Magnetotransport anisotropy effects in epitaxial magnetite (Fe_3O_4) thin films

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Epitaxial films of magnetite (Fe_3O_4) have been grown on $SrTiO_3$ (100) and sapphire (α-Al_2O_3, 0001) substrates by pulsed laser deposition, and they exhibit crystal orientations of [100] and [111], respectively. Films of both orientations show a clear Verwey transition near 120 K. The temperature dependence of magnetoresistance of the films is examined at fields up to 8.5 T applied parallel to the film plane. The [100] oriented films show a pronounced extremum in magnetoresistance (MR) at the Verwey transition temperature (T_v), while no such feature is observed in the case of the [111] oriented films. However, the [111] oriented films exhibit a sudden change in the MR dependence on T at T_v and films of both orientations show a rapid increase of MR as the temperature is decreased below about 110 K. An attempt is made to analyze these data in terms of the known anisotropy in magnetostriction and optical magnon-phonon dispersions, and the small polaron band and hopping conductivity mechanisms. [S0163-1829(98)03014-8]

Magnetite (Fe_3O_4) is perhaps one of the most studied oxides over the past 50 years because of its rather unique and interesting set of transport and magnetic properties.[1–4] It is a cubic inverse spinel compound with tetrahedral sites occupied by Fe^{3+} ions and octahedral sites shared by Fe^{2+} and Fe^{3+} ions. The moments of the Fe^{3+} ions on octahedral and tetrahedral sites are opposite to each other and the net moment arises only from the Fe^{2+} ions; the arrangement being termed as ferrimagnetic. The presence of Fe^{2+} and Fe^{3+} ions on octahedral sites leads to a fairly low electrical resistivity[3] in this compound at room temperature due to carrier hopping between the Fe^{2+} and Fe^{3+} ions, the typical value in well characterized single crystals being lower than 10 mΩ cm at 300 K. As the temperature is lowered, the resistivity undergoes an increase of about two orders of magnitude at 120 K with a concomitant decrease in the magnetic moment.[5] This transition, known as Verwey transition,[2] has been a subject of many experimental and theoretical investigations over the past decades.[1–20] It is attributed to charge ordering of Fe^{2+} and Fe^{3+} ions below the Verwey transition temperature.

Several past studies on magnetite have addressed crystal symmetry issues on which an emphasis is placed in the present work. Heaps[15] measured the magnetostriction of a single-crystal magnetite. In a field of 5 kOe the fractional longitudinal changes in length in the [100], [110], and [111] directions were found to be -4, $+30$, and $+12\times10^{-6}$, respectively, the corresponding transverse changes being $+4$, -44, and -28×10^{-6}, respectively. Bickford[16] studied the temperature dependence of the ferromagnetic resonance absorption in a synthetic single crystal of magnetite, and from measurements of the resonance magnetic field along the [100] and [110] directions, he obtained the first-order anisotropy constant K_1. He found that above 130 K (which is above T_v) the anisotropy constant is negative, indicating that [111] is the easy direction of magnetization, while below 130 K, [100] is the easy direction of magnetization.

More than 50 years ago, Masumoto and Shirakawa[17] had studied the magnetoresistance (MR) and intensity of magnetization of single-crystal magnetite at fields up to 1.2 T at 273, 178, and 78 K. At 273 and 178 K (both above T_v), they found the MR to increase as a function of field, rapidly at low fields and slowly at higher fields. On the other hand, at 78 K (below T_v), they found the MR to increase linearly with the field over the entire range studied. Interestingly, similar behavior has also been reported by Xiong *et al.* in colossal magnetoresistance (CMR) materials.[21] Subsequently, Feng and co-workers[18] performed a high field (2–10 T) study of magnetoresistance in polycrystalline thin-film samples of Fe_3O_4 and reported the observation of a very broad -7.5% MR peak at about 130 K. Recently, Gridin, Hearne, and Honig[19] have reported an interesting observation of a very sharp MR extremum in single-crystal $Fe_{3(1-\delta)}O_4$, $\delta=0.0006$. The MR was found to peak to -17% at T_v in a field of 7.7 T with a full width at half maximum of the peak of about 0.53 K. On lower and higher sides of the peak the negative MR values were found to be 35 and 0.5%, respectively, in the same field. In these studies the external field (applied perpendicular to the current direction) was not along any high symmetry direction in the cubic lattice. Thus, the issue of magnetotransport anisotropy in Fe_3O_4 has remained unaddressed so far. It is also important to mention here that the subject of magnetoresistance in oxide systems has acquired a renewed significance during the past few years with the emergence of colossal magnetoresitance manganite materials.[20–29] Since in these CMR materials, as well as in the case of Fe_3O_4, a coupling between lattice, magnetism, and transport is suggested with a possible role of small polarons,[7–9,27,28] it is of interest to explore whether any interesting parallels can be drawn between their behaviors from comparison of magnetoresistance systematics.

