmagnetic ground state, Γ_1 , of the Pr^{3+} ion. Therefore, we have investigated the crystal-field splitting in $\text{PrD}_{1.95}$ by means of inelastic neutron experiments. The

experiments were carried out at the triple-axis machines HB2 and HB3 at the HFIR, ORNL, using constant Q scans ($Q = 1.8, 2.6$ and 3.5).

Fig. 1 shows the excitation spectrum at 10 K ($Q = 2.6$). Three clear excitations are observed at 9, 20 and 41 meV, in agreement with the previous work of Knorr et al. /5/. However, in contrast to these authors we believe that only the 41 meV peak arises from crystal-field excitations between the ground state and the first excited state. Fig. 2 shows the temperature dependence of the 20 and 41 meV peaks for 10 and 170 K. The integrated intensities, I , for the various temperatures are summarized in Table I. Since the intensities of the 9 meV (not shown) and 20 meV peaks slightly grow with increasing temperatures, these peaks are ascribed by us to the high density of states at the zone boundary for the TA and LA phonons, respectively. These values agree with the dispersion curves obtained on single crystals of Ce dideuteride /6/, which is only slightly lighter than $\text{PrD}_{1.95}$. On the other hand, in Fig. 2 the intensity of the 41 meV peak is seen to decrease by raising the temperature. As follows from Table I, the intensity at 295 K is reduced by a factor of nearly two compared with the 10 K data. Since its intensity decreases also with increasing momentum transfer, the 41 meV is attributed to crystal-field excitations.

In view of this very high value of the crystal-field splitting, the ordering temperature $T_N = 2.3$ K seems to be an order of magnitude too large for a van Vleck induced magnetic ordering. Therefore we believe that the ground state of the Pr^{3+} ions has to be magnetic, probably Γ_5 , whereas the nonmagnetic ground state, Γ_1 , seems to be unlikely.

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TABLE I

T[K]	I(20 meV)	I(41 meV)
10	555(37)	300(36)
70	662(54)	285(50)
170	694(74)	241(45)
295	—	164(27)

Table 1: Temperature dependence of the integrated intensities of the 20 and 41 meV peaks in $\text{PrD}_{1.95}$.

FIGURE CAPTIONS

Fig. 1 Excitation spectrum of $\text{PrD}_{1.95}$ at 10 K
($E_{\text{final}} = 14.9 \text{ meV}$).

Fig. 2 Temperature dependence of the 20 meV (a) and
41 meV (b) peaks in $\text{PrD}_{1.95}$.

Fig 1

Fig 2

(a)

COUNTS VS ENERGY

(b)

COUNTS VS ENERGY