CHAPTER 4

A COMPARATIVE STUDY OF ANISOTROPIC MAGNETISM AND MAGNETOTRANSPORT PROPERTIES OF EPITAXIAL AND POLYCRYSTALLINE \( \text{Nd}_{0.51}\text{Sr}_{0.49}\text{MnO}_3 \) THIN FILMS

4.1 INTRODUCTION

As described in the previous Chapter 3, among the doped rare earth manganites [10,263], \( \text{Nd}_{1-x}\text{Sr}_x\text{MnO}_3 \) shows strong electron correlations and phase coexistence over a wide temperature range with the occurrence of paramagnetic (PM), ferromagnetic (FM), antiferromagnetic (AFM) and charge/orbital ordered (CO/OO) phases in the composition range \( 0.48 \leq x \leq 0.51 \) [223–226]. At \( x = 0.50 \) the bulk material undergoes a transition from PM to the FM phase at \( T_C \sim 250 \) K followed by a transition to a charge-ordered insulating (COI) phase at \( T_{CO} \sim 150 \) K, which is accompanied by a real-space ordering of the \( d_{(x^2-y^2)} \) and \( d_{(3z^2-r^2)} \) orbitals and the Mn\(^{3+}\) and Mn\(^{4+}\) ions in a checkerboard pattern referred to as the correlated electronic (CE) state. The CE state is composed of zigzag ferromagnetic chains of Mn\(^{3+}\) and Mn\(^{4+}\) ions that are coupled in an AFM manner in the normal direction. Phase separation (PS) between the CE and FM phases occurs in the range \( 0.48 < x < 0.51 \) [10]. It has been recently shown that a small deviation of the order of 1% in Sr doping concentration (e.g., at \( x = 0.49 \)) has a strong influence on the respective phase fraction of the competing phases [231,264]. It has been shown that the AFM-COI phase and the concomitant metal–insulator (MI) transition at \( T_{CO} \) that appear in bulk samples are modified in thin films, indicating the important role of strain induced by film/substrate lattice mismatch [118,230,231,265,266]. It is argued that strain prevents the occurrence of the lattice distortions necessary to accommodate and drive the CO state [230]. Besides all these properties, anisotropic magnetoresistance (AMR) also affected by strain and disordered in manganites thin films [267-274]. To understand the magnetic anisotropy in manganites system, we explored some references [118,265-275], which is explained in next Section 4.2.
CHAPTER 4

4.2 MAGNETIC ANISOTROPY: SOME BASIC ASPECTS

When the internal energy depends on the direction of its spontaneous magnetization with respect to the crystallographic axes, the magnetic material is said to possess magnetic anisotropy. Magnetically anisotropic materials will align its moment with one of the easy axis, and easy axis can be defined as an energetically favorable direction of spontaneous magnetization that is determined from the different sources of magnetic anisotropy, like magneto crystalline anisotropy, shape anisotropy, magnetic surface and interface anisotropy etc., the most important type of anisotropy is the magneto crystalline anisotropy which is caused by the spin orbit (SO) interaction of the electrons. The electron orbitals are linked to the crystallographic structure, and due to interaction of orbitals with spin, orbitals are aligned along a well-defined crystallographic axis. Therefore a magnetic material is easier to magnetize in a particular direction (i.e. easy magnetization axis). The exchange energy is found to be larger than magneto crystalline energy, but the magnetization’s direction is only determined by the anisotropy because the exchange interaction just tries to align the magnetic moments parallel, no matter in which direction.

Figure 4.1: Definition of the direction cosines (taken from [275]).

The magnetization direction $\mathbf{m} = \mathbf{M}/|\mathbf{M}|$ relative to the coordinate axis can be given by the direction cosine $\alpha_i$ as $\mathbf{m} = (\alpha_1, \alpha_2, \alpha_3)$ with, $\alpha_1 = \sin \theta \cos \phi$, $\alpha_2 = \sin \theta \sin \phi$, $\alpha_3 = \cos \theta$. These relations fulfil the condition: $\alpha_1^2 + \alpha_2^2 + \alpha_3^2 = 1$, which is often used below. The magneto crystalline energy per volume $E_{\text{crys}}$ can be described by a power series expansion of the components of the magnetization:

$$E_{\text{crys}} = E_0 + \sum b_{ij} \alpha_i \alpha_j + \sum b_{ijk} \alpha_i \alpha_j \alpha_k + \sum b_{ijkl} \alpha_i \alpha_j \alpha_k \alpha_l + \ldots + 0(h)$$

(4.1)
The higher order terms $O(h)$ with at least the fifth order in ‘$\alpha$’ are very small and can usually be neglected. Since there is no energy difference for oppositely magnetized systems, that is, $E(M) = E(-M)$ or $E(\alpha_i) = E(-\alpha_i)$ therefore, no odd terms of $\alpha_i$ occur in the series expansion that now looks like,

$$ E_{\text{crys}} = E_0 + \sum b_{ij} \alpha_i \alpha_j + \sum b_{ijkl} \alpha_i \alpha_j \alpha_k \alpha_l $$

This is the general equation of energy density for different crystallographic systems like cubic, tetragonal, hexagonal etc.

