Chapter-III

Magnetotransport Studies on GdBa$_2$Cu$_3$O$_z$ Superconductor
3.1 Introduction

High temperature superconductors (HTSC) have attracted special attention in last twenty years mainly due to their negligibly small resistance and exclusion of magnetic flux below superconducting transition temperature \(T_C\). In comparison with conventional type-I superconductors, type-II HTSC has attracted much interest of researchers and scientists essentially due to their higher \(T_C\) values well above liquid nitrogen (LN\(_2\)) temperature (i.e. 77K). This success in finding high temperature superconductivity in type-II superconductors promoted more interest and attention in research and development on cuprate based superconductors as a new challenge both from fundamental as well as application point of views.

HTSC based thin films have attracted extraordinary consideration because of their potential applications in electronic devices such as high \(T_C\) transistors [1], resistive switching device [2], field effect devices [3, 4], field induced tunable device [5], spin injection device [6], superlattices [7], etc. In last decade, varieties of fundamental studies have been carried out in superconductor based thin film devices and heterostructures for their practical applications [2 – 7]. Zhang et al [2] have studied different states of resistance and switching between them in YBa\(_2\)Cu\(_3\)O\(_{6+x}\) / Nb-doped SrTiO\(_3\) heterostructures and different states of resistance and their switching have been discussed on the basis of trapping and detrapping process via oxygen vacancies near the interface of YBa\(_2\)Cu\(_3\)O\(_{6+x}\) / Nb-doped SrTiO\(_3\) and conducting filaments through the junction barrier. Matthey et al [3] have studied NdBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) / SrTiO\(_3\) film for its electric field dependent superconducting transition temperature, mobile carrier density and in-plane penetration depth using field effect device geometry while Salluzzo et al [4] have studied the superconductor based field effect device for its electronic phase separation and electric field induced doping effect in Nd\(_{1+x}\)Ba\(_{2-x}\)Cu\(_3\)O\(_{7-\delta}\) film grown on single crystalline (100) SrTiO\(_3\) substrate.
Since the discovery of superconductivity, for researchers and scientists, it is very curious question to justify on effect of externally applied magnetic field on the transport and electrical behavior of HTSC. Upon application of magnetic field, resistive transition gets broadened along with the shift in $T_C^0$ (at which resistivity becomes negligibly small) towards lower temperature [8 – 15]. Almost all families of superconductors have been studied for their magnetic field dependent resistivity broadening in superconducting transition behavior including Y123 [8], Tl1234 [9], Bi2212 [10, 11], Fe based [12 – 15], etc. Xiaojun et al [8] have studied simultaneous effect of temperature and magnetic field on activation energy in YBa$_2$Cu$_3$O$_{7-\delta}$ (Y123) superconducting epitaxial thin films wherein they have employed thermally excited flux creep model and Arrhenius law to derive activation energy values. Khurram et al [9] have exploited thermally activated flux flow (TAFF) mechanism to understand dissipation mechanism in Tl$_{1.4}$Ca$_2$Ba$_2$Cu$_3$O$_{12-\delta}$ (Tl1234) superconductors and have shown that flux pinning characteristic depends on the microstructure of the samples. Similarly, Sharma et al [11] have investigated the role of grain morphology in deriving activation energy and its dependence on the temperature and applied magnetic field for sol-gel grown Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) superconductors. Recently, much attention has been devoted to the findings of and investigations on superconductivity and other related properties of Fe based superconductors [12 – 15]. Furthermore, magnetic field effects on the transport and electrical properties, especially resistivity and resistive broadening behavior, in SmFeAsO [12], Fe$_{1+y}$(Te$_{1+x}$S$_x$)$_2$ [13], S-doped FeTe [14], and FeSe0.9-xMx (M = Si and Sb) [15] superconductors have been studied. TAFF mechanism is used to understand resistive broadening characteristic under applied magnetic field, pinning energy variation with temperature - field, flux motion with temperature - field, and temperature - field dependent activation energy in Fe based superconductors [13 – 15]. Also, Pandya et al have demonstrated the fluctuations in excess conductivity analysis of resistive broadening and its dependence on applied magnetic field for doped FeTe and FeSe superconductors [14, 15].

GdBa$_2$Cu$_3$O$_Z$ (Gd123) is one of the interesting system of REBa$_2$Cu$_3$O$_Z$ (RE123; RE is the rare earth element/s and Y) HTSC family exhibiting onset superconducting
transition temperature \( (T_{\text{onset}}) \sim 92.7\,\text{K} \) \cite{16}. Extensive studies have been carried out on pure and doped Gd123 superconductor based compounds in different forms such as polycrystalline bulk, thin film, composite, etc for substitutional effects, Raman studies, dimensionality aspects, flux dynamics, etc \cite{16–27}. Mohammadizadeh et al \cite{21} have studied effect of Pr-substitution on flux dynamics in Gd(Ba\(_{2-x}\)Pr\(_x\))Cu\(_3\)O\(_7\) superconductors. Under doped and over doped R\(_{1-x}\)Ca\(_x\)Ba\(_2\)Cu\(_3\)O\(_7\) (R = Gd and Y) superconductors have been studied for intergranular flux pinning effect using magnetic susceptibility measurements and TAFF mechanism for evaluating activation energy in the compounds \cite{22}. Mohammadizadeh et al \cite{23} have investigated magnetic field effect on the resistive behavior in GdBa\(_2\)Cu\(_3\)O\(_7\\pm\delta\) superconductors and found thermally activated flux creep mechanism responsible for the resistive broadening.

