# Evolution of the metallic state in LaNiO$_3$/LaAlO$_3$ superlattices measured by $^8$Li $\beta$-detected NMR

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Using ion-implanted $^8$Li $\beta$-detected NMR, we study the evolution of the correlated metallic state of LaNiO$_3$ in a series of LaNiO$_3$/LaAlO$_3$ superlattices as a function of bilayer thickness. Spin-lattice relaxation measurements in an applied field of 6.55 T reveal two equal amplitude components: one with metallic ($T$ linear) $1/T_1$ and a second with a more complex $T$ dependence. The metallic character of the slow relaxing component is only weakly affected by the LaNiO$_3$ thickness, while the fast component is much more sensitive, exhibiting the opposite temperature dependence (increasing toward low $T$) in the thinnest, most magnetic samples. The origin of this bipartite relaxation is discussed in terms of electronic phase separation.

## I. INTRODUCTION

The perovskite rare-earth metal nickelates ($R$NiO$_3$) are an important example of a metal-insulator transition (MIT) in a strongly correlated system [1]. The MIT can be tuned by increasing the size of the rare-earth metal cation $R^{3+}$ which increases the Ni-O-Ni angle [1], and hence the electron hopping integral and conduction bandwidth. By this means, the MIT is suppressed to $T = 0$ K between $R =$ Pr and La. The prevailing view is that LaNiO$_3$ is a conventional metal characterized by a $T^2$ resistivity [2] with some evidence of strong correlations such as enhanced carrier mass and magnetic susceptibility. However, in superlattices (SLs) with insulating interlayers of LaAlO$_3$, it can be driven insulating and antiferromagnetic when very thin [2 unit cells (u.c.)] [3,4], clearly demonstrating that LaNiO$_3$ remains close to quantum critical. This motivated the current experiment to better understand the correlated metallic state and how it changes as it approaches the thickness-controlled MIT.

NMR is an essential tool for studying strongly correlated electron materials, such as the high-$T_c$ cuprates [5]; however, weak signals generally preclude its use for thin films, with a few important exceptions [6–11]. An alternative is $\beta$-detected NMR ($\beta$-NMR) [12], where a spin-polarized

radioisotope is implanted into the sample and the resulting $\beta$-decay is used to detect the NMR similar to muon spin rotation ($\mu$SR). $\beta$-NMR is a powerful local probe of metals [12], including strongly correlated Sr$_2$RuO$_4$ [13] and LaNiO$_3$ [14].

Here, $\beta$-NMR is used to study the evolution of the metallic state of LaNiO$_3$ with decreasing thickness in a series of LaNiO$_3$/LaAlO$_3$ SLs. Spin-lattice relaxation (SLR) measurements reveal two components: one with $T$ linear relaxation below 200 K and a second with a more complex $T$ dependence, indicating LaNiO$_3$ is not microscopically homogeneous but instead is phase separated. The slow relaxing component retains a metallic character similar to bulk LaNiO$_3$, independent of thickness. In contrast, the fast relaxing component depends on thickness, deviates strongly from metallic, and exhibits a low $T$ upturn, consistent with the emergence of magnetic ordering.

## II. SAMPLES

Using pulsed laser deposition, the SLs were deposited on (LaAlO$_3$)$_{0.3}$(SrAlTaO$_6$)$_{0.7}$ (LSAT) substrates and annealed in an O$_2$ rich atmosphere, similar to the methods of Ref. [3]. The deposition begins with $n(=2, 3, 4.10)$ u.c. of LaNiO$_3$ followed by an insulating interlayer of 2 u.c. of LaAlO$_3$. The stacking sequence ($n$‖2) was repeated 20 times for each SL (see Table I), terminating with LaAlO$_3$. The crystallinity, interface sharpness, and layer thicknesses were verified using x-ray diffraction. Prior to the $\beta$-NMR experiments, the samples ($5 \times 5 \times 0.5$ mm$^3$) were mounted on Al$_2$O$_3$ crystals with Ag paint for compatibility with the spectrometer's cold-finger cryostat. For further details on the sample characterization and growth, see the Supplemental Material [15].

