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Pressure-induced linear enhancement of the superconducting transition in $Nd_{0.8}Sr_{0.2}NiO_2$ thin films

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Abstract

We report the pressure (P) effect on the superconducting transition temperature T_c and the upper critical field $\mu_0 H_{c2}$ of infinite-layer $Nd_{0.8}Sr_{0.2}NiO_2$ thin films by measuring the electrical transport properties under various hydrostatic pressures to 4.6 GPa. At ambient pressure, it shows the clear superconducting transition with $T_c \sim 10$ K. Based on the evolution of resistance $R(T)$, we found that the T_c is monotonically enhanced to ∼14 K upon increasing pressure to 2.9 GPa. The constructed temperature–pressure phase diagram indicates that the calculated slope dT_c/dP is about 1.14 K GPa^{-1} and the superconducting T_c shows no signatures of saturation with pressure. It thus gives the possibility to further enhance T_c by employing higher pressures or heterostructure engineering. In addition, the normalized slope of upper critical field $\mu_0 H_{c2}(0)$ implies that the electron correlations are gradually decreasing with pressure, which exhibits an opposite evolution with superconducting T_c. Our work further confirms the positive pressure effects in nickelate superconductors and gives more insight to further enhance its superconducting transition temperature.

Keywords: thin films, nickelates, unconventional superconductivity, high pressure, $Nd_{0.8}Sr_{0.2}NiO_2$

1. Introduction

Recently, the experimental observation of superconductivity in infinite-layer nickelate $Nd_{1-x}Sr_xNiO_2$ thin films has received considerable attention [1], and it further stimulates the discovery of superconductivity in other rare-earth substituted infinite-layer nickelate $La_{1-x}(Ca/Sr)_xNiO_2$ [2, 3] and $Pr_{1-x}Sr_xNiO_2$ [4] systems. As can be seen from the constructed phase diagram of $(La/Pr/Nd)_{1-x}Sr_xNiO_2$ [2, 4–6] and $La_{1-x}Ca_xNiO_2$ [3], the superconducting phase is next to the weakly insulating state in the underdoped and overdoped regimes, which is different from the cuprates [7]. However, the corresponding bulk materials display an insulating behavior with decreasing temperature gradually, and no indication of superconductivity emerges even when the pressure increases to 50 GPa [8]. Therefore, a question has been raised to whether the experimental observation of superconductivity originates from the heterostructure or epitaxy strain between the thin

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films and the $SrTiO_3$ (STO) substrates. Despite many efforts in experimental investigations and theoretical calculations have been devoted to understanding its superconducting mechanism [9–19], a consensus has not yet been reached due to the grant challenges in material synthesis.

Aiming at exploring more Ni-based superconductors, researchers have succeeded in synthesizing new nickelate superconductors, such as $Nd_6Ni_5O_{12}$ thin film on $NdGaO_3$ substrate with $T_c \sim 13$ K [20], and $Pr_{0.82}Sr_{0.18}NiO_2$ thin film on $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ substrate with $T_c \sim 15$ K [21, 22]. While many efforts have been paid to explore more Ni-based superconductors with higher T_c, the reported highest T_c remains lower than 20 K at ambient pressure. Thus, an important issue in this field is how to further enhance the superconducting T_c. As indicated by the theoretical calculations, the in-plane compressive lattice strain would raise the superconducting T_c [23], and then it was evidenced in experiments where the STO substrate was substituted by $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ substrate with smaller in-plane lattice parameters [21, 22]. On the other hand, the in-plane lattice constant of the reported nickelate thin films is locked by the STO substrate with $a \sim 3.91$ Å, while the maximum T_c in nickelates at similar optimal hole doping exhibits an inverse correlation to the c-axis constant, such as $T_c = 9$ K for $La_{0.8}Sr_{0.2}NiO_2$ ($c \approx 3.44$ Å) [2], $T_c = 10$ K for $La_{0.82}Ca_{0.18}NiO_2$ ($c \approx 3.385$ Å) [3], $T_c = 11$ K for $Nd_{0.775}Sr_{0.225}NiO_2$ ($c \approx 3.375$ Å) [5, 6] and $T_c = 14$ K for $Pr_{0.82}Sr_{0.18}NiO_2$ ($c \approx 3.36$ Å) [4]. Moreover, the application of high pressure to the superconducting nickelate thin films follows this trend that pressure can enhance the superconducting T_c of $Pr_{0.82}Sr_{0.18}NiO_2$ thin films to over 30 K [24]. However, whether this approach is applicable to other nickelate superconductors is still needed to explore. So, it is worthy to investigate the evolution of superconducting properties in other infinite layer nickelates by employing high pressure.