Keeping in mind all the above aspects of superconductivity and results of the studies on Gd123 based superconductors, in this chapter result of magnetotransport studies have been presented on polycrystalline GdBa\(_2\)Cu\(_3\)O\(_7\) (Gd123) superconducting compound and its thin films having two different thicknesses, i.e. 100 and 200nm, grown on single crystalline (100) oriented SrTiO\(_3\) (STO) substrates using pulsed laser deposition (PLD) technique. Effect of applied magnetic field and film thickness on the resistive broadening and other superconducting parameters and their comparison with bulk Gd123 superconductor have been discussed in detail.

### 3.2 Experimental Details

Polycrystalline bulk GdBa\(_2\)Cu\(_3\)O\(_7\) (Gd123) superconductor was synthesized by conventional solid state reaction (SSR) route by stoichiometric mixing of respective metal oxides (Gd\(_2\)O\(_3\) and CuO) and carbonates (BaCO\(_3\)) subjected to pre-heating (at 400\(^\circ\)C for 03hrs) separately prior to calcination at 920\(^\circ\)C for 24hrs twice with intermediate grindings. Well calcined powder was palletized (10mm and 20mm in diameter) after proper grinding followed by sintering at 950\(^\circ\)C for 24hrs. Final product, i.e. well sintered pellet was then annealed at 500\(^\circ\)C for 24hrs under oxygen environment.
Well annealed pellet of Gd123 superconductor was characterized by XRD measurements at room temperature Cu Kα radiation to confirm its single phasic nature. Well annealed pellet (20mm diameter) was used as a target in pulsed laser deposition (PLD) technique to grow GdBa$_2$Cu$_3$O$_7$ (Gd123) films having two different thicknesses (i.e. 100 and 200nm) on single crystalline high quality (100) SrTiO$_3$ (STO) substrates. The parameters used during the deposition are given in the table below:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laser used</td>
<td>KrF Excimer</td>
</tr>
<tr>
<td>Targets</td>
<td>Gd$_1$Ba$_2$Cu$_3$O$_7$</td>
</tr>
<tr>
<td>Wavelength</td>
<td>248nm</td>
</tr>
<tr>
<td>Laser energy</td>
<td>~ 200mJ/cm$^2$</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>5Hz</td>
</tr>
<tr>
<td>Substrate temperature</td>
<td>~ 780°C</td>
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<tr>
<td>Oxygen partial pressure</td>
<td>~ 400mTorr</td>
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The phase purity and structure of the films were determined by performing the XRD measurements at room temperature using Cu Kα radiation. Transport properties were investigated by taking the R–T measurements (temperature range: 2 – 300K) under various applied magnetic fields (0, 1, 2, 4, 5, 6, 8, 10, 12 and 14T; field was applied perpendicular to the film ab plane) using d.c. four probe technique in PPMS (Quantum design).

### 3.3 Structural Properties

Figure 3.1 shows room temperature XRD pattern of polycrystalline bulk GdBa$_2$Cu$_3$O$_7$ (Gd123) polycrystalline bulk superconductor collected using Cu Kα radiation. It is confirmed that Gd123 sample is single phasic in nature without any detectable impurities within the measurement range studied. It can be seen that, sample possesses orthorhombic structure having lattice parameters a = 3.8325Å, b = 3.8954Å and c = 11.6558Å with unit cell volume V = 174.011Å$^3$. PLD grown Gd123 films were
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characterized by XRD using Cu Kα radiation to confirm the phases present and crystalline nature. Figure 3.2 shows room temperature XRD patterns of PLD grown (a) 200nm and (b) 100nm GdBa$_2$Cu$_3$O$_7$ (Gd123/STO) films grown on single crystalline (100) SrTiO$_3$ substrates. Figure 3.2 (c) shows the XRD pattern of pristine STO (100) substrate for reference.

**Figure 3.1**: XRD pattern of GdBa$_2$Cu$_3$O$_7$ polycrystalline bulk superconductor

**Figure 3.2**: XRD patterns of (a) Gd123/STO (200nm) film, (b) Gd123/STO (100nm) film and (c) pristine STO substrate. Insets: Enlarged views of (200) peaks. * indicates XRD peaks of Gd123 film confirming polycrystalline growth
Insets of figure 3.2 (a) and (b) show an enlarged view of (200) reflections of film and substrate revealing a small separation between the two peaks [which is absent in the inset of figure 3.2 (c)]. This separation indicates the presence of lattice mismatch ($\delta$) between the film and the substrate. The lattice mismatch can be calculated using the formula: $\delta$ (%) = $[(d_{\text{substrate}} - d_{\text{film}}) / d_{\text{substrate}}] \times 100$ and for presently studied Gd123/STO film, $\delta$ is found to be $\sim +0.25\%$ for 200nm Gd123 film while it increases to $\sim +1.36\%$ for 100nm film thickness. Positive sign of $\delta$ in both the films indicates the nature of strain is tensile. In addition, three peaks at $\sim 30.54^\circ$, 38.43$^\circ$ and 54.85$^\circ$ are observed in the XRD patterns of figure 3.2 (a) and (b) confirming the polycrystalline growth of 100nm and 200nm Gd123/STO films onto the single crystalline (100) STO substrate which may be due to the higher thicknesses of the films.

