Complex magnetism and magnetoresistance of Gd-Tb-Dy-Ho-Lu hexagonal high-entropy alloy

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Hexagonal high-entropy alloys (HEAs) composed solely of rare earth (RE) elements [1–3] are special in this class of multicomponent metallic alloys because great similarity in the chemical properties of RE elements allows for an almost complete mutual solubility. Pure metals from the heavy half of the RE series possess a hexagonal close-packed (hcp) structure at room temperature (with the exception of Yb). The hcp lattice parameters a and c change only slightly along the RE series due to the lanthanide contraction of atomic radii r_i toward heavier RE elements, so that the associated lattice deformation, which contributes to the enthalpy of mixing ΔH_{mix}, can be to a good approximation neglected. The binary mixing enthalpies ΔH_{mix}^{ij} for any pair *i* and *j* of the heavy-RE elements are also zero, indicating that the elements mix ideally. This is by definition an ideal solid solution, in which the intermolecular forces are the same between every pair of molecular kinds and there is no enthalpy of mixing, $\Delta H_{mix} = 0$. Hexagonal HEAs produced from the heavy-RE elements are thus prototypes of an ideal HEA.

Great chemical similarity of the RE elements and their almost complete mutual solubility imply that the electronic properties of RE-based HEAs can be predictably tuned with composition, but since the RE elements possess disparate magnetic properties, their random mixing on an almost undistorted hcp lattice may result in unprecedented magnetic behavior. We have determined magnetic phase diagram and magnetoresistance of a Gd-Tb-Dy-Ho-Lu "ideal" hexagonal HEA. The phase diagram is very rich, containing an antiferromagnetic (AFM) state, a field-induced ferromagnetic (FM) state above the AFM-to-FM spin-flop transition, a low-temperature spin-glass state and a discontinuous (1st-order) metamagnetic transition at $T = 2$ K. The complex (H,T) phase diagram is a result of competition between the periodic potential arising from the electronic band structure that favors periodic magnetic ordering, the substitutional-disorder-induced random local potential that favors spin-glass-type spin freezing in random directions, the Zeeman interaction with the external magnetic field that favors spin alignment along the field direction and the thermal agitation that opposes any spin ordering. Magnetoresistance (Fig. 1) reflects complexity of the (H,T) phase diagram. Its temperature dependence can be explained by a continuous weakening and final disappearance of the periodic potential upon cooling that leads to the destruction of long-range ordered periodic magnetic structures. Magnetoresistance is large only at temperatures, where the AFM and field-induced FM structures are present and exhibits a maximum at the critical field of the AFM-to-FM transition. Within the AFM phase, the

magnetoresistance is positive with a quadratic field dependence, whereas it is negative with a logarithmic-like field dependence within the field-induced FM phase. At lower temperatures, the long-range periodic spin order "melts" and the magnetoresistance diminishes until it totally vanishes within the low-temperature spin glass phase. The magnetoresistance is asymmetric with respect to the field sweep direction, reflecting nonergodicity and frustration of the spin system.

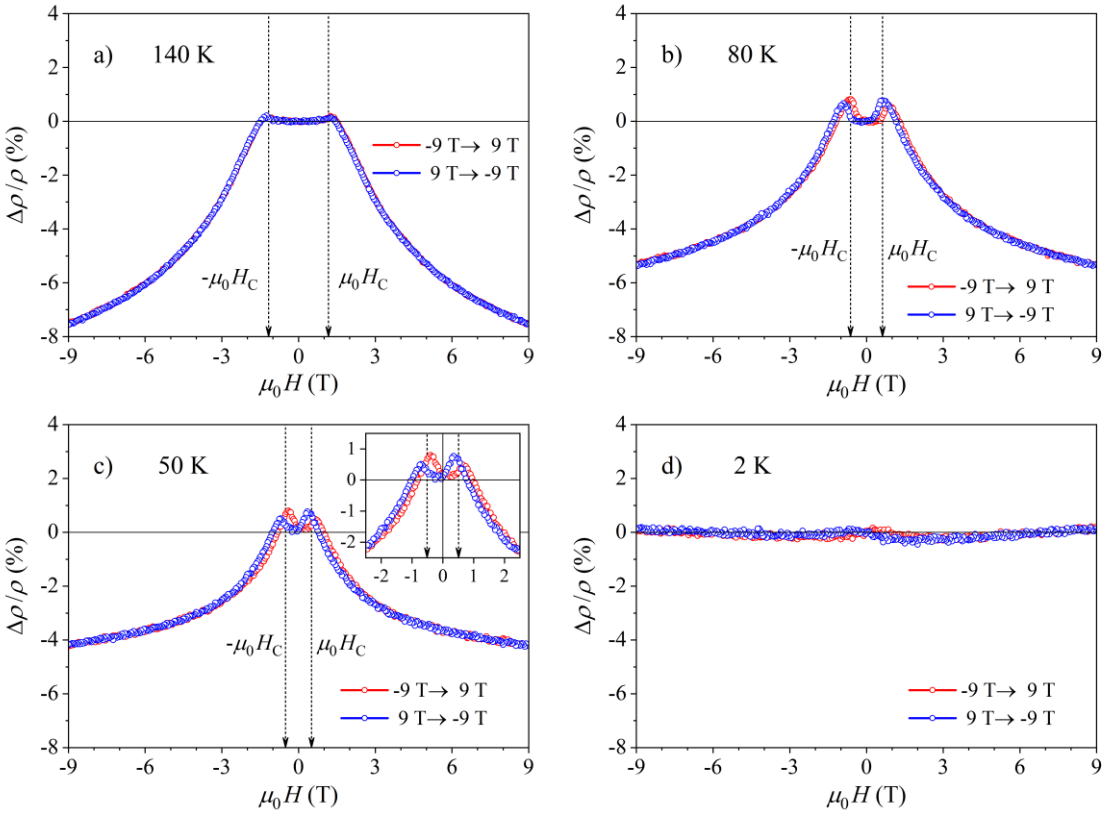

Figure 1. Magnetoresistance $\Delta\rho/\rho$ of the Gd-Tb-Dy-Ho-Lu hexagonal HEA at different temperatures below the Néel temperature T_N: **(a)** 140 K, **(b)** 80 K, **(c)** 50 K, and **(d)** 2 K. At each temperature, two $\Delta\rho/\rho$ curves were measured, where the first one was recorded by sweeping the field from –9 T to 9 T and the second one in a reversed sweep. Dashed vertical arrows mark the critical field values $\pm\mu_0 H_c$ at which the external field induces the AFM-to-FM spin-flop transition. The inset in panel (c) shows the asymmetry of the $\Delta\rho/\rho$ curves for the two field sweep directions on an expanded scale.

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