Pressure-induced monotonic enhancement of T_c to over 30 K in the superconducting $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin films

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Abstract

The successful synthesis of superconducting nickelate thin films with the highest $T_c \sim 15$ K has reignited great enthusiasms on this class of potential analogue to high- T_c cuprates suggested decades ago. To pursue higher T_c is always an important task in studying new superconductors. Here we report for the first time the effect of pressure on the superconducting properties of infinite-layer $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin films by measuring electrical resistivity under various pressures in a cubic anvil cell apparatus. We find that the onset of superconductivity, T_c^{onset} , can be enhanced monotonically from ~ 18 K at ambient pressure to ~ 31 K without showing signatures of saturation upon increasing pressure to 12.1 GPa in the presence of liquid pressure transmitting medium. This encouraging result indicates that the T_c of infinite-layer nickelates superconductors can be further raised up by applying higher pressures or strain engineering in the heterostructure films. In addition to the pressure effect, we also discussed the influence of stress/strain on the superconducting properties of the nickelate thin films.

Introduction

The discovery of high- T_c superconductivity in $\text{La}_{1.85}\text{Ba}_{0.15}\text{CuO}_4$ at 1986 by J. G. Bednorz and K. A. Muller [1] has triggered exhaustive experimental and theoretical studies on the cuprates with an aim to find more superconductors with higher T_c and to unveil the underlying mysteries mechanisms. After over 30 years of endeavor, the highest T_c in cuprates, ~ 135 K at ambient [2] and ~ 164 K under pressure [3], remains to be much lower than room temperature, and the mechanism is still an enigma. Alternatively, as the nearest neighbor to copper in the element periodic table, the infinite-layer nickelates such as doped LaNiO_2 and NdNiO_2 showing the great similarities in both crystal structures and electronic configurations to cuprates have been considered and explored extensively as potential high- T_c superconductors ever since the early 1990s [4–13]. Unfortunately, superconductivity was not observed in the synthesized powder and thin-film nickelates samples until very recently.

In 2019, Li *et al.* reported the experimental observation of superconductivity with $T_c = 9\text{--}15$ K in the hole-doped infinite-layer $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ thin films obtained by soft-chemistry topotactic reduction from the corresponding perovskite phase [14]. From the experimentally constructed superconducting phase diagram, the observed $T_c(x)$ has a non-monotonic evolution with doping x , which is similar to the hole-doped cuprates [15–17]. This discovery has reignited the enthusiasms on the nickelates and immediately attracted intensive investigations recently. In contrast to the superconducting thin-film samples, the bulk samples exhibit an insulating ground state, and no indications of superconductivity was observed even under pressures up to 50 GPa [18]. It has thus raised the question whether the observed superconductivity correlates intimately with the heterostructure or epitaxy strain between the thin films and the substrate. For the parent LaNiO_2 and NdNiO_2 , X-ray absorption spectroscopy (XAS) and resonant inelastic X-ray scattering (RIXS) confirmed the nominal $3d^9$ electronic configuration, but revealed a reduced hybridization between Ni $3d$ orbitals and O $2p$ orbitals, and an enhanced coupling between Ni $3d$ states

and La/Nd 5*d* states [19]. Notably, the electron energy loss spectroscopy (EELS), high-resolution XAS and RIXS experiments further revealed that the doped holes reside on the Ni sites, forming the low-spin d^8 state [20, 21]. These latter observations are different from those in cuprates [22, 23]. Recent STM experiments uncovered a mixed *s*- and *d*-wave superconducting gap feature on the rough surface of Nd_{1-x}Sr_xNiO₂ thin films [24], and these results were reproduced by employing *ab initio* treatment on different terminated surfaces [25, 26]. As a summary from theoretical side, three main perspectives on nickelates have been proposed, including the cuprate-like correlated single-Ni-orbital $d_{x^2-y^2}^2$ band [23, 25, 27–32], the Ni-3*d*-multiorbital effects [33–37], and the Kondo physics between Ni-3*d*-orbital and Nd-5*d*-orbital [38–40]. So far, consensus has not yet been reached about the superconducting mechanism. This is partially attributed to the great challenges in the materials' synthesis and thus the relatively poor samples' quality of these infinite-layer nickelates.