In the present work we have examined the anisotropy in the magnetoresistance of Fe_3O_4 (magnetite) thin films. We prepared [100] and [111] oriented epitaxial thin films on (100) oriented $SrTiO_3$ and (0001) oriented sapphire substrates and measured the resistivity at fields up to 8.5 T applied along the film plane. We find a discontinuity in the temperature dependence of MR at T_v for films of both the orientations, however, an MR extremum is observed only in

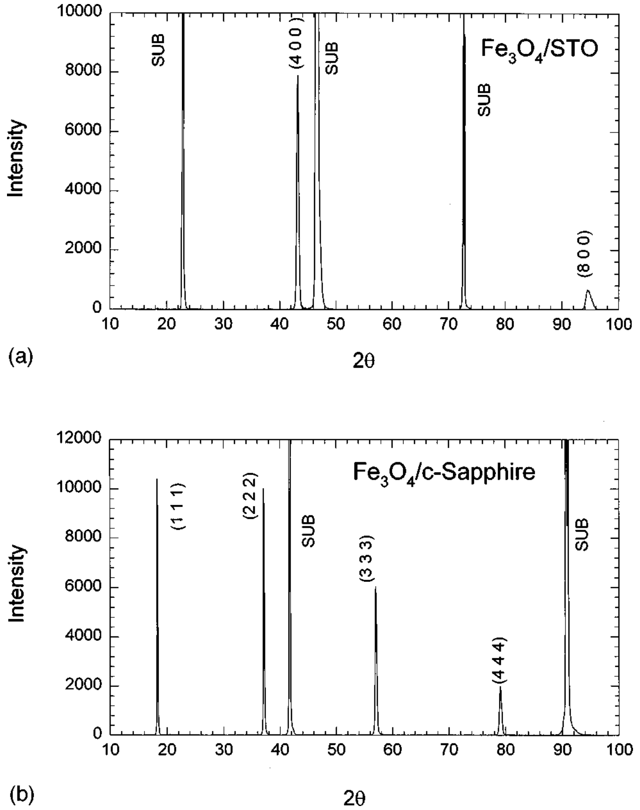

FIG. 1. X-ray diffraction patterns for the Fe_3O_4 films deposited on (a) $SrTiO_3$ (100) and (b) sapphire (0001) substrates.

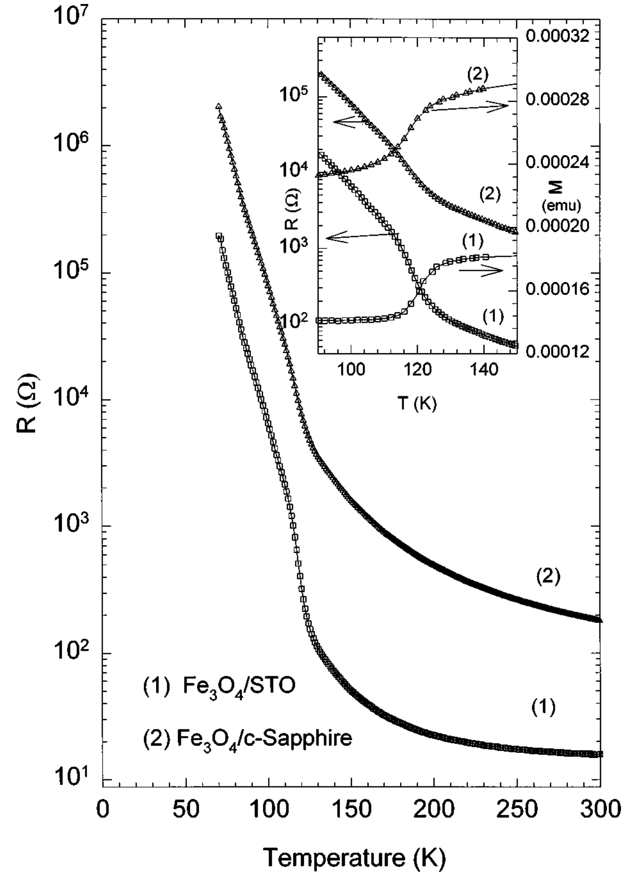

FIG. 2. Dependence of resistance on temperature for the Fe_3O_4 films deposited on $SrTiO_3$ (100) and sapphire (0001) substrates. The inset shows weak field (0.2 Oe) magnetization data and the resistance on expanded temperature scale.

