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Pulse and quasi-static remagnetization peculiarities of $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal

V.T.Dovgii, A.I.Linnik, V.I.Kamenev, V.Yu.Tarenkov, S.L.Sidorov, B.M.Todris, V.I.Mikhailov, N.V.Davideiko, T.A.Linnik

Donetsk Institute for Physics and Engineering, National Academy of Sciences of Ukraine, 72 R.Luxemburg Str., 83114 Donetsk, Ukraine

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The hysteretic behavioral features of magnetization and resistance upon remagnetization under quasistatic (up to 9 T) and pulse (up to 14 T) magnetic fields have been investigated. The relaxation processes of magnetization and resistance after the effect of 9 T magnetic field have also been studied. The mechanism of remagnetization of the antiferromagnetic insulating-ferromagnetic metallic (AFM/I–FM/M) phases and the existence of high conductivity state of the sample after removal of the magnetizing field is proposed for low temperatures. The mechanism is caused by structural transition, which is induced by magnetic field (due to magnetostriction), and slow relaxation of the FM-phase (greater volume) to the equilibrium AFM-phase (smaller volume) after the field removal. Remagnetization of $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal under pulse field at low temperatures (18 K) has shown that time of the AFM/I → FM/M phase transition was lower than time of the return FM/M → AFM/I phase transition by 6 orders of magnitude.

Исследованы гистерезисные особенности поведения намагниченности и сопротивления при перемагничивании в квазистатическом (до 9 Т) и импульсном (до 14 Т) магнитном поле. Изучены процессы релаксации намагниченности и сопротивления после воздействия магнитного поля 9 Т. Для низких температур предложен механизм перемагничивания фаз антиферромагнитной изолирующей — ферромагнитной металлической (AFM/I–FM/M) и существования высокопроводящего состояния образца после снятия намагничивающего поля. Механизм обусловлен структурным переходом, который индуцирован магнитным полем (за счет магнитострикции), и медленной релаксацией FM-фазы (большего объема) к равновесной AFM-фазе (меньшего объема) после снятия поля. Показано, что для температуры 18 К при импульсном перемагничивании время фазового перехода AFM/I → FM/M на 6 порядков меньше времени фазового перехода FM/M → AFM/I.

Особливості імпульсного і квазістатичного перемагнічування монокристала $Nd_{0.5}Sr_{0.5}MnO_3$. *В.Т.Довгий, О.І.Лінник, В.І.Каменєв, В.Ю.Таренков, С.Л.Сидоров, Б.М.Тодрис, В.И.Михайлов, Н.В.Давидейко, Т.О.Ліннік.*

Досліджено гістерезисні особливості поведінки намагніченості і опору при перемагнічуванні у квазістатичному (до 9 Т) і імпульсному (до 14 Т) магнітному полі. Вивчено процеси релаксації намагніченості і опору після впливу магнітного поля 9 Т. Для низьких температур запропоновано механізм перемагнічування фаз антиферомагнітної ізолюючої — феромагнітної металевої (AFM/I–FM/M) і існування високопровідного стану зразка після зняття поля, що намагнічує. Механізм обумовлений структурним переходом, який індукований магнітним полем (за рахунок магнітострикції), і повільною релаксацією FM-фази (більшого об'єму) до рівноважної AFM-фази (меншого об'єму) після зняття поля. Показано, що для температури 18 К при імпульсному перемагнічуванні час фазового переходу AFM/I → FM/M на 6 порядків менше часу фазового переходу FM/M → AFM/I.

1. Introduction

The study of rare earth manganites with perovskite structure is an important field of solid-state physics. Interactions between magnetic, electric and structural parametres of these compounds are interesting as for strong-correlated systems [1–3]. The nature of magnetic and electric interactions in these materials is not completely clear now and is a subject of wide discussions. The compounds are interesting as materials that show colossal magnetoresistance (CMR) and can be used in magnetic recording and reading devices, as high-sensitivity sensors of magnetic field, current, temperature, and pressure [3, 4].

Half-substituted manganites exhibit spin, charge, and orbital ordering. This ordering can be suppressed by magnetic field due to the transition from the antiferromagnetic semiconductor state to the ferromagnetic metallic state [5, 6].

