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ABSTRACT

We report measurements of the Meissner screening profile in a Nb(300 nm)/Al₂O₃ thin film using ^8Li β -detected nuclear magnetic resonance (β -NMR). The NMR probe ^8Li was ion-implanted into the Nb film at energies ≤ 20 keV, corresponding to mean stopping depths comparable to Nb's magnetic penetration depth λ . ^8Li 's strong dipole-dipole coupling with the host ^{93}Nb nuclei provided a "cross-relaxation" channel that dominated in low magnetic fields, which conferred indirect sensitivity to the local magnetic field via the spin-lattice relaxation (SLR) rate $1/T_1$. From a fit of the $1/T_1$ data to a model accounting for its dependence on temperature, magnetic field, and $^8\text{Li}^+$ implantation energy, we obtained a magnetic penetration depth $\lambda_0 = 51.5(22)$ nm, consistent with a relatively short carrier mean-free-path $\ell = 18.7(29)$ nm typical of similarly prepared Nb films. The results presented here constitute an important step toward using ^8Li β -NMR to characterize bulk Nb samples with engineered surfaces, which are often used in the fabrication of particle accelerators.

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I. INTRODUCTION

The transition metal Nb is a type-II superconductor with a transition temperature $T_c \approx 9.25$ K¹ that is highest among the pure elements. Accompanying this accolade is the highest lower critical field $B_{c1} \approx 170$ mT¹ of any known superconductor, making Nb

uniquely suited for applications where superconductivity is required in the presence of (relatively) large magnetic fields. One such example is its use in superconducting radio frequency (SRF) cavities,^{2,3} which make up many accelerator beamlines (for charged particles) across the globe. In the pursuit of optimal accelerating

efficiency and cryogenic operation economy, there is great interest in *engineering* Nb to remain in the Meissner state up to the highest magnetic fields possible. This can be achieved empirically by chemically modifying its surface through baking^{4,5} or doping,^{6,7} as well as by coating it with thin layers of other superconducting (and insulating) materials.^{8–13}

To better understand how these microscopic modifications affect Nb's superconducting properties, an experimental technique capable of interrogating the *local* electromagnetic fields in the near surface region is essential. Similarly, as the material properties are expected to vary below the surface, *spatial* resolution is also a requirement. In reality, there are few experimental techniques capable of simultaneously satisfying these criteria. Perhaps, the most widely known is low-energy muon spin rotation (LE- μ SR),^{14,15} which uses implanted positive muons μ^+ as subsurface magnetic probes. Indeed, LE- μ SR has been used to study Meissner screening in Nb, revealing: the importance of non-local electrodynamics^{16,17} and strong-coupling corrections¹⁸ in “clean” samples;¹⁹ the dependence of the magnetic penetration depth on film preparation technique;²⁰ as well as an “anomalous” modification to the screening profile for select surface treatments.²¹ As the latter result is controversial,^{22,23} it is imperative that alternative means of study be developed to corroborate such findings. To this end, here we employed a lesser-known (but closely related) technique—ion-implanted β -detected nuclear magnetic resonance (β -NMR)—to study the superconducting properties of Nb below its surface. Thorough accounts of the β -NMR technique can be found in several recent^{24,25} and older^{26–31} review articles, with additional details given in several technical overviews.^{32–34}

In ion-implanted β -NMR,^{24,25} a radioactive spin probe is “inserted” into a material of interest and its nuclear magnetic resonance (NMR) response is monitored through the anisotropic property of β -decay (see, e.g., Ref. 35), analogous to the detection scheme used in LE- μ SR.^{14,15,36} Just as in “conventional” NMR using stable nuclei (see, e.g., Refs. 37–40), β -NMR is sensitive to the electronic and magnetic properties of the “host” material; however, its sensitivity (per nucleus) is greater by a factor of $\sim 10^{10}$, allowing for the study of situations typically inaccessible by NMR (e.g., thin films,^{41–44} heterostructures,^{45–48} or isolated impurities^{49,50}). Supplementing this feature is the ability to control the implantation energy that the probe radioisotope is ion-implanted, allowing for it to be “placed” at specific depth ranges. This further extends the types of problems that can be studied, especially those where *depth-resolution* is essential. For example, the depth-resolution afforded by ^8Li β -NMR has been used to investigate: molecular dynamics in glassy polystyrene;^{51–53} the magnetism of Mn_{12} single-molecule magnets;⁵⁴ surface magnons in antiferromagnetic Fe_2O_3 ;⁵⁵ finite size effects in Pd thin films;⁴³ as well as the superconductor NbSe_2 in both the vortex⁵⁶ and Meissner⁵⁷ states.