In addition to Nd_{1-x}Sr_xNiO₂ thin films, superconductivity has also been achieved in other doped rare-earth nickelates, such as La_{1-x}(Ca/Sr)_xNiO₂ [41, 42] and Pr_{1-x}Sr_xNiO₂ [43]. The phase diagrams of (La/Pr/Nd)_{1-x}Sr_xNiO₂ and La_{1-x}Ca_xNiO₂ thin films are all featured by the superconducting phase adjacent to the weakly insulating state in the underdoped and overdoped regimes [16, 17, 41–43]. In addition, the doping-dependent generalized superconducting dome was found to shift upon changing the rare-earth cations. However, a local T_c valley structure of the superconducting phase was not observed in the Pr_{1-x}Sr_xNiO₂ [43], which is strictly different from the situation in Nd_{1-x}Sr_xNiO₂ and La_{1-x}(Ca/Sr)_xNiO₂ [16, 17, 41, 42]. These comparisons highlight the important role of the rare-earth cations in addition to the lattice strain applied by the SrTiO₃ substrate in determining the superconducting state of the nickelates. Although much recent effort has been devoted to the infinite-layer nickelates, the reported highest T_c^{onset} remains lower than 15 K [14, 16, 17, 41–45]; it thus becomes an important issue to further enhance the T_c of the superconducting nickelates to higher temperatures. In this regard, the application of high pressure that has been widely employed to raise T_c of cuprates [3] and iron-pnictides unconventional superconductors [46–49] should be a primary choice. To the best of our knowledge, however, this approach has not been applied to the superconducting nickelate thin films so far. We are thus motivated to investigate the effect of pressure on the superconducting properties of the infinite-layer nickelates.

In this work, we have performed detailed transport measurements on the infinite-layer Pr_{0.82}Sr_{0.18}NiO₂ thin films by using the palm-type cubic anvil cell (CAC) under various hydrostatic pressures up to 12.1 GPa. We observe a positive pressure effect on the superconducting $T_c(P)$, which increases monotonically from ~ 18 K at ambient to ~ 31 K at 12.1 GPa without leveling off. This result is quite encouraging and should promote further endeavors to raise the T_c of this class of new superconductors. During the course of this study, we noticed that the superconducting nickelate thin films are very sensitive to the pressure conditions or the pressure transmitting medium (PTM). So we also discussed the influence of stress/strain on the superconducting properties of the nickelate thin films.

Experimental Details

The $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_3$ films were deposited on TiO_2 -terminated SrTiO_3 (001) substrates at 650 °C with 150 mtorr oxygen partial pressure using a pulsed laser deposition system [50, 51]. The laser energy density of 1.2 J/cm^2 and a frequency repetition of 4 Hz were provided by a XeCl excimer laser with 308 nm central wavelength. Subsequently, a SrTiO_3 epitaxial capping layer was deposited at 600 °C. After the deposition, each film was vacuum-sealed in a glass tube (vacuum of 4×10^{-4} torr) with 0.2 g CaH_2 , and kept for 60 minutes at 290 °C to obtain the superconductivity of the $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ film. The X-ray diffraction θ - 2θ symmetric scans of the $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_3$ film and the $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ ones were obtained by a Rigaku SmartLab (8 kW) high-resolution x-ray diffractometer, as shown in Fig. S1.

Standard four-probe method was used to measure temperature-dependent resistivity of the $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin films with the electric current applied within the ab -plane. The magnetic field is applied parallel to the c -axis. We employ a palm-type CAC to measure its resistivity under various pressures up to 12.1 GPa with the glycerol as the liquid PTM, and to 6.6 GPa with the Daphne 7373 as the liquid PTM [52]. For comparison, we also performed similar measurements with solid PTM, h-BN. The pressure values inside the CAC were estimated from the pressure-loading force calibration curve pre-determined by measuring the T_c of Pb at low temperatures.

Results And Discussions

Figure 1(a) shows the temperature dependence of in-plane resistivity $\rho(T)$ for a $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin film (#1) under various pressures up to 12.1 GPa with glycerol PTM. At ambient pressure, $\rho(T)$ exhibits a typical metallic behavior in the normal state prior to the superconducting transition below $\sim 18 \text{ K}$. As can be seen, the magnitude of normal-state resistivity shows complex and non-monotonic evolutions with increasing pressure; it first decreases considerably from 0 to 2.5 GPa, increases gradually until 6.8 GPa, and then decreases again at higher pressures. At $P > 4.6 \text{ GPa}$, a broad hump feature appears in $\rho(T)$ between 50 K and 250 K, similar to the situation seen in the Fe-based superconductors due to the incoherent-to-coherent crossover of the $3d$ electrons [53–56]. These high-pressure results resemble the observations in the Sr/Ca-doped infinite-layer nickelate thin films [14, 16, 17, 41–43], in which the defects or cation-site disorders dominate the scattering processes in resistivity. Although the application of high pressure does not introduce extra disorders in principle, the stress/strain upon compression might partially deteriorate the delicate thin films and enhance the carrier scattering as discussed below. In the presence of liquid PTM, fortunately, the superconducting transition remains visible up to at least 12.1 GPa, which is the highest pressure in the present study.