the case of the (100) oriented film.[30]

The films used in our studies were prepared by pulsed excimer laser deposition using α-Fe_2O_3 as the target. The depositions were carried out at a substrate temperature of 570 C in vacuum of better than 1×10^{-5} Torr. The thickness of the films on both the substrates was about 2000 Å. In Figs. 1(a) and 1(b) are shown the x-ray-diffraction data for the films deposited on $SrTiO_3$ (100) and α-Al_2O_3 (0001) substrates, respectively. It can be clearly seen that the films exhibit (100) and (111) orientations normal to the film plane on $SrTiO_3$ and α-Al_2O_3, respectively. The full width at half maximum for the rocking curve on the (400) peak of Fe_3O_4 on $SrTiO_3$ is $\sim0.2°$ while that on the (111) peak of Fe_3O_4 on α-Al_2O_3 is $\sim0.1°$, indicating a high degree of orientational quality of the films. The results of Rutherford backscattering channeling (minimum yield of a few percent) and transmission-electron microscopy studies (to be discussed in a separate paper) also establish that the films are of high epitaxial quality. Atomic force microscopy data show the rms roughness over $10\,\mu\text{m}\times10\,\mu\text{m}$ area of ~1.8 nm for both the films showing the high surface quality of the films.

In Fig. 2 are shown the data for resistivity dependence on temperature. It is clear that both films show a sharp change in resistivity at ~120 K, the Verwey transition temperature. The transition is of course not as sharp as obtained in some single crystals that are carefully processed for strain release and fixation of oxygen stoichiometry.[5,6,9] Substrate related strains and small departures from the precise Fe:O stoichiometry may be responsible for the observed broadening. The change in the resistivity at the Verwey transition is smaller for (111) than (100) oriented film. In the inset is shown the resistivity data on the expanded scale near 120 K along with the low residual field (0.2 Oe) magnetization data. The sharp decrease in the magnetic moment signifies the expected feature of the Verwey transition. It is to be noted from these data that the transition is broader for the [111] oriented film on α-Al_2O_3 than the [100] film on $SrTiO_3$. This can once again be attributed to the differing strain fields in the two cases. On [0001] sapphire the deposited Fe_3O_4 film has [111] orientation normal to the film plane and hence the [110] edge of its cubic unit cell (*d* value for the 220 plane is 2.967 Å) is the key parameter for the evaluation of the global strain *vis a vis* the 2.75 Å distance between the Al-Al neighbors on the (0001) surface. This mismatch is 7.9% and the strain in the film is compressive. In the case of the growth of (100) oriented Fe_3O_4 on (100) $SrTiO_3$ (STO), comparison of the *d* value of 1.952 Å for the 200 plane of STO and 2.0993 Å for the 400 plane of Fe_3O_4 gives the mismatch of 7.55% and of the same sign as for the case of sapphire. Thus, the global in-plane strains in the two samples are similar. It should be emphasized however that Fe_3O_4 has an inverse spinel structure with a rather peculiar arrangement of Fe and O atoms and hence epitaxial films on (0001) sapphire and (100) STO would differ in microscopic strain fields. Also, homogeneous compressive strains of comparable magnitudes in (100) and (111) directions are bound to be magnetically inequivalent in so far as anisotropy aspects are concerned since the Fe-O networks are not similarly placed *vis a vis* the two orientations.