According to the previous report on the $Pr_{0.82}Sr_{0.18}NiO_2$ thin films, the thin films would be deteriorated under higher pressures, especially when the liquid pressure transiting medium solidifies [24]. Moreover, the metallic behavior of resistance at ambient pressure will immediately change to the insulating behavior by employing the solid pressure transiting medium under a smaller pressure, which indicates that strong strain/stress will enhance the carrier scatterings [24]. In this work, we have performed detailed transport measurements on the infinite-layer $Nd_{0.8}Sr_{0.2}NiO_2$ thin films by using the piston cylinder cell (PCC) and cubic anvil cell (CAC) under various hydrostatic pressures up to 4.6 GPa, due to the sensitive response of nickelate thin film to the pressure conditions.

Our results show that the superconducting T_c exhibits a monotonic enhancement under high pressure, and the slope of $T_c(P)$ calculated from the constructed temperature–pressure phase diagram is about 1.14 K GPa^{-1} without showing any saturation. Such a pressure coefficient of T_c is consistent with previously reported high-pressure data of $Pr_{0.82}Sr_{0.18}NiO_2$ [24]. Therefore, our results indicate that the superconducting T_c in infinite-layer nickelate superconductors can be further enhanced by heterostructure engineering or physical/chemical pressures.

2. Experimental details

The STO (001) substrates of the size 5 × 5 mm^2 were pre-annealed at 900 °C with an oxygen partial pressure of 1×10^{-5} Torr, and then the $Nd_{0.8}Sr_{0.2}NiO_3$ thin films were deposited on the STO substrates at 650 °C with 150 mTorr oxygen partial pressure by using a pulsed laser deposition system. The laser pulse energy was 960 mJ cm^{-2} and the frequency was 4 Hz. After finishing the growth process, the precursor nickelate films were wrapped in the aluminum foil and sealed with 0.1 g CaH_2 powder in quartz tubes (vacuum better than 1×10^{-5} Torr). The reduction was carried out at a temperature of 290 °C for 5 h, with the heating and cooling rates of 10 °C min^{-1}. More details can be found in the [25]. The obtained infinite-layer $Nd_{0.8}Sr_{0.2}NiO_2$ thin films was cut into three pieces of the size 0.5 × 0.3 mm^2 with the STO substrate for transport measurements, and the three samples were labeled with S1, S2 and S3. Here, the standard four probe method was used to measure the temperature-dependent resistance of $Nd_{0.8}Sr_{0.2}NiO_2$ thin films with the electrical current applied within the ab-plane. We employ the PCC (for samples of S1 and S2) and palm-type CAC (for sample S3) to measure its resistance under various hydrostatic pressures up to 4.6 GPa. The Daphne 7373 and glycerol were used as the pressure transmitting medium in PCC and CAC, and the pressure values inside the pressure cell were estimated by measuring the T_c of Pb at low temperatures.