3.4 Transport Properties

To understand the transport properties of presently studied Gd123 bulk and thin films, temperature dependence of resistance has been measured in the temperature range: 2 – 300K under different applied magnetic fields between 0 and 14T. Figure 3.3 shows the variation in resistivity with temperature under 0T field for Gd123 bulk and thin films. It is clearly seen that, all the three samples exhibit superconductivity at low temperature. Bulk Gd123 shows $T_c^{\text{onset}} \sim 94.26\text{K}$ which decreases to 77.85K (100nm) and 89.88K (200nm) in Gd123 films. Decrease in $T_c^{\text{onset}}$ in thin films of Gd123 may be ascribed to the presence of lattice mismatch (interface strain) in the films which is completely absent in bulk. 200nm Gd123 film possesses lower strain values resulting in enhanced $T_c^{\text{onset}} \sim 89.88\text{K}$ in 200nm Gd123 film. Resistivity is higher in lower thickness Gd123 film which gets suppressed in 200nm Gd123 film (in the range of $10^{-6} \ \Omega\text{cm}$) which is mainly due to lower strain at the film – substrate interface in higher thickness film. Bulk Gd123 sample exhibit zero resistivity (in the range of $10^{-6} \ \Omega\text{cm}$) below temperature $T_0 \sim 83.82\text{K}$ which gets suppressed to 37.09K (100nm) and 68.84K (200nm) under 0T field. Mean field transition temperature ($T_c^{\text{mf}}$), maximum in $d\rho / dT$, is found to decrease from 92.81K (bulk) to 71.60K (100nm) and 78.85K (200nm) under zero applied magnetic field.
Figure 3.3: Variation in resistivity with temperature under 0T field for Gd123 bulk and thin films. Inset: Temperature dependence of resistivity of Gd123 bulk and thin films under 14T field

Inset of figure 3.3 shows temperature dependence of resistivity for all the three samples under study measured under 14T applied magnetic field. All the three samples exhibit superconductivity under 14T field with comparatively larger values of resistivity and reduced values of $T_0$ and $T_{C^{mf}}$ while $T_{C^{onset}}$ remains unchanged with applied field indicating the field induced resistive broadening behavior of all the samples under study. Under 14T, values of $T_0$ are ~ 62.20, 03.20 and 26.15K for bulk, 100nm and 200nm Gd123 samples, respectively, while values of $T_{C^{mf}}$ are ~ 88.58, 37.60 and 52.13K, respectively, for bulk, 100nm and 200nm Gd123 samples.

Figure 3.4 shows the variation in resistivity with temperature (range: 2 – 100K) measured under different applied magnetic fields for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film.
Figure 3.4: Variation in resistivity with temperature (range: 2 – 100K) for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film measured under different applied magnetic fields. Inset: Temperature dependent resistivity with temperature (range: 2 – 300K) measured under different applied magnetic fields.
It is seen that for all the three Gd123 samples, resistivity increases significantly with increase in applied magnetic field below their respective $T_{C_{\text{onset}}}$ while above $T_{C_{\text{onset}}}$, resistivity changes marginally with field. As discussed above, for all the three samples, $T_{C_{\text{onset}}}$ remains unchanged with applied magnetic field while $T_0$ and $T_{C_{\text{mf}}}$ continuously decrease with increase in applied magnetic field. Effect of applied magnetic field on the variation in $T_0$ and $T_{C_{\text{onset}}}$ leads to an existence of resistivity broadening in all the three samples. Figure 3.5 shows the temperature induced variation in derivative of resistivity with respect to temperature ($d\rho / dT$) (enlarged views) under different applied magnetic fields for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film.

As shown in figure 3.5, a sharp peak at (a) 92.81K for bulk Gd123, (b) 71.60K for 100nm Gd123 film and (c) 78.85K for 200nm Gd123 film is observed in $d\rho / dT$ vs. $T$ plots under zero applied field which has already been recognized as a mean field $T_C$ ($T_{C_{\text{mf}}}$). Values of $T_{C_{\text{mf}}}$ for all the three samples under different applied magnetic fields are shown in figure 3.5. Also, one can notice that, with increase in applied field, sharpness of the peak decreases suggesting the field induced resistive broadening effect in all the three samples. Maximum broadening in $d\rho / dT$ vs. $T$ plots can be seen in 100nm Gd123 film which becomes comparatively sharper in 200nm Gd123 film while maximum peak sharpness in $d\rho / dT$ vs. $T$ plots has been observed for bulk Gd123 sample. This can be attributed to the higher strain value in 100nm film which gets suppressed in higher thickness film while bulk Gd123 is strain free system. For all the three samples, above 100K (not seen here) no appreciable field effect on the normal state resistance is observed.

Observation of field induced suppression in $T_0$ and $T_{C_{\text{mf}}}$ and resistivity broadening in the presently studied samples can be ascribed to the two possible mechanisms: (i) penetration of magnetic field lines into the grain boundary regions and (ii) motion of vortices along with existing defects which causes energy dissipation within the material, wherein the defects are in the form of oxygen vacancies in the CuO chains affecting directly the carrier concentration and chain ordering and hence superconducting behavior of the film [28].
Figure 3.5: Temperature induced variation in derivative of resistivity with respect to temperature (d\(\rho\)/dT) (enlarged views) under different applied magnetic fields for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film.
Figure 3.6 shows the variation in (a) $T_0$ and (b) $T_{C_{mf}}$ with applied magnetic field for bulk Gd123 and 100nm and 200nm Gd123 films. As discussed above, both, $T_0$ and $T_{C_{mf}}$ decrease with increase in applied magnetic field. It can be noted that $T_0$ and $T_{C_{mf}}$ vary slightly with applied field in bulk Gd123 while magnetic field becomes much effective in modifying the values of $T_0$ and $T_{C_{mf}}$ in Gd123 films (both 100nm & 200nm).

![Graph showing variation of $T_0$ and $T_{C_{mf}}$ with applied magnetic field](image)

**Figure 3.6:** Variation in (a) $T_0$ and (b) $T_{C_{mf}}$ with applied magnetic field for bulk Gd123 and 100nm and 200nm Gd123 films
3.5 Magnetotransport Properties

It is very important to note that, in Gd123 films, effect of applied magnetic field is much stronger than bulk Gd123 which can be ascribed to the presence of strain at the interface which is non-stoichiometric in nature and is incorporated with large oxygen vacancies which, act as pinning centers, resulting in large flux pinning in thin films as compared to bulk sample. Also, more strained 100nm film may possess large number of oxygen vacancies resulting in strong pinning effect and hence effective resistivity broadening in 100nm film as compared to higher thickness 200nm film. This suggests a control of magnetic field over the values of $T_0$ and $T_c^{mf}$ in Gd123 films as compared to bulk counterpart.