For a cubic symmetry the energy density can be expressed in terms of the direction cosines and the anisotropy constants as,

$$ E_{\text{crys}}^c = K_0 + K_1 (\alpha_1^2 \alpha_2^2 + \alpha_1^2 \alpha_3^2 + \alpha_2^2 \alpha_3^2) + K_2 (\alpha_1^2 \alpha_2^2 \alpha_3^2) + \cdots $$

The coefficients $K_i$ are called the magneto crystalline anisotropy constants and are functions of the coefficients $b_i$, ...

In case of a tetragonal system due to the reduced symmetry only the indices 1 and 2 are indistinguishable. In this case the energy density can be given by

$$ E_{\text{crys}}^t = K_0 + K_1 \cos^2 \theta + K_2 \cos^4 \theta + K_3 \sin^4 \theta (\sin^4 \varphi + \cos^4 \varphi) $$

$$ = K'_0 + K'_1 \sin^2 \theta + K'_2 \sin^4 \theta + K'_3 \sin^6 \theta + K'_4 \sin^8 \theta \cos 4\varphi $$

For a hexagonal symmetric system, the energy density is given by,

$$ E_{\text{crys}}^h = K_0 + K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta + K_4 \sin^8 \theta \cos 6\varphi $$

For tetragonal and hexagonal systems the magneto crystalline energy is related to a cylindrical symmetry up to terms of the second or forth order, respectively. The energy is only dependent on the angle ‘$\theta$’ between the magnetization direction and the z-axis. Therefore, we find a uniaxial symmetry. The azimuthal angle ‘$\varphi$’ characterizes the anisotropy concerning the basal plane and thus the energy in order to rotate the magnetization in a plane perpendicular to the z-axis. The terms $\cos 4\varphi$ and $\cos 6\varphi$ in equation 4.4 and 4.5, reflect the four- and six-fold symmetry of the tetragonal and hexagonal basal planes, respectively.

In general, it is found that the coefficients (crystal anisotropy constants) depend on material and temperature and that the constants $K_1$ and $K_2$ are sufficient for a good agreement between experiment and calculation. The sign and ratio of the constants determine the easy magnetization axis or the preferred axis, i.e. crystallographic directions in which the magnetization is aligned without external magnetic field. It can be identified by calculating the minimum energy of $E_{\text{crys}}$. 

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Magneto crystalline anisotropy is not exhibited by polycrystalline samples which have no preferred orientation of the grains. It has an overall isotropic behaviour concerning the energy being needed to magnetize it along an arbitrary direction. But if the sample is not spherical then one or more specific directions occur which represent easy magnetization axis which is caused by the shape, this phenomenon is called shape anisotropy.

The relationship \( \mathbf{B} = \mu_0 (\mathbf{H} + \mathbf{M}) \) only holds inside an infinite system. A finite sample exhibits poles at its surfaces which lead to a stray magnetic field outside the sample. This occurrence of a stray field results in a demagnetizing field inside the sample. The energy of a sample in its own stray field is given by the stray field energy,

\[
E_{\text{str}} = -\frac{1}{2} \int \mu_0 \mathbf{MH}_d \, dV
\]

with \( \mathbf{H}_d \) being the demagnetizing field inside the sample. The calculation is rather complicated for a general shape. It becomes easier for symmetric objects like ellipsoid. For thin magnetic films and multilayers the stray energy is given by,

\[
E_{\text{str}} = K_0 + K_{\text{shape}} \sin^2 \theta
\]

with \( K_{\text{shape}} \propto -M^2 < 0 \). \( K_0 \) and \( K_{\text{shape}} \) are anisotropy constants. The stray field energy reaches its minimum value at \( \theta = 90^\circ \). This means that the shape anisotropy favours a magnetization direction parallel to the surface (i.e. within the film plane). It is found that the shape anisotropy dominates the magneto crystalline anisotropy which results in an in-plane magnetization for thin film systems.

Magnetic surface and interface anisotropies play a crucial role in the low-dimensional systems like thin films. At interfaces due to the broken symmetry the anisotropy energy contains terms with lower order in \( \alpha \) which are forbidden for three-dimensional systems. Therefore the effective anisotropy constant \( K^{\text{eff}} \) is divided into two parts, describing the volume and the surface contributions:

\[
K^{\text{eff}} = K^V + 2 \frac{K^S}{d}
\]

where, \( K^V \) is the volume dependent magneto crystalline anisotropy constant and \( K^S \) is the surface dependent magneto crystalline anisotropy constant. The factor of two comes here because of the creation of two surfaces. The second term exhibits an inverse dependence on the thickness \( d \) of the system. Therefore, it is only important for thin films. Thus, the volume contribution always dominates for thick films with a magnetization being within the film plane. The relative amount of the surface contribution increases with decrease in thickness.
followed by a spin reorientation transition towards the surface normal below critical thickness \(d_c\).

