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In situ study of the temperature-dependent magnetoresistance of ultrathin epitaxial Fe films on GaAs(1 0 0)

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Abstract

An in situ magnetoresistance apparatus was used to carry out electrical transport measurements for various temperatures (100–300 K) on ultrathin iron films (1.0–8.0 ML) epitaxially grown on GaAs substrate. At 250 K, we observed a "GMR-like" magnetoresistance for a 2.5 ML thick Fe film attributed to the tunneling between the superparamagnetic clusters. Further cooling to 100 K established the onset of ferromagnetic ordering of the clusters giving rise to an anisotropic magnetoresistance signal. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Magnetoresistance—thin films; Clusters; Tunneling; Superparamagnetism

Spin-dependent transport phenomena in magnet/semiconductor heterostructures have recently attracted a strong interest. Although the injection of spin-polarised charge carriers from a magnetic semiconductor into a non-magnetic semiconductor has been demonstrated already [1, 2], the injection of spin-polarised electrons from a ferromagnetic metal directly into a semiconductor is still a great challenge. Key issues are the interface quality and the size and lateral separation of the ferromagnetic metal contacts.

In this contribution, we report the transport properties of epitaxial Fe nanoclusters grown on atomically clean semi-insulating GaAs (1 0 0) substrates. The separation is well below the spin diffusion length giving rise to the possibility of spin-dependent electron transport between magnetic clusters. In addition our in situ magnetoresistance (MR) set-up allowed us to vary the measurement temperature between 100 and 300 K enabling us to study the temperature characteristics and the evolution of the magnetoresistance with thickness of 1.0–8.0 ML thick films.

After a brief etching of the GaAs in H_2SO_4 solution, the substrate was inserted into the ultrahigh vacuum system with a base pressure of 4×10^{-10} mbar and annealed to 550°C for 30 min. Fe was then grown at room temperature at a rate of about 1 ML/min. At a chosen Fe thickness the growth process was interrupted and magnetoresistance measurements were carried out at various temperatures.

The in situ MR set-up used for these experiments consists of a rotatable sample holder fitted to a liquid nitrogen cryostat and an eight pin-probe located in the centre of the poles of an electromagnet (see Fig. 1). The pin-probe and electromagnet (MR apparatus) can be moved towards the sample in order to electrically contact the ultrathin film using small spring-loaded pins. Four pins at a time are arranged in a "cross-type" way in order to allow standard four-terminal measurements along two different crystallographic orientations. Moreover, by rotating the sample (with the probe) in the gap of the magnet by an angle of 90°, longitudinal and transverse magnetoresistance measurements can be carried out.

In the ultrathin film regime, Fe forms three-dimensional single-crystal clusters on GaAs with an average diameter of a few nanometers. Each cluster behaves as a "giant" spin where the total spin moment is of the order of 2000 μ_B. Moreover, all clusters have a common easy axis direction determined by the interface-induced and crystal anisotropy. The thickness range under investigation is also of particular interest since a change in

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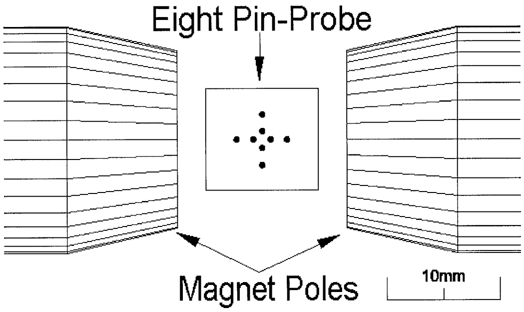

Fig. 1. Drawing of the main parts of the in situ MR apparatus.

magnetotransport processes can be expected from a film at the percolation threshold.

For films thinner than 4 ML we observed a non-metallic negative temperature coefficient of the resistivity ρ when the samples were cooled down from room temperature to 100 K. In particular, the resistivity of a 1 ML Fe film is very well fitted by the expression $\rho \propto \exp[(C/T)^{1/2}]$, where C is a constant depending on the tunneling barrier height and T is the temperature (see Fig. 2). The observed temperature dependence is a strong indication that the main mechanism for electron conduction is thermally activated tunneling between metallic clusters [3,4].

The investigations of the Fe films with in situ MR showed clearly that these iron clusters undergo a three-step phase transition when cooled down from room temperature to 100 K due to their small size. Above 330 K, only a very weak MR response was found for the 2.5 ML Fe film indicating that the majority of the iron islands are non-magnetic with a Curie temperature of about 330 K.