To understand the effect of applied magnetic field on transition temperatures and hence to make clear an existence of field induced resistive broadening effect in all the three samples, following temperature differences [(a) $\Delta T_1$ and (b) $\Delta T_2$ & $\Delta T_3$] have been calculated and plotted in figures 3.7 (a) and (b).

It is can be seen that $\Delta T_1 = [T_c^{mf} - T_0]$ varies non-monotonically with applied magnetic field [figure 3.7 (a)]. Importantly, $\Delta T_1$ is found to be highest in 100nm Gd123 film which may be ascribed to the large strain at film – substrate interface. As shown in figure 3.7 (b), $\Delta T_3 = [T_{C_{onset}} - T_0]$ monotonically increases with increase in applied magnetic field while its values, throughout the field studied, are higher in 100nm Gd123 film while $\Delta T_3$ values are lower in bulk Gd123 sample as compared to 200nm Gd123 film which can be attributed to larger strain value in 100nm film and strain free state in bulk sample. In addition, enhancement in $\Delta T_3$ with applied field is mainly due to field induced resistive broadening effect. In fact, $\Delta T_1$ and $\Delta T_3$ parameters directly signify the resistive broadening effect in presently studied samples. To illustrate it more clearly, in figure 3.7 (b), $\Delta T_2 = T_{0(H=0)} - T_{0(H=H)}$ is plotted as a function of applied field $H$ which is found to increase with applied field confirming the field induced resistive broadening effect in all the samples.
Figure 3.7: Variation in (a) $\Delta T_1$ and (b) $\Delta T_2$ & $\Delta T_3$ with applied magnetic field for bulk Gd123 and 100nm and 200nm Gd123 films (equations are given) (c) Straight line fitted plots of $\Delta T_2$ vs. $H^{2/3}$ for bulk Gd123 and 100nm and 200nm Gd123 films. Inset: $\Delta T_3$ vs. $H$ plots fitted by equation: $\Delta T_3 = A \times H^n$ for all the three samples.
Figure 3.7 (c) shows the straight line fitted $\Delta T_2$ vs. $H^{2/3}$ plots indicating the linear dependence of $\Delta T_2$ on $H^{2/3}$ for all the three samples with error values of $\sim$ 04% (bulk), 10% (100nm film) and 10% (200nm film) indicating an increase in the value of slope from bulk Gd123 to 100nm to 200nm Gd123 films. Inset of figure 3.7 (c) shows the variation in $\Delta T_3$ with applied field, fitted using equation $\Delta T_3 = A \times H^n$, where $A$ is the proportionality constant and $n$ is the exponent. Equation is found to be fitted well with calculated $\Delta T_3$ data only above $H = 2T$ (as indicated by violet colored arrow) and the value of $n$ increases from bulk Gd123 to 100nm to 200nm Gd123 films, as shown in figure 3.7 (c).

Various models and mechanisms, as discussed in introduction part, have been reported to explain the magnetic field induced resistive broadening in HTSC [11, 29 – 32]. In the present case, following two aspects have been observed: (i) no change in $T_{C_{onset}}$ with applied magnetic field and (ii) $\Delta T_2$ scales as $\Delta T_2 \sim H^{2/3}$. These two features are the essential conditions for the TAFF mechanism, caused by the flow of vertices which are thermally activated. Hence, resistivity can be written as $\rho = \rho_0 (H, T) \exp (-U_0 / K_B T)$ [33], where $\rho_0$ is the residual resistivity for normal state of superconductor and $U_0$ is the pinning energy, which can be calculated from the slope of the linear part of $\ln (\rho / \rho_0)$ vs. $T^{-1}$ plots. Manifestation of superconducting transition broadening (resistive broadening) in the presence of applied magnetic field can be interpreted as energy dissipation (i.e. pinning energy, $U_0$) caused by vortex motion. Further discussion in this regards is limited to the temperature region: $T_0$ and $T_{C_{onset}} (\Delta T_3)$.

Figure 3.8 shows the variation in normalized resistivity ($\rho_n$) (of normal state) (in logarithmic scale) $\ln (\rho / \rho_n)$ with temperature inverse showing straight line fits slope of curve value with applied field for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film.
Figure 3.8: In (ρ / ρn) vs. T⁻¹ plots under various applied magnetic fields for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film. Black straight lines are fitted lines for each magnetic field in the respective ΔT₃ region.
Using the slope of the above curves, calculated value pinning energy ($U_0$) is plotted as a function of applied magnetic field in figure 3.9 for all the three samples. It can be clearly seen that, with increase in applied field, pinning energy decreases for all the three samples. Pinning energy is found to be larger for 200nm Gd123 film as compared to bulk Gd123 sample at lower applied fields ($\leq 4$T) (as indicated by violet colored arrow) while for higher field values ($> 4$T), pinning energy for bulk becomes higher than 200nm film. For lower thickness film, having higher film – substrate strain, possess lowest pinning energy value throughout the field range studied. The values of $U_0$ are in the range of μeV for 100nm film which can be attributed to a strong effect of magnetic field in modifying resistive behavior (resistive broadening) mainly due to disordered state of film – substrate interface having large number of oxygen vacancies and its non-stoichiometric nature.