Apart from colossal magnetoresistance (CMR), another important magnetotransport property of doped rare earth manganites is anisotropic magnetoresistance (AMR). The anisotropic magnetoresistance (AMR) of a material provides a measure of the dependence of the resistivity on the direction of the magnetization. This phenomenon has been mainly studied in polycrystalline or amorphous materials, where it depends only on the angle between the current and the magnetization [266–274].

**4.3 PRESENT WORK**

The AMR of manganites thin film can be utilized for storing memory in two different directions (in-plane and out-of-plane) within the same used area. Nonspherical charge distribution possesses an orientation dependent energy due to its interaction with the surrounding lattice electric field, and through the spin–orbit interaction this leads to an angular dependent contribution from the total moment to the overall energy of the system, resulting in magneto-crystalline anisotropy [266]. In manganite thin films the non-spherical charge distribution around Mn ions gets further modified by substrate induced strain and lattice defects [118,265]. A reduction in dimensionality (e.g., in thin films) of such a system enhances the easy axis magnetic anisotropy and a decrease in electrostatic screening, which favors directional tunneling of electron through the insulating stripe domain walls formed at the phase-separated FM domain boundaries [272]. In manganites both strain and magneto-crystalline anisotropy are reasoned for the occurrence of AMR near the insulator–metal transition temperature (\(T_{IM}\)) but detailed explanation for this is still debatable [267]. Apart from the strain and hence the film thickness other factors such as the structural defects, spin disorder, nature of the magnetic ground state, phase coexistence, etc. are also expected to play a crucial role. So far the low field anisotropic magnetoresistance (LF-AMR) has not been explored and investigated in manganites with mixed ground state having strong phase coexistence. In this Chapter, we are going to present a comparative study of magnetism and various magnetotransport properties of epitaxial and polycrystalline thin films of nearly half-doped manganite \(\text{Nd}_{0.51}\text{Sr}_{0.49}\text{MnO}_3\) (NSM49), we have also studied the impact of compressive strain and polycrystallinity on the out-of-plane AMR in NSM49 thin films that show strong phase coexistence.
Thin films were prepared by on axis DC magnetron sputtering on single crystal (001) oriented LaAlO$_3$ (LAO) and (100) oriented Yttria stabilized ZrO$_2$ (YSZ) substrates from a stoichiometric Nd$_{0.51}$Sr$_{0.49}$MnO$_3$ target synthesized by the solid state reaction route. Synthesis processes for both the films are same as explained in Chapter 3. The details of the deposition conditions are given in Section 2.4 of Chapter 2. In order to see the effect of strain we have prepared 30 and 100 nm thick films on LAO single crystal, while the thickness of the film on single crystal YSZ (film is polycrystalline in nature) was kept 100 nm for comparative study with the film of same thickness 100 nm on single crystal LAO (film is epitaxial in nature).

Several characterization techniques have been used to study different properties of both varieties of films. The cationic composition of films was probed by energy dispersive X-ray spectroscopy (EDS or EDX) analysis and the structure was investigated by X-ray diffraction (XRD). Temperature dependent DC magnetization was measured by a commercial SQUID magnetometer (MPMS-XL) and the temperature (77–300 K) and magnetic field (H \leq 3.4 kOe) dependent electrical transport was measured by the four-contact method.

We have studied the some magnetotransport properties such as anisotropic magnetoresistance (AMR) and magnetoresistance (MR), in two measurement configurations in both of which the current is applied parallel to the film surface. In the first one (designated IN) the current is along the film surface and the magnetic field is applied parallel to the current (H$_{\parallel}$ and H$_{\parallel}$). In the second one (designated OUT) the magnetic field is applied normal to the film surface and is hence perpendicular to the current (H$_{\perp}$ and H$_{\perp}$).

In the following sections the 30 and 100 nm thick films on LAO will be denoted by L30 and L100, respectively, while the 100 nm film on YSZ will be referred to as Z100. Detail explanation with results and discussion of these characterizations given in next section.

**4.4 RESULTS AND DISCUSSION**

**4.4.1 ELEMENTAL ANALYSIS**

Energy-dispersive X-ray spectroscopy (EDS or EDX) analysis was carried out at several places on the films and the average cation composition was found to be Nd/Sr/Mn: 0.507/0.493/0.982, which is very close to the target composition. Good chemical homogeneity of the films is evidenced by very small spatial variation in the cationic composition.