The $\rho(T)$ data below 35 K are displayed in Fig. 1(b) to highlight the evolutions of the superconducting transition. Here, we define the $T_c^{\text{onset},1}$ (red up-pointing arrow) as the temperature where $\rho(T)$ starts to deviate from the extrapolated normal-state behavior, the $T_c^{\text{onset},2}$ (black down-pointing arrow) and T_c^{offset} as the interception between two straight lines below and above the superconducting transition. These two onset temperatures, $T_c^{\text{onset},1}$ and $T_c^{\text{onset},2}$, should correspond to the temperatures where the superconducting state (or Cooper pairs) just appears and grows up quickly, respectively. At ambient

pressure, the $\rho(T)$ shows a high superconducting transition with $T_c^{\text{onset},1} \approx 18$ K, $T_c^{\text{onset},2} \approx 16.3$ K, which are consistent with the previous report [43]. It should be noted that there is a small residual resistance below $T_c^{\text{offset}} \approx 12.2$ K, and it should be ascribed to the inhomogeneity of the small-sized sample for high-pressure measurements. When we increase pressure to 2.5 GPa, both the normal-state and the residue resistivity decrease considerably, and the superconducting transition moves to higher temperature with $T_c^{\text{onset},1} \approx 20.9$ K, $T_c^{\text{onset},2} \approx 18.9$ K, and $T_c^{\text{offset}} \approx 15$ K. Upon increasing pressure from 2.5 to 4.6 GPa, however, the normal-state and the residue resistivity exhibit an anomalous increase by $\sim 50\%$ at 35 K and $\sim 200\%$ at 2 K as seen in Fig. 1(b). But the superconducting transition at 4.6 GPa remains relatively sharp and is further enhanced to $T_c^{\text{onset},1} \approx 23.9$ K, $T_c^{\text{onset},2} \approx 21.5$ K and $T_c^{\text{offset}} \approx 17.5$ K. For $P \geq 6.8$ GPa, the superconducting transition is broadened up considerably featured by a long tail with a large residual resistivity. As discussed below, this should be attributed to the degradation of the delicate thin-film samples in the presence of substantial stress/strain upon compression. Fortunately, the utilization of the relatively soft liquid PTM, though solidified under pressure and cooling down, still preserves the metallic behavior and allows us to monitor the onset of the superconducting transitions that continues to increase with pressure. This can be seen clearly from the normalized resistivity $\rho(T)/\rho(35$ K) in Fig. 1(c). The $T_c^{\text{onset},1}$ and $T_c^{\text{onset},2}$ are enhanced gradually from 26 K and 23.1 K for 6.8 GPa to 31 K and 28.6 K at 12.1 GPa.

To check the reproducibility of the above results, we measured another sample (#2) that shows a slightly higher T_c at ambient pressure in a separate run up to 6.6 GPa with Daphne 7373 as the liquid PTM. As seen in Fig. 2(a), this sample shows similar evolutions for both the normal-state and the residual resistivity as a function of pressure. But an obvious upturn appears above the superconducting transition at 6.6 GPa. Fig. 2(b) shows the enlarged view of $\rho(T)$ data below 30 K to track the evolutions of the superconducting transition for the #2 sample. We can see that the superconducting T_c increases gradually with pressure from ambient to 6.6 GPa. However, the residual resistivity exhibits a prominent enhancement above 2.4 GPa, which can be seen more clearly in Fig. 2(c) by normalizing the resistivity $\rho(T)/\rho(30$ K). At 6.6 GPa, the residual resistivity below the superconducting transition increases to the value that is about half of the normal-state value. This should be induced by the solidification of the liquid PTM at high pressures and low temperatures, which will be discussed in detail below.

Figure 3 shows the pressure dependences of $T_c^{\text{onset},1}$ and $T_c^{\text{onset},2}$ obtained from the above resistivity measurements on the #1 and #2 samples under high pressures. The positive pressure effect on T_c can be visualized clearly, and the onset of T_c is raised up monotonically from below 20 K at ambient to above 30 K at 12.1 GPa without showing any sign of saturation. This means that the T_c of these nickelate superconductors can be further enhanced by higher pressures, and our present study has thus pushed the T_c limit of superconducting nickelates to over 30 K. A closer inspection of $T_c(P)$ for #1 reveals a slight slope reduction around ~ 5 GPa from $dT_c/dP \approx +1.28$ K/GPa to $+0.93$ K/GPa, see the dashed lines in Fig. 3. As discussed below, such a subtle change of dT_c/dP might arise from the degradation of the thin-film sample in the presence of stress/strain upon compression.