![Figure 3.9: Variation in pinning energy ($U_0$) with applied magnetic field for Gd123 bulk and 100nm and 200nm Gd123 films. Insets: (a) straight line fitted plots of $U_0$ vs. $H^{-1}$ and (b) straight line fitted plots of $U_0$ vs. $H^{1/2}$](image)
To understand the effect of temperature on the pinning energy, two different possibilities can be considered: (i) $U_0$ is proportional to $H^{-1}$ (confirming independence of $U_0$ on temperature) and (ii) $U_0$ is proportional to $H^{-1/2}$ (confirming dependence of $U_0$ on temperature). Inset (a) of figure 3.9 shows the variation in $U_0$ with $H^{-1}$ fitted into straight line indicating that $U_0$ varies linearly with field inverse at higher field region while for lower field values, it does not follow $U_0 \propto H^{-1}$ relation. Inset (b) of figure 3.9 shows the plots of $U_0$ vs. $H^{-1/2}$ for all the three samples with straight line fitting throughout the field range studied suggesting that $U_0$ is strongly dependent on temperature (in the temperature window: $\Delta T_3$) which can be correlated with the thermal activation of flux lines in the presently studied bulk Gd123 and 100nm and 200nm Gd123 films.

In order to understand the effect of temperature on the pinning energy ($U_0$), variation in $U_0$ [$U_0 = -T \ln (\rho / \rho_n)$] with temperature under different applied magnetic fields in $\Delta T_3$ temperature range window has been plotted in figure 3.10 for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film. Straight line has been fitted to a small temperature region well below their $T_0$ such as 2 – 6K for bulk Gd123, 2 – 4.5K for 100nm Gd123 film and 20 – 70K for 200nm Gd123 film. Slope of these straight lines can be understood in the context of barrier height ($\gamma$): $\gamma = U_0 / K_B T$ [34]. It is found that the slope decreases with increase in applied field suggesting the field induced reduction in barrier height for all the three samples. Figure 3.11 shows the variation in barrier height ($\gamma$) with applied magnetic field for all the three samples.
Figure 3.10: Temperature dependent pinning energy under various applied magnetic fields with the straight line fits for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film.
Barrier height ($\gamma$) signifies the mean height of energy barrier that has to be overcome by a magnetic flux vortex during its motion which results into the rise in resistance (if, it overcomes). It is clearly seen that, values of barrier height for all the applied fields are higher for 200nm Gd123 film which can be attributed to the lower strain in higher thickness film (200nm) while values of $\gamma$ are smaller for 100nm Gd123 film throughout the field range studied mainly due to its higher strain at the film – substrate interface. The values of $\gamma$ can also be correlated with the values of transition temperatures for both the films understudy, since higher the values of $\gamma$, larger the values of transition temperatures and stronger the superconductivity.

Figure 3.11: Field induced variation in barrier height ($\gamma$) with applied magnetic field for Gd123 bulk and 100nm and 200nm Gd123 films and their non-linear fittings using power law equation: $\gamma = b \times H^m$
In addition, figure 3.11 also shows the non-linear fittings to $\gamma$ vs. $H$ plots of all the three samples fitted using power law equation: $\gamma = b \times H^{-m}$ for $1 - 14T$ field range. Fitted parameters (values of $b$ and $m$) are given as the inset of figure 3.11. Values of $b$ and $m$ are higher for 200nm Gd123 film while lower for 100nm Gd123 film. By comparing the results of non-linear fittings shown in figure 3.7 and figure 3.11, the reduction rate of $\gamma$ with field (parameter: $m$; figure 3.11) is higher than the rate of increase in $\Delta T_3$ (parameter: $n$; figure 3.7), for all the three samples, suggesting faster decrease in barrier height as compared to the emergence of resistive broadening ($\Delta T_3$) with applied magnetic field.