**4.4.2 STRUCTURAL CHARACTERIZATION**

The X-ray diffraction (XRD) data shows that the target material is of single phase with a distorted perovskite structure ($Pbnm$-space group). The cell parameters are found to be,
a ~ 5.477 Å (a/√2 ~ 3.873 Å ), b ~ 5.449 Å (c/√2~3.853 Å ) and c ~ 7.649 Å (b/2~ 3.824 Å ). The average in-plane lattice parameter of the bulk target, a_{av} = (a/√2 + b/2)/2 ~ 3.863 Å, is larger than the in-plane lattice parameters of the LAO substrate used (a_{LAO} = 3.798 Å). Thus, Nd_{0.51}Sr_{0.49}MnO_3 films on LAO are expected to grow with in-plane compressive strain. In contrast the large difference between the lattice parameters of the target and the YSZ substrate is expected to result in polycrystalline growth.

The estimated out-of-plane (OP) lattice parameter of L30 film is about a_{op} = 3.918 Å, which is considerably larger than that of the bulk target (a_c = 3.824 Å). The elongated OP lattice constant confirms the presence of in-plane compressive strain. In the case of L100, each (001) diffraction maximum splits into two, the first of which coincides with that of the L30, while the second corresponds to a smaller OP lattice constant (3.867 Å) as shown in Figure 4.2.

![Figure 4.2: XRD graph of NSM49 target, L30, L100 and Z100 thin films, arrows on L30, and L100 graphs shows the material peaks.](image)

This suggests that the strain is only partially relaxed and non-uniform in the thicker film, viz. L100. The film on YSZ (Z100) is also of single phase but polycrystalline in nature. However, the occurrence of only selected peaks {(0ℓ0) and (212)} shows some degree of preferential
orientation in these films. The near equality of the OP of Z100 and the bulk target suggests the absence of any strain effect due to the substrate.

4.4.3 MAGNETIC AND ELECTRICAL PROPERTIES

To study the magnetic and electrical properties of these films we employed some characterizations techniques mentioned in subsection 4.4.2. The paramagnetic–ferromagnetic (PM–FM) transition was determined from the zero field cooled (ZFC) magnetization data taken at a constant magnetic field \( H = 500 \) Oe applied parallel to the film plane. The FM transition is observed at \( T_C \approx 200, 225 \) and \( 260 \) K in L30, L100 and Z100, respectively which have been shown in Figure 4.3. In these graphs we can see that multiple transitions are observed at \( T < T_C \) in all films. At \( T < T_C \), FM to A-AFM phase is observed, which is followed by a transition to the CO state at \( T_{CO} \). A detailed analysis of the zero field cooled (ZFC) and field cooled (FC) magnetization data of these films reveals that the magnetic ground state is dominantly FM, where the CO-AFM clusters are embedded in the ferromagnetic matrix.

![Figure 4.3](image)

**Figure 4.3:** Variation of magnetization with temperature of all the films in ZFC mode.

Such an admixture of AFM (superexchange) and FM (double exchange) phases (interactions) could cause the occurrence of cluster glass. This, as revealed by the divergence in the ZFC–FC magnetizations indeed is the case in all the three films [264, 276].
The electron transport property i.e. temperature dependent resistivity, of all the films is in very good agreement with the magnetization data as shown in Figure 4.4. The insulator–metal transitions for all films are observed at $T_{IM} \approx 170$ K, 230 K and 140 K for L30, L100 and Z100 thin films, respectively.

![Figure 4.4: Variation of resistivity with temperature for all the films.](Image)

In order to check the magnetic anisotropy in these films the isothermal $M$–$H$ loops were measured with $H$ applied parallel to the film plane surface ($H_{||}$) as well as normal to the film plane ($H_{\perp}$). The representative $M$–$H$ loops of L100 and Z100 are shown in Figure 4.5. Among the two epitaxial films the in-plane $M$–$H$ loop of the 100 nm film (L100) is relatively narrow and more upright, suggesting better FM features as suggested also by the ZFC-$M$–$T$ data. The in-plane ($H_{||}$) coercivity of L30 and L100 is $H_{C||} \approx \pm 876$ and $\pm 390$ Oe, respectively. L30 and L100 show in-plane ($H_{||}$) saturation magnetization $M_{S||} \approx 505$ and 760 emu/cm$^3$, respectively. In the polycrystalline film on YSZ (Z100) the in-plane ($H_{||}$) $M$–$H$ loop is symmetric about the $H$-axis and coercivity and saturation magnetization are found to be $H_{C||} \approx \pm 910$ Oe and $M_{S||} \approx 805$ emu/cm$^3$, respectively. When the magnetic field was applied normal to the films surface ($H_{\perp}$), the corresponding out-of-plane $M$–$H$ loops were found to be tilted from the vertical by angle $\Phi$. The value of the tilt angle was found to be $\Phi \approx 27^\circ$ in case of the single crystalline films (L30 and L100) and $\Phi \approx 17^\circ$ for the polycrystalline film (Z100). Since a large value of angle $\Phi$ indicates a smaller out-of-plane magnetization as compared to
the in-plane magnetization, it could be regarded as a measure of the degree of anisotropy. The out-of-plane coercivity ($H_{c\perp}$) and saturation field were enhanced appreciably, while the corresponding saturation moment ($M_{s\perp}$) was reduced. For example, the typical observed values were, $H_{c\perp} \approx 670$ Oe, $H_{c\perp} \approx 8.5$ kOe and $M_{s\perp} \approx 550$ emu/cm$^3$ for the 100 nm film on LAO. Similar variation trend was also observed in L30 and Z100 also.