3. Results and discussions

Figure 1 shows the temperature dependent resistance $R(T)$ of $Nd_{0.8}Sr_{0.2}NiO_2$ thin films under various pressures up to 1.56 GPa by using the PCC. At ambient pressure, the resistance $R(T)$ curves of sample S1 and S2 exhibit the typical metallic behavior when the temperature gradually cools down. As we can see that the normal state resistance $R(T)$ first shows an increase with increasing pressure gradually to 0.31 GPa and then it decreases again when the pressure further increases to 1.56 GPa, figures 1(a) and (c). As is displayed in figures 1(b) and (d), we can see the enlarged view of resistance $R(T)$ below 20 K for S1 and S2. Here, we determine the $T_c^{0.9Rn}$ as the transition temperature at the 10% resistance drop of the normal state resistance R_n, T_c^{mid} as the transition temperature at the 50% resistance drop of the normal state resistance R_n and T_c^{zero} as the transition temperature at zero resistance (R_n is the resistance above the superconducting transitions). At ambient pressure, the resistance $R(T)$ shows the superconducting transition with $T_c^{0.9Rn} \approx 7.8$ K, $T_c^{mid} \approx 5.8$ K and $T_c^{zero} \approx 4$ K for S1 and $T_c^{0.9Rn} \approx 7.7$ K, $T_c^{mid} \approx 5.8$ K and $T_c^{zero} \approx 3.4$ K for S2, which is a little bit lower than the previous reports [1, 5, 6]. This would be induced by the local inhomogeneities originating from the reduction process [24]. When we gradually increase the pressure to 1.56 GPa, the superconducting

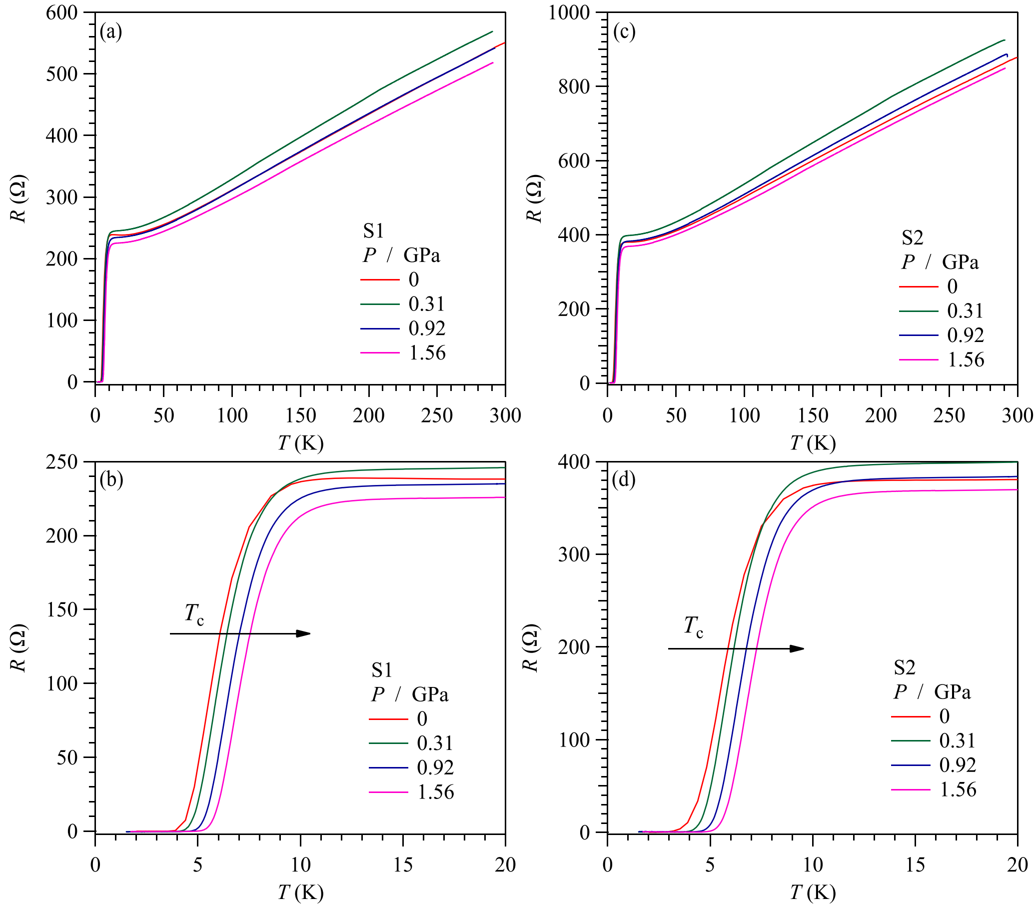

Figure 1. Temperature dependence of resistance $R(T)$ of $Nd_{0.8}Sr_{0.2}NiO_2$ thin films under various pressures up to 1.56 GPa for (a) sample S1 and (c) sample S2. The resistance $R(T)$ curves below 20 K for (b) sample S1 and (d) sample S2, illustrating the variation of the superconducting transition temperatures with pressure. The arrows in (b) and (d) show the evolution of superconducting transition of S1 and S2.