In order to estimate the upper critical field ($H_{C2}$) of (a) Gd123 bulk, (b) 100nm Gd123 film and (c) 200nm Gd123 film, $H - T$ diagram from 10%, 50% and 90% criteria of normal state resistivity ($\rho_{\text{norm}}$) for Gd123 film is shown in figure 3.12. Values of the slopes $[dH / dT]$; obtained from the straight line fitted (green lines) $H - T$ diagram of figure 3.12 (a), (b) and (c)] for 10, 50 and 90% $\rho_{\text{norm}}$ are $-0.812$, $-2.094$ and $-7.365$ for bulk Gd123, $-0.342$, $-0.455$ and $-1.645$ for 100nm Gd123 film and $-0.443$, $-0.816$ and $-6.820$ for 200nm Gd123 film, respectively. $H_{C2}$ can be calculated using Werthamer – Helfand – Hohenberg (WHH) theory using the formula: $H_{C2} = 0.693T_c \left(\frac{dH}{dT}\right)_{T_c}$ [15], where mean field $T_c$ ($T_{C_{\text{mf}}}$) is used for these calculations. Maximum calculated $H_{C2}$ (under zero applied field) for presently studied bulk Gd123 is $\sim 474$ T which decreases up to $\sim 82$ T in 100nm film while for 200nm film, it becomes $\sim 373$ T. Suppression in $H_{C2}$ can be attributed to the presence of strain in the films at the interface while higher strained 100nm film has lower $H_{C2}$ as compared to 200nm film. The value of $H_{C2}$ obtained from the linear extrapolation of $H - T$ plots (figure 3.12 (a), (b) and (c); for 90% $\rho_{\text{norm}}$ results) to $T = 0K$ is $\sim 690$ T (for bulk Gd123), $\sim 122$ T (for 100nm Gd123 film) and $\sim 583$ T (for 200nm Gd123 film). Difference between the values of $H_{C2}$ obtained from WHH theory and linear extrapolation is mainly due to the fact that in WHH theory, $T_{C_{\text{mf}}}$ is considered while in linear extrapolation, $T_{C_{\text{onset}}}$ is considered.
Figure 3.12: H – T diagram for calculating upper critical field (\(H_{c2}\)) from 10%, 50% and 90% criteria of normal state resistivity (\(\rho_{\text{norm}}\)) and straight line fits (green lines) for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film. Field dependent variation in \(H_{c2}\) in the context of 10%, 50% and 90% criteria of normal state resistivity (\(\rho_{\text{norm}}\)) for (d) bulk Gd123, (e) 100nm Gd123 film and (f) 200nm Gd123 film
It is possible to calculate the coherence length ($\xi$), for presently studied bulk Gd123 and 100 and 200nm Gd123 thin films under different applied fields, using the formula [given by Ginzburg – Landau (GL) theory]: $U_0 c Hc^2 \xi^3 / 8\pi$, where $\xi^3 = \xi_{ab}^2 \times \xi_c$ ($\xi_{ab}$ and $\xi_c$ are the coherence lengths across ab plane and c direction). Variation in (a) $\xi_{ab}$ and (b) $\xi_c$ with applied magnetic field is shown in figures 3.13, 3.14 and 3.15 for bulk Gd123, 100nm and 200nm Gd123 films, respectively, under the consideration of 10%, 50% and 90% $\rho_{\text{norm}}$ criteria. For Bulk Gd123, under 10% $\rho_{\text{norm}}$ criterion (0T), calculated $\xi^3 = \xi_{ab}^2 \times \xi_c = 480\text{Å}^3$. In order to calculate $\xi_{ab}^2$, one can use the formula: $\xi_{ab}^2 = \varphi_0 / 2\pi Hc^2$ and calculated value of $\xi_{ab}^2 \sim 631\text{Å}^2$ indicates that in plane (across ab plane) coherence length, $\xi_{ab}$, is $\sim 25.12\text{Å}$ [figure 3.13 (a)] while out of plane (along c direction) the coherence length ($\xi_c$) becomes $\sim 0.76\text{Å}$ [figure 3.13 (b)]. For 10% $\rho_{\text{norm}}$ (0T), values of calculated $\xi_{ab}$ and $\xi_c$ are $\sim 44.06\text{Å}$ and $\sim 5.28 \times 10^{-4}\text{Å}$ for 100nm film which become $\sim 36.87\text{Å}$ and $2.74\text{Å}$ for 200nm film, respectively. It is important to note that $\xi_c$ is lower than inter planar spacing along c-axis ($\sim 3.9\text{Å}$) indicates the 2D (or 1D) nature of superconductivity for all the three samples.

Figure 3.13: Variation in (a) $\xi_{ab}$, (b) $\xi_c$, (c) $\lambda_{ab}$ and (d) $\lambda_c$ with applied magnetic field under 10%, 50% and 90% criteria of normal state resistivity ($\rho_{\text{norm}}$) for bulk Gd123
Figure 3.14: Variation in (a) $\xi_{ab}$, (b) $\xi_c$, (c) $\lambda_{ab}$ and (d) $\lambda_c$ with applied magnetic field under 10%, 50% and 90% criteria of normal state resistivity ($\rho_{\text{norm}}$) for 100nm Gd123 film.

Figure 3.15: Variation in (a) $\xi_{ab}$, (b) $\xi_c$, (c) $\lambda_{ab}$ and (d) $\lambda_c$ with applied magnetic field under 10%, 50% and 90% criteria of normal state resistivity ($\rho_{\text{norm}}$) for 200nm Gd123 film.

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In addition, the anisotropy factor, $\gamma' = \xi_{ab} / \xi_c$, is found to be ~33 under 10% criterion of normal state resistivity ($\rho_{\text{norm}}$) for bulk Gd123 which increases to ~83447 in 100nm Gd123 film while decreases to ~13 in 200nm Gd123 film indicate that 200nm film has lower anisotropy mainly due to its three dimensional disorder free state and lower strain at the film – substrate interface while for 100nm film, anisotropy factor is found to be very large which can be attributed to the larger strain at the interface. With increase in applied magnetic field, $\xi_c$ decreases for all the three samples which can be correlated with the field induced penetration effect and hence suppression in superconductivity. One can also estimate the penetration depth for all the Gd123 bulk and thin film samples using the formula: $U_0 \sim \phi_0^2 \xi / 48\pi^2 \lambda^2$; where coherence length, $\xi$, will be treated separately for ab plane ($\xi_{ab}$) and c-axis ($\xi_c$) in order to calculate penetration depth, $\lambda$, across ab plane ($\lambda_{ab}$) and c-axis ($\lambda_c$), respectively. Variation in (c) $\lambda_{ab}$ and (d) $\lambda_c$ with applied field for 10%, 50% and 90% $\rho_{\text{norm}}$ criteria is shown in figure 3.13 (bulk Gd123), figure 3.14 (100nm Gd123 film) and figure 3.15 (200nm Gd123 film). For 10% $\rho_{\text{norm}}$ ($H = 0$T) in Gd123 bulk sample, $\lambda$ ($H = 0$T, $T = 0$K) is found to be ~374nm ($\lambda_{ab}$) and ~65nm ($\lambda_c$) which increases ~33$\mu$m ($\lambda_{ab}$) and ~114nm ($\lambda_c$). For 200nm Gd123 film, these values get suppressed up to ~350nm ($\lambda_{ab}$) and ~95nm ($\lambda_c$). With applied field, $\lambda_{ab}$ and $\lambda_c$ increase which can be ascribed to the field induced penetration effect resulting in the suppression in superconductivity through the breaking of cooper pairs in all the three samples. In addition, penetration depth across ab plane ($\lambda_{ab}$) is larger than that along c-axis ($\lambda_c$) mainly due to the geometrical measurements, i.e. magnetic field is applied in the direction perpendicular to the film ab plane. Inset of figure 3.13, figure 3.14 and figure 3.15 show the variation in distance between the vertices [calculated using the formula: $a_0 = 1.07 (\phi_0 / H)^{1/2}$, $\phi_0$ is the magnetic flux quantum and H is applied field] with applied field and the distance is found to decrease with increasing field line density. Values of $a_0$ is similar for all the samples understudy, since it is not dependent upon any variable parameters but it only depends upon applied field values.

