Pis'ma v ZhETF, vol. 88, iss. 11, pp. 873 – 876

Observation of the magnetic domain structure in $Cu_{0.47}Ni_{0.53}$ thin films at low temperatures

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Submitted 28 October 2008

We report on the first experimental visualization of domain structure in films of weakly ferromagnetic $Cu_{0.47}Ni_{0.53}$ alloy with different thickness at liquid helium temperatures. Improved high-resolution Bitter decoration technique was used to map the magnetic contrast on the surface of the films well below the Curie temperature T_{Curie} ($\sim$ 60 K). In contrast to magnetic force microscopy, this technique allowed visualization of the domain structure without its disturbance while the larger areas of the sample were probed. Maze-like domain patterns, typical for perpendicular magnetic anisotropy, were observed. The average domain width was found to be about 100 nm.

PACS: 75.60.Ch, 75.70.–i

The interplay between superconductivity and ferromagnetism leads to a number of interesting phenomena [1, 2], which can be utilized in various applications. The interest of investigating the domain structure of weakly ferromagnetic $Cu_{1-x}Ni_x$ ($x \sim 0.5$) alloys, in particular, is caused by their active use in thin film superconductor (S)/ ferromagnet (F) heterostructures. The most promising microelectronic devices based on such heterostructures are basic elements of digital rapid single flux quantum (RSFQ) and quantum logic circuits. Apart from that, using $Cu_{1-x}Ni_x$ films as weak ferromagnets in fundamental S/F system properties research looks quite promising, which was confirmed by numerous theoretical and experimental investigations [2 – 7]. The physical properties of CuNi alloys are relatively well known. The average magnetic moment and the Curie temperature of uniform CuNi alloys decrease linearly with Ni concentration and both approach zero at $\sim$45 at.% Ni content. The magnetism of CuNi films is weaker than that in bulk material. Although $Cu_{1-x}Ni_x$ films with Ni concentration close to the critical value seem to be good candidates for using them in S/F proximity systems, at the same time they have several disadvantages. One of them is that the film structure is very sensitive to the fabrication conditions. In particular, the homogeneity of sputtered films is not ideal, (at least close to $x = 0.5$) since there is a tendency of Ni-rich clusters forming [8]. But features of CuNi films domain structure, which have huge influence on the transport and magnetic properties of S/F systems [6] have never been revealed. In this paper we present results of studying magnetic domains in thin films of $Cu_{0,47}Ni_{0,53}$ (hereafter called CuNi) alloy.

In the past decade magnetic force microscopy (MFM) has become a well-established technique for observation of the distribution of magnetic domains with submicron resolution [9 – 14]. It is widely used, except for the cases when MFM magnetic probe might bring distortions into the scanned image. That can happen, for instance, because of the sample local magnetization change by the probe itself during the scan. It's known that even well below the Curie temperature, the coercive field for thin films of some ferromagnets (CuNi in particular) might become almost zero, so using even magnetically soft MFM probes can disturb the picture of local magnetization when performing scans. Therefore, to study domain structure in such films we used the improved low temperature Bitter decoration technique [15], which also has such additional advantages as high spatial resolution and magnetic sensitivity. The Bitter technique is based on the deposition of fine dispersed magnetic nanoparticles, driven by the stray field gradients in the vicinity of magnetic material, at places where the magnetic field is higher. Decoration patterns can be examined by means of scanning electron microscopy (SEM). The patterns provide no information about the magnitude of the magnetization, yet in materials even with low stray fields, Bitter patterns can quickly yield information about the size and shape of domains of various types that might be present.

For our experiments $Cu_{0.47}Ni_{0.53}$ thin films were grown by RF-sputtering in Ar atmosphere of $P_{Ar} = 4 \cdot 10^{-2}$ mbar on silicon substrates at room temperature. The deposition rate was 0.25 nm/s. The Cu and

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Ni contents in the sputtered films were determined by Rutherford Backscattering (RBS) analysis. It confirmed that the Ni concentration in sputtered films was of the same value as in used CuNi targets.

