Local properties at the boundaries of irradiated regions in LCMO CMR films.

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Magnetic and magneto-transport properties and magnetization patterns in La$_{2/3}$Ca$_{1/3}$MnO$_3$ (LCMO) films irradiated with heavy ions through a mask to yield a sharp boundary between irradiated and un-irradiated areas were studied. It is found that this boundary enhances locally the resistance and magneto-resistance (MR) and shifts the maximum of MR to larger temperatures. Magneto-optical observations reveal a strong local anisotropy at the boundary tilting magnetic moments from the film plane and producing strong magnetic inhomogeneity responsible for the increase of magneto-resistance.
The treatment of the mechanism of colossal magneto-resistance (CMR) is based on the concept of spin polarized transport which implies that spin scattering on different magnetic inhomogeneities gives an essential contribution to the resistance [1]. Application of large enough fields that smooth out these magnetic inhomogeneities results in reduction of the spin scattering and thus in the negative magneto-resistance. The fact that in manganites the same carriers perform both charge transfer and magnetic exchange between the ions, i.e. work as transport and magnetic order agents simultaneously, makes the magneto-resistance especially large in these materials. Most of the experiments confirm such a physical picture emphasizing the role of magnetic inhomogeneities. The magneto-resistance value in CMR which is much larger in films and ceramic samples compared to that in high quality single crystals of the same composition [2] is a direct proof of this picture. A specific effect of natural and artificial grain boundaries as strong spin-scattering centers in perovskite manganites that could be used for tailoring local magnitude of the magneto-resistance for applications in novel magneto-electronic devices has been widely discussed in literature [3].

In this paper we present an attempt to introduce another type of boundary into CMR films resulting from the spatially modulated irradiation. It is revealed that a boundary between irradiated and un-irradiated areas brings about a local magnetic anisotropy assisting the tilt of magnetic moments from the film plane which gives a considerable contribution to the magneto-resistance of the sample. This anisotropy can be associated with the mismatch of strains in the un-irradiated part of the film stretched by the substrate and additional deformations induced in the irradiated part. It is found that the magneto-resistance maximum near $T_C$ is shifted to higher temperatures and the value of magneto-resistance at high $T$ becomes larger at the boundary compared to both irradiated and un-irradiated areas. These results offer a new way of tuning the local magneto-transport parameters for possible CMR applications by the inhomogeneous irradiation of the samples.
Here we present magnetic, transport and magneto-optical data obtained for a 60 nm thick epitaxial La$_{2/3}$Ca$_{1/3}$MnO$_3$ film on (001) STO substrate. Qualitatively similar results were obtained for films on LAO and NGO substrates and will be published elsewhere. The films were grown using pulsed laser deposition at a small oxygen pressure [4]. They were half covered with a thick golden screen and irradiated with 1.1 GeV Pb-ions to a dose of irradiation: $1.1 \times 10^{11}$ ions/cm$^2$ (~300 Å columnar defect spacing). The gap between the sample and the screen was $\leq$2 microns so the irradiation boundary width should not be larger than that. Gold contacts were deposited onto sample allowing resistivity measurements in the irradiated, in the non-irradiated section and across the boundary, respectively (see sketch in Fig.2). The magnetic structure in different areas of the samples was imaged using the magneto-optical indicator technique [5].

The ion irradiation of oxides is known to produce columnar defects characterized by amorphous core of 5 – 10 nm diameter [6]. In the distorted region the electronic spectrum is expected to be modified and the mean free path is reduced inducing changes in both transport and magnetic properties. Additional magnetic inhomogeneities on the scale of the relaxation radius of the strains around columnar defects could be also expected.

However, it turned out that irradiation on the contrary smoothes out the magnetic inhomogeneities in the films. Fig.1 illustrates magneto-optic patterns around the boundary of irradiation. In the irradiated area (on the right) the contrast revealed at remagnetization of the sample in the horizontal field along the in-plane easy axis is much smoother than in the un-irradiated region (on the left). The strongest inhomogeneity is revealed at the boundary (bright line) where magnetic moments are tilted from the film plane. This picture can be explained by the relaxation of the film-substrate mismatch stresses in the irradiated area and appearance of the strain mismatch at the boundary between this area and the un-irradiated part of the sample.
An essential role of the boundary in the magneto-transport response of the film is revealed by the resistance and magneto-resistance measurements illustrated in Figs. 2 and 3. In both irradiated and un-irradiated areas the maximum of $\rho(T)$ in the fields below 1kOe is observed at ~265K which practically coincides with the Curie point of ~263K determined from interpolation of the sharp decrease of $M(T)$ measured by SQUID in the field of 1 kOe (see Fig. 2a). This maximum drops down and shifts to larger temperatures in higher fields ($T_{\text{max}}$ ~277K at 10 kOe). It is clear from Fig. 2a that irradiation results in the increase of the resistivity, consistent with previous reports [6]. However, the value of the resistivity measured across the boundary turns out to be even larger than in the irradiated area. Accounting for a small relative volume of the boundary in ~1.2 mm gap between the voltage contacts one can conclude that the boundary introduces a substantial increase of the resistance, exceeding that in the irradiated region.

