Influence of gold diffusion-doped on phase formation, superconducting and microstructure properties of Bi$_{1.8}$Pb$_{0.35}$Sr$_{1.9}$Ca$_{2.1}$Cu$_3$O$_y$ superconductors

O Ozturk$^1$, M Akdogan$^2$, C Terzioglu$^2$, and A Gencer$^3$

$^1$ Department of Physics, Faculty of Arts and Sciences, Kastamonu University, 37600 Kastamonu-Turkey.
$^2$ Department of Physics, Faculty of Arts and Sciences, Abant Izzet Baysal University, 14280 Bolu-Turkey.
$^3$ Department of Physics, Faculty of Arts and Sciences, Ankara University, 06100 Ankara-Turkey.

E-mail: terzioglu_c@ibu.edu.tr

Abstract. We report on low-field magnetic properties of gold diffusion-doped Bi$_{1.8}$Pb$_{0.35}$Sr$_{1.9}$Ca$_{2.1}$Cu$_3$O$_y$ superconducting bulk samples by performing ac susceptibility measurements. The undoped samples were prepared by the standard solid-state reaction method. Doping of Bi$_{1.8}$Pb$_{0.35}$Sr$_{1.9}$Ca$_{2.1}$Cu$_3$O$_y$ was carried out by means of Au-diffusion during sintering from an evaporated gold film on pellets. To investigate the effect of gold-diffusion and diffusion-annealing duration on transport, magnetic and microstructure properties of the superconducting samples we performed magnetoresistivity, scanning electron microscopy (SEM) and X-ray diffraction (XRD) measurements. The ac susceptibility as a function of temperature measurements were carried out at different values of the ac magnetic field amplitudes ($H_{ac}$) in the range between 20 A/m and 320 A/m for 211 Hz. The imaginary part of ac susceptibility measurements is used to calculate intergranular critical current density $J_c(T_p)$ using the Bean Model. $J_c(T_p)$ is seen to increase from 60 A cm$^{-2}$ to 90 A cm$^{-2}$ with increasing diffusion-annealing time from 10 h to 50 h. The peak temperature, $T_p$, in the imaginary part of the ac susceptibility is shifted to a lower temperature with decreasing diffusion-annealing duration as well as increasing ac magnetic fields. The force pinning density ($\mathcal{J}(0)$) increased with increasing diffusion annealing time. The value of $T_c$ in gold-diffused samples, in comparison with the undoped samples, increased from 100 ± 0.2 K to 104 ± 0.2 K. It was observed that the value of $T_c$ of the gold-doped samples enhanced with further increasing diffusion-annealing duration. XRD patterns and SEM micrographs are used to obtain information about Bi-2223 phase ratio, lattice parameters and grain size of the samples. Gold doping enhanced the formation high-$T_c$ phase and increased the grain size. The possible reasons for the observed improvements in transport, microstructure and magnetic properties due to Au diffusion and diffusion-annealing time were discussed.

* Cabir Terzioglu, Department of Physics, Faculty of Arts and Sciences, Abant Izzet Baysal University, 14280 Bolu-Turkey.
1. Introduction

The discovery of high temperature superconducting materials has generated great interest of the scientific community for many reasons [1]. Since high-$T_c$ cuprate superconductors are chemically unstable and highly reactive, most materials including the noble metals and oxides react with these superconductors when they come into contact. Bi-based superconductors have attracted the attention of many researchers for its high transition temperature, critical current density and easy synthesis [2-8]. Bi–Sr–Ca–Cu–O system is an important member of the high-$T_c$ superconductor families with an orthorhombic structure and it has been found at least in three phases with atomic ratios in the order of Bi, Sr, Ca, Cu, namely (2201), (2212) and (2223) with zero resistivity transition temperatures of 20 K, 85 K and 110 K, respectively [9]. In these series, Bi-2223 is the most attractive because of having the highest superconducting transition temperature.