In this work, we report the results of studying the structural, magnetic, and transport properties of $Nd_{0.5}Sr_{0.5}MnO_3$ single crystals in the wide temperature range (6...400 K) under quasi-static magnetic field up to 9 T or pulse magnetic field up to 14 T. The processes of magnetization and resistance relaxation after the effect of 9 T magnetic field are also analyzed.

The purpose of this work was to reveal the nature of the hysteretic behavior of the field dependences of resistance and magnetization at low temperatures in pulse and quasi-static magnetic fields up to 14 T, as well as to establish the reasons for the high conductivity sample state after removal of the magnetic field.

2. Experimental

$Nd_{0.5}Sr_{0.5}MnO_3$ single crystals were grown by floating zone melting upon radiative heating. The lattice parameters and crystallographic axis directions were determined by DRON-2 X-ray diffractometer (NiKα-radiation) from the positions of (600), (060), and (008) lines. The samples under study had the orthorhombic structure *Pnma* with the lattice constants $a = 0.54780$ nm, $b = 0.54308$ nm, and $c = 0.76116$ nm. The degree of distortion in a–b plane of the crystal is sufficiently small (a and b axes are different by 0.9 %). Therefore, under crystallization these crystallographic directions are not met resulting in twins formation in the sample. This becomes apparent in bifurcation of the X-ray reflections. However the half-width of the rocking curves for all directions did not exceed 1.5°, which is indicative of a good quality of the crystals grown. It should be noted that twinning is a typical feature of manganite single crystals with distorted perovskite structure [7, 8].

The field and temperature dependences of the magnetization $M(H)$ and resistance $\rho(H)$ were measured by means of PPMS-9 setup in quasi-static magnetic field and their relaxation was studied. The sample resistance was determined by the four-probe method. As the preliminary experiments showed, the resistance depended weakly on both the mutual direction of the magnetic field and the measuring current and their orientations with respect to the crystallographic axes. Thus, the resistance was measured in the following geometry: the measuring current was directed along c axis of the crystal, and the magnetic field was oriented perpendicularly to the current. The dependences $M(H)$ were analyzed in pulsed magnetic field in the temperature range of 18...200 K using induction technique. In each measuring cycle, the sample was subjected to bipolar pulse of the magnetic field with total width of 1.5 ms. In all experiments, the magnetic field was oriented perpendicularly to c crystallographic axis.

3. Results and discussions

$Nd_{0.5}Sr_{0.5}MnO_3$ single crystals undergo the following transformations upon cooling (from room temperature): paramagnetic insulator (PM/I)-ferromagnetic metal (FM/M) (255 K) and ferromagnetic metal (FM/M) (255–150 K)-antiferromagnetic insulator (AFM/I) (<150 K) [6]. In this paper, experiments on pulsed and quasi-static remagnetization carried out in the temperature range below 150 K, i.e, in area of the existence of antiferromagnetic insulator. Investigation of the temperature dependences of magnetization and resistance were carried out in the range of 6...300 K. In this case all of the above phase transitions have been detected. Note that the phase transition metal-insulator transition at $T = 150$ K for our single crystal, apparently, can be attributed to the Mott type metal-insulator transition [9].

Fig. 1 shows the field dependences of magnetization $M(H)$ and resistivity $\rho(H)$ (in the inset) at temperature of 10 K under quasi-static field (curves *1–4*) and at temperature of 18 K under pulsed field (curves *5–7*). Curves *1–3* correspond to successive

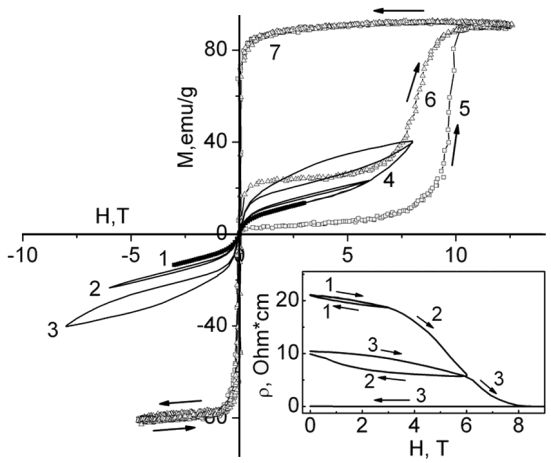

Fig. 1. Field dependences of magnetization $M(H)$ and resistivity $\rho(H)$ (in the inset) of a $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal at a temperature of 10 K in a quasi-static field (curves *1–4*) and at a temperature of 18 K in a pulsed field (curves *5–7*). The directions of variation in the magnetic field are shown by arrows.