While LE- μ SR and β -NMR are largely complementary techniques,⁵⁸ differences in probe properties (e.g., radioactive lifetime) make β -NMR much more amenable to the measurement of phenomena transpiring over longer timescales, such as spin-lattice relaxation (SLR) (in non-magnetic materials). SLR is driven by stochastic fluctuations in the local electromagnetic field at the probe's stopping site, which dynamically “re-orient” the probe's spin, returning the polarization of the ensemble to ~ 0 (i.e., its thermal

equilibrium value). When the “host” contains an abundance of isotopes with non-zero nuclear spin, the implanted probe couples to the “bath” of host spins via the dipolar interaction, providing a relaxation “channel” with sensitivity to the static component of the local field. This form of “cross-relaxation” (see, e.g., Ref. 59) has been used to study the magnetic penetration depth in the multi-band superconductor NbSe_2 ,⁵⁷ as well as order-parameter fluctuations in superconducting Pb and Ag/Nb thin films,⁶⁰ by way of the β -NMR probe ^8Li . Here, we applied and extended this methodology to extract the Meissner screening profile in a $\text{Nb}(300\text{ nm})/\text{Al}_2\text{O}_3$ thin film. Specifically, in a 20 mT applied magnetic field, we measured the temperature and implantation energy (i.e., depth) dependence of implanted ^8Li 's SLR rate $1/T_1$, revealing a sharp increase in relaxation at temperatures below T_c , whose magnitude increases deeper below the material's surface. By contrast, this depth-dependence vanishes in the normal state, consistent with the expected signature for Meissner screening.^{57,60} From a fit of the data to a model accounting for $1/T_1$'s dependence on temperature, implantation energy, and magnetic field, we determined that the screening profile is well-described by a simple London model^{61,62} with a magnetic penetration depth exceeding Nb's “intrinsic” value. The result is consistent with similarly prepared films, known to possess relatively short carrier mean-free-paths.

The rest of the manuscript is organized as follows. Details of the β -NMR experiments are given in Sec. II, along with information on the preparation of our Nb sample (Sec. II B). Our results and analysis are presented in Sec. III, with a description of our data given in Sec. III A, followed by an account of our analysis approach (Sec. III B), and the modeling of the extracted results (Sec. III C). We discuss our findings and their implications in Sec. IV, and a summary is given in Sec. V.

II. EXPERIMENT

β -NMR experiments were performed at TRIUMF's isotope separator and accelerator (ISAC) facility in Vancouver, BC, Canada. The probe radioisotope ^8Li (nuclear spin $I = 2$; gyromagnetic ratio $\gamma/(2\pi) = 6.302\,21(7)\text{ MHz T}^{-1}$;^{63,64} electric quadrupole moment $Q = +31.4(2)\text{ mb}$;⁶⁵ radioactive lifetime $\tau_\beta = 1.2096(5)\text{ s}$;⁶⁶ and mass $m_{^8\text{Li}} = 8.022\,486\,24(5)\text{ u}$ ^{67,68}) was generated from an isotope production target (e.g., Ta foil stacks)⁶⁹ irradiated with a $\sim 500\text{ MeV}$ proton beam at a current on the order of $\sim 20\text{ }\mu\text{A}$. The target, heated in excess of $\sim 2000\text{ K}$ to accelerate thermal out-diffusion of ^8Li , was coupled to a surface ion source⁶⁹ and a mono-energetic $\sim 20\text{ keV}$ beam of $^8\text{Li}^+$ was extracted with an intensity of $\sim 10^7$ ions/s. A high-resolution magnetic dipole mass separator was used to ensure the isotopic purity of the beam, which was transported through a high vacuum beamline via electrostatic optics. During transport, the ion beam was neutralized in-flight and subsequently spin-polarized by collinear optical pumping with circularly polarized resonant laser light,³² yielding a high degree of nuclear spin-polarization ($\sim 70\%$ ^{70,71}). The highly polarized beam was then re-ionized and (electrostatically) steered to the spectrometer,^{24,33} where the probe $^8\text{Li}^+$ ions were implanted into the Nb thin film, which was affixed to a holder compatible with the spectrometer's cold-finger cryostat. The energy of the ion beam was controlled by biasing the spectrometer's platform to high positive voltage