As part of a study of doping, the effects of additions of oxides such as SiO$_2$, Al$_2$O$_3$, MgO, ZrO$_2$ and 3d transition metal oxides on YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) [10-11] and (Bi,Pb)$_{2}$Sr$_{2}$Ca$_{2}$Cu$_{3}$O$_{y}$ (BPSCCO) [12] have been investigated. Among the metals, Au and Ag have been found to be non-poisoning to YBa$_2$Cu$_3$O$_{7-x}$ [13], while Ag has been found to be the only metal element that is non-poisoning to the BSCO system [14]. It has been reported that Au doping reduces the superconductivity of Bi(Pb)SrCaCuO [14-15], which is unexpected since Au and Ag are chemically very similar. Dzhafarov et al. [15] investigated the effect of Au diffusion combined with slow and fast cooling on $T_c$ and $J_c$ in Bi-2223 system and found that Au diffusion promotes formation of the high-$T_c$ (Bi-2223) phase and increases significantly $J_c$. In the resent work [16], the role of diffusion-annealing time on the mechanical properties of gold diffusion-doped Bi-2223 superconductors was investigated. That work indicated that the Au-doping and diffusion-annealing duration increased the Vickers microhardness, Young’s modulus, yield strength and fracture toughness of the samples. It was found that the microhardness values decreased with increasing applied load. We also investigated the indentation size effect (ISE) by using different models such as Meyer’s law, Hay-Kendall approach and modified proportional specimen resistance model. The Hay-Kendall approach was sufficient for describing the observed ISE [17]. Besides, the gold diffusion in Bi$_{1.8}$Pb$_{0.35}$Sr$_{1.9}$Ca$_{2.1}$Cu$_{3}$O$_{7-x}$ has been studied over the temperature range of 500-830 °C using the technique of successive removal of thin layers and calculation of the lattice parameter c from XRD patterns [18].

In the present work, we reported the results of ac susceptibility, magnetoresistivity, SEM and XRD measurements in gold diffusion-doped Bi-2223 system. The possible reasons for the observed improvements in transport, magnetic and microstructure properties due to Au diffusion and diffusion-annealing duration were discussed.

2. Experimental details

Superconducting Bi$_{1.8}$Pb$_{0.35}$Sr$_{1.9}$Ca$_{2.1}$Cu$_{3}$O$_y$ samples were prepared by the standard solid-state reaction method. The weighed powders of Bi$_2$O$_3$ (99.99%), PbO (99.9+%), SrCO$_3$ (99.9+%), CaCO$_3$ (99+%) and CuO (99+%) in the cation ratio of Bi: Pb: Sr: Ca: Cu = 1.8:0.35:1.9:2.1:3 were mixed by grinding machine for 24 h. After milling, the mixed powders were calcined in air at 700, 750 and 800 °C for 24 h. At every calcined temperature, the sample cooled to room temperature to grind. The sample and then cooled down to room temperature. The heating and cooling rates of the temperature were chosen to be 10 and 3 °C min$^{-1}$, respectively. The Au evaporation (thickness of about 50µm) on one face of the samples was carried out using an AUTO 306 Vacuum Coater (EDWARDS). Doping of Bi$_{1.8}$Pb$_{0.35}$Sr$_{1.9}$Ca$_{2.1}$Cu$_{3}$O$_y$ was carried out by means of Au-diffusion during sintering from an evaporated gold film on pellets. The Au layered superconducting samples were annealed at 830 °C for 10 h (G1), 20 h (G2) and 50 h (G3) (Fig. 1). At the end of this run, Au diffusion was carried out by means of Au-diffusion during sintering from an evaporated gold film on pellets. The Au layered superconducting samples were annealed under the same conditions.
Ac susceptibility measurements on the samples were performed using a 7130 AC susceptometer of Lake Shore with a closed cycle refrigerator at 211 Hz frequency [19-20]. The ac susceptibility as a function of temperature (40–120 K) at fixed ac magnetic field amplitude ($H_{ac}$) in a range from 20 to 320 A/m was performed on the samples. The ac magnetic field was applied along the longest dimension of the samples. The measurements of ac susceptibility are commonly used to determine magnetic and superconducting properties of materials. In particular, the ac susceptibility measurement is useful in distinguishing between inter- and intra-grain properties. At high ac fields, two loss peaks can usually be seen in the imaginary part of susceptibility data; a broad peak at low temperature (coupling losses) due to the motion of inter-granular (Josephson) vortices [21] and a narrower peak (intrinsic losses) due to the motion of intra-granular Abrikosov vortices near $T_c^{onset}$ [22]. We observed only one peak (inter-granular) in each of the $\chi''$ vs. $T$ plots at low fields, but at high fields (160 and 320 A/m) one more peak (intragranular) was observed. From the $\chi'$ and $\chi''$ vs. $T$ plots, the values of the $T_c^{onset}$ (diamagnetic onset temperature) and $T_p$ (temperature corresponding to $\chi''$ peak) were obtained, respectively. Bean critical-state model [23] is used for determination of inter- and intra-grain critical current densities from ac susceptibility measurements.