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TABLE I. Summary of the superlattice samples.

| $n$ | Composition | Bilayer thickness | Total thickness |
|---|---|---|---|
| 10 | $[(LaNiO_3)_{10}(LaAlO_3)_2]_{20}$ | 4.6(5) nm | 92(1) nm |
| 4 | $[(LaNiO_3)_4(LaAlO_3)_2]_{20}$ | 2.6(5) nm | 52(1) nm |
| 3 | $[(LaNiO_3)_3(LaAlO_3)_2]_{20}$ | 2.0(5) nm | 40(1) nm |
| 2 | $[(LaNiO_3)_2(LaAlO_3)_2]_{20}$ | 1.5(5) nm | 30(1) nm |

## III. METHODS

To measure the $\beta$-NMR, a spin-polarized radioactive $^8Li^+$ ion beam was implanted into the sample. The measured $\beta$-decay asymmetry is proportional to the average longitudinal nuclear spin polarization [12,16], with a proportionality constant $A_0$ depending on the detector geometry and decay properties. The asymmetry was obtained by combining the count rates in two opposing scintillation detectors placed along the polarization axis. The experiments were performed at the ISAC facility at TRIUMF in Vancouver, Canada [12]. The NMR isotope $^8Li$ has nuclear spin $I = 2$, gyromagnetic ratio $\gamma/2\pi = 6.3016$ MHz T$^{-1}$, nuclear electric quadrupole moment $Q = +32.6$ mb, and radioactive lifetime $\tau = 1.21$ s. The implantation energy was 4.9 keV, corresponding to a mean depth of $\sim$21 nm with a straggle of 11 nm [17].

To measure the SLR, the asymmetry was monitored both during and after the 4 s beam pulse, during which it approaches a dynamic steady-state, while afterward it relaxes to near zero. Unlike conventional NMR, the $^8Li$ is hyperpolarized in flight by optical pumping, so no radio frequency (RF) field is required. The SLR rates were measured from 5 to 300 K in an applied field of $B_0 = 6.55$ T normal to the surface.

## IV. RESULTS

Figure 1 compares the SLR data for different SLs. The relaxation is easily measurable in all cases. Generally, we expect it to be fastest in the (presumably most magnetic) $n = 2$ SL, and slower for the more metallic samples (larger $n$). To quantify its evolution, a model relaxation function is required. The simplest choice that provides a good fit is a biexponential, to which we add a nonrelaxing term to account for a small fraction of $^8Li$ stopping in the substrate due to range straggling. Specifically, at time $t$ after an $^8Li$ arriving at time $t'$, the polarization follows

$$R(t,t') = f_{SL}[(1 - f_f)e^{-\lambda_s(t-t')} + f_f e^{-\lambda_f(t-t')}] + (1 - f_{SL}) \tag{1}$$

where $\lambda_i$ ($i = s, f$) are the SLR rates. $f_{SL}$ is the fraction of $^8Li$ in the SL, while $(1 - f_{SL})$ is in the substrate. Within the SL, $f_f$ is the fast relaxing fraction and $(1 - f_f)$ is slow relaxing. The only $T$ dependent parameters in the fits were the two rates. Biexponential relaxation appears intrinsic to $^8Li$ in LaNiO$_3$, and a similar analysis was necessary for a bulk single crystal [14]. Interestingly, the fast relaxing fraction $f_f = 50\%$ is independent of LaNiO$_3$ thickness, and in agreement with the bulk
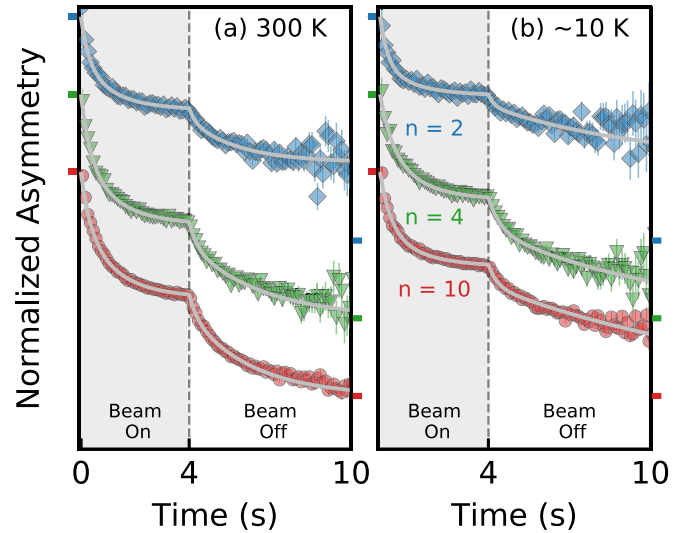


FIG. 1. Examples of $^8Li^+$ SLR data in $[(LaNiO_3)_n(LaAlO_3)_2]_{20}$ with $B_0 = 6.55$ T at two temperatures for $n = 2, 4$ and 10. The lines are fits to Eq. (1) convoluted with the 4 s beam pulse indicated by the shaded area. The data has been normalized and for clarity, offset vertically. The colored $y$-axis ticks on the left (right) represent the full (zero) asymmetry for each SL.