To further confirm the superconducting transitions and also to characterize the superconducting properties of the $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin films under pressure, we studied the effect of magnetic fields on resistivity at each fixed pressure. Fig. 4(a) shows the $\rho(T)$ curves under various magnetic fields at ambient pressure. All $\rho(T)$ data under different pressures for #1 up to 12.1 GPa and for #2 up to 6.6 GPa are shown in Fig. S2 and S3, respectively. As seen in Figs. 4(a), S2 and S3, the superconducting transition is gradually suppressed to lower temperatures and the transition width is broaden up with increasing magnetic fields. Here, we used the criteria of 50% ρ_n to define the superconducting transition temperature as T_c^{mid} , and plotted the temperature dependences of $\mu_0 H_{c2}(T_c^{\text{mid}})$ in Fig. 4(b). Then, we can estimate the zero-temperature upper critical field $\mu_0 H_{c2}(0)$ by employing the empirical Ginzburg-Landau (G-L) formula, i.e., $\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0)(1-t^2)/(1+t^2)$, where $t = T/T_c$ represents the reduced temperature. The fitting results are indicated by the broken lines in Fig. 4(b). Unlike the monotonic enhancement of $T_c(P)$, the $\mu_0 H_{c2}^{\text{GL}}(0)$ exhibits a non-monotonic evolution with pressure. As show in Fig. 4(c), the obtained $\mu_0 H_{c2}^{\text{GL}}(0)$ first increases quickly from ~ 36 T at 0 GPa to 76 T at 4.6 GPa with a slope of about 8.7 T/GPa, and then it decreases nearly linearly to ~ 62.3 T with a smaller slope of -1.8 T/GPa up to 12.1 GPa. As discussed below, the reduction of $\mu_0 H_{c2}^{\text{GL}}(0)$ above 4.6 GPa might be correlated with the degradation of the superconducting state. However, in the plot of normalized $\mu_0 H_{c2}(T)/\mu_0 H_{c2}^{\text{GL}}(0)$ versus $T_c^{\text{mid}}/T_c^{\text{mid}}(0)$, Fig. 4(d), all the data at various pressures almost fall onto one single straight line, which indicates that the dominant pair-breaking effect actually remains unchanged under high pressures [57].

Before discussing the effect of pressure on T_c , it is necessary to first clarify the influence of pressure conditions on the nickelate thin-film samples. As we known, the liquid PTM will be solidified under pressure and/or upon cooling down, which would unavoidably produce some stress/strain on the sample. Considering the delicate nature of these nickelate thin films, this effect should be particularly severe and degrade the superconducting properties gradually. For the glycerol and Daphne 7373 used in the present study, they were reported to solidify above ~ 6 GPa and ~ 2.4 GPa at room temperature [58, 59]. This explains why the normal-state and residual resistivity experience an anomalous enhancement at $P > 6$ GPa for #1 and at $P > 2.4$ GPa for #2. Meanwhile, the accumulation of the stress/strain accompanying the solidification of PTM under high pressures would lead to the gradual degradation of the thin film and thus the superconducting transition. This explains the observed broaden-up of the superconducting transition with a long tail as well as the gradual reduction of the resistivity drop at the transition at $P > 4.6$ GPa for #1 and at $P > 2.4$ GPa for #2. In addition, the obvious degradation of the superconducting transition by stress/strains can also explain the observed slope change of $T_c(P)$ in Fig. 3 and the anomalous reduction of $\mu_0 H_{c2}(0)$ shown in Fig. 4(c). If this is the case, $T_c(P)$ might be raised more efficiently and reach a higher T_c than 31 K at 12.1 GPa under a better hydrostatic pressure condition.

To verify the above hypothesis about the influence of stress/strain, we performed comparative high-pressure resistivity measurements on the $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin film (#3) in CAC by using the solid PTM, h-BN, which is expected to produce a stronger stress/strain than the liquid one. As shown in Fig. S4, the

$\rho(T)$ is immediately altered to an insulating-like behavior under pressure of 2 GPa, and the magnitude of resistivity increases significantly with further increasing pressure to 4 GPa. This comparison also highlights that the liquid PTM, though solidified under pressure, remains relatively soft so that it can preserve the metallic behavior and allows us to see the evolution of the superconducting transition.