In order to understand the transport mechanism and effect of magnetic field on the conduction in normal state of Gd123 superconducting bulk and films, all $\rho - T$ plots were fitted using the power law equation: $\rho (T) = \rho_0 + bT^2$ for all the three samples understudy.
It was found that transport mechanism in the normal state of Gd123 bulk and films does not follow this power law. Later on, \( \rho (T) = \rho_0 + \rho_1 T^n + \rho_2 \exp(-T_0/T) \) law has been successfully fitted to normal state of all the \( \rho - T \) plots under various applied magnetic fields for bulk Gd123 (fitting temperature range: 100 – 300K), 100nm Gd123 film (fitting temperature range: 80 – 150K) and 200nm Gd123 film (fitting temperature range: 100 – 200K), as shown in figure 3.16. In this law, first term is residual resistivity (resistivity at 80K in the case of 100nm film while resistivity at 100K for the case of bulk and 200nm film) while second term is related to inter-band charge carrier scattering. Third, exponential term, in the formula is related to the phonon assisted inter-band scattering or inter-band umklapp scattering mainly due to the consequential effect of two or more interacting waves of lattice, charge carriers, etc. In the phonon assisted inter-band scattering, characteristic energy (temperature), \( T_0 \), corresponds to the energy of a phonon (with minimum wave vector) required to scatter an electron between bands [36] while in umklapp scattering it corresponds to the energy of a phonon (with wave vector) matching a particular dimension of a single sheet of the fermi surface [37]. In practice, \( T_0 \) does not depend on the temperature but depends on the applied magnetic field which signifies the energy of phonons near the boundary zones.
Figure 3.16: Typical normal state resistivity fits to $\rho(T) = \rho_0 + \rho_1 T^n + \rho_2 \exp(-T_0/T)$
(temperature range: 100 – 300K for bulk Gd123, 80 – 150K for 100nm Gd123 film and 100 – 200K for 200nm Gd123 film)
Figure 3.17: Variation in parameters (a) $n'$ and (b) $T_0$ with applied magnetic field for bulk Gd123, 100nm Gd123 film and 200nm Gd123 film
Variation in parameters (a) $n'$ and (b) $T_0$ with applied magnetic field for bulk Gd123, 100nm Gd123 film and 200nm Gd123 film is shown in figure 3.17. It can be seen from figure 3.17 (a) that, for all the three samples, power law exponent ($n'$) increases with increasing applied magnetic field indicting the field induced enhancement in the inter-band charge carrier scattering in Gd123 superconductor. Parameter $n'$ increases from bulk Gd123 to 100nm and 200nm Gd123 films, throughout the field range studied. Also, for all the three samples, $n'$ vs. $H$ plots have been fitted non-linearly using equation: $n' = c \times H^l$ and it is found that $c$ increases while $l$ decreases from bulk Gd123 to 100nm Gd123 film to 200nm Gd123 film. It indicates that the rate of enhancement in inter-band charge carrier scattering exponent ($n'$), i.e. parameter $l$, decreases from bulk Gd123 to 100nm to 200nm Gd123 films.

Magnetic field induced enhancement in inter-band charge carrier scattering and characteristic phonon energy results into the degradation in transport in bulk Gd123, 100nm Gd123 film and 200nm Gd123 film. Also, magnetic field induced resistive broadening effect observed for presently studied three samples prompts one to study magnetoresistance (MR) behavior of the samples. It is interesting to calculate the temperature dependent variation in MR using the formula: $\text{MR} (%) = \left[ (\rho_H - \rho_0) / \rho_0 \right] \times 100$, where $\rho_H$ and $\rho_0$ are the resistivity with and without applied magnetic field, $H$, respectively. Figure 3.18 shows the variation in MR with temperature under different applied fields for (a) bulk Gd123 (range: 2 – 300K), (b) 100nm Gd123 film (range: 2 – 150K) and (c) 200nm Gd123 film (range: 2 – 200K). Insets in figure 3.18 show an enlarged view of MR vs. $T$ plots for all the three samples understudy.
Figure 3.18: Variation in MR with temperature under different applied fields for (a) bulk Gd123 (range: 2 – 300K), (b) 100nm Gd123 film (range: 2 – 150K) and (c) 200nm Gd123 film (range: 2 – 200K). Insets: Enlarged view of MR vs. T plots for all the three samples.
It is interesting to note that, all the samples exhibit positive MR ($\rho_H > \rho_0$) throughout the temperature and field range studied indicating better field sensitivity of the bulk and thin films of Gd123 superconductor. Importantly, for all the three samples, positive MR increases with increase in applied magnetic field. It can be seen that, for bulk Gd123, MR remains almost constant up to ~ 62K after which it increases with temperature up to ~ 82K and then starts to decrease with temperature [figure 3.18 (a)]. The peak in MR vs. T plots observed for bulk Gd123 remains unchanged with applied magnetic field. From figure 3.18 (b), it can be seen that MR decreases continuously with temperature up to ~ 72K above which it remains almost invariant with temperature for 100nm Gd123 film. For 200nm Gd123 film, similar to bulk Gd123, peak in MR vs. T plots at ~ 61K is seen which shifts towards higher temperature with decrease in applied magnetic field. It can be clearly seen that, bulk Gd123 shows maximum MR (%) ~ $1.7 \times 10^5$ % at ~ 82K under 14T field which gets suppressed in 100nm Gd123 film. Value of maximum MR in 100nm Gd123 film is found to be ~ $1.4 \times 10^4$ % at ~ 2K under 14T field while for 200nm Gd123 film, maximum MR is found to be ~ $2.5 \times 10^8$ % at 61K under 14T field. Presently observed values of MR for Gd123 bulk and thin films are much higher than that observed for manganite based thin films [38, 39]. Further insights into the MR studies on present samples require more detailed investigations.