The measurement of dc resistivity was performed with the four-probe method on all samples. Both voltage and current contacts were made with silver paint. Magnetoresistivity measurements were made under different dc magnetic field (0, 0.1, 0.2, 0.3 and 0.5 T) in the temperature rage between 60 K and 130 K. The magnetic field was applied parallel to the current direction by an electromagnet. The magnetic field was applied parallel to the current direction. The external dc magnetic field was provided by an electromagnet. The transition temperature $T_c$ was determined as the temperature at which zero resistivity was achieved.
XRD data were taken using a Rigaku D/Max-IIIC diffractometer with CuKα radiation in the range 2Θ = 4°- 60° with a scan speed of 3°/min and a step increment of 0.02° at room temperature. XRD data was taken from powder of the samples. Phase purity and the lattice parameters were obtained from these XRD patterns. The accuracy in determining the lattice parameter c was ± 0.001 Å. The surface morphologies of the Au diffusion-doped and pure samples were studied by using a Philips XL30 SFEG scanning electron microscope.

3. Results and discussion

The samples produced are named as G0 (undoped sample annealed at 830 °C for 10 h), G1 (gold diffused sample annealed at 830 °C for 10 h), G2 (gold diffused sample annealed at 830 °C for 20 h), and G3 (gold diffused sample annealed at 830 °C for 50 h).

3.1. Ac susceptibility

We have investigated the effect of Au diffusion and diffusion-annealing duration on some physical properties of Bi$_{1.8}$Pb$_{0.35}$Sr$_{1.9}$Ca$_{2.1}$Cu$_3$O$_y$ high-T$_c$ superconductors using the low field ac magnetic susceptibility method. The temperature and magnetic field dependence of the complex ac susceptibility, $\chi(T) = \chi'(T) + i\chi''(T)$, of the samples are shown in Fig. 2. $\chi'$ and $\chi''$ measure the amount of shielding and the amount of energy loss due to the induced current, respectively. As seen from the real parts of the plots, the onset of critical temperature, $T_c^{\text{onset}}$, appeared at 110 K at different magnetic field values for the all samples. They show field independent behaviors until 102 K. This temperature is the increasing point of the imaginary part. The susceptibility-temperature curves shifted to lower temperature, considerably increased the transition width and also decreased the shielding fraction of the superconducting phase in the samples with increasing the ac field amplitudes and decreasing the diffusion-annealing duration as depicted in Figs. 2 and 3. This similar effect of the diffusion-annealing time and the applied external dc field in transition width is revealed for the dc resistivity measurements in the current study.
Figure 2. The temperature dependence of ac susceptibility of the (a) $G_0$, (b) $G_1$, (c) $G_2$ and (d) $G_3$ samples for 20, 40 and 80 A/m.