![Image of isothermal M–H loop measured at 4 K of (a) L100 and (b) Z100 IN and OUT, respectively, indicate that the field is applied in the plane and normal to the plane of the films.](image)

**Figure 4.5:** Isothermal M–H loop measured at 4 K of (a) L100 and (b) Z100 IN and OUT, respectively, indicate that the field is applied in the plane and normal to the plane of the films.

The M–H loops described above show that the magnetization easy axis lies in the plane of the films. The difference in the in-plane and out-of-plane magnetic behaviors of the present films can be understood in terms of the different types of anisotropies that are at play, such as, magneto-crystalline, shape and surface anisotropies [275]. The magnetic anisotropy is generally defined as the energy required to rotate the magnetization direction from the easy to the hard axis. Magneto-crystalline anisotropy originates from the interaction between the electronic spin and orbital degrees of freedom. The electronic orbitals are linked to the crystallographic structure and their interaction with the spins causes the latter to preferentially align along well defined crystallographic axes. Therefore, there are directions in space (generally referred to as the easy axis) along which a magnetic material is easier to be magnetized than in other ones. However, polycrystalline samples without a preferred granular orientation do not possess any magneto-crystalline anisotropy. But, an overall isotropic
behaviour concerning the energy needed to magnetize it along an arbitrary direction is only
given for a spherical shape. For non-spherical shapes there are one or more specific
directions, solely caused by the shape, which represent easy magnetization axes. This
anisotropy is known as shape anisotropy. The direction of magnetization is determined by the
competing magneto-crystalline and shape anisotropies. The constant characterizing magneto-
crystalline anisotropy is found to be smaller than that characterizing the shape anisotropy and
hence the latter dominates the former, which results in an in-plane magnetization for thin film
systems. In low dimensional systems such as thin films, the anisotropies are affected further
by the broken symmetry at the interfaces and hence additional contributions that are forbidden
in three dimensional cases arise. Therefore effective anisotropy constant (\(K_{\text{eff}}\)) can be
regarded as consisting of two parts, one related to the volume (\(K^V\)) and the other related to the
surface (\(2K^S/d\)) contribution. The surface contribution exhibits an inverse dependence on the
thickness (d) of the system (such as thin films) and hence becomes the dominant contribution
in thin films. The relative amount of surface contribution increases with decreasing film
thickness and below a critical value a spin reorientation towards the normal to the surface
occurs. The energy of a sample in its own stray magnetic field is called the stray energy and is
given in equation 4.6.

In the present case, the shape anisotropy is the most dominant factor in determining
magnetic anisotropy. The surface anisotropy contribution is expected to be larger in L30 than
in L100. Thus the more upright M–H loop, smaller values of \(H_C\) and \(H_S\), and larger \(M_S\) that
suggest better FM features in L100 (as compared to L30) could be caused by reduced
contribution from the surface anisotropy. In case of polycrystalline samples without any
preferred growth, the grains do not possess magneto-crystalline anisotropy. Instead the shape
anisotropy is known to play the dominant role. In partially oriented polycrystalline film such
as Z100 in the present case both magneto-crystalline anisotropy and shape anisotropy are
expected to be present, of which the latter is the dominant one. We would like to suggest that
presence of grain boundaries could also lead to enhanced surface anisotropy contribution in
the polycrystalline film (Z100). In addition to the magnetic anisotropies at play, the intrinsic
nature of the compound, the strong phase fluctuation caused by the occurrence of multiple
magnetic phases, such as FM, A-type AFM and CE-type AFM-CO in the vicinity of half
doping, could be one of the factors determining the easy axis to be in the plane of the film.
We must point out that out-of-plane easy axis has also been reported in compressively
strained thin films of prototype manganites, e.g., \(\text{Pr}_{0.67}\text{Sr}_{0.33}\text{MnO}_3\) [121]. However, the
thickness of these films (5–15 nm) is much smaller than that of the one studied in the present case and at such lower thicknesses spin reorientation along the out-of-plane direction could be one of the possible reasons for the out of-plane easy axis.

4.4.4 OUT OF PLANE ANISOTROPIC MAGNETORESISTANCE

In this section we are going to present some magnetotransport properties such as anisotropic magnetoresistance (AMR) and magnetoresistance (MR) by applying an external magnetic field (H) along the in-plane and out-of-plane to the film for both varieties of films.