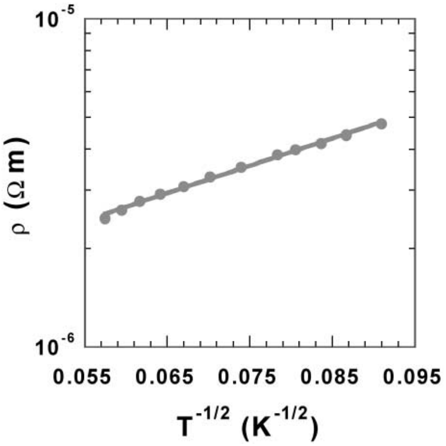

Fig. 2. Temperature dependence of the resistivity for a 1 ML Fe film.

For temperatures between 200 and 330 K ferromagnetic ordering within each cluster occurs but no ferromagnetic interaction between clusters can be established. The film is then predominantly in a superparamagnetic state. For this phase we found a negative giant magnetoresistance ("GMR-like" response) for 2.5–3.0 ML thick Fe films. In Fig. 3a a typical longitudinal MR loop is shown for a 2.5 ML thick film at 250 K.

When the samples were further cooled below 200 K the Fe islands couple ferromagnetically. The longitudinal MR response in the low-temperature regime changes from a "GMR-like" to an anisotropic magnetoresistance (AMR) dominated signal, as can be seen in Fig. 3b for a 2.5 ML Fe film at 135 K. The increase of the resistivity with an increasing magnetic field is due to the AMR

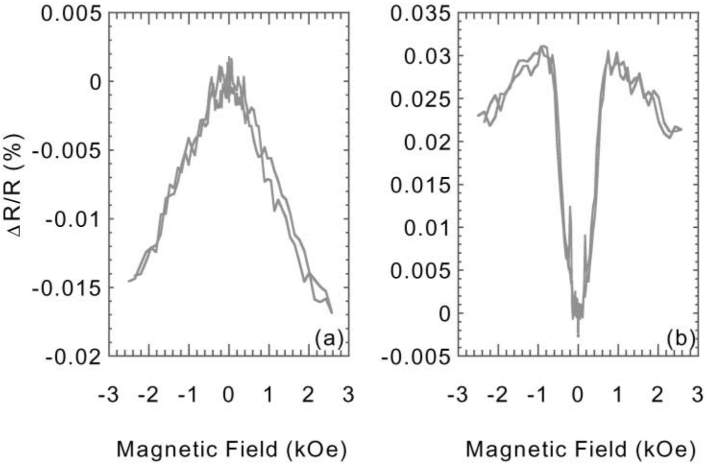

Fig. 3. Magnetoresistance loop taken from a 2.5 ML Fe film on GaAs at two different temperatures. (a) "GMR-like" response at 250 K, (b) AMR response at 135 K.

effect of the ferromagnetically ordered Fe clusters. For magnetic fields larger than 1 kOe the resistivity again decreases with an increasing applied field. This can be explained in terms of a superimposed AMR with a "GMR-like" signal. Due to a distribution in cluster size there is a coexistence of ferromagnetic ordered clusters with superparamagnetic clusters at a fixed temperature. At higher fields the "GMR-like" effect of the superparamagnetic clusters dominates the MR response and therefore decreases the resistivity.

This change in sign of the magnetoresistance marks clearly the vanishing of the superparamagnetic phase and the onset of ferromagnetic ordering between clusters. From the measured blocking temperature and by fitting a Langevin function to the MOKE loops we estimated the island diameter to be of the order of 5 nm, which is in agreement with STM images obtained from 2.3 ML Fe on InAs for which a similar growth mode is found [5]. The observed superparamagnetic to ferromagnetic transition is in agreement with MOKE results already reported by Xu et al. [6]. After deposition of more than 3 ML the transition from the superparamagnetic to the ferromagnetic phase is complete, the "GMR-like" signal vanishes and the AMR signal is clearly established, as expected for a continuous film.

In summary, in situ magnetoresistance was used to investigate the electrical transport properties of ultrathin epitaxial Fe films deposited on GaAs(1 0 0) substrate. For the cluster phase it was found that the main mechanism for electron conduction is thermally activated tunneling between the Fe nanoparticles. These clusters are predominately superparamagnetic below room temperature ($T \approx 250\,\mathrm{K}$) giving rise to a "GMR-like" magnetoresistance. At low temperatures ($T \approx 100\,\mathrm{K}$) the Fe clusters establish ferromagnetic order and therefore, exhibit the well-known anisotropic magnetoresistance.

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