[14]. For further details on the analysis, see the Supplemental Material [15] (and Refs. [18–23] therein).

The resulting rates $(1/T_1)_i \equiv \lambda_i$ are shown as a function of temperature in Fig. 2. The slow rate in Fig. 2(a) remains linear (metallic) for all the SLs, with a slope similar to bulk LaNiO$_3$ [14]. Closer inspection reveals that (1) above $\sim$200 K (marked by the vertical arrow), $1/T_1$ becomes sublinear; (2) the SLs are all faster relaxing than the bulk, and this enhancement is mostly due to a finite $n$-independent intercept as $T \to 0$; and (3) the slope changes systematically with $n$, but not monotonically, as the shallowest slope is at $n = 4$. Below 200 K, we fit $(1/T_1)_s$ to a line to obtain the values shown as a function of $n$ in Fig. 3.

In contrast, the fast rate in Fig. 2(b) is much more strongly enhanced over the bulk (by a factor of 3–5 at 200 K). Similarly, there is a linear region (above $\sim$100 K), with a slope not far from the bulk, but this linearity does not persist to low $T$. Instead, it shows an upturn below 50 K, which is most dramatic at $n = 2$. At the MIT, we expect to lose the metallic Korringa relaxation, but as in other $R$NiO$_3$, the insulating state contains Ni local moments. The increasing SLR at low $T$ is consistent with relaxation from fluctuations of the Ni spins which slow and eventually freeze.

Remarkably, we find no obvious signal from the LaAlO$_3$. Like LSAT [24], the relaxation in bulk LaAlO$_3$ is extremely slow [25]. However, the LaAlO$_3$ is very thin, and any $^8Li$ at the interface will retain some coupling to the adjacent LaNiO$_3$, enhancing $1/T_1$ compared to the bulk and making the distinctly LaAlO$_3$ volume fraction rather small. The $^8Li^+$ site energy in the LaAlO$_3$ layer may also be higher than LaNiO$_3$ due to the higher valent Al$^{3+}$, resulting in a bias against stopping in this layer. In the thin SLs, the LaAlO$_3$ fraction may be included in the substrate term, while in the
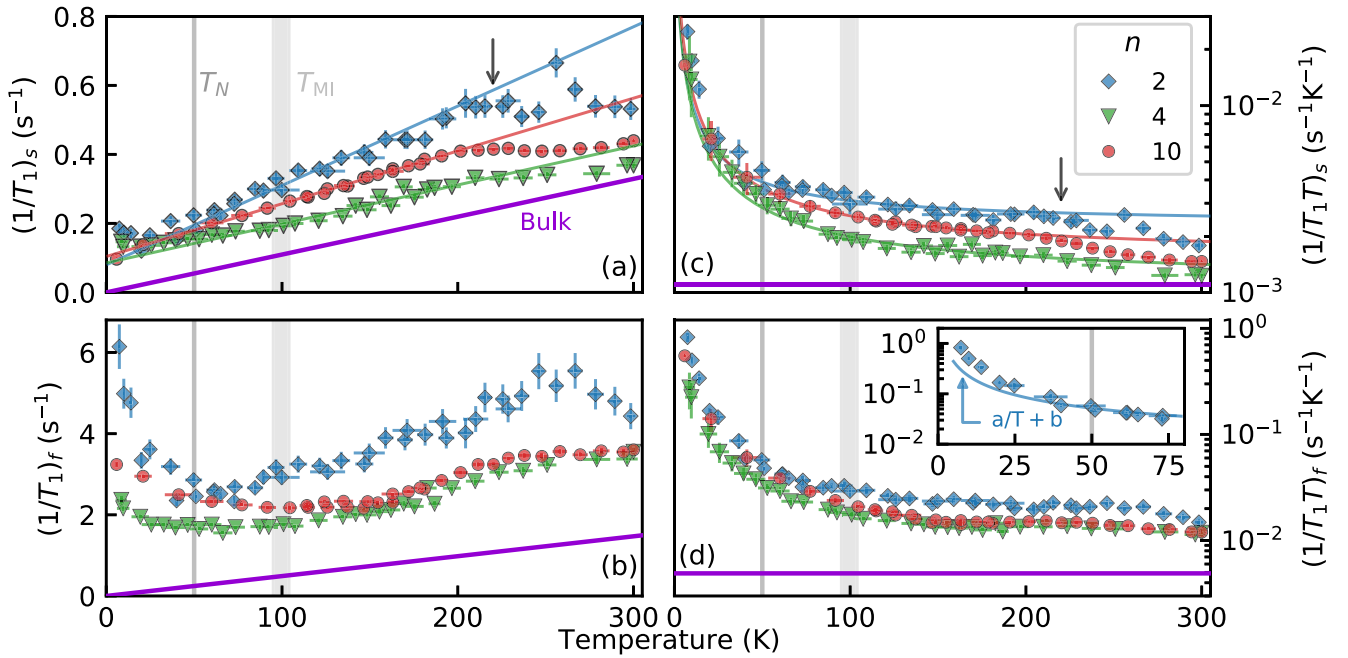
FIG. 2. The slow [(a), (c)] and fast [(b), (d)] $^8$Li SLR rates and $(T_1 T)^{-1}$ as a function of temperature at $B_0 = 6.55\,\text{T}$ in $[(\text{LaNiO}_3)_n (\text{LaAlO}_3)_2]_{20}$ for $n = 2$, 4, and 10. Below $\sim 200\,\text{K}$, $(1/T_1)_s$ appears linear with a nonzero intercept (fits shown with solid lines). The vertical arrow at $\sim 200\,\text{K}$ indicates the sublinear deviation. The shaded region and the vertical line illustrate the reported $T_{\text{MI}}$ and $T_N$ in an $n = 2$ SL on SrTiO$_3$ [3]. For reference, the bulk Korringa slope is shown as the solid purple line [14]. The inset of panel (d) illustrates the low $T$ upturn for $n = 2$, which spans more than an order of magnitude. The data for the $n = 3$ SL has been omitted to avoid clutter; however, the trend is very similar to $n = 2$.