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(up to 20 keV), electrostatically decelerating the $^8\text{Li}^+$ from their transport energy (within the final few centimeters of their trajectory) before implantation. Note that, in the lead-up to implantation, the ^8Li spin-polarization is parallel to the film's surface and the applied field, but perpendicular to the beam's momentum (see, e.g., Ref. 33).

An essential aspect of this approach is the spatial (i.e., depth) resolution obtained through ion-implanting our β -NMR probe ^8Li into Nb. Ion-implantation is a controlled, non-equilibrium means of introducing impurities in solids, which finds use (on an industrial scale), for example, in the electrical doping of semiconductors (see, e.g., Ref. 76). During implantation, the “injected” ion loses energy through its interaction with the host's electrons and nuclei, amounting to a series of (dampened) collisions that thermalize the projectile. The slowing process is stochastic, resulting in a *distribution* of stopping positions for a mono-energetic projectile. This process can be simulated accurately using Monte Carlo methods (see, e.g., Refs. 77 and 72), and the results from such simulations are used to connect the probe's NMR response with a spatial region of the sample, just as in LE- μSR .^{78,79} Several implantation profiles for 2 to 20 keV $^8\text{Li}^+$ stopping in Nb, simulated using the SRIM Monte Carlo code,^{72,73} are shown in Fig. 1.

A. Measurements

In each β -NMR measurement, the ^8Li spin-polarization was monitored after implantation through the anisotropy of its radioactive β -decay.³⁵ Specifically, the experimental asymmetry A (proportional

to the average longitudinal spin-polarization p_z) was measured by combining the β -rates in two opposed scintillation counters.²⁴ In our measurements, the sense of circular polarization (left and right) of the pumping laser light was also alternated,³² producing either “positive” or “negative” helicity in the $^8\text{Li}^+$ beam (i.e., its nuclear-spin polarization is aligned or counter-aligned with the beam direction). Data were collected separately for each helicity and then combined to remove detection systematics (see, e.g., Refs. 28 and 24). Explicitly, the well-known “four-counter” method was used,

$$A = \frac{1 - r}{1 + r} \equiv A_0 p_z, \quad (1)$$

where

$$r \equiv \sqrt{\frac{(L_+/R_+)}{(L_-/R_-)}}$$

is the geometric mean of the ratio of rates in the opposing counters L and R for the $\pm$ polarization senses. Note that the proportionality constant between A and p_z , denoted as A_0 in Eq. (1), depends on both the experimental geometry and the details of the β -decay (on the order of $\sim 10\%$ here).

SLR measurements were performed by monitoring the transient decay of spin-polarization both during and following a short pulse of beam of duration Δ (typically ~ 4 s). During the pulse, the