Figure 3. The temperature dependence of ac susceptibility of the $G_0$, $G_1$, $G_2$ and $G_3$ samples for 80 A/m.
The maximum peak of $\chi''$ appears at a temperature $T_p$ where the intergranular field penetrates the centre of sample. As seen in Figure 2, $T_p$ values of the samples shifted to lower temperature and the intensity of the imaginary parts of susceptibility slightly increased with increasing the ac field amplitude. The imaginary parts of the $G0$ sample show peaks at 90, 88, 79, 69 and 64 K for the fields 20, 40, 80, 160 and 320 A/m, respectively. The other samples ($G1$, $G2$ and $G3$) showed the same behaviour and corresponding data are tabulated in Table 1. The shift is related to the pinning force strength. It was found that the overall susceptibility curves of the samples shifted to higher temperatures and increased the shielding fraction of superconducting phase for all ac field amplitudes with increasing the diffusion-annealing duration as shown in Figure 3 for 80 A/m. A similar behaviour is seen for the other field values. At large ac field amplitudes two loss peaks are usually found; a broad peak at low temperature and a narrower peak near $T_c^{\text{onset}}$. We did not see any intrinsic loss peaks in our samples for 20, 40 and 80 A/m because of the low value of the applied field. This may be due to small grain size and no flux penetration into the grains [24]. For higher field values namely for 160 and 320 A/m the intragranular (intrinsic loss) peak is clearly observed (Fig.4).

Table 1. Peak temperature $T_p$, and applied ac magnetic field amplitude $H_{ac}$ of the samples.

| Samples | $T_p$(K) | $H_{ac}$ (A/m) |
|---------|----------|----------------|
| $G0$    | 90       | 20 88 79 69 64 |
| $G1$    | 92       | 40 89 83 74 66 |
| $G2$    | 95       | 80 91 85 76 67 |
| $G3$    | 98       | 160 94 89 79 70 |

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Figure 4. The temperature dependence of ac susceptibility of the (a) G0, (b) G1, (c) G2 and (d) G3 samples for 160, and 320 A/m.

Figure 5. Inter-granular peak temperature $T_p$ versus ac field amplitude for the G0, G1, G2 and G3.

In order to study the effect of Au diffusion and diffusion-annealing time on the intergranular pinning force, the peak temperature dependence as a function of ac magnetic field, $H_{ac}$, has been investigated. Fig. 5 exhibits graph of the peak temperature vs. ac field amplitudes. As can be clearly seen from the figure, each set of data shows an excellent linear relationship for all the samples. Müller critical state model assumes a magnetic flux independent pinning force densities, and $\alpha_{j}$ and $\alpha_{k}$ for inter- and intragranular vortices described by the relation [25]:

\[ J_{\alpha} = J_{\alpha}^{\text{Müller}}(T) \]

\[ g_{\alpha} = g_{\alpha}^{\text{Müller}}(T) \]
where $U$ is 
\[
\left[ \frac{\mu_0 \mu_{\text{eff}}(0)}{2a \alpha_j(0)} \right],
\]
a is the length of the samples, $\mu_{\text{eff}}(0)$ is the effective permeability of the ceramic, and $\alpha_j(0)$ is the intergranular pinning force density. The slope of each line is proportional to $(\alpha_j(0))^{-1/2}$ and the vertical intercept of each line corresponds to the peak temperature, $T_{p0}$, at zero ac field amplitude. From a least squares fit of this expression using Eq. (1) to the data $T_{p0}$ and $U$ were extracted for all samples and tabulated in Table 2. Gold doping of the sample (G1) increased $T_{p0}$ from 90 K to 92 K and $U$ decreased from 0.090 and 0.086, in comparison with the undoped sample (G0). As can be seen from the table, the values of $T_{p0}$ increased while $U$ decreased with increasing diffusion-annealing time. A decreasing trend in $U$ means that the values of $(\alpha_j(0))$ increase with increasing the diffusion-annealing time, being consistent with our previous transport critical current density measurement such as $J_c$ increased with increasing the diffusion-annealing time [18]. A decreasing trend in pinning force of the samples with decreasing diffusion-annealing time is attributable to grater voids and defects as will be confirmed by SEM results.