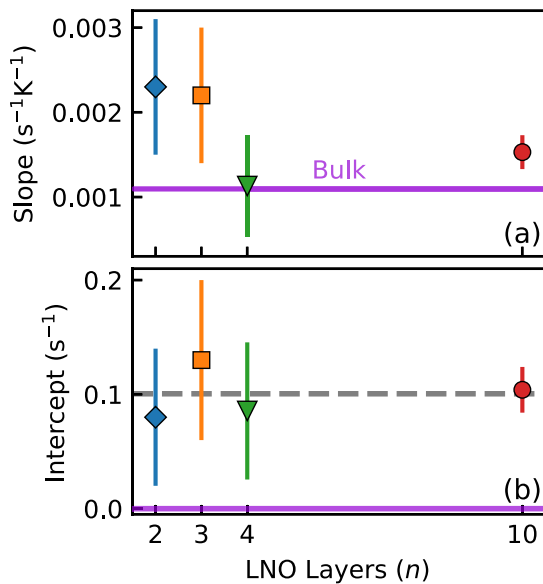thicker SLs it may make a small contribution to the slow component.



FIG. 3. (a) Slopes from the linear region of the slow component as a function of the LaNiO$_3$ thickness $n$. The bulk slope is shown as the solid purple line. The $n = 4$ and 10 values are scattered around the bulk, while the thinner SLs ($n = 2$ and 3) are significantly enhanced. (b) The $T \rightarrow 0$ intercepts appear independent of $n$, with average value shown by the dashed gray line.

## V. DISCUSSION

Having outlined the results, we now discuss their implications. Like the implanted muon in $\mu$SR, $^8$Li$^+$ is typically located at a high-symmetry interstitial site in the host lattice. In perovskite oxides, this site (denoted $P$) is midway between adjacent $A$-site ions at the center of a square $B$O plaquette [14,24,26]. Analogous to the implanted muon and native NMR nuclei, $^8$Li is coupled via the hyperfine interaction to the electronic system of the host. However, it is much longer lived than the muon, making it sensitive to phenomena on longer timescales such as Korringa relaxation.