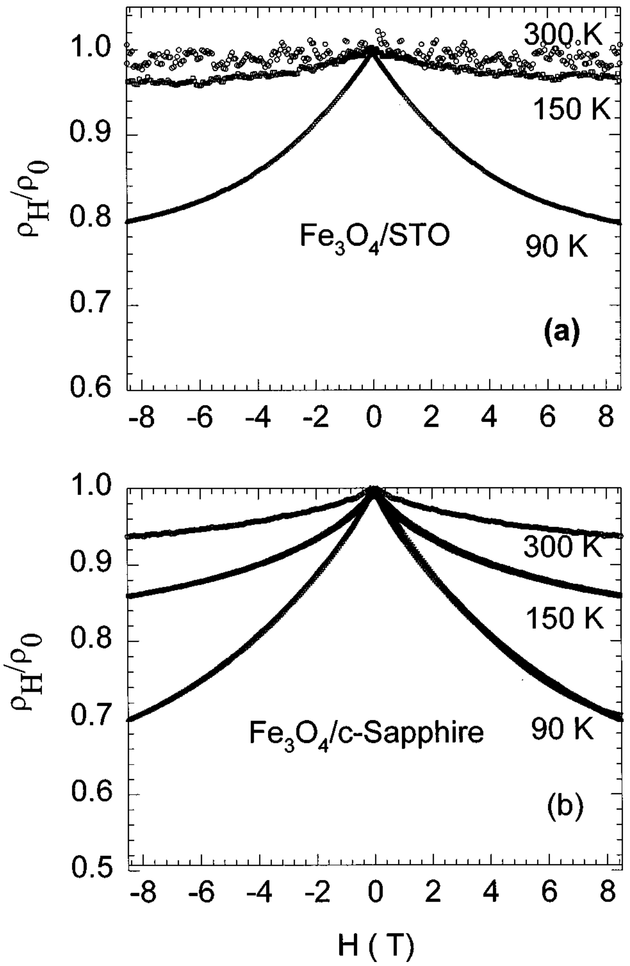

FIG. 3. Dependence of ρ_H/ρ_0 on H (T) for the Fe_3O_4 films deposited on $SrTiO_3$ (100) and sapphire (0001) substrates.

In Fig. 3 are shown the data on the dependence of MR on magnetic field at different temperatures. The field was applied in the film plane without any specific choice of azimuth. Care was taken to implement adequate magnetic cycling to achieve stable values for resistivity in applied field. It may be seen that the MR is higher at room temperature for the [111] oriented films than the [100] oriented films. Below T_v, the films of both orientations show a rapid increase of MR with decrease in temperature and the field dependence is also similar in the two cases. The nature of the field dependence observed for the Fe_3O_4 samples bears a resemblance with the dependence seen in the case of CMR samples below T_c. It is interesting to note that in both these materials a major role of small polarons[7–9,27,28] has been suggested to explain the observed transport features. Below T_c the CMR material is a ferromagnetic metal, while above T_c it exhibits a paramagnetic semiconductor characteristics. On the other hand, Fe_3O_4 is a ferrimagnetic semiconductor from room temperature down to T_v (120 K) and below T_v it is a ferrimagnetic insulator. The observed field dependence of MR therefore appears to be a characteristic feature of polarons in a strongly correlated magnetically polarized system. Indeed, Teresa *et al.*[28] have suggested that the transport in CMR can be described by a narrow polaronic band (strong electron-phonon interaction) along with the magnetic character of the carrier (ferromagnetic cluster) that allows the broadening of the polaronic band under the presence of an applied magnetic field. This qualitative picture is however insufficient to explain the precise nature of the field dependence of resistivity. Therefore, in the spirit of the arguments propounded in previous papers[29–31] on the nature of field dependence observed in the case of CMR materials, we fit our data to an expression $\rho_H = \rho_0\ [1 + \alpha H + \beta H^2]$. The data fit very well to this form and the values of α and β for the 90 K measurements on the Fe_3O_4 films on STO (Sapphire) turn out to be $\alpha = -0.0617\ (-0.049)\ T^{-1}$ and $\beta = 3.157\times 10^{-3}\ (3.06\times 10^{-3})\ T^{-2}$. Equally good fits are also obtained for the data at other temperatures. Very recently, Gong *et al.*[30] have also used a similar dependence for Mott activation energy to explain the transport behavior of (100) oriented Fe_3O_4 film grown on (100) MgO substrate. The rapid rise in MR with decrease in temperature below T_v is intriguing and the corresponding explanation would need a better insight into the magnetic character of the nature of carrier transport below T_v.