The temperature dependence of inter-grain and intra-grain critical current density is often determined via the temperature dependence of the inter and intra-grain maximum in the imaginary part, $\chi''(T)$ of the ac susceptibility using the Bean model [24,25]. The inter-grain critical current density, $J_c^{\text{inter}}(T_p)$, was calculated for our samples using the relation $J_c^{\text{inter}}(T_p) = H_{ac}/a$ for the sample having cross section of the rectangular bar shaped, like $2ax2b$ where $a < b$. $J_c^{\text{inter}}(T_p)$ is the inter-granular critical current density and $H_{ac}$ is the amplitude of applied field at $T_p$. The determined $J_c^{\text{inter}}(T_p)$ at 77 K for the samples is listed in Table 2. As seen from the table, the value of $J_c^{\text{inter}}(T_p)$ increases with increasing diffusion-annealing time. This result is consistent with our previous transport critical current density measurements on the same samples [18]. Moreover, we observed the same evidence for this behavior in $\rho$ - $T$ transport measurements in the samples such as $T_c$ increases with increasing diffusion-annealing time. For intra-grain critical current density, we take the average grain size instead of $a$. For higher field values namely for 160 and 320 A/m the intra-granular (intrinsic loss) peak is clearly observed (Fig.4). In this work, we are not interested in calculating the intra-grain critical current density values. But it is clear that the intra-granular critical current densities are expected to be higher than the inter-granular critical current densities since intra-granular peak appears at high temperature.

### Table 2. The extracted values of $T_{p0}$, $U$ and $J_c^{\text{inter}}(T_p)$ of the samples.

| Samples | $J_c^{\text{inter}}$(A/cm²) (at 77 K) | $T_{p0}$ (K) | $U$ ($(\alpha_j(0))^{-1/2}$) |
|---------|-------------------------------------|--------------|-----------------------------|
| G0      | 35                                  | 90.34        | 0.094194                    |
| G1      | 60                                  | 93.17        | 0.093293                    |
| G2      | 72                                  | 95.46        | 0.089810                    |
| G3      | 90                                  | 97.70        | 0.085977                    |

### 3.2. XRD and SEM analyses

Fig. 6 shows the XRD patterns from the surfaces of (a) undoped Bi1.8Pb0.35Sr1.9Ca2.1Cu3Oy, (b) Au-diffused Bi1.8Pb0.35Sr1.9Ca2.1Cu3Oy annealed at 830 oC for 10 h, (c) Au-diffused Bi1.8Pb0.35Sr1.9Ca2.1Cu3Oy annealed at 830 oC for 20 h, and (d) Au-diffused Bi1.8Pb0.35Sr1.9Ca2.1Cu3Oy annealed at 830 oC for 50 h. Some of the miller indices are indicated in the figure. The calculated
lattice parameter $c$ from the $(00\ell)$ peaks of the XRD data is given in Table 3. The diffusion doping of the sample by gold was exhibited by an increase of the lattice parameter $c$ of the sample (0.19%) for the Au-doped sample (G1) in comparison with that for the undoped sample (G0). The impurity phase (Ca$_2$PbO$_4$) and $(002)L$ peaks are observed around $2\theta = 17.66^\circ$ and $5.83^\circ$ in the G0 sample, respectively. In the G1, G2, G3 samples, $(008)H$ peak is observed around $2\theta = 19^\circ$ while the $(008)L$ peak is appeared in the G0 sample. As can be seen from the table the lattice parameter $c$ increases with increasing diffusion annealing time.

### Table 3. Critical temperature $T_c^{\text{offset}}$, lattice parameter $c$, relative volume fraction, and room temperature resistivity of the samples.

| Samples | Critical Temperature $T_c^{\text{offset}}$ (K) | Lattice Parameter $c$ (Å) | Volume Fraction (%) | 2212 | 2223 |
|---------|----------------------------------|----------------|-------------------|------|------|
| G0      | 100 ± 0.2                        | 36.960         | 33                | 67   |
| G1      | 104 ± 0.2                        | 37.030         | 23                | 77   |
| G2      | 105 ± 0.2                        | 37.190         | 14                | 86   |
| G3      | 106 ± 0.2                        | 37.206         | 11                | 89   |