First, the magnetic properties of $Cu_{0.47}Ni_{0.53}$ films structured in the shape of narrow bridges with thickness d_F ranging from 5 to 30 nm were studied by measuring anomalous Hall voltage $V_{\rm Hall}$, which is proportional to the film magnetization [16]. Fig.1a shows the dependence of $V_{\rm Hall}$ corresponding to the saturation magnetization of the sample with a particular thickness on the sample thickness. In all cases the applied magnetic field was perpendicular to the film surface. Typical temperature dependence of the Hall voltage for the CuNi film with $d_F = 22$ nm is presented in Fig.1b. For all film thicknesses it appeared to be non linear with a weakly pronounced saturation at low temperatures and tail-like behavior close to the $T_{\rm Curie}$. $T_{\rm Curie}$ for different samples was estimated by extrapolating the linear part of the $V_{\rm Hall}(T)$ dependence as presented in Fig.1b. In order to estimate the field range for the most effective magnetic domain decoration the hysteresis magnetization loops were measured as well. Results of magnetization reversal for $Cu_{0.47}Ni_{0.53}$ film with $d_F = 20$ nm are presented in Fig.2. The measurements were performed at 4.2 K well below the $T_{\rm Curie}$ (60 K) of the sample. Several field sweeps were performed with different values of maximum field in the range of 150–700 Oe as shown in Fig.2. The coercive field $H_{\rm Coer}$ for those sweeps was found in the range between 50–150 Oe.

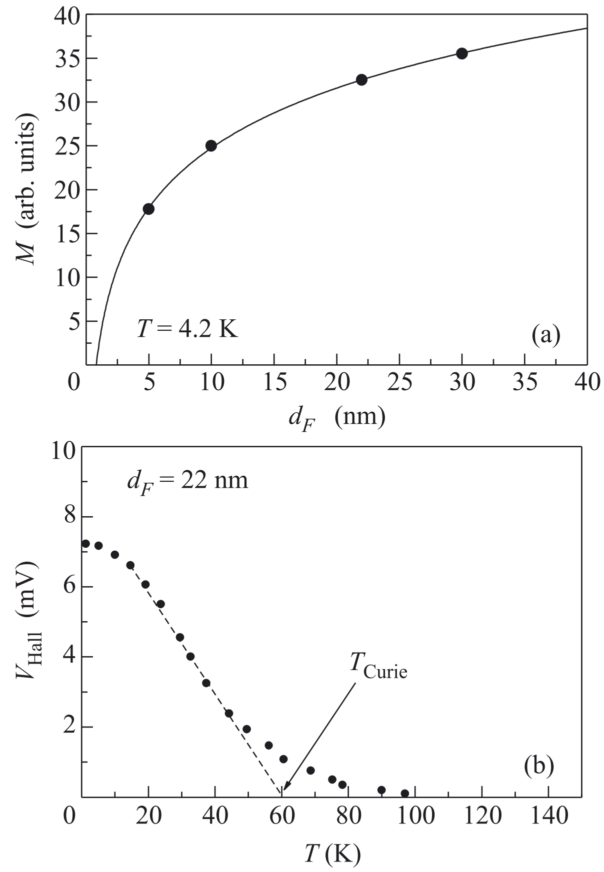

Fig1. (a) Saturation magnetization M as function of the film thickness d_F for $Cu_{0.47}Ni_{0.53}$. The dependence has a logarithmic like behavior. The line between experimental points serves to guide an eye. (b) Anomalous Hall voltage $V_{\rm Hall}$ dependence with temperature T for $Cu_{0.47}Ni_{0.53}$ film with $d_F = 22$ nm

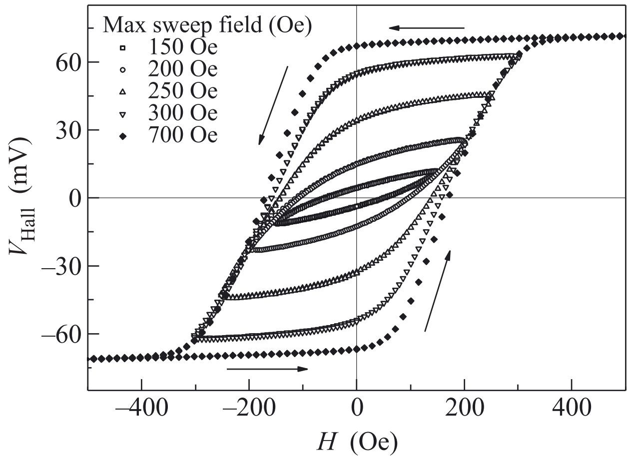

Fig.2. Hall voltage $V_{\rm Hall}$ as function of applied magnetic field H for $Cu_{0.47}Ni_{0.53}$ film with $d_F = 20$ nm measured at 4.2 K. Different curves correspond to different maximal sweep fields H as indicated. For all curves H was perpendicular to the film surface. Arrows show the direction of magnetic field sweep