3.6 Temperature Coefficient of Resistivity (TCR)

To explore the potential applications of presently studied Gd123 bulk and thin films, temperature coefficient of resistance (TCR) (which can be useful for the development of temperature sensor based devices) was calculated at mean field transition temperature ($T_{C_{mf}}$), as a function of temperature under different applied magnetic fields for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film, is shown in figure 3.19. TCR has been calculated using the formula: $\text{TCR} (\%K^{-1}) = \left[\frac{1}{\rho (T_{C_{mf}})}\right] \times \left[\frac{d\rho}{dT}\right] \times 100$. Inset of figure 3.19 (a) shows an enlarged view of TCR vs. T plots under different applied fields for the better clarity purpose.
Figure 3.19: Variation in temperature coefficient of resistance (TCR) with temperature under different applied magnetic fields for (a) bulk Gd123, (b) 100nm Gd123 film and (c) 200nm Gd123 film. Inset of (a) shows an enlarged view of TCR vs. T plots under different applied fields for bulk Gd123 for the better clarity purpose.
For bulk Gd123, a sharp peak in TCR vs. T plots can be seen near its corresponding mean field transition temperature ($T_{C_{mf}}$) [figure 3.5 (a)] which shifts towards lower temperature with applied magnetic field and peak becomes broader under higher applied fields. For 100nm Gd123 film, broad peak in TCR, near its corresponding $T_{C_{mf}}$ [figure 3.5 (b)], shifts towards lower temperature with increase in applied magnetic field. 100nm Gd123 film exhibits double peak like behavior in TCR vs. T plots under all the applied magnetic fields studied. This may be ascribed to large structural strain at the film – substrate interface as well as strong resistive broadening behavior observed for lower thickness film [figure 3.4 (b)]. In 200nm Gd123 film, peak in TCR vs. T (H) plots, in vicinity of its corresponding $T_{C_{mf}}$, is sharper than that of lower thickness Gd123 film and the sharpness of the peak gets reduced with increase in applied magnetic field and peak becomes broader at higher applied fields [figure 3.19 (c)]. The peak in TCR vs. T plots shifts towards lower temperature. All the three samples understudy exhibit positive TCR which gets suppressed upon increase in applied magnetic field. Value of TCR decreases from bulk Gd123 (maximum ~ 66.50%K$^{-1}$; @ ~ 93K & 0T) to 100nm Gd123 film (maximum ~ 07.23%K$^{-1}$; @ ~ 71K & 0T) followed by enhancement in the values of TCR upon increase the film thickness ~ 200nm (maximum ~ 31.37%K$^{-1}$; @ ~ 79K & 0T). It is worth to note that, for all the three samples, temperature sensitivity (TCR) is almost zero below their corresponding $T_0$ and it is very small in value above their $T_{C_{onset}}$ suggesting remarkable temperature sensitivity of presently studied Gd123 superconductors (bulk and thin films) is only possible within the temperature window: $T_0 < T < T_{C_{onset}}$. On one hand, in the present study, the understanding of various responsible charge transport mechanisms, superconducting parameters, magnetic field effects and film thickness effects on the superconductivity and its transport properties have been discussed mainly in resistive broadening region ($T_0 < T < T_{C_{onset}}$) and normal state resistivity, on the other hand, very interesting observation of large temperature sensitivity and its dependence on applied magnetic field and film thickness have been investigated for the same temperature window, which makes this study quantitative as well as qualitative from fundamental as well as potential application point of view.
Conclusions

Transport and magnetotransport properties of solid state reaction (SSR) grown high temperature superconducting (HTSC) Gd₁Ba₂Cu₃O₇ (Gd123) bulk and pulsed laser deposited (PLD) grown Gd123 films, with two different thicknesses (100nm and 200nm), have been studied in detail.

Investigations on the transport properties of all the three samples using temperature dependent resistivity under different applied fields show the field induced reduction in transition temperatures, $T_0$ and $T_{C_{mf}}$, while $T_{C_{onset}}$ remain invariant with applied field resulting in an existence of resistive broadening effect in Gd123 bulk and thin films. For all the samples, it is observed that, magnetotransport in the resistivity broadening region follows the thermally activated flux flow (TAFF) mechanism and pinning energy is found to reduce with increase in field. Pinning energy ($U_0$) is found to be higher in 100nm film while lower in 200nm film as compared to value with that for bulk Gd123. Temperature dependence behavior of $U_0$ suggests that, barrier height ($\gamma$) decreases exponentially with applied magnetic field for all the three samples. Werthamer – Helfand – Hohenberg (WHH) theory suggests that, bulk Gd123 possesses large $H_{C2}$ ~ 474T while 200nm (100nm) film possesses $H_{C2}$ ~ 373T (~ 82T). All the three samples follow 2D (or 1D) nature of superconductivity instead of most conventional 3D behavior of superconductivity.

Normal state transport, in three samples, has been understood using the $\rho (T) = \rho_0 + \rho_1 T^n + \rho_2 \exp (- T_0/T)$ power law expression suggesting the important role of phonon assisted inter-band scattering and / or umklapp scattering. On application of magnetic field, the phonon and charge carrier (electron) waves get modified which changes the normal state resistivity to the higher values. Observation of large MR and TCR near the mean field transition temperature ($T_{C_{mf}}$) and their dependence upon bulk and film thickness for Gd123 superconductor points towards of potential applications of Gd123 and other oxide based HTSC thin films and devices.
References


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