Proceedings of the European Conference "Physics of Magnetism '99", Poznań 1999

CRYSTAL-FIELD ORIGIN OF THE GIANT MAGNETOCRYSTALLINE ANISOTROPY OF $PrRu_2Si_2$

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$PrRu_2Si_2$ shows ferromagnetism below 14 K with the ordered moment of $2.7\mu_B$. It exhibits an enormous magnetic anisotropy at 4.2 K with the anisotropy field of about 400 T. We have attrributed the magnetism of $PrRu_2Si_2$ to the Pr^{3+} ions. Thus we performed calculations of the fine electronic structure of the Pr^{3+} ion in the tetragonal symmetry, relevant to $PrRu_2Si_2$, taking into account crystal-field and inter-site exchange interactions. Our calculations reproduce well the zero-temperature moment, the single-crystalline magnetization curves, and giant anisotropy field as 400 T. The magnetocrystalline-anisotropy energy K_1 of 59 J/cm^3 is the largest known anisotropy — the anisotropy energy of the $Nd_2Fe_{14}B$ supermagnet amounts to 12.5 J/cm^3 only. Unfortunately, this giant anisotropy is confined to low temperatures only which prohibits its technical applications in the permanent-magnet industry.

PACS numbers: 71.70.–d, 71.20.Eh

$PrRu_2Si_2$ exhibits a giant magnetocrystalline anisotropy about 400 T [1, 2]. The aim of this contribution is to find the origin of this giant anisotropy, in the current literature as the source of the anisotropy three main factors are mostly discussed [1]: the anisotropic exchange, the hybridization, and the single-ion mechanism. We are by years in favour of the single-ion mechanism [3, 4], via the crystal-field effect, of the magnetocrystalline anisotropy and it was very intriguing that the authors of Ref. [1] have claimed that such big anisotropy cannot be obtained as the crystal-field effect. Moreover, our interest to $PrRu_2Si_2$ has raised as the preceding compound, $CeRu_2Si_2$, exhibits the pronounced heavy-fermion phenomena. It turns out that we can provide the crystal-field explanation for this giant magnetocrystalline anisotropy, that provides also a quite consistent description of other magnetic and electronic (m–e) properties.

$PrRu_2Si_2$ is an intermetallic compound. It crystallizes in a tetragonal structure of $ThCr_2Si_2$-type and exhibits the ferromagnetic ordering below 14 K [1, 2, 5, 6]. The magnetization curves measured on a single-crystalline sample [1], which reveal this giant magnetocrystalline anisotropy, are shown by experimental points in

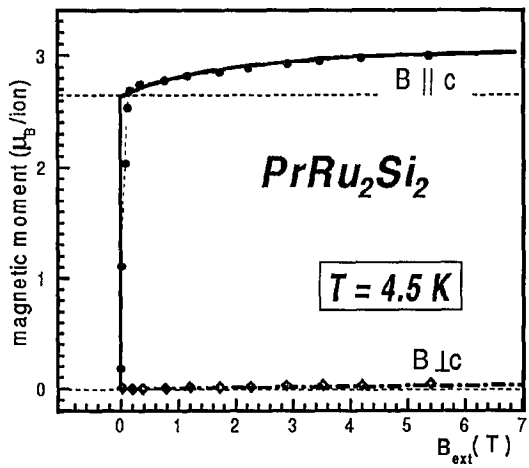

Fig. 1. Calculated magnetization curves at 4.5 K (lines) on single-crystalline $PrRu_2Si_2$ along main crystallographic directions of the tetragonal unit cell. The points denote experimental results from Ref. [2].

Fig. 1. From this figure one can see that the magnetic moment is directed along the tetragonal c-axis and it amounts to $2.7\mu_B$/f.u. [1, 2]. Inelastic-neutron-scattering (INS) experiments [2, 5] reveal local excitations with the energies of 2.25 and 29 meV providing a strong argument for the existence of the localized CEF-like levels.

In order to understand the magnetic properties of $PrRu_2Si_2$ we have applied the individualized-electron model for rare-earth compounds [7, 8]. According to this model f electrons are keeping their individuality also being placed into intermetallic, in which the energy of localized $4f$ electrons is sufficiently deep below the Fermi level and ionic compounds. In intermetallics there are also itinerant electrons originating from outer electrons of all constituting elements. Thus in intermetallics there coexists a few (here only 2 are specified) physically important electronic subsystems: f electronic subsystem(s) and conduction–electron (c–e) subsystem. In the situation without the hybridization of $4f^n$ states and c–e states these two subsystems are described by completely different theoretical approaches: localized-electron and itinerant-electron models. Ru and Si are not magnetically and electronically, below at least 1 eV, active. The f electron subsystem exhibits a discrete energy spectrum associated with bound states of the highly-correlated electron system f^n, f^2 in the present case, whereas the conduction electrons are forming a band. The interactions between them, important for the low-temperature magnetism, go basically via the spin polarization [7].