magnetization and demagnetization of the sample under quasi-static field up to 3, 6, and 8(9) T. The magnetization curves demonstrate coexistence of the FM and AFM phases. Each time after reaching the equilibrium state the initial portions of the $M(H)$ curves go along the same curve *4* and correspond (under the weak fields) to magnetization of the existing FM phase, whereas the hysteretic portions of $M(H)$ under the strong fields correspond to remagnetization of the AFM/I–FM/M phases.

Remagnetization under pulse field differs significantly from that under quasi-static field. Magnetization of the sample, which was initially in the equilibrium state, begins along curve *5*, and the AFM/I–FM/M transition occurs sharply under pulsed field of about 10 T. Demagnetization and remagnetization under negative field occur along curve *7* until the field becomes zero. Repeated remagnetization in 3–5 min after the end of the first cycle begins along curve *6*. The AFM/I–FM/M transition manifests itself under pulsed field of about 8 T. Then remagnetization occurs again along curve *7*.

The field dependences of resistivity $\rho(H)$ (Fig. 1, inset) under the quasi-static field also exhibit hysteretic behavior; as can be seen, there is sharp decrease in the resistance under 9 T magnetic field, this high-conductivity state remaining after magnetic field removal. Note that, during magnetization up to 3 T, dependence $\rho(H)$ has a very weak hysteresis and is reversible (curve *1–1*). At the same time, after magnetization to 6 T, the sample resistance is much decreased and does not recover the initial value after the field removal (curve 2–2). The subsequent magnetization cycle to 9 T began from this new value, yielded a larger hysteresis, and led to the above-mentioned high-conductivity state (curve *3–3*). Comparison of the dependences $M(H)$ and $\rho(H)$ reveals the contradiction: the magnetization curves are closed and the sample is demagnet ized under zero field, whereas the resistivity curves are open (except for the cycle 1–1) and the low resistivity state is retained with increase of magnetic field up to 9 T and successive decrease to zero.

Fig. 2 and 3 show, respectively, the temperature dependences of the magnetization $M(T)$ and resistivity $\rho(T)$ after switching off 8(9) T magnetic field. The $\rho(T)$ curve was recorded immediately after the field removal, and the $M(T)$ curves were recorded both immediately after the field removal (Fig. 2, curve *2*) and after 24 h (Fig. 2, curve *1*). It can be seen in Fig. 2 that behavior of curve 1 is in good agreement with the known literature data [7]. At the same time, the dependences $\rho(T)$ (Fig. 3) and $M(T)$ (Fig. 2, curve *2*) at low temperatures demonstrate recovery of the equilibrium M and ρ values, which is typical of the AFM state. Since the $M(T)$ and $\rho(T)$ were recorded at very slow increase of the temperature (~1 K/min), it can be assumed that contribution of the temperature in the relaxation process is small at the low temperatures. Therefore, the same data were used to construct the

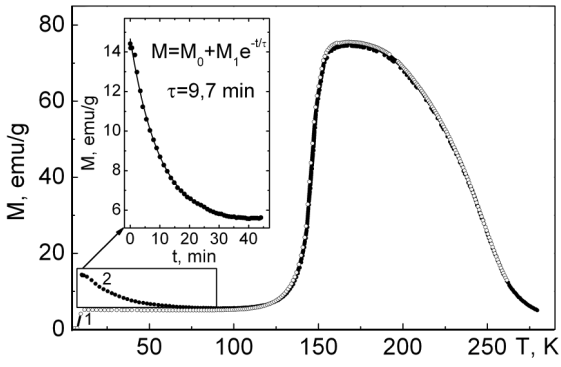

Fig. 2. Temperature dependences of magnetization $M(T)$ of a $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal (measured in a 0.5 T field): (*1*) 24 h after removal of 8 T field and (*2*) immediately after a decrease in the field from 8 to 0.5 T. The relaxation curve of the initial portion of the dependence $M(T)$ is shown in the inset (indicated by a rectangle).