transitions almost parallelly move to higher temperatures with $T_c^{mid}\sim 7.3$ K and $T_c^{zero}\sim 5.5$ K for S1 and $T_c^{mid}\sim 7.1$ K and $T_c^{zero}\sim 5.2$ K for S2.

To further track the evolution of superconducting transition in $Nd_{0.8}Sr_{0.2}NiO_2$ thin film, we perform the resistance measurements for sample S3 to higher pressures by employing CAC. As displayed in figures 2(a) and (b), we can clearly see that the superconducting transition temperatures are $T_c^{0.9Rn}\approx 8.7$ K and $T_c^{mid}\sim 6.5$ K at ambient pressure. Moreover, the residual resistance appears at low temperatures, which may originate from the sample inhomogeneity which is induced in the topotactic reduction process. With gradually increasing pressure to 2.9 GPa, the normal state resistance shows an increase, and the superconducting transition were enhanced to $T_c^{0.9Rn}\approx 11.3$ K and $T_c^{mid}\sim 9.1$ K. When we further increase the pressure to 4.6 GPa, the normal state resistance exhibits a vertical shift, and displays an upturn below 100 K. But we can still observe the superconducting transition starting at ∼13 K with a weak resistance drop. Compared to the high-pressure results of $Pr_{0.82}Sr_{0.18}NiO_2$, the emergent insulating behavior in the resistance of $Nd_{0.8}Sr_{0.2}NiO_2$ sample at 4.6 GPa should correlated with the lack of protection by the STO caping layer deposited on the thin film, which is explicitly described in the synthesized process [25] and will be discussed in detail below.

Based on the obtained $T_c^{0.9Rn}$ and T_c^{mid} from the transport measurements by employing the PCC and CAC, we construct the superconducting temperature–pressure phase diagram of $Nd_{0.8}Sr_{0.2}NiO_2$ thin films as displayed in figure 3(a). It explicitly describes the gradual enhancement of the superconducting transition temperature, and we can calculate the slope $dT_c/dP\sim 1.14$ K GPa^{-1} as indicated by the pink dashed lines. It is consistent with previous reports on $Pr_{0.82}Sr_{0.18}NiO_2$ thin films under high pressures, figure 3(b). Noticeably, the enhancement of superconducting T_c in infinite-layer nickelates would receive more attention both on theoretical calculations and experiments to further raise the superconducting T_c and explore the superconducting mechanism.

Figures 4(a) and (b) display the temperature dependence of resistance $R(T)$ under various magnetic field at ambient pressure and 2.9 GPa. As we can see, the superconducting transition of $Nd_{0.8}Sr_{0.2}NiO_2$ thin film is gradually suppressed to lower temperatures and the transition width becomes more broad with increasing the magnetic field gradually. Here, we use the 50% drops in $R(T)$ to define the upper critical field as $\mu_0H_{c2}(T_c^{mid})$, and the temperature dependence

