Magnetic states of Rare Earth Metals at High Pressure

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The cycle of papers consists of four works.

The rare earth metal compounds are strongly correlated electronic systems, which demonstrate a reach variety of physical phenomena, including Kondo effect, valence instability, non-conventional superconductivity, quantum critical phenomena, giant magnetostriction, various magnetic states. The indirect nature of magnetic exchange interactions between localized moments by means of conduction electrons in combination with crystal field effects, hybridization with electronic states of outer electronic shells and additional factors leads to formation of numerous magnetic states, from simple ferromagnetic to complex modulated antiferromagnetic ones with incommensurate character [1-4].

In addition, the elemental rare earth metals demonstrate a rich structural polymorphism under variation of high pressure, but features of the long range magnetic order in pressure induced phases were not well studied yet. The important insights into microscopic mechanisms of the formation of physical properties could by reached by high pressure studies, since different response of competing factors on pressure application allows to distinguish their contributions. Due to extremely high complexity of neutron scattering studies at high pressure, the pressure behavior of magnetic states of rare earth metals and their modification caused by structural phase transitions remains poorly explored.

The present cycle of works includes results of detailed and systematic studies of magnetic and crystal structure of model representatives of rare earth metal family: terbium Tb [1], gadolinium Gd [2], holmium Ho [3] and thulium Tm [4] by means of neutron diffraction in a wide pressure and temperature ranges. In Tb, a structural phase transition from the initial hexagonal closed packed (hcp) structure to the Sm-type rhombohedral structure was observed at pressures above 4 GPa [1]. It is accompanied by the ferromagnetic (FM) – antiferromagnetic (AFM) phase transition as well. For the first time the magnetic structure of the rhombohedral pressure induced phase of Tb was determined. It has a complex modulated commensurate AFM character. Based on performed DFT calculations, a theoretical model of the pressure induced structural phase transition was constructed.

In the case of gadolinium, being a very strong neutron absorber, the isotopically substituted sample ^{160}Gd (isotope enrichment of 95 %) was studied. At ambient conditions, a coexistence of the hcp phase and Sm-type rhombohedral phase was already observed [2]. Under high pressure, the hcp phase was suppressed gradually and above 6 GPa only the Sm-type phase was found. The long range ferromagnetic order formed in the hcp phase is also suppressed rapidly was a pressure coefficient of the Curie temperature dT_C/dP = -6.3(1) K/GPa. In the Sm-type phase below the Néel temperature of about 100 K (evaluated for 9 GPa), the long range antiferromagnetic order is formed, similar to those found for Tb.

In the hcp phase of Tm, at ambient pressure below T_N = 52 K the incommensurate magnetic state with a longitudinal sinusoidal modulation and propagation vector $q = (0, 0, q_z)$ was formed. The q_z value increases on cooling from 0.279(2) and reaches the commensurate value of 2/7 at the temperature T_1 = 24 K. The new magnetic arrangement is composed of structural blocks involving three or four hexagonal layers with magnetic moment directions parallel within a given block and oppositely alternating from block to block along the c axis. Under high pressure, the T_N value increases slightly up to 57 K at 5.5 GPa with a pressure coefficient dT_N/dP = 0.9 K/GPa. The pressure dependence of the T_1 temperature is more pronounced with a pressure coefficient dT_I/dP = 2.9 K/GPa. The arrangement of commensurate magnetic structures formed below T_1 is strongly pressure dependent. A formation of the complex magnetic states composed of structural blocks involving three or four hexagonal layers with magnetic moment signs alternating from block to block along the c axis with q_z = 7/23 at 2 GPa and q_z = 7/22 at 5.5 GPa was observed [3].

In contrast to Tm, in Ho at ambient pressure below T_N = 127 K the incommensurate helical state with a propagation vector $q = (0, 0, q_z)$ is evidenced. On temperature lowering, the q_z value decreases from 0.270 (at 125 K) to 5/26 at T_L = 20 K. Below the lock-in transition temperature T_L, a commensurate cone structure is formed. Upon lattice compression, a gradual suppression of the low temperature cone structure was found and just a helical state was observed above 4 GPa. The lock-in transition temperature grows up to about 50 K (at 8.7 GPa) with a coefficient of $dT_L/dP \approx$ 3.4 K/GPa and q_z component evolves towards 1/4. Simultaneously, the Néel temperature decreases with a pressure coefficient $dT_N/dP \approx$ -2.2 K/GPa. The magnetic P-T phase diagram of Ho is constructed.

The obtained results are important for formation of contemporary basics in the field of rare earth metals magnetism and they have stimulated further research of international scientific groups in this direction.

List of papers

1. D. P. Kozlenko, V. Yu. Yushankhai, R. Hayn, M. Richter, N. O. Golosova, S. E. Kichanov, E. V. Lukin, and B. N. Savenko, Pressure-induced structural transition and antiferromagnetism in elemental terbium, Physical Review Materials 5, 034402 (2021), DOI: 10.1103/PhysRevMaterials.5.034402.

2. N.O. Golosova, D.P. Kozlenko, E.V. Lukin, S.E. Kichanov, B.N. Savenko, High pressure effects on the crystal and magnetic structure of 160Gd metal, Journal of Magnetism and Magnetic Materials, 540, 168485 (2021). https://doi.org/10.1016/j.jmmm.2021.168485

3. N.O. Golosova, D.P. Kozlenko, E.V. Lukin, S.E. Kichanov, B.N. Savenko, Pressure tuning of magnetic states in elemental thulium, Journal of Magnetism and Magnetic Materials, 560, 169662 (2022), https://doi.org/10.1016/j.jmmm.2022.169662

4. N.O. Golosova, D.P. Kozlenko, E.V. Lukin, S.E. Kichanov, B.N. Savenko, High pressure effects on magnetic states of elemental holmium, Journal of Magnetism and Magnetic Materials, 580, 170971 (2023), https://doi.org/10.1016/j.jmmm.2023.170971