After excluding the extrinsic factors, we can briefly discuss the positive pressure effect on $T_c(P)$ of $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin films, which is the main finding of the present work. First, the contraction of the lattice parameter under pressure should favor a higher T_c when comparing to the effect of chemical pressure with nearly similar optimal hole doping. For the reported nickelate thin films, the in-plane lattice constants $a \sim 3.91$ Å seems to be locked by the SrTiO_3 substrate [14, 41–43], but the c -axis constant shows an inverse correlation to the optimal T_c , i.e. $T_c = 9$ K for $\text{La}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ ($c \approx 3.44$ Å) [41], $T_c = 10$ K for $\text{La}_{0.82}\text{Ca}_{0.18}\text{NiO}_2$ ($c \approx 3.385$ Å) [42], $T_c = 11$ K for $\text{Nd}_{0.775}\text{Sr}_{0.225}\text{NiO}_2$ ($c \approx 3.375$ Å) [14, 16, 17], and $T_c = 14$ K as for $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ ($c \approx 3.36$ Å) [43]. If this trend is followed under physical pressure, the contraction of c -axis under pressure should lead to an enhancement of T_c . Secondly, we notice that the initial slope of $\mu_0 H_{c2}(T)$ in Fig. 4(b) experiences an obvious enhancement from 0 to 2.5 GPa, when we believe that the influence of stress/strain is negligible. Based on the single-band model, the initial slope of $\mu_0 H_{c2}(T)$ is related to the effective mass m^* of charge carriers via the relationship of $-(1/T_c)[d\mu_0 H_{c2}/dT]_{T_c} \propto (m^*)^2$ [57, 60]. From a linear fitting to $\mu_0 H_{c2}(T)$, we can extract the normalized slope $-(1/T_c)[d\mu_0 H_{c2}/dT]_{T_c}$, which increases from ~ 0.2 T/K² at 0 GPa to ~ 0.23 T/K² at 2.5 GPa, Fig. 4(c). Such a pressure-induced enhancement of effective mass m^* or the electron correlations should also contribute to the observed positive pressure effect on T_c . Further theoretical studies on the electronic structures under pressure are needed to have a better understanding on the importance of electronic correlations, hybridization between Ni-3*d* and rare-earth 5*d* electrons, etc.

Conclusion

In summary, we have performed a high-pressure study on the superconducting $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin films by employing the cubic anvil cell apparatus. Our results reveal that its T_c increases by ~ 13 K from ~ 18 K at 0 GPa to ~ 31 K at 12.1 GPa without showing any signature of saturation. It means that there is still much room for further raising the T_c of the superconducting nickelates. This finding is encouraging and should promote more studies to further raise the T_c of superconducting nickelates. We have also explained the positive pressure effect on $T_c(P)$ in terms of the lattice contraction and mass enhancement. In addition, the influence of stress/strain under pressure is also discussed.

Declarations

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Figures

Figure 1

(a) Temperature dependence of resistivity $\rho(T)$ of Pr_{0.82}Sr_{0.18}NiO₂ thin film (#1) under various pressures up to 12.1 GPa by using the glycerol PTM. Inset of (a) shows the crystal structure of PrNiO₂.

(b) Variation of the superconducting transition under high pressures. (c) Variation of normalized resistivity $\rho/\rho(35\text{K})$.

Figure 2

(a) Temperature dependence of resistivity $\rho(T)$ of $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{NiO}_2$ thin film (#2) under various pressures up to 6.6 GPa by using the Daphne 7373 PTM. (b) Variation of the superconducting transition under high pressures. (c) Variation of normalized resistivity $\rho/\rho(30\text{ K})$.

Figure 3

Temperature-pressure phase diagram of $\text{Pr}_{0.82}\text{Sr}_{0.18}\text{Ni}_2$ thin films. Pressure dependences of the superconducting transition temperatures $T_{c, \text{onset}, 1}$ and $T_{c, \text{onset}, 2}$ are determined from the $\rho(T)$.

Figure 4

(a) Temperature dependences of the resistivity with magnetic fields up to 9 T at 0 GPa. (b) Temperature dependences of the upper critical field $\mu_0 H_{c2}$ at different pressures, where $\mu_0 H_{c2}$ values are determined by using the criteria 50% pn. The broken lines represent the Ginzburg–Landau (GL) fitting curves. (c) Pressure dependence of the zero-temperature upper critical field $\mu_0 H_{c2}(0)$ obtained from the empirical GL fitting and the normalized slope $-(1/T_c)[d\mu_0 H_{c2}/dT]|T_c$. (d) Normalized T/T_c dependence of normalized $\mu_0 H_{c2}(T)/\mu_0 H_{c2}^{GL}$ under various pressures.

Supplementary Files

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