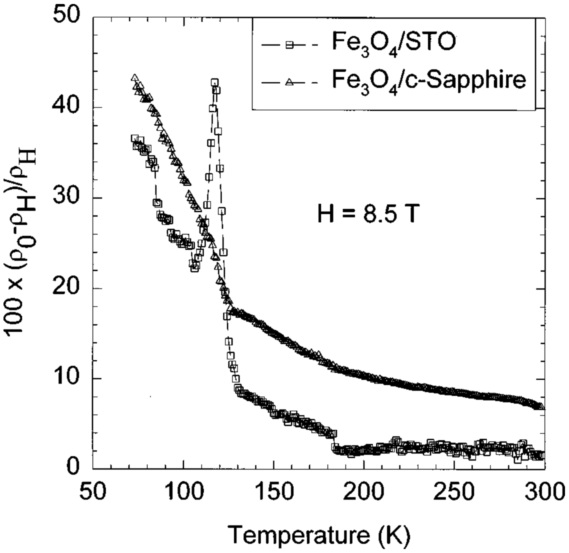

FIG. 4. Temperature dependence of magnetoresistance $[100\times(\rho_0-\rho_H)/\rho_H]$ for the Fe_3O_4 films deposited on $SrTiO_3$ (100) and sapphire (0001) substrates.

In Fig. 4 is shown the temperature dependence of MR $[100\times(\rho_H-\rho_0)/\rho_H]$ for the [100] and [111] oriented films. It can be seen that from 300 K down to about 180 K, the [100] oriented film on $SrTiO_3$ shows no significant MR and no significant change in MR value with decrease in temperature. Below 180 K the MR begins to rise with decreasing temperature and a fairly sharp extremum in its value occurs at the Verwey transition temperature (T_v). A significant rise in MR is observed as the temperature is lowered below T_v. The [111] oriented film on sapphire exhibits a considerably higher MR at 300 K as compared to the [100] case. The MR is seen to rise slowly from 300 K down to 180 K and somewhat more rapidly from 180 K down to T_v. The extremum at T_v is absent for the [111] case. The sharp discontinuity in the slope of the MR curve near T_v is however noteworthy. The MR shows a rapid increase below T_v similar to the [100] case.

It is also interesting to note that below T_v the magnetoresistance shows a rather sharp increase with decrease in tem-

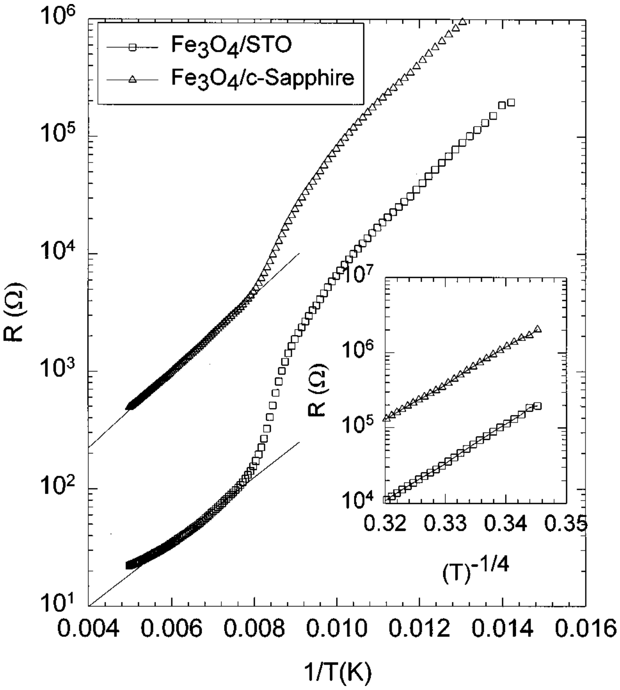

FIG. 5. Dependence of resistance on $1/T$(K) for the Fe_3O_4 films deposited on $SrTiO_3$ (100) and sapphire (0001) substrates. Inset shows the dependence of resistance on $(T)^{-1/4}$ for temperature range below T_v.