The magneto-resistance, $MR$, defined as $(\rho(H,T) - \rho(0,T))/\rho(0,T)$ for all three regions measured as a function of temperature in a field of 1 kOe is shown in Fig. 2b. It is remarkable, that the maximum $MR(T)$ values in the irradiated and un-irradiated areas (Fig. 2b) occur at the same temperature, which increases with $H$ (e.g. 247K at 1 kOe and 252K at 10 kOe). When measured across the boundary the maximum of $MR(T)$ is always shifted to higher $T$ (e.g. 250K at 1 kOe and 255K at 10 kOe). As a result the high temperature magneto-resistance across the boundary is noticeably larger (especially when accounting for its relative small volume) than in both neighboring regions.

On the field dependencies of $MR$ (Fig. 3a) one can reveal a small field region of positive magneto-resistance and large field region of negative. This can be explained by the dominating positive contribution to the $MR$ due to the nucleation of domains and magnetic inhomogeneities (that increase the resistivity) during remagnetization from a remanent state. In contrast, larger fields suppress the inhomogeneities (decrease the resistivity) and approach
the film to the saturated state. The magneto-resistance in the irradiated area changes from positive to negative at noticeably smaller fields in accordance with smoothed magnetic inhomogeneities revealed in Fig.1. At the same time in the boundary region the MR(H) curve is slightly wider and its maximum is shifted to slightly larger H corresponding to the stronger local inhomogeneity at the boundary. Larger fields are needed to polarize this region and appropriate MR value at T<T_c becomes slightly smaller than in the neighboring areas. Actually, absolute changes of the resistivity Δρ(H) across the boundary are larger than in the other regions. A small difference of the MR value across the boundary compared to the un-irradiated area (Fig.3a) may be referred to a small relative volume of the boundary between the voltage contacts and its larger ρ(0) entering the denominator of MR. The hysteresis of the MR curves in Fig.3a shows a direct correspondence to the magnetization hysteresis in the ferromagnetic state (Fig. 3b). It disappears and the MR acquires a purely negative sign above T_c. At T>T_c the MR value of the boundary becomes larger than that of the irradiated and un-irradiated sections (Fig. 3a) which could be interesting for applications as a way of improving high temperature CMR parameters.

In conclusion, we performed a spatially inhomogeneous irradiation of LCMO films and found that the boundary between irradiated and un-irradiated areas induces a strong local distortion of magnetic moments which essentially modifies the magnetoresistance of the film by shifting its MR(T) maximum to larger T and increasing the high-temperature value of MR. The effect of the boundary which is stronger than just effect of homogeneous irradiation can be associated with the strain mismatch between irradiated and un-irradiated parts of the film.

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Figure captions

Fig.1 Magneto-optical patterns around the boundary between irradiated (right) and un-irradiated (left) area. Bright contrast at the boundary reveals strong normal fields due to magnetic moments tilted from the film plane.

Fig.2 (a) Temperature dependence of the resistivity in the irradiated, un-irradiated areas and across their boundary in the fields of 0 and 10 kOe. The inset shows the temperature dependence of the magnetic moment of the film measured in 1 kOe. M(T) curve did not change noticeably after irradiation. The scheme of contacts on the sample is shown in the bottom. (b) Temperature dependence of the magneto-resistance, MR = (ρ(1kOe)-ρ(0))/ρ(0).

Fig.3 (a) Field dependence of the magneto-resistance in the irradiated, un-irradiated areas and across their boundary at 200K (bottom field scale, left y-scale) and 265 K (top field scale and right y-scale). (b) Magnetization hysteresis in the film at 200K.
References


Fig. 1  V.K. Vlasko-Vlasov et al.
Fig. 2  V.K. Vlasko-Vlasov et al.
Fig. 3  V.K. Vlasko-Vlasov et al.