We have attributed the magnetic properties of $PrRu_2Si_2$ to be predominantly due to the Pr ions. In Fig. 2 the energy level scheme of the Pr^{3+} ion in $PrRu_2Si_2$ related to the lowest Hund's rules ground multiplet $^3H_4$ is presented. It contains 5 singlets and 2 doublets as one expects for the action of the tetragonal CEF interactions [4, 9]. The 9-fold degeneracy is fully removed, as the ground state is a singlet in the form $\Gamma_{t1}^{(1)} = 0.70|+4\rangle + 0.11|0\rangle + 0.70|-4\rangle$.

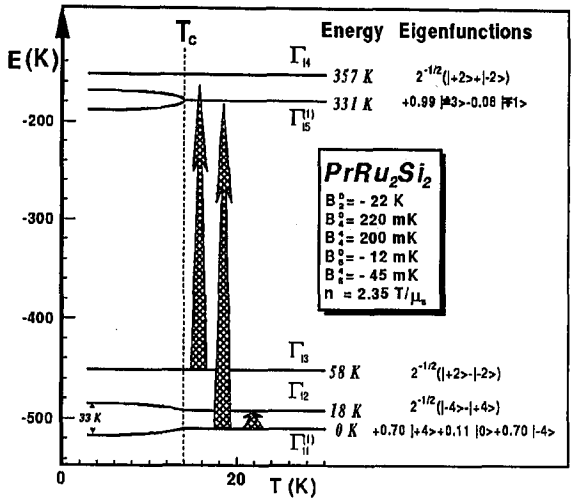

Fig. 2. Energy level scheme of the Pr^{3+} ion in $PrRu_2Si_2$. Under the action of the tetragonal CEF interactions the 9-fold degenerate ground multiplet is split into 5 singlets and 2 doublets — here only 6 lowest states are shown, next are at 991 K (dublet, $\Gamma_{t5}^{(2)}$) and 1507 K (singlet, $\Gamma_{t1}^{(2)}$). Three arrows indicate the allowed INS excitations. They are assigned to those experimentally detected in Refs. [2] and [5].

The magnetism of this singlet can be relatively easily induced by spin-dependent interactions despite its general non-magnetic character. Such charge-formed ground state is necessary in order to reproduce the experimentally observed large magnetic moment as well its direction. The general Hamiltonian has the following form [10]:

$$H = \sum_{n=1}^{6} \sum_{m=0}^{n} B_n^m \hat{O}_n^m + ng^2\mu_B^2 \left(-J\langle J\rangle + \frac{1}{2}\langle J\rangle^2 \right) + g\mu_B J B_{\text{ext}}.$$

The second term accounts for the intersite spin-dependent interactions and is indispensable for the formation of the magnetic state. The last one allows us to calculate the effect of external field, for calculation of the paramagnetic susceptibility.

The formation of the magnetic state at 14 K is seen in Fig. 2 as the discontinuity in the temperature dependence of the energy-level positions. At 0 K $PrRu_2Si_2$ experiences the molecular field of 6.4 T originating from the intersite RKKY interactions. Two arrows indicate the allowed INS excitations that have been indeed measured.

In Fig. 3 the full magnetization curves, at 4.2 K, calculated within our CEF approach are presented. These calculations reveal that the giant anisotropy, even above 400 T, can be realized by the crystal-field interactions. Unfortunately, this anisotropy occurs only at low temperatures due to the low value of the Curie temperature of 14 K.

In conclusion, we have shown that the giant anisotropy field as large as 400 T observed in $PrRu_2Si_2$ [1, 2] can be accounted for by crystal-field interactions. We are quite convinced in our crystal-field explanation for this giant

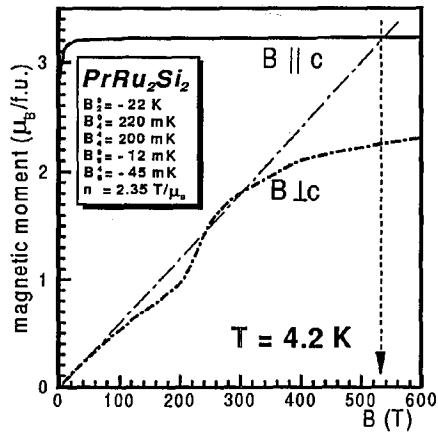

Fig. 3. Full magnetization curves, at 4.2 K, calculated within the CEF approach. It reveals that the giant anisotropy, even with larger anisotropy field than 400 T, can be caused by crystal-field interactions.

magnetocrystalline anisotropy as it provides also a quite consistent description of other magnetic and electronic properties, like temperature dependence of the paramagnetic susceptibility and of the heat capacity. Moreover, the observation of localized CEF-like states by different experimental techniques closes the internal consistency of our approach. Experimentally-observed energy separations by the inelastic neutron scattering [2] are in good agreement with the obtained energy level scheme. The calculated neutron transition probabilities are in agreement with the INS spectrum obtained by Mulders et al. [2].

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