Special consideration should be given to the CuNi thin film decoration procedure. The sequence of the entire experiment can be described as follows. The sample was initially cooled in zero magnetic field down to 4.2 K. During the decoration the temperature of the sample increased (of about 3–4 K) up to the decoration temperature T_d (the temperature measured by the resistive thermometer in the end of the iron evaporation process). The first series of decoration procedures was performed at magnetic fields $H_{\rm dec}$ = 100, 250, 300 Oe, on the virgin curve of the hysteresis loop (see Fig.2). For each new field value a new sample (geometrically identical to the previous one) was used. Additional experiment was done to make sure that multiple domain situation occurs after the magnetization switch. For that the magnetic field was gradually increased from 0 to 300 Oe and then swept back to $H_{\rm dec} = -150$ Oe, which corresponds to $-H_{\rm Coer}$ for this value of maximum sweeping field, as shown in Fig.2. In all cases the applied magnetic field was perpendicular to the sample surface.

Fig.3 presents the distribution of the iron particles mapping magnetic contrast related to the domain structure on the surface of the sample obtained with SEM. Fig.3a, b, c show the domain structure for decoration fields $H_{\rm dec} = 100$, 250 and 300 Oe respectively. That, as it has been mentioned before, corresponds to the evolution of the domain state on the virgin curve of the hysteresis loop. At the lowest applied field $H_{\rm dec} = 100$ Oe the decorated domain structure implies practically demagnetized state on the surface of the sample, which is believed to be perpendicular to the spontaneous magnetization axis. Domains form a maze-like pattern with a typical domain width of about 100 nm. Increasing the decorating field $H_{\rm dec}$ up to 250 Oe as indicated in Fig.3b results in widening of the positive (magnetization is pointed up) domains. Degradation of the pattern quality occurs because of the decrease of the local field gradients on the film surface. The domain structure (i.e. decorated magnetic contrast) almost disappears when approaching $H_{\rm dec} = 300$ Oe, see Fig.3c. A maze-like domain structure shows up clearly again, as it can be seen from Fig.3d after magnetization switching at the $H_{\rm dec} = H_{\rm Coer} = -150$ Oe.

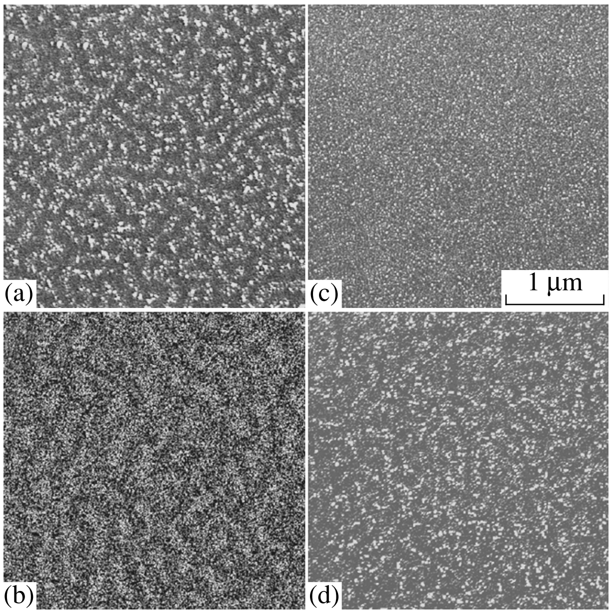

Fig.3. Evolution of the domain structure with external magnetic field applied perpendicular to the film plane: (a) $H = 150$ Oe, (b) $H = 250$ Oe, (c) $H = 300$ Oe, (d) $H = -150$ Oe