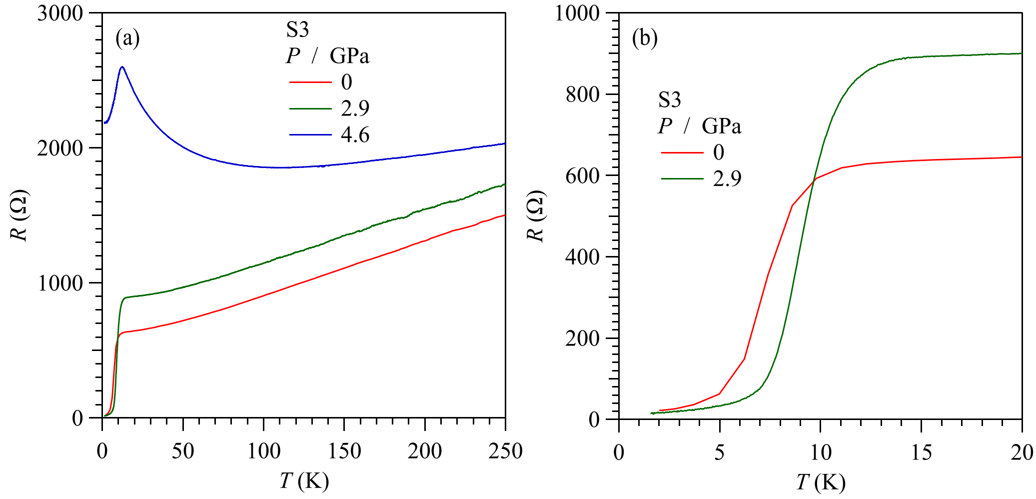

Figure 2. (a) Temperature dependence of resistance for sample S3 under various pressures up to 4.6 GPa by employing CAC. (b). The resistance $R(T)$ curves below 20 K for better illustration of the superconducting transition with pressure.

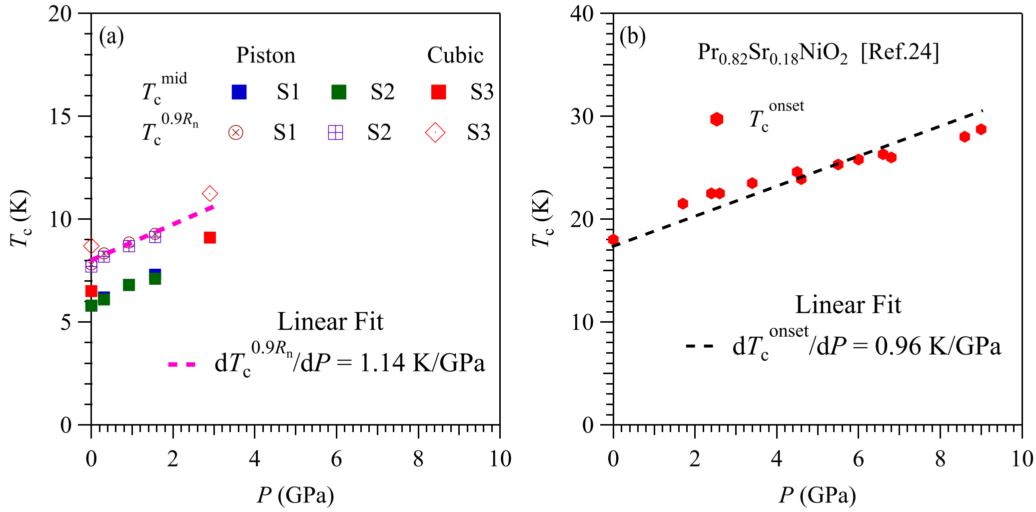

Figure 3. Temperature–pressure phase diagram of $Nd_{0.8}Sr_{0.2}NiO_2$ thin films (a) and $Pr_{0.82}Sr_{0.18}NiO_2$ thin films (b).

of $\mu_0 H_{c2}(T_c^{\mathrm{mid}})$ was shown in figure 4(c). Then, we further calculate the zero-temperature-limit upper critical field $\mu_0 H_{c2}(0)$ by using the empirical Ginzburg–Landau formula $\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0)(1 - t^2)/(1 + t^2)$, where t represents the reduced temperature T/T_c. The fitting results are indicated by the broken lines in figure 4(c). The $\mu_0 H_{c2}(0)$ exhibits a monotonic enhancement with pressure, which is similar to the evolution of $T_c(P)$, figure 4(d). On the other hand, we also extract the normalized initial slope of upper critical field—$(1/T_c)\ |dH_{c2}/dT|_{T_c}$ which is related to the effective mass m^* based on the single-band model. As indicated in figure 4(d), it decreases from $\sim$0.24 T K^{-2} at 0 GPa to $\sim$0.19 T K^{-2} at 2.9 GPa, implying that the effective mass m^* of charge carriers or the electron correlations exhibit a pressure-induced reduction. In this sense, our high-pressure results are consistent with the previously reported data in $Pr_{0.82}Sr_{0.18}NiO_2$ thin films [24].