With only one site, biexponential relaxation is unexpected. In bulk LaNiO$_3$, where both rates are $T$ linear, we attributed it to two subtly different $P$ sites in an unconventional distorted perovskite structure [14]. This was reasonable, since the rates differed only in magnitude, not temperature dependence. However, it is incompatible with the present data where they exhibit distinct $T$ dependences. With no evidence for other sites, we instead propose that the electronic state of LaNiO$_3$ (even in a metallic single crystal) is *intrinsically inhomogeneous*, i.e., microscopically separated into two equally abundant phases with distinct electronic properties. Electronic phase separation is prevalent in many correlated electron conductors [27], including stripe order in the closely related (but 2D) hole-doped La$_2$NiO$_4$ [28]; however, this is the first evidence for it in a 3D perovskite metal.

Separation into distinct metallic states is rather uncommon, but it does occur in the Kondo lattice compound YbRh$_2$Si$_2$, where $^{29}$Si spin-lattice relaxation becomes biexponential near the field-induced quantum critical point [29]. Interestingly,

here, the phase fraction (and not the relaxation rate) evolves with magnetic field and temperature, but converges toward $f_f = 0.5$ ($R = 1$ in their notation) at the lowest temperatures. LaNiO$_3$ has been suggested to have a Kondo lattice character with a much larger energy scale [30]. The phase separation revealed in our data persists at least up to 300 K, even in the single crystal. So, unlike YbRh$_2$Si$_2$, where the phenomenon is limited to low $T$, we expect lattice dynamics to have some influence.

In contrast, phase separation into metallic and insulating phases is known in other $R$NiO$_3$ [31,32], but only in the vicinity of the MIT. There, the insulating volume fraction varies from 0 to 1 through a narrow coexistence region around the transition. In contrast, our volume fraction is constant over the whole temperature range, including in the metallic limit of the single crystal [14]. If this phenomenon is related, then the metal-insulator phase separation must reflect some underlying inhomogeneity present in the high-temperature metallic phase. This is likely structural in origin. This is observed, for example, in NdNiO$_3$, where the formation of insulating regions is correlated with step-edges in the sample and the microstructure of this phase separation is stable through temperature cycling [31]. Candidate sources of structural heterogeneity are competing distorted perovskite structures (octahedral rotation pattern) or bond disproportionation (octahedral size) [1,33]. We note the precise structure of LaNiO$_3$ remains elusive. Although it is considered rhombohedral, recent neutron scattering results indicate a lower local symmetry [34,35]. It has also been suggested to contain nanoscopic monoclinic insulating pockets [34], which is inconsistent with the two distinct metallic phases we find in bulk [14].

In summary, the biexponential relaxation strongly suggests *microscopic electronic phase separation* in LaNiO$_3$ that persists up to at least 300 K. The phase separation is static on the second timescale, robust across multiple LaNiO$_3$ samples, including a single crystal [14], thin film [14], and SLs, and has not yet been observed with other experimental techniques. Confirming it may require other local probes, such as conventional NMR, or scanning probe methods that could directly image its spatial distribution [32].

In similar SLs, low energy $\mu$SR (in zero and low fields) finds a broad distribution of static fields (150 G width) below 50 K at $n = 2$ which pervade the entire sample volume [3], seemingly inconsistent with our results. However, $1/T_1$ does not probe the static fields directly, instead being determined by field fluctuations at the Larmor frequency (41.27 MHz). On the other hand, static fields could propagate out from an incomplete magnetic volume fraction into the nonmagnetic phase, provided the two were intimately mixed at the nanoscale [36]. This may explain why the scale of the internal fields is more than $10\times$ smaller than bulk $R$NiO$_3$ [37]. Alternatively, in analogy with YbRh$_2$Si$_2$, we may have stabilized a distinct phase-separated ground state with the large $B_0$ field.

Now we turn to what the evolution of $1/T_1$ can tell us about the MIT at small $n$. In a conventional metal, the Korringa $T$ linear dependence is a consequence of Fermi statistics and a density of states (DOS) that is smooth and featureless on the scale of $kT$. The Korringa slope is then proportional to the square of the DOS at the Fermi level $\rho(E_F)$. Considering