XRD measurements provide data about lattice parameters, average crystallite size and phase purity for Au diffused Bi-2223 bulk samples. The values of grain sizes can be estimated from XRD measurements by using Scherrer-Warren formula [26],

$$D = \left( \frac{0.94\lambda}{B \cos \theta} \right),$$

where $D$ is the crystallite size in nm, $\lambda$ (15.418 nm) is the wavelength of x-ray in nm, $\theta$ is the angle of intensity peak and $B$ is the full width at half maximum (FWHM) of the same intensity peak. With increasing diffusion-annealing time, the intensity of the XRD peaks increases while their positions shift towards smaller angles. This phenomenon indicates that average crystallite size becomes greater as confirmed by SEM investigations. The grain sizes calculated from XRD patterns are about 212, 230, 235 and 272 nm for G0, G1, G2 and G3 samples, respectively.

The relative volume fractions of the Bi-2223 and Bi-2212 phases in the samples were calculated from the peak intensities of the XRD results, using the following expressions [27,28]:

$$f_{(2223)} = \frac{\sum I_{H(hkl)}}{\sum I_{H(hkl)} + \sum I_{L(hkl)}},$$

and

$$f_{(2212)} = \frac{\sum I_{L(hkl)}}{\sum I_{H(hkl)} + \sum I_{L(hkl)}},$$

where $I_{H(hkl)}$ and $I_{L(hkl)}$ are the intensities of the $(hkl)$ diffraction lines for Bi-2223 and Bi-2212 phases, respectively (Fig.6).

The relative volume fractions of the phases for all the samples are given in Table 3. As can be seen from the table and from the XRD results one will notice that the volume fraction of Bi-2212 phase decreases while Bi-2223 phase increases with increasing the diffusion-annealing time.

In order to determine the grain sizes and possible precipitation of gold at the grain boundaries the surface morphology of the undoped and Au doped samples was studied by SEM. Fig. 7 represents surface micrographs for (a) the undoped (G0) and (b) gold doped sample annealed at 830 oC for 10 h.
The microstructure of G0 sample is remarkably different from that of G1 sample. Tiny spherical gold particles on the surface of the G1 sample can be seen (Fig. 7b). They are absent on the surface of G0 sample. A broad grain size distribution can be seen for the G1 sample. The average grain sizes calculated using image processing are about 250, 300, 325, 375 nm for the G0, G1, G2 and G3 samples, respectively. These values are consistent with grain sizes calculated from XRD peaks. It is observed that the grain connectivity is worsened gradually with decreasing diffusion-annealing time. The surface of the G1 sample is more uniform with better alignment of grains. Grain size of the gold-doped samples is greater. SEM photograph of G1 shows better crystallinity in comparison with the G0. These results indicate that the surface morphology of the sample is improved with gold diffusion. In the undoped sample flake-type grains were observed and these flake-type grains are believed to be due to the low-\(T_c\) phase (Bi-2212) [29,30], being consistent with calculation of the relative volume fraction of the phases in the present study. The flake-type grains are less dominant in sample G1 in comparison with sample G0 while the composition of the needle-type grains grew gradually. These needle-type grains correspond to the high-\(T_c\) phase (Bi-2223) [29-31]. The increase of \(J_c\) in Bi(Pb)-Sr-Ca-Cu-O by Au diffusion may be caused by the increase of grain size.

![Image](image_url)

**Figure 6.** The XRD patterns for the G0, G1, G2 and G3. The peaks indexed L(hkl) and H(hkl) represent the Bi-2212 and Bi-2223 phases, respectively.
3.3. Magnetoresistivity measurements