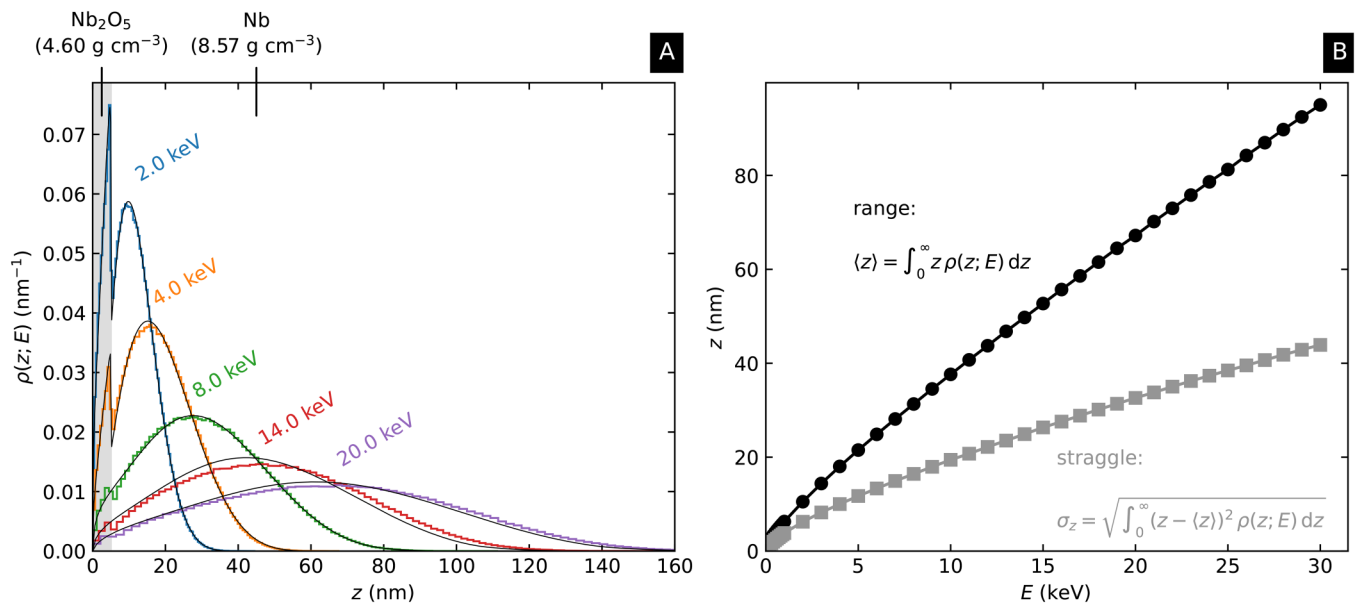


FIG. 1. Results from simulations of 10^6 $^8\text{Li}^+$ ions implanted in a $\text{Nb}_2\text{O}_5(5\text{ nm})/\text{Nb}$ target, calculated using the SRIM Monte Carlo code^{72,73} by aid of the Python package `pysrim`.⁷⁴ (a): Typical $^8\text{Li}^+$ stopping profiles at select implantation energies E are shown, represented here as histograms (with 100 bins) of the stopping probability $\rho(z; E)$ vs depth below the surface z . The thin black lines denote fits to a phenomenological model for $\rho(z; E)$ [Eqs. (15) and (16)—see Sec. III C], capturing all features of the stopping profiles at each energy. Note that the abrupt change in $\rho(z; E)$ at the $\text{Nb}_2\text{O}_5/\text{Nb}$ boundary is related to the different layer densities (indicated in the inset). The thickness of the surface pentoxide oxide layer (5 nm) is typical for Nb (see, e.g., Ref. 75). (b): Moments of the stopping profiles. As E increases, so does the $^8\text{Li}^+$ range and straggle (i.e., the mean, $\langle z \rangle$, and standard deviation, σ_z).

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polarization approaches a steady-state value, while after the pulse, it relaxes to essentially zero. At the edge of the pulse, there is a discontinuity in the slope, characteristic of β -NMR SLR data acquired in this manner.^{24,25} This bipartite behavior can be described quantitatively by considering a uniform distribution of probe arrival times (i.e., a time-independent $^8\text{Li}^+$ production rate). Explicitly, for an arrival time t' , the temporal evolution of nuclear spin-polarization at time $t > t'$ follows

$$p_z(t) = p_0 \times \begin{cases} \frac{\int_0^t \exp[-(t-t')/\tau_\beta] R(t', t') dt'}{\int_0^t \exp[-t'/\tau_\beta] dt'}, & t \leq \Delta, \\ \frac{\int_0^\Delta \exp[-(t-t')/\tau_\beta] R(t', t') dt'}{\exp[-(t-\Delta)/\tau_\beta] \int_0^\Delta \exp[-t'/\tau_\beta] dt'}, & t > \Delta, \end{cases} \quad (2)$$

where $p_0 \approx 70\%$ is the degree of nuclear spin-polarization produced from optical pumping,^{70,71} and $R(t, t')$ is a “relaxation” function describing the decay of p_z (e.g., an exponential in the simplest case). Note that, unlike in conventional NMR, no transverse radio frequency (RF) field B_1 is required for measurement of the SLR, meaning that there is no intrinsic spectral resolution of the relaxation, which represents the SLR of the *entire* ^8Li population (i.e., not just those with observable resonances). A typical SLR measurement required ~ 30 min, corresponding to $\sim 10^8$ total decay events.