The magnetotransport measurements were carried out in temperature range 77–300 K and magnetic field (H \leq 3.4 \text{kOe}) applied parallel (H \parallel I) and normal (H \perp I) to the transport current. The low field AMR was calculated from the expression described earlier. The LFMR is defined in chapter 1, equation 1.1.

The representative temperature dependences of the LFMR measured at H = 3.4 \text{kOe} parallel to the current (IN) and perpendicular to it (OUT) are shown in Figure 4.6.

![Figure 4.6: Temperature dependence of low field magnetoresistance (LFMR) measured at H = 3.4 \text{kOe} applied parallel (IN) and perpendicular (OUT) to the transport current.](image)

L30 shows a large LFMR \approx 35\%, which in the thicker film (L100) decreases to \sim 25\%. In the current perpendicular to the current configuration the LFMR is drastically...
reduced in all the films. The peak in MR–T curves in both the films is observed below $T_{\text{IM}}$. In contrast to the epitaxial films, films on YSZ show temperature dependence akin to those of the polycrystalline manganites [11], in which LFMR (at $T = 78$ K and $H = 3.4$ kOe LFMR $\sim 26\%$) decreases monotonically as temperature increases.

Our one of the main objective of this chapter is to study this magnetotransport property which is low field anisotropic magnetoresistance (AMR). The temperature dependence of the low field AMR (LF-AMR) of all the films is plotted in Figure 4.7.

![Figure 4.7](image)

**Figure 4.7:** Temperature dependence of anisotropic magnetoresistance (AMR) at constant field of 3.4 kOe.

L30 and L100 show maximum LF-AMR $\approx -15\%$ and $-13\%$ at $T (< T_{\text{IM}}) = 130$ and 200 K, respectively. The LF-AMR of Z100 increases monotonously as temperature is lowered, which is similar to that of the ferromagnetic alloys [118].

We have also measured the isothermal AMR near the temperature corresponding to the highest MR, as a function of magnetic field. The magnetic field dependence of the AMR measured at temperatures in the vicinity of the peak MR is shown in Figure 4.8. As seen in **Figure 4.8**, AMR–H shows a peak in all the three films. In L30, AMR peaks to 18% at $H = 1.55$ kOe. In the thicker film (L100) AMR decreases considerably and acquires the maximum value $\approx 14\%$ at $H = 2.8$ kOe. In contrast to the single crystalline thin films the polycrystalline film on YSZ shows the highest AMR, which peaks to $\sim 20\%$ at $H = 1.7$ kOe (at 78 K).
must mention here that in the vicinity of $T_{IM}$, where AMR peaks in the two single crystalline films (L30 and L100), the AMR–H curves did not show any appreciable hysteresis. Hysteresis is pronounced only in the lower temperature regime in these films but LFMR as well AMR is considerably reduced. In contrast, the polycrystalline film (Z100) showed appreciable hysteresis at $T<100$ K and is clearly seen in the AMR–H curve measured at 78 K (Figure 4.8). Among the epitaxial thin films the thinner one (L30) has a higher degree of structural and magnetic disorder that results in lower FM and IM transitions. This is caused by the large in-plane compressive strain and the occurrence of the magnetically disordered dead layer at the substrate film interface.

![Figure 4.8](image)

**Figure 4.8:** Magnetic field dependence of anisotropic magnetoresistance (AMR) for L30, L100 and Z100 films at constant temperature 130, 200 and 78 K, respectively.

The increased film thickness and the associated strain relaxation dilute the role of the dead layer in the thicker film (L100). In the present case higher AMR (~18%) in the fully strained film (L30) as compared to the partially strained one (14% in L100) suggests that larger compressive strain may lead to higher AMR. The consequences of the magnetically disordered dead layer with respect to anisotropy are not very clear. However, the present results demonstrate that it is not detrimental to AMR. The higher value of AMR in the case of more disordered L30 film suggests that disorder, in general, may be an important factor in determining the magnitude of AMR. This is also supported by the results on the
polycrystalline film on YSZ (Z100), which has the highest degree of disorder among the three films. As seen in Figure 4.7 and Figure 4.8, Z100 shows the highest AMR among the three films.

Large LF-AMR in the current films could also be attributed to the intrinsic nature of \( \text{Nd}_{0.51}\text{Sr}_{0.49}\text{MnO}_3 \). In \( \text{Nd}_{1-x}\text{Sr}_x\text{MnO}_3 \) the occurrence of PM, FM, AFM and CE type CO/OO phases around \( x = 0.48–51 \) makes its phase diagram multicritical [10]. For example the large drop in the resistivity of the NSM49 films in the present study is a consequence of bicritical nature. Just below the bicritical point the FM phase appears abruptly with a sudden disappearance of the PM component. However, the CE type CO/OO clusters may still be present in small fraction in both PM and FM phases. These aspects are discussed in detail in Refs. [264,276]. Such intrinsic phase coexistence is not found in standard CMR materials such as \( \text{Pr}_{0.70}\text{Sr}_{0.30}\text{MnO}_3 \) and \( \text{La}_{0.70}\text{Ca}_{0.30}\text{MnO}_3 \) [10,11].

