

Theoretical study of magnetism in Cd₆Tb 1/1 periodic cubic approximant of a Tsai-type quasicrystal

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From the beginning of the discovery of quasiperiodicity in intermetallic compounds magnetic properties were in the center of interests. The possibility of exotic behaviour due to intrinsic quasiperiodicity was high. The magnetic structure was investigated in numerous phases bearing Rare-Earth elements as possessing localized magnetic moments. The first long-range magnetic order was discovered in Im-3 Cd₆Tb 1/1 cubic approximant crystal [1]. Tb atoms are grouped in icosahedral shell of Tsai-type rhombictriacontahedral clusters being structural units. The orientation of magnetic moments for vertex- and body-centered clusters is antiparallel constituting antiferromagnetism. The transition temperature is 24 K with effective magnetic moment equal to 9.8 μ_B . The aim of the study is to deliver the theoretical justification on the existence of long-range magnetic order in Cd₆Tb based on the electronic structure calculations.

Density functional theory calculations were performed using Quantum Espresso [2,3]. PBE exchange correlation functional was chosen with PAW pseudopotentials. Energy cutoffs for wavefunctions and charge density were set to 125 Ry and 625 Ry, respectively. Selfconsistent calculations were done in Γ point only and density of states on a 4³ k-point grid.

Tb-4f electrons form atomic-like states near the Fermi energy which slow the convergence and require large energy cutoffs. Firstly, we considered the experimental crystal structure. Scalar-relativistic calculations fail to predict the correct ground state because ferromagnetic configuration is slightly favoured by 0.1meV/atom and the magnetic moments equal 6.0 μ_B per Tb atom are too small. Nonmagnetic structure has higher total energy by 55 meV/atom. The Fermi surface is built mainly from Tb-4f states with small contributions from Tb-5d and Cd-5p. DFT+U calculations using Cococcioni's simplified approach [4] with single parameter $U = 8$ eV increased the magnetic moments by 0.4 μ_B . The inclusion of the spin-orbit coupling favours antiferromagnetic configuration with a large energy difference of over 300 meV/atom against the ferromagnetic one. Magnetic moments converged slowly to approximately 8.2 μ_B . Then we used the cell with relaxed volume and fixed experimental atomic positions. The lattice constant increased by 0.9 a_B and the magnetic moments converged to 9,2 μ_B . Atomic position relaxation was omitted because of large computational time. Overall, our calculation indicate the importance of the spin-orbit coupling and volume relaxation in the confirmation of the antiferromagnetism in the Cd₆Tb cubic approximant.

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This work was supported by the National Science Centre Poland, grant no. UMO-2022/47/I/ST3/00340 and by Polish high-performance computing infrastructure PLGrid (Cyfronet AGH) grant no. PLG/2024/017306.