We have investigated the effect of Au diffusion and diffusion-annealing time on the superconducting properties performing dc resistivity measurements under different dc magnetic fields in the range of zero and 0.5 T. The temperature dependence of magnetoresistivity for all samples (G0, G1, G2 and G3) is shown in Fig. 8. As seen from the figure above 114 K the behaviour of the temperature dependence of the resistivities of the samples is metallic. The room temperature resistivity decreases with increasing the diffusion-annealing time. The zero-resistivity transition temperatures at zero dc fields are 100 K for G0, 104 K for G1, 105 K for G2 and 106 K for G3 sample. It is observed that the zero resistivity transition temperature of the G1 (104 K) is higher than that of G0 sample (100 K), being in agreement with literature [15]. The increased value of $T_c$ in the Au diffusion-doped samples compared with the G0 in our investigations can be interpreted as a result of gold diffusion in inter-grain boundaries. This in turn causes the increase of inter-grains contact surface or decreasing the inter-grain resistivity as can be seen in Fig. 8. It was observed that the $T_c$ of gold-doped samples increases from 104 K to 106 K with increasing diffusion-annealing time. From the above results, it was inferred that the annealing time has the positive effect on the $T_c$ value of the samples. The increase of $T_c$ in Bi(Pb)-Sr-Ca-Cu-O by Au diffusion may also be caused by the increase of grain sizes and their orientations, by improved coupling between Au-diffused superconducting grains, and by the increased number of flux pinning centres due to the presence of Au in the inter-grain regions.
agreement with literature, XRD patterns obtained from the G0 sample has the characteristic impurity peak due to Ca$_2$PbO$_4$ while the gold-doped samples do not have the impurity peak. Presence of this impurity peak in G0 sample is consistent with the experimental finding that the gold-doped samples had lower normal state resistivity and higher zero resistivity transition temperature (Table 3). It was concluded that formation of Ca$_2$PbO$_4$ will disturb Pb stoichiometry of the sample which in turn will elevate phase segregation from Bi-2223 to Bi-2212. On the other hand, the transition temperature and the lattice parameter c increase with increasing the diffusion-annealing time. This behaviour is due to the change in the Meissner effect as a function of diffusion-annealing time, and is also consistent with the results of the XRD and ac susceptibility measurements.

Fig. 8 displays the temperature dependence of resistivity at magnetic fields of 0, 100, 200, 300 and 500 mT for all the samples. We observed that the broadening of the resistivity transition width increases with increasing external dc magnetic field and with decreasing the diffusion-annealing time, being consistent with our ac susceptibility measurements. It was also observed that, with increasing dc magnetic field up to 0.5 T, $T_c$ ranges from 100 K to 67 K for G0, 104 K to 70 K for G1, 105 K to 71 K for G2, and 106 K to 75 K for G3 sample.
(b) 

(c)
Figure 8. The temperature dependence of electrical resistivity for the (a) \(G0\), (b) \(G1\), (c) \(G2\) and (d) \(G3\) samples under various applied dc magnetic field up to 0.5 T.

4. Conclusion
In conclusion, \(\text{Bi}_{1.8}\text{Pb}_{0.35}\text{Sr}_{1.9}\text{Ca}_{2.1}\text{Cu}_3\text{O}_y\) samples were prepared by solid-state reaction method. The effect of gold diffusion and diffusion-annealing time on the undoped and gold-doped samples was investigated by performing ac susceptibility, magnetoresistivity, XRD and SEM measurements. Our investigations revealed that the gold-diffusion might have improved the coupling of the grains, leading to enhancement of the microstructure and superconducting properties of \(\text{Bi}_{1.8}\text{Pb}_{0.35}\text{Sr}_{1.9}\text{Ca}_{2.1}\text{Cu}_3\text{O}_y\) samples. When gold-doped \(\text{Bi}_{1.8}\text{Pb}_{0.35}\text{Sr}_{1.9}\text{Ca}_{2.1}\text{Cu}_3\text{O}_y\) samples are compared with the undoped sample, following statements are concluded:

- The peak temperature shifted to higher temperature.
- The inter-granular pinning force density increased.
- The inter-granular critical current density also increased.
- The lattice parameter \(c\) increased.
- The relative volume fraction of Bi-2223 phase increased.
- The surface morphology of the microstructure is improved.
- The needle-like grains grew gradually.
- The critical transition temperature increased.
- Room temperature resistivity decreased.

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