![Figure 4.9](image_url)

**Figure 4.9:** Magnetic field dependence of low field magnetoresistance (LFMR) measured at \( T = 77 \) K with \( H \) applied parallel (IN) and perpendicular (OUT) to the transport current.

Hence one of the factors contributing to large AMR could be the intrinsic multicritical nature of \( \text{Nd}_{0.51}\text{Sr}_{0.49}\text{MnO}_3 \). In order to understand the origin of the peaks in the AMR–\( H \) curves, we separately measured the in plane and out-of-plane LFMR and the experimental data are plotted in Figure 4.9. The in-plane and out-of-plane LFMR show different magnetic field dependences. In the epitaxial films (L30 and L100) the in-plane LFMR rises with magnetic field and no saturation like effect is seen. On the other hand, the in-plane LFMR of
The polycrystalline thin film shows a sharper rise at smaller magnetic fields and then saturation like behavior is seen. This behavior is typical to the polycrystalline manganites [11].

The out-of-plane LFMR of all the films, irrespective of the crystallinity and nature, has nearly the same magnetic field dependence. In this configuration, LFMR increases slowly at smaller magnetic field and then rises more rapidly with nearly linear magnetic field dependence. This difference in the in-plane and out-of-plane LFMR could be due to the difference in nature of the coupling of magnetization with the applied magnetic field. The strength of coupling of magnetization depends on the angle between the magnetization vector of the film and the applied magnetic field. In our case the magnetization vector of the film is oriented along the film plane (see Figure 4.5), and as H is in the plane of the film the coupling is strong and when it is normal to the film the coupling is weak [121,271,275]. The temperature and field dependences of LFMR and AMR and the variations of their magnitude could also be understood in terms of the magnetic domain motion under the action of an applied magnetic field [277]. When the magnetic field is in the plane of the film, because of the in-plane easy axis, the strong in-plane coupling of magnetization with applied H results in maximum magnetic domains alignment along the field direction and resistance is lowered. When the magnetic field is applied normal to the plane (out-of-plane), much larger magnetic field is required to rotate and align all the domains along the magnetic hard axis (coupling of magnetization weaker) [121,272]. Consequently the out-of-plane LFMR is smaller and also has different field dependence than that of the in-plane. This causes a large AMR as well as a peak in the AMR–H curves observed in the present films. As discussed above, the thickness dependence of LFMR and AMR could be due to the change in the domain wall width with variation in film thickness. The decrease in LFMR with increasing film thickness suggests that the domain wall thickness at lower film thickness is smaller. The large LF-AMR in the polycrystalline films (Z100) could be caused by the dominance of the shape over the magneto-crystalline anisotropy.

Our investigations shows that in single crystalline film on LAO substrate the AMR, (i) decreases with increasing film thickness, (ii) shows a peak just below the $T_C/T_{IM}$, and (iii) peak becomes narrower with increased film thickness. The AMR values observed in the present study are slightly higher than observed in case of thin films of nearly similar composition. Recently it has been reported by Egilmez et al. [97], that in the Nd$_{0.55}$Sr$_{0.45}$MnO$_3$ the maximum MR and AMR is expected to be $\sim 30\%$ and $\sim 7\%$, respectively. The
composition and bandwidth dependence of AMR has recently discussed by Egilmez et al. [97] in their review that provides a detailed discussion of the AMR in manganites. Our results show that the MR and AMR of the single crystalline films are dependent on the strain state of the film. The film under larger compressive strain shows higher MR and AMR values than the thicker (~100 nm) film which is under smaller compressive strain. R. Patterson et al., have studied the MR and AMR properties of vacuum annealed La_{0.65}Ca_{0.35}MnO_{3−δ} films across the metal-insulator transition [270]. They have attributed the observed increase in resistivity and the concomitant decrease in the MR and AMR out peaks to the epitaxial strain relaxation resulting from thermal treatment and oxygen vacancy creation. Egilmez et al., have studied the thickness dependence of the AMR in La_{0.65}Ca_{0.35}MnO_{3} thin films and observed a decrease in the LFMR as well as AMR with increasing film thickness [271]. Their data suggests that an increasing epitaxial lattice strain that develops as the film thickness is decreased is responsible for an increase of both the MR and AMR. The results relating to the AMR magnitude, the thickness and strain dependence of AMR, etc., in present study shows a very good agreement with these reported results. O’Donnell et al., also have reported nearly similar AMR values as in the present case in their MBE grown La_{0.7}Ca_{0.3}MnO_{3} thin films [268]. In case of the polycrystalline film the absence of AMR peak and monotonous increase in MR and AMR values with temperature lowering suggests that the disorder (grain boundaries and higher density of defects like dislocations, stacking faults, etc.) could be the main reason for this. In this case, as shown by Liu et al. [278], the anisotropy in the spin polarized tunnelling could play important role.