perature for films of both the orientations. It is known that above T_v, the transport in Fe_3O_4 exhibits an activated behavior, while below T_v, it has a rather complex character.[7,32–34] Drabble and co-workers[32] showed that below about 70 K the transport appears to be characterized by a variable range hopping (VRH) mechanism[33,34] [$R = R_0 \exp(a/T)^{1/4}$]. Graener *et al.*[33] observed a similar VRH-type behavior from 100 K down to 40 K. However, Pai and Hoing[34] showed that in carefully prepared high purity single crystals, the behavior between about $0.5T_v$ (i.e., 60 K) and T_v is activated type[32] while below $0.5T_v$ it exhibits the VRH character. In Fig. 5 we show the $\ln R$ vs $1/T$ data and in the inset we show the $\ln R$ vs $(1/T)^{1/4}$ plots for $T<0.9T_v$ for films of both orientations. The activation energy values for $T>T_v$ are found to be 52.7 and 62.7 meV for films on STO and sapphire, respectively. These values are close to the value reported for the case of carefully prepared single-crystal samples.[3,5] Under an applied field of 8.5 T the activation energy decreases to 51.7 and 60.7 meV for films on STO and sapphire, respectively. From the VRH fits to the data below T_v we find the values of parameter ''a'' in the VRH formula to be 2.25×10^8 K and 1.823×10^8 K for the films on STO and sapphire, respectively. These values are close to the value of 1.61×10^8 K obtained by Gong *et al.*[30] in their very recent work on (100) oriented Fe_3O_4 films on (100) MgO. In the magnetic field of 8.5 T, these values change to 2.02×10^8 K and 1.67×10^8 K for films on STO and sapphire, respectively.

In order to elucidate the possible causes of the observed magnetotransport and the corresponding anisotropy behavior, it is necessary to discuss the prevalent physical picture concerning transport in Fe_3O_4 below, near, and above the Verwey transition, and the possible influence of an applied magnetic field on the attendant phenomena.

Ihle and Lorenz[7] have emphasized the role of two strong interactions in this system in the context of transport, viz. the electron-phonon interaction that should lead to the formation of small polarons (SP's) and the intersite coulomb interaction which should result into a strong SP-SP correlation. They argue that below T_v, and over some temperature range above T_v, the transport should occur by polaronic band conduction due to the dominance of the SP-SP correlation, while beyond several degrees above T_v, the transport should occur via hopping of small polarons. Within the physical picture provided by Ihle and Lorenz,[7] the application of magnetic field should cause broadening of the small polaronic band leading to enhanced transport in the films of both orientations. As discussed below, the magnetostriction anisotropy and the anisotropy of charge ordering at T_v could cause the differences observed between the [100] and [111] oriented cases in so far as the modification to the polaronic states is concerned.

Heaps[15] has shown that the magnetostriction in Fe_3O_4 exhibits a significant degree of anisotropy and the behavior of the (100) and (111) systems of planes is opposite of each other. When the field is applied along the [100] and [111] directions, there is an expansion and contraction, respectively, along the field direction. The sign of the transverse magnetostriction is opposite to the longitudinal one, as expected. In the case of our [100] and [111] oriented films, the magnetostriction effect should therefore cause distinctly different modifications of internal stresses and correspondingly the phonon spectra. Due to the coupling between the phonon and magnon modes expected and suggested in this system,[5] the coupled phonon-magnon modes would then be differently affected in the films of the two orientations upon the application of the magnetic field. Moreover, the ordering which occurs at T_v has its own lattice and symmetry *vis a vis* that of the space lattice itself and it is very different when seen along the [100] and [111] directions. Thus, given the magnetoelastic coupling in the system, application of magnetic field along these two directions should cause different changes in the nearest- and the next-nearest-neighbor interaction energies that are responsible for the short- and long-range ordering effects in the lattice and consequently the broadening of the SP-SP correlation band and also the observed decrease in the SP hopping energy. Indeed, the polaronic hopping energy is proportional to the inverse square of the optical phonon frequency that would be affected by the application of the magnetic field via coupled phonon-magnon modes. Decrease of the activation energy in our case reflects the stiffening of the magnon and, correspondingly, the phonon-magnon mode due to the applied magnetic field.

Aragon[5] has made a detailed analysis of magnetization and exchange in stoichiometric and nonstoichiometric magnetite over the regimes that show the first- and the second-order Verwey transitions. Since in the present work we have examined the transport in the presence of magnetic field, Aragon's discussion pertaining to magnons, charge and magnetic ordering is of relevance to the analysis of our data. Using the fundamental results of spin-wave theory in the harmonic approximation and from the low-temperature magnetization dependence on T, Aragon has obtained the value of the exchange constant $J_{AB}/K_B = 23$ K, where A and B

correspond to the Fe^{3+}/Fe^{2+} ions on tetra and octahedral sites, respectively. Remarkably, the straight line dependence of M on $T^{3/2}$ is seen by Aragon almost to the onset of the first-order Verwey transition implying that optical magnons are not appreciably excited in this case. He has therefore attributed the small (about 0.1%) decrease in magnetization invariably observed at T_v to the partial condensation of these modes. Iizumi *et al.*[14] have already provided an evidence of condensation of optical phonon modes at T_v, consistent with the proposal of Yamada wherein the coupling between charge density, i.e., the distribution of the Fe^{2+} and Fe^{3+} ions at the octahedral sites, and these phonons has an essential role in causing the lattice instability at the Verwey transition.