Fig. 3(a), there is a twofold increase in the slow component's slope for $n < 4$, consistent with an effective narrowing of the conduction band on approaching the 2D limit. However, for all $n$, we find a finite intercept, inconsistent with simple metallic behavior. More generally, for a correlated metal susceptible to magnetic order, the Moriya expression [38] relates $1/T_1$ to the imaginary part of the generalized susceptibility $\chi(\boldsymbol{q}, \omega)$,

$$\frac{1}{T_1 T} = \frac{4 k_B}{\hbar} \sum_{\boldsymbol{q}} \frac{|A(\boldsymbol{q})|^2}{(\gamma_e \hbar)^2} \frac{1}{\hbar \omega_0} \chi''(\boldsymbol{q}, \omega_0), \qquad (2)$$

where $\gamma_e$ is the electron gyromagnetic ratio, $\omega_0$ is the NMR frequency, and $A(\boldsymbol{q})$ is the hyperfine form factor which acts as a filtering function on magnetic fluctuations at wave vector $\boldsymbol{q}$. For a conventional metal, the summation in Eq. (2) is independent of $T$ for $kT \ll E_F$. A finite intercept implies that, in addition to a constant part, the sum contains a term proportional to $1/T$. This is seen clearly in the corresponding plot of $1/(T_1 T)$ (on a log scale) in Fig. 2(c). Such a Curie-like term suggests a population of dilute uncoupled local moments, as has been considered near the MIT in doped semiconductors [39]; however, we find no evidence for the emergence, at low temperature, of a broad distribution of $1/T_1$ that should arise from dilute spins. Interestingly, this term appears independent of $n$, so it is probably not caused by Ni moments at the LaAlO$_3$ interface.

Aside from the intercept, the slow relaxing phase exhibits a metallic character that is not very strongly modified from the bulk, even for the smallest $n$. In contrast, the fast relaxing phase exhibits a much more dramatic sensitivity to thickness. In the bulk, the factor of 4 between the Korringa slopes in the fast and slow phases [14] would imply a factor of 2 in $\rho(E_F)$ in an uncorrelated metal. In the SLs, compared to the slow component, $(1/T_1)_f$ shows a $10\times$ larger $T$ independent term (vertical shift) and an upturn below $\sim$50 K with $(1/T_1)_f$ doubling between 50 K and 4 K at $n = 2$ [inset of Fig. 2(d)]. Similar $n = 2$ SLs have a zero-field Néel transition at $T_N \approx 50$ K [3,4] near the upturn and an ordering wave vector $\boldsymbol{q}_{\text{AF}} = (1/4, 1/4, 1/4)$ [4]. The $P$ site hyperfine form factor $A(\boldsymbol{q}_{\text{AF}}) \neq 0$, so $^8$Li senses fluctuations at $\boldsymbol{q}_{\text{AF}}$. However, even the thicker SLs with $n = 4$ and 10 (that are thought to remain nonmagnetic and metallic) show a similar, albeit muted feature. The upturn implies a term in Eq. (2) increasing more steeply than $1/T$ at low $T$. This is likely related to magnetic ordering, but probably modified by the strong applied field (cf. the transition in Mn-doped Bi$_2$Te$_3$ [40]). Note the vertical scale in Fig. 2(d) spans an order of magnitude more range than the slow component in Fig. 2(c).

Finally, we return to the sublinearity in $(1/T_1)_s$ above 200 K (vertical arrow in Fig. 2). The deviation is subtle, sample dependent, and does not vary monotonically with $n$, as it is smallest at $n = 4$. A similar deviation was observed in bulk LaNiO$_3$, although there it had the opposite sense for the slow and fast components. This feature is probably related to small changes in the Ni−O−Ni angle around 200 K [34], which also coincides with a decrease in the carrier density from RF conductivity [35]. In addition, the magnetic susceptibility crosses over from a low temperature Pauli-like regime to a high temperature Curie-Weiss dependence [41] at about

200 K. It is reasonable to expect such a small structural change would differ between a crystal, SL, and film, and it may even depend on the precise thermal history.

## VI. CONCLUSION

$\beta$-NMR spin-lattice relaxation measurements of implanted $^8$Li in a series of LaNiO$_3$/LaAlO$_3$ SLs reveal biexponential relaxation, consistent with single crystal LaNiO$_3$ [14]; but, in contrast, the two components exhibit very different temperature dependences. Thus, we propose LaNiO$_3$ is electronically phase separated. The slow relaxing phase is metallic, not strongly modified from the bulk, and persists to low $T$, even for the thinnest ($n = 2$) SL. As $n \to 2$, the slope and the

DOS are enhanced consistent with band narrowing toward the 2D limit. In contrast, the fast relaxing phase is much more sensitive to $n$ and appears magnetic at low $T$. Any microscopic understanding of the electronic properties of LaNiO$_3$ and its MIT in SLs must take this phase separation into account.

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