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# Exchange coupling in Eu compounds

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**Abstract**

Using ab initio electronic structure calculations employing the LDA + $U$ method we have investigated the exchange coupling in materials containing Eu in $Eu^{2+}$ formal valency including: (i) insulating ferromagnets (EuO, EuS, EuSe, EuTe), and (ii) elemental metal BCC Eu. The total energies calculated with constrained magnetic structures were mapped on the Heisenberg Hamiltonian in order to investigate the thermodynamic properties. The mechanisms of inter-site exchange, which differ between the studied systems, are discussed.


*Keywords:* Rare earth; Exchange interaction; Electronic structure

The magnetism of most rare-earth (RE) materials is governed by localized magnetic moments of 4f orbitals interacting indirectly through the delocalized valence electrons, which gives rise to a variety of magnetic behaviors [1]. The ab initio electronic structure methods based on density functional theory (DFT) [2] and the standard semi-local approximations [3,4] have notorious problems in dealing with the strong correlations within the 4f shell, in particular the filling of these state is often incorrect due to their unphysical location at the Fermi level. Here we use the LDA + $U$ functional [5] which achieves the splitting between the occupied and unoccupied f band through an additional orbital-dependent term and yields proper band filling.

The Wien2k [6] implementation of the full-potential linearized augmented-plane-waves method (FLAPW) with LDA + $U$ functional is employed to study the exchange interactions in different types of materials containing Eu: (i) insulating Eu monochalcogenides EuX, and (ii) metallic BCC Eu. The common feature of these systems is the occupation of Eu 4f shell with seven electrons. The many-body groundstate of a half-filled shell is uniquely determined by the intra-shell exchange interaction (1st Hund's rule) and thus

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the multiplet effects are not important for low energy physics, which makes the LDA + $U$ description quite realistic.

The Eu monochalcogenides crystallizing in rock-salt structure exhibit ferromagnetic ordering unusual for an insulator (EuSe and EuTe only at elevated pressure) [7]. We have mapped ground-state energies obtained for different collinear magnetic structures onto Heisenberg Hamiltonian with exchange up to second neighbors and evaluated the ordering temperatures using the RPA approximation [8]. The calculated Curie and Néel temperatures as a function of lattice constant are shown in Fig. 1. The trend toward ferromagnetism in the compounds with decreasing atomic number of the chalcogen atom and with applied pressure is well captured by the calculations. The results are qualitatively similar but are rather sensitive to the value of $U$ [9], which points to an important role of kinematic exchange effects and which is to be contrasted with our next example.

Elemental Eu crystallizing in body-centered cubic (BCC) structure is a metal with a spin spiral magnetic groundstate below 91 K [13,14]. We have used the frozen magnon (spin spiral) approach where no limitations on the spatial range of exchange interactions are imposed. The exchange constants in reciprocal space representation, which correspond directly to the energies of the spin-spiral states, are shown in Fig. 2. Both the propagation vector $\mathbf{q}$ of the ground state spin-spiral and the ordering temperature of 112 K obtained with the RPA formula [15] compare well to the experiment and calculations of Turek et al. [15]. The origin of the spin-spiral groundstate is discussed in Ref. [12].

Contrary to EuX case the exchange constants in BCC Eu are insensitive to the value of $U$. This fact points to different mechanism of inter-site exchange. In a metallic system such as BCC Eu the atomic f–d exchange gives rise to inter-site coupling through the Ruderman–Kittel–Kasuya–Yoshida (RKKY) mechanism. The f–d exchange depends only of the polarization of the f shell, which is independent of $U$ over the range of realistic values. The Bloembergen–Rowland mechanism, which is the analog of RKKY for insulating systems, is much weaker and kinematic effects, which involve hopping to and from f states, become important. The contribution of the kinematic effects depends on the energy of the f bands relative to the rest of the system, which is how the dependency on $U$ enters.

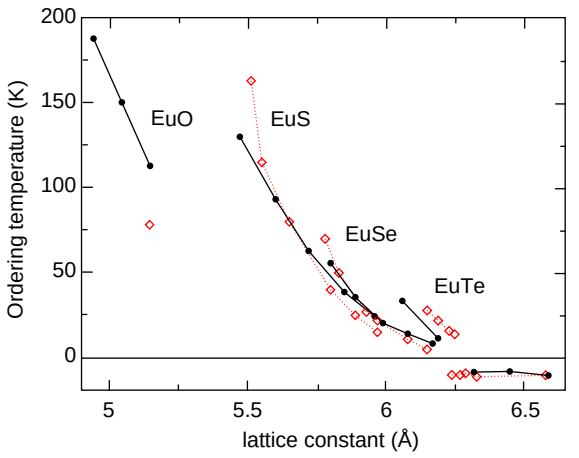


Fig. 1. The Curie and Néel (shown negative) temperatures of Eu monochalcogenides calculated with $U = 6$ eV (full dots) compared to experimental values [10,11] (diamonds). The lines serve as guides to eyes.

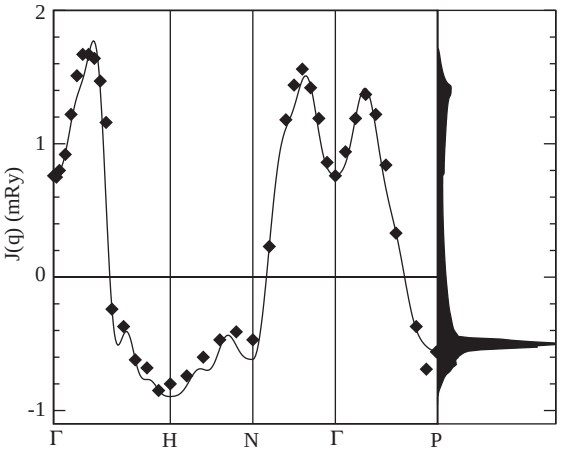


Fig. 2. The q-dependent exchange constants calculated along the high symmetry lines (diamonds) and the fit obtained from values on a regular mesh throughout the Brillouin zone. Density of modes is shown in the right panel. For details see Ref. [12].

In conclusion, using Heisenberg Hamiltonian with coupling constants obtained from electronic structure calculations we were able to obtain magnetic ground states and ordering temperatures of both metallic and insulating systems containing Eu in agreement with experiment.

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