The appearance and absence of the MR peak at T_v in the [100] and [111] oriented films, respectively, reflects a distinctly different nature of lattice instability and the process of long-range ordering in the two cases under applied field, as the temperature is lowered. Gridin, Hearne, and Honig[19] have discussed the appearance of the sharp MR peak observed by them in their single-crystal specimen using the established facts regarding the transport sytematics in Fe_3O_4 and thermodynamic arguments. Thus, the resistivity very near T_v is characterized by the relation

$$\rho = \rho_0 \exp(\varepsilon / k_b T),$$

where ε is the activation energy (enthalpy) per Fe_3O_4 formula unit that corresponds to the charge exchange between Fe^{2+} $(3d^6)$ and Fe^{3+} $(3d^5)$ ions on the octahedral site. The activation energy should be a function of T and H. Using the definition of MR as $\phi = [R(T,H) - R(T,0)]/R(T,0)$, Gridin and co-workers[19] show that near T_v, $\phi \sim (\varepsilon_H - \varepsilon_0)/k_b T$. Tracing the discontinuous change in enthalpy to the corresponding change in entropy (σ) via $\Delta\varepsilon_H = \Delta\sigma_H T_v$, Gridin and co-workers[19] obtain an approximate expression for ϕ^*, the discontinuous change in ϕ at T_v, as

$$\phi^* \sim \Delta M H / k_b \delta T_{1/2}.$$

Here, ΔM is the discontinuous change in the magnetization per Fe_3O_4 formula unit at T_v and $\delta T_{1/2}$ is the full width at half maximum of the MR peak at T_v. Thus, in this model, the height of the sharp peak in MR near T_v should be directly proportional to the degree of discontinuous change in the magnetization per formula unit and inversely proportional to the width of the transition. The change of the average magnetic moment extracted from their MR data was found to be of the order of 0.1%, which matches with the percentage dip observed in the magnetization at the Verwey transition. As discussed earlier, Aragon has attributed the small decrease in magnetization at T_v to the partial condensation of optical magnon modes. The observed anisotropy in MR at the Verwey transition in our films should then correspond to the anisotropic features of the optical magnon or the corresponding phonon-magnon dispersion. Since partial condensation of such modes apparently causes the instability at the Verwey transition leading to long-range ordering of Fe^{2+} and Fe^{3+} ions, the influence of a magnetic field on transport phenomena is expected to be different in the two cases.

The precise mechanism behind the occurrence of the MR extremum at the Verwey transition in the case of the (100) oriented Fe_3O_4 film is not clear at this time. As stated earlier, Gridin, Hearne, and Honig[19] have discussed the appearance of the MR extremum observed in their single-crystal specimen based on thermodynamic arguments. While these arguments provide some insight into the attendant phenomena, they fail to give the desirable microscopic picture. Our demonstration of the anisotropy of the MR extremum seems to suggest that the said extremum is related to the extreme sensitivity of the mixed state in the transition region (order-disorder of Fe^{2+} and Fe^{3+} ions) to the magnetic field due to coupled phonon-magnon states. More work is clearly needed to resolve these issues.

In conclusion, we have studied the anisotropy of magnetotransport in epitaxial thin films of Fe_3O_4 deposited on $SrTiO_3$ (001) and sapphire (0001) substrates. It is shown that for field applied in the film plane, the MR shows a sudden change in its dependence on temperature at the Verwey transition T_v for films of both (100) and (111) orientations. However, the sharp MR extremum is seen only for the (100) oriented film. The magnetoresistance is also seen to rise sharply with decrease in temperature below T_v in films of both orientations. The nature of the magnetic field dependence of resistivity resembles the behavior observed in CMR materials below T_c. These magnetotransport data further emphasize the role of polaronic states and phonon-magnon effects in the context of the carrier transport in this system.

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