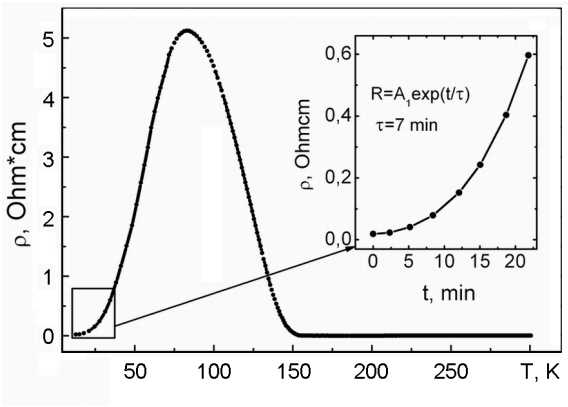

Fig. 3. Temperature dependence of resistivity $\rho(T)$ of a $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal. The relaxation curve of the initial portion of the dependence $\rho(T)$ is shown in the inset (selected by a rectangle).

time dependences of $M(t)$ and $\rho(t)$ for the lowest temperatures, where these are well described by the exponential dependence. Relaxation time constants at these regions were $\tau \approx 9.7$ min and $\tau \approx 7$ min for magnetization and resistivity, respectively (see insets in Fig. 2 and 3).

According to the neutron diffraction data and the results of magnetic investigations, there are three phases in $Nd_{0.5}Sr_{0.5}MnO_3$ manganite at $T < 150$ K: AFM of CE type (~60 %), AFM of A type (~20 %), and FM (~20 %) with a unit cell volume $V_{CE} = 159.1$ Å^3 (monoclinic symmetry), $V_A = 158.65$ Å^3, $V_{FM} = 159.4$ Å^3 (orthorhombic symmetry) [9]. At the same time, at $T > 150$ K, the main (>80 %) phase is FM with the largest unit cell volume. Our experiments showed that, at temperatures of about 10 K under magnetic field, the AFM/I–FM/M transition occurred as a result of the lattice transformation of $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal due to magnetostriction. Under the quasi-static magnetic field, the rate of field change appears to be comparable with the rate of the lattice transformation and the AFM/I–FM/M transition occurs gradually (Fig. 1). At the same time, apparently, under the pulse field, the rate of field variation signally exceeds the lattice transformation rate; therefore, the AFM/I–FM/M transition occurs only when the field value achieves the level at which only the FM/M phase can exist. This conclusion is confirmed by the transition sharpness (Fig. 1, curve *5*). Remind that total duration of the bipolar pulse remagnetization is ~ 1.5 ms. Therefore, the field strength at which the rearrangement of the crystal lattice and the transition AFM/I–FM/M with destruction of charge ordering is achieved for 0.3...0.4 ms. From this we can conclude that the transition itself occurs in time that does not exceed 0.3 ms. While the field decreases and becomes negative, the sample behaves as purely FM medium, exhibiting a high magnetic moment. This is especially obvious for the pulsed field experiment (Fig. 1, curve *7*). Under quasi-static field, one can observe a hysteresis, the cause of which is that the sample has enough time to partially relax to the equilibrium state. As a result, after repeated magnetization under the positive or negative field, the sample has smaller M value. The hysteresis value is proportional to the field to which the sample is magnetized. The obvious reason for this is that part of the AFM/I phase, which constantly increases, passes to the FM/M state with increase of the field. Metastability of the FM/M state is evidenced by curve *6* in Fig. 1. As was noted above, this curve was obtained in a new cycle of pulse remagnetization in 3–5 min after the end of the previous cycle. Obviously, the sample was partially relaxed to the AFM/I phase for this time; however, part of the sample retained the FM/M phase with lower magnetic moment, to which the sample was magnetized after repeated remagnetization. Afterward, the complete AFM/I–FM/M transition occurs under the weaker pulsed field (about 8 T). Thus, recovery of the equilibrium lattice of the phase AFM/I with recovery of the charge ordering takes place many minutes at temperatures of 10...20 K after removal of the magnetic field. This is about 6 orders of magnitude higher than the transition to the metastable phase FM/M during magnetization of the crystal. Mentioned relaxation is strongly temperature dependent and at higher temperatures the process is faster.