By performing the high-pressure resistivity measurements on $Nd_{0.8}Sr_{0.2}NiO_2$ thin films, we track the evolutions of the superconducting transition T_c and the upper critical fields $\mu_0 H_{c2}$ and construct the phase diagram of $Nd_{0.8}Sr_{0.2}NiO_2$ thin films as discussed above. First, we reveal the positive pressure effect on the superconducting transition of $Nd_{0.8}Sr_{0.2}NiO_2$. It tells us that we can further raise its superconducting transition T_c under higher pressures. According to the reported data of the infinite-layer nickelate thin films, the in-plane lattice constants seem to be locked by the STO substrate with $a \sim 3.91$ Å, while the maximum superconducting T_c exhibits an inverse evolution with the lattice constant change of c-axis [26]. It is in good agreement with the high-pressure results of $Pr_{0.82}Sr_{0.18}NiO_2$ thin films [24]. These findings deserve further experimental investigations to explore the optimal superconducting T_c in the infinite-layer nickelate systems.

On the other hand, theoretical calculations predict that the in-plane compressive strain would enhance the coupling between the $3d$ orbitals of Ni and the $2p$ orbitals of O, change the band structure and thus would

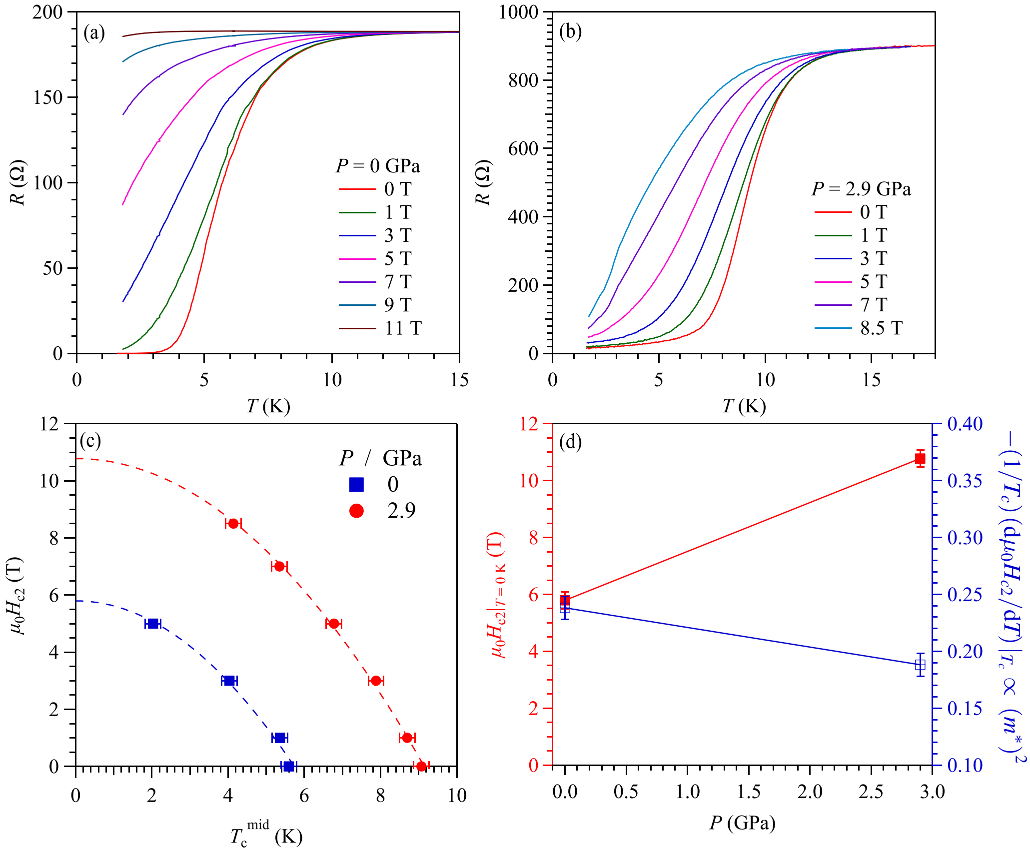

Figure 4. (a) Temperature dependence of resistance at ambient pressure with various magnetic fields up to 11 T. (b) Temperature dependence of resistance at 2.9 GPa with various magnetic fields up to 8.5 T. (c) Temperature dependences of the upper critical field $\mu_0 H_{c2}$ at 0 GPa and 2.9 GPa, where $\mu_0 H_{c2}$ values are determined by using the criteria of 50% R_n. The broken lines represent the Ginzburg–Landau (G–L) fitting. (d) Pressure dependence of the upper critical field $\mu_0 H_{c2}(0)$ and the normalized slope—$(1/T_c)[d\mu_0 H_{c2}/dT]|_{Tc}$.

raise the superconducting transition temperature [23]. More recently, the infinite-layer nickelate thin film superconductors $Nd_{1-x}Sr_xNiO_2$ and $Pr_{0.8}Sr_{0.2}NiO_2$ were successfully synthesized on another substrate $(LaAlO_3)_{0.3}$ $(Sr_2AlTaO_6)_{0.7}$. As we know, it has a smaller in-plane lattice constant $a = 3.868$ Å than the widely used STO substrate with $a = 3.905$ Å. Based on the transport measurements, the superconducting transition T_c shows an obvious enhancement [21, 22]. It indicates that the superconducting T_c can be further enhanced to higher temperatures by employing stronger compressive strain. Moreover, this assumption was further evidenced by the high-pressure measurements that the superconducting T_c exhibits the linear enhancement with pressure in $Pr_{0.82}Sr_{0.18}NiO_2$ [24]. The observed reduction of