Quadrupolar ordering in $PrFe_4P_{12}$ skutterudite

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Rare-earth filled skutterudite compounds are intensively studied in the last few years because of their diverse and complex behavior. $PrRu_4P_{12}$ shows a metal-insulator phase transition at about 60K which is accompanied by also a structural transition. Skutterudite compounds with Nd or Eu show ferromagnetism, while exotic superconducting phases were found in $PrOs_4Sb_{12}$ discovering the first Pr-based heavy fermion superconductor.

We are particularly interested in the nature and main property of the ordered phase of $PrFe_4P_{12}$ skutterudite. This compound shows a non-magnetic phase transition at 6.5K which can be suppressed by external magnetic field. The crystal field level scheme of $PrFe_4P_{12}$ has not been determined yet experimentally, although the low-lying scheme is responsible for the low-temperature behavior, e.g. the possible orderings. The CEF splittings in the $PrRu_4P_{12}$ and $PrOs_4Sb_{12}$ are clearly known by neutron diffraction experiments. But the interpretation of the neutron scattering spectra of $PrFe_4P_{12}$ is not unambiguous: polycrystalline samples do not show clear peaks and the peaks appearing in the single crystalline samples spectra become clear only below the transition temperature. Thus, the question of the CEF scheme is left open. The most fascinating recent experimental finding on the $PrFe_4P_{12}$ skutterudite is the appearance of a high-field phase very sharply located around the (111) magnetic field direction [1]. This result can be a key point in the identification of the low-energy level scheme and also the low-field ordered phase. Additionally, it was also found that the electrical resistivity shows an enhancement around the (111) field direction [2].

We found that only the Γ_1–$\Gamma_4^{(1)}$ low-lying scheme gives a level crossing point in the ground state uniquely for the (111) field direction which can be responsible for the appearance of the high-field phase. Accepting that the low-field phase is the antiferro-quadrupolar ordered phase of the Γ_3 moments, which is supported by experimentally, we examined this AFQ ordering model in the case of tetrahedral symmetry with the Γ_1–$\Gamma_4^{(1)}$ low energy scheme. We found that this model can lead to the appearance of the new high-field phase for $\mathbf{H}\|(111)$ in the case when the energy separation between the singlet and triplet states is small. With the introduction of ferro-type interactions between the dipoles and $\mathcal{T}^{\beta}$ octupoles the measured phase boundary can be reproduced qualitatively. Changing the ferro-type interaction parameters, either two separated phases or continuous phase boundary can be obtained. We found parameter sets where the two phases have the same $\mathbf{q} = (1, 0, 0)$ ordering vector. We also found another set where the low-field phase has $\mathbf{q} = (1, 0, 0)$ while the high-field phase has $\mathbf{q} = 0$.

We calculated the electrical resistivity from the crystalline electric field states. With this very simple picture we can reproduce qualitatively the measured resistivity enhancement around the (111) field direction near the level crossing point at low temperatures.

References

[1] T. Tayama et al., J. Phys. Soc. Japan **73**, 3258 (2004).
[2] E. Kuramochi et al., Acta Physica Polonica B**34**, 1129 (2003).