Interplay between charge localization and magnetic ordering in amorphous Gd_xSi_{1-x}

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Abstract

A new theoretical model and its experimental verification are presented, describing the anomalous transport and magnetic properties of amorphous gadolinium silicon. Our explanation reported here presumes high disorder of the system; this leads to a redistribution of electron density, causing regions with high electron concentration (drops) to appear. The magnetic ordering is more favourable in these drops than in the volume. To verify the true magnetic state of the drop (AFM or FM) the “local” experimental method, electron spin resonance, was used, together with transport measurements. Direct experimental observation of local ferromagnetic ordering in drops is reported.

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The anomalous transport and magnetic properties of amorphous gadolinium silicon has been the object of controversial discussion since 1996 [1,2]. There are three temperature ranges with different magnetic and transport properties. At \( T > 50 \text{K} \) the temperature dependence of conductivity in a-Gd_xSi_{1-x} coincides with that of its non-magnetic structural analogue Y_xSi_{1-x}. At \( T < 50 \text{K} \) a significant difference in their behaviour has been observed: in a-Gd_xSi_{1-x} the conductivity diminishes with temperature much quickly than in a-Y_xSi_{1-x}. This fact points to the magnetic nature of the phenomenon. Below 5K, the material shows a spin glass like freezing [2]. In the intermediate \( (5 \text{K} < T < 50 \text{K}) \) temperature range of the low field magnetization obeys qualitatively the Curie–Weiss law but with low Curie constant and effective temperature \( \theta \). Large negative magnetoresistance is found below 50K.

To explain these properties, we take into consideration the high disorder of the system, leading to a redistribution of electron density and causing regions with high electron concentration (drops) to appear. Magnetic ordering is more favourable in the drops than in the volume. There are however two different possibilities for the ordering: ferromagnetic (FM) or antiferromagnetic (AFM). To choose the real scenario and verify the true magnetic state of the drops, a “local” experimental method, electron spin resonance (ESR), was used, together with conductivity and Hall effect measurements.

According to the standard RKKY approach, local FM ordering of Gd moments appears in the drops at temperatures below \( T_{loc} \sim 50 \text{K} \). That corresponds to an effective Curie temperature \( \theta(x) \), estimated from measurements of magnetization \( \mu(T) \) at high \( T \). This value is typical for a dilute magnetic material: \( \theta(x) \approx x \theta(I) \). Fig 1 shows the temperature dependence of magnetization \( \mu \) obtained as a double integrated ESR line intensity. In contrast to low field magnetization measurements [2] we observe drastic growth of magnetization obtained from...
ESR ($B_0 \approx 3500\text{ Oe}$) at temperature below 50 K. This growth and obvious increase of ESR resonance field $B_{R_{\text{perp}}}$ for normal orientation of magnetic field relative to sample surface directly indicate the appearance of FM drops. According to Ref. [3]:

$$B_{R_{\text{perp}}} = B_0 + 4\pi fM;$$

$f$ is the part of FM ordered Gd atoms, $M$ is the saturation magnetization in drop. We observe the shift of resonance field to be about 150 Oe, corresponding to $f \approx 0.03$. Additional magnetization caused by FM drops at $T < 50\text{ K}$ is $\Delta M \approx S g S g \mu_B L/(K S g \mu_B B_0(k_B T))$, where $L(y) = \coth(y) - 1/y$, $S = 7/2$ is Gd$^{3+}$ spin, $g = 2$ is $g$-factor, $\mu_B$ is Bohr magneton, $k_B$ is Boltzman constant. We estimate the characteristic number of Gd atoms inside the drop to be of order of $K \approx 30–100$. This value of $K$ gives the characteristic diameter of the drops to be equal to 1.5–2.5 nm.

We suggest that spin polarization of electron states, caused by local ferromagnetic ordering, leads to the splitting of energy sublevels for electrons with spin up and down inside the drops. The lowering of occupied spin-up sublevel increases spin polarization of the drop and also results in lowering of the Fermi level, which enhances the tendency towards electron localization. The Fermi energy $E_F$ decreases with temperature and reduces itinerant electron concentration outside the drops. That explains the temperature dependence of the conductivity $\sigma(T)$ of Gd$_{0.14}$Si$_{0.86}$ in the region between 5 and 50 K and its difference from $\sigma(T)$ of Y$_{0.14}$Si$_{0.86}$. It is clearly demonstrated in Fig. 2(a,b) that the conductivity change with temperature and magnetic field $\Delta \sigma(T,B)$ is proportional to the variation of electron concentration $\Delta n(T,B)$. The metal–insulator transition (MIT) is observed in samples with $x < 0.14$ at temperatures below 5 K, when $E_F$ crosses the mobility edge. We suppose that AFM ordering also appears at low temperature due to the coupling of FM drops via dipole–dipole interaction, so the total moment of the system remains zero.

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References