effective mass m^* or the electron correlations further implies that the compressive strain changes the band structure. In this regard, our finding of pressure reducing the electronic correlations is consistent with the theoretical calculations based on the one-band Hubbard model that the decrease of onsite interaction U would lead to a continuous increase of superconducting T_c [17]. Anyway, more experiments and theoretical calculations should be performed to further explore the key role of electronic correlations, hybridization between Ni-3d and rare-earth 5d electrons or the cation size in determining the superconducting T_c in the infinite-layer nickelate thin films.

Finally, we give a brief discussion on the pressure-induced insulating behavior of the resistance at 4.6 GPa. In this work, the $Nd_{0.8}Sr_{0.2}NiO_3$ thin films were deposited on the STO substrates without STO capping layers [25]. During the reduction process, the apical oxygen of the octahedra in $Nd_{0.8}Sr_{0.2}NiO_3$ thin film were removed and the infinite-layer $Nd_{0.8}Sr_{0.2}NiO_2$ exposed to the surroundings directly. According to the previous reports on the $Pr_{0.82}Sr_{0.18}NiO_2$ thin films with the STO capping layer, the thin films would be deteriorated under higher pressures, especially when the liquid pressure transiting medium solidifies. Moreover, the metallic behavior of resistance at ambient pressure changes to the insulating behavior immediately by employing the solid pressure transiting medium under a moderate pressure, which indicates that the strong strain/stress would introduce the dislocations/defects and enhances the carrier scatterings [24]. Without the STO capping layer, the $Nd_{0.8}Sr_{0.2}NiO_2$ thin film is more sensitive to the pressure conditions and eventually shows the insulating behavior with a relative strong strain/stress at lower pressures.

4. Conclusion

In summary, we have performed high-pressure electrical transport measurements on $Nd_{0.8}Sr_{0.2}NiO_2$ thin films by using the

PCC and CAC. The constructed temperature–pressure phase diagram reveals a linear enhancement of superconducting T_c with a slope ~1.14 K GPa^{-1} and no saturation was observed. Therefore, our work further confirms the positive pressure effects on the infinite-layer nickelates superconductors and provides the possibility to further enhance its T_c by heterostructure engineering or physical/chemical pressure. In addition, our results support the predictions of the theoretical calculations and would motivate further exploration of more nickelate superconductors.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

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