Interplay between structure and magnetism in the magneto-caloric material $Gd_5(Si_xGe_{1-x})_4$

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Extended Abstract

Materials that exhibit the magneto-caloric effect (MCE) are of great interest as their temperature can be manipulated through the application and removal of magnetic fields. Their main application is in magnetic refrigeration, such as in the adiabatic demagnetization of paramagnetic substances to achieve milli-Kelvin temperatures in laboratories around the world. Since induced paramagnetism is significant only at very low temperatures, the effective harnessing of magnetic entropy in paramagnets is limited to very low temperature applications. The use of magneto-caloric materials for near-room-temperature refrigeration requires magnetic ordering transitions near room temperature. While many materials qualify, the second-order paramagnetic-ferromagnetic (PM-FM) transition that can be triggered in ferromagnetic materials by the application of a magnetic field just above their zero-field Curie ordering temperature results in too gradual a change in magnetization to yield a significant MCE (the latter depends on dM/dT). The remarkable feature of the $Gd_5(Si_xGe_{1-x})_4$ class of materials is that their PM-FM transition coincides with a first-order martensitic structural transition [1]. The coupled, first-order magneto-structural transition (also known as magnetic-martensitic) induced by an applied magnetic field yields a sharp change in magnetic and structural entropy, both of which can be harnessed for magnetic refrigeration applications near room temperature. The martensitic transition involves large shear displacement (≈ 1Å) of Gd-containing nanoslabs relative to one another, resulting in the breaking (reforming) of Ge(Si)-Ge(Si) covalent bonds that provide connectivity between these slabs and the concomitant disappearance (appearance) of ferromagnetic ordering. The profound effect of this martensitic structural transition on the magnetism of this material is the focus of this study.

We explore the nature of the magnetic-martensitic phase transformation in $Gd_5(Si_xGe_{1-x})_4$ ($x \leq 0.5$) through x-ray magnetic circular dichroism (XMCD) and x-ray absorption fine structure (XAFS) measurements as a function of composition, temperature, applied magnetic field and pressure. XMCD results at the Gd L_3 and Ge K edges, which probe the Gd 5d and Ge 4p orbital character of the conduction band, are presented. The role of spin-polarization in these bands in mediating the exchange interactions between localized Gd 4f moments, and how these interactions are modified by the structural transition, will be discussed.

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[1] V. K. Pecharsky and K. A. Gschneidner Jr., Advanced Materials **13**, 683 (2001).

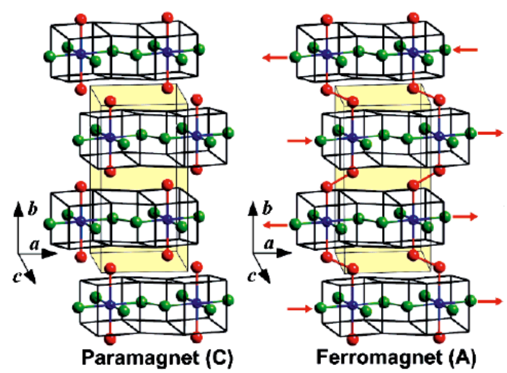

Fig. 1. Crystal structure modifications connected by a martensitic phase transformation. The arrows indicate the direction of slab displacement. The Gd-containing slabs are connected (disconnected) in the ferromagnetic (paramagnetic) phase. (adapted from Ref. 1).

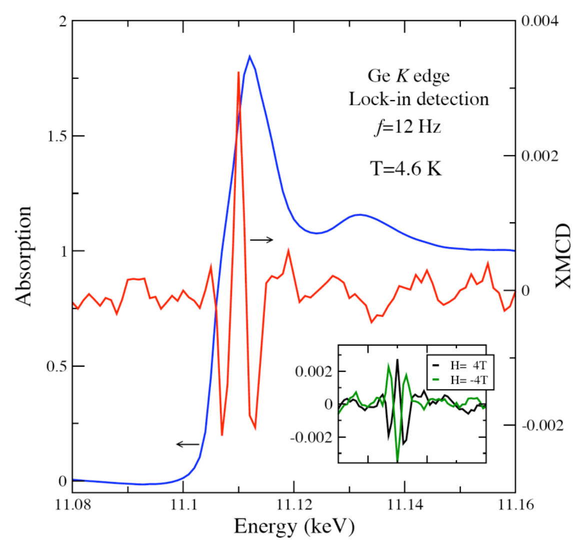

Fig. 2. Ge K-edge x-ray absorption and magnetic circular dichroism for x=0.125.