Fig. 4 shows the T–H diagram in order to compare the influence of pulse and quasi-static magnetic field on AFM/I↔FM/M magnetic transitions in $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal. The wide region (inclined shading to the right) corresponds to critical magnetic fields of phase transition at pulse remagnetization (right points are the AFM/I–FM/M transition, the left points are the return FM/M–AFM/I transition). The narrow region (cross shading) corresponds to critical magnetic fields of phase transition at quasi-static remagnetization (the fields of return FM/M–AFM/I transition are shown by shaped line). We can see from

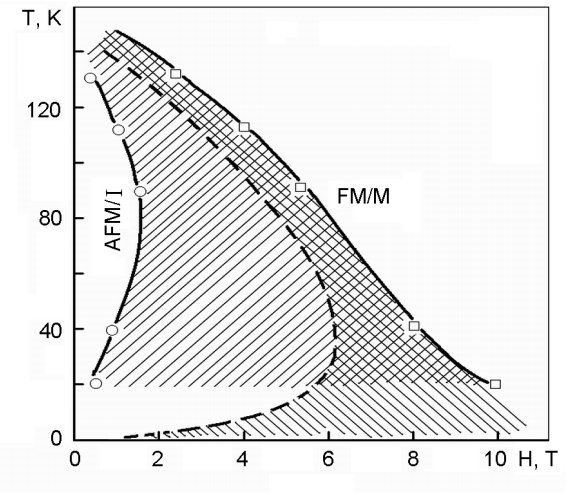

Fig. 4. T–H diagram of a $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal in the pulse field. The dashed line is a phase transition boundary AFM/I in the quasi-static field.

the phase diagram that the area of existence of metastable phase FM/M at remagnetization under the pulse field is much wider, than under the quasi-static one.

Tunnel spectroscopy experiments with $Nd_{0.5}Sr_{0.5}MnO_3$ show that the gap in the density of states at the charge ordering temperature is ~ 300 meV. This gap is much wider than the energy of charge ordering (~ 12 meV) and 9 T magnetic field (~ 1.8 meV) [10, 11]. Therefore, it is unclear how the field of several Tesla can destroy the charge-ordered state on this energy scale.

Apparently, the AFM → FM transition is possible because of the existence of several phases, both the magnetic and structural, as well as induction by the magnetic field due to magnetostriction of the structural transformation with increase in the unit-cell volume. This transition is clearly accompanied by the destruction of the charge ordering and disappearance of the gap in the density of states, as is in the case of the spontaneous transition to the FM-state at temperature of 150 K. After removal of the magnetic field, the crystal structure slowly relaxes to the equilibrium structure typical of the AFM phase. The nonequilibrium FM phase existing near 10 K for several minutes leads to the high-conductivity state of the sample.

4. Conclusions

Remagnetization of $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal under pulse field at low temperatures (18 K) has shown that time of the AFM/I → FM/M phase transition was lower than time of the return FM/M → AFM/I phase transition by 6 orders of magnitude. The same relationship of times took place at destruction and restoration of the charge ordering.

Energy of the magnetic field 9 T (~ 1.8 meV) is not enough to implement the transition AFM/I → FM/M and the destruction of the charge ordering (kT_{CO}~ 12 meV, Δ_{CO}~ 300 meV). Therefore the described effect is obviously due to structural transition that is induced by the magnetic field caused by magnetostriction. This is accompanied by the charge ordering destruction and increase of the cell volume.

Existence of the nonequilibrium FM phase at 10 K for many minutes causes conducting state of the sample after removal of the magnetic field due to slow relaxation of the nonequilibrium FM-phase (with increased lattice volume) into the equilibrium AFM-phase (with smaller lattice volume).

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