The understanding of the occurrence of a peak in the AMR in the vicinity of T_C/T_{IM} in manganites is still vague. The conventional theories in which the anisotropic scattering of extended metallic conduction electrons causes the AMR and that account for the AMR in metals and their alloys have failed to describe the same in manganites. As proposed by O’Donnell et al., the magnetotransport anisotropies are instead due to a local spin–orbit induced orbital deformation that influences the local hopping conduction process characteristic of manganites near T_{IM}. The importance and relevance of such orbital deformations in manganites has been demonstrated in several manganite thin films grown by MBE and PLD [267,279]. The overlap between neighbouring ions is changed by the orbital deformation as the magnetization is rotated, which in turn modulates the conductivity through a changing hopping probability. In this regard the carrier localization and the JT effect plays a very important role as pointed out by Srivastava et al. [280]. The reduced AMR, deep in the
FM region has been shown to be consistent in magnitude with that predicted by conventional metallic theory. As far as the peak in the AMR – T near $T_C/T_{IM}$, is now believed to be caused by the occurrence of the spin fluctuations that are enhanced in region of strong competition between the DE mediated FM and SE mediated AFM phases. Here the JT distortion which favours SE and enhances carrier localization is expected to play crucial role. The FM-DE (spin) and J-T distortion (orbit) are delicately balanced near $T_C/T_{IM}$. As mentioned earlier, the dominance of the shape anisotropy results in an in-plane easy axis and hence application of even a small magnetic field parallel to the film plane destroys this balance in favour of the FMM and the resistivity decreases sharply. Further, owing to the anisotropic nature of the enhanced J-T distortions, the magnetic field response of the out-of-plane and in-plane components may be different. This is because the anisotropic J-T distortions could alter the spin-orbit interaction of the system via changing the magnetic interaction through the manganese ions.

It has also been suggested that bandwidth and disorder including quenched disorder plays a very important role in determining the AMR of manganites. It has been reported recently by Srivastava et al. [280], that the AMR in manganites is strongly related to the 'e$_g$' electron bandwidth and hence carrier localization. They have observed that the AMR measured at $H = 4$ kOe increases with increasing bandwidth (and hence carrier localization) and approaches as high as ~60 % in Nd$_{0.55-x}$Sm$_x$Sr$_{0.45}$MnO$_3$ at $x = 0.45$. They have contributed this to, (i) enhanced spin fluctuations, (ii) the anisotropic nature of the J-T distortions, and (iii) strong spin-orbit coupling due to unquenching of the orbital angular momentum of $t_{2g}^3$ and $e_{1g}$ configuration in the strong carrier localization regime (higher $x$). For example in the present study the activation energy for small polaron hopping ($E_A$) is found to be ~138 meV and ~88 meV, [264] for the 30 nm and 100 nm films respectively. This clearly shows that carrier localization is much stronger in case of the thinner 30 nm film. As mentioned earlier this particular film also shows AMR as well as LFMR higher than the 100 nm film. This shows that the carrier localization plays important role in the AMR and LFMR in manganites. Thus, the enhancement in the AMR could be attributed to (1) enhanced carrier localization, (2) anisotropic nature of the J-T distortion, and (3) the enhanced spin-orbit coupling due to unquenching of the 3d orbitals and associated angular momentum.
4.5 SUMMARY

In the present chapter, the in-plane and out-of-plane LFMR and out-of-plane AMR in single crystalline and polycrystalline Nd$_{0.51}$Sr$_{0.49}$MnO$_3$ thin films have been studied and compared.

- In all the films the magnetic easy axis lies in the plane of the film. The presence of magnetic anisotropy in the polycrystalline film shows that the shape anisotropy is dominant over the magneto crystalline anisotropy.

- All films show large LFMR when the applied magnetic field is parallel to the plane of the film (H\parallel Plane) and H\parallel I. The LFMR is lowered considerably when H \perp plane and H \perp I.

- The large LF-AMR (13–15% at 3.4 kOe) in both varieties of films suggests that the shape anisotropy dominates over the surface and magnetocrystalline anisotropies.

- The LFMR and the LF-AMR of the single crystal films show a weak dependence of the films on film thickness (and hence the degree of strain) and could be related to the thickness dependent variation in the domain wall thickness.

- The AMR in single crystalline films shows a peak near the T$_{\text{IM}}$. In contrast the polycrystalline film shows a monotonous increase in AMR on lowering the temperature.

- The magnetic field dependence of AMR shows a peak. Large AMR observed in the present thin films could be attributed to the occurrence of multiple magnetic phases that causes strong phase fluctuations.