The results of investigations reveal several advantages of using CuNi alloys, which justify effectiveness of their utilization in Josephson SFS junctions. Firstly, for this particular ferromagnetic material the exchange energy is relatively small ($E_{ex}/k_B \sim 800$ K and $T_{\rm Curie} \sim \sim 60$ K correspondingly). That implies the superconducting order parameter decay length ξ_{F1} and the period of its spatial oscillation ξ_{F2} can be of the order of several nanometers instead of ~ 1 nm for the weak link of Josephson SFS junction made of non-diluted ferromagnet. Therefore, making Josephson SFS junctions with comparatively thick F-layers using simple thin film technologies becomes possible. Second the domain structure of CuNi films has a spatial period of about 0.1 μm, as the decoration experiments showed, which provides a good averaging of the net magnetization in F-layer. This, in its turn, allows to fabricate submicron ($\sim 0.2-0.3\,\mu$m) SFS sandwiches without having the undesired macroscopic stray fields. The final remark should be made about the possibility to manipulate the Josephson characteristics of SFS junctions (critical current, phase difference) with external magnetic field up to 20 Oe without disturbing the domain structure of F-layer, since it has a strong perpendicular anisotropy.

Several notes have to be made on the decoration technique. Clearly, in order to obtain the highest possible resolution of the method special care has to be taken of the decorating particle size as well as of their magnetic properties [17]. It is important to remind that when the applied field is higher than the local stray fields pointing up from the film surface, the magnetic particles become polarized along the applied field before landing on the ferromagnet surface [18].

Therefore, the particles aggregate to the positive domain areas, in which magnetization is aligned in the direction of an external magnetic field while negative domain areas are free from iron particles. Our additional experiments demonstrated that actual particle size distribution strongly depends on the buffer He pressure as was determined by SEM. Typical size distribution of the particle size is presented in Fig.4. The highest resolution in the range of 10-100nm was reached when the average particle size was about 10 nm, which was reached at $(2-3) \cdot 10^{-2}$ Torr of buffer He. The serious limitation of the decoration method lies in the fact that magnetic properties of the decorating particles strongly depend on their size: the saturation magnetization decreases with particle size (about 25% of that in bulk iron for 10 nm size particles [19]). Increasing the particle size implies larger magnetic moment and thus higher magnetic sensitivity, but the spatial resolution of the pattern gets reduced. Those particles with larger magnetic moment tend to form irregular clusters on the sample surface due to large interparticle interaction.

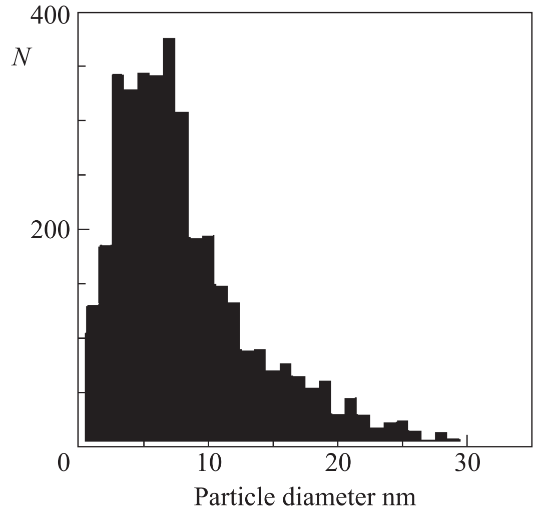

Fig.4. Size distribution of Fe decoration particles. N- number of Fe particles on the scanned area of $2\mu m^2$. He gas pressure was $\sim 2 \cdot 10^{-2}$ Torr

Recently the perpendicular magnetic anisotropy in dc-magnetron sputtered $Ni_{60}Cu_{40}/Cu$ multilayers was detected by hysteresis loop measurements for CuNi layer thickness between 4.2 nm and 34 nm [20]. However, in those experiments the features of the domain structure of CuNi films were not revealed.

In summary, the improved Bitter technique allowed visualizing the domain structure of weakly ferromagnetic $Cu_{0.47}Ni_{0.53}$ on large area (tens of square millimeters) at low temperatures. The image of the magnetic contrast on the surface of $Cu_{0.47}Ni_{0.53}$ films was seen for the first time. It was experimentally shown that thin CuNi films tend to have small scale domain structure. The films with thickness in the range of 10–30 nm have perpendicular magnetic anisotropy, which results in maze-like domain patterns and nearly rectangular hysteresis loop. The characteristic domain structure scale is found to be about 100 nm. We are grateful to V.V. Ryazanov and L.S. Uspenskaya for helpful discussions and L.G. Isaeva for helping to prepare evaporators. This work is supported by RFBR (# 07-02-00174), Joint Russian-Israeli project MOST-RFBR (# 06-02-72025) and InQubit, **Ltd.**

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