B. Sample

A 300 nm Nb thin film from a previous β -NMR experiment⁸⁰ was employed in this study. The film was grown ($1 \text{ \AA s}^{-1}$ deposition rate) by RF sputtering a 99.9% Nb target (Goodfellow) onto an $8 \times 12 \times 0.5 \text{ mm}^3$ epitaxially polished Al_2O_3 (0001) substrate (Honeywell) at $\sim 50^\circ\text{C}$ in 3 mTorr of Ar ($25 \text{ cm}^3 \text{ min}^{-1}$ flow), resulting in a high degree of orientation in the $\langle 110 \rangle$ direction [confirmed by x-ray diffraction (XRD)]. The film had a bulk critical temperature $T_c \approx 8.9 \text{ K}$, as determined from magnetometry measurements. Further characterization details are given elsewhere.⁸⁰

III. RESULTS AND ANALYSIS

This section is divided into three parts. First, we present the raw SLR data and describe their features in Sec. III A. Subsequently, we consider the analysis of the SLR data and show the extracted SLR rates ($1/T_1$) in Sec. III B. Finally, in Sec. III C, we construct a model for $1/T_1$ that allows for the extraction of the magnetic penetration depth and apply it to the results.

A. SLR data

Typical ^8Li SLR data in Nb at low temperatures and magnetic fields are shown in Fig. 2. In all cases, the SLR is orders of magnitude faster than in fields on the order of $\sim 1 \text{ T}$,⁸⁰ typical of measurements in the field regime where low-frequency fluctuations of the “host” nuclear spins provide the dominant relaxation mechanism

(see, e.g., Refs. 57 and 81). The relaxation, however, while fast relative to the “natural” timescale of the probe ($\sim 1/\tau_\beta$), remains observable for all conditions investigated here. From the temperature dependent measurements at a constant implantation energy $E = 19.9 \text{ keV}$ (i.e., $\langle z \rangle \approx 70 \text{ nm}$), it is apparent that, upon cooling the sample from the normal state ($T > T_c$) to the Meissner state ($T < T_c$), the SLR rate increases (as evidenced by the increased haste in the decay of the SLR “curves”). This SLR behavior is consistent with that observed in other superconductors.^{57,60} Similarly, from measurements at constant temperature $T = 3.5 \text{ K}$ (well into the Meissner state), it is clear that as the implantation energy increases (equivalent to ^8Li sampling deeper below Nb’s surface), so too does the SLR rate. Note that equivalent measurements in the normal state ($T > T_c$) show no such dependence on E , suggesting that this SLR response is a result of Nb’s superconductivity. Interestingly, we note that the data in Fig. 2 include measurements both where the applied field is zero during cool-down (zero-field-cooled) and when it is 20 mT (field-cooled), showing no appreciable differences (see below).

B. Quantifying $1/T_1$

To quantify the observations outlined in Sec. III A, we consider a model to determine the SLR rate $1/T_1$. First, we remark that, in contrast to measurements at higher fields,⁸⁰ the decay of the SLR “curves” is *non-exponential* at all measured conditions. This is not unprecedented for a spin $I = 2$ nucleus, though it is difficult to identify the exact underlying cause (e.g., Refs. 82–85), which may be complicated by a fundamental change in the character of the SLR at low magnetic fields (see, e.g., Ref. 81). This detail is compounded by the fact that there are *two* $^8\text{Li}^+$ stopping sites in Nb at low temperatures.⁸⁰ While an atomistic model of the stopping sites may illuminate the matter, it is still forthcoming.⁸⁶ Consequently, we elected for a pragmatic approach and fit the SLR data using a (phenomenological) stretched exponential “relaxation” function. For a uniform distribution of probe arrival times (i.e., a time-independent $^8\text{Li}^+$ production rate), $R(t, t')$ from Eq. (2) takes the form

$$R(t, t') = \exp \left[- \left(\frac{t - t'}{T_1} \right)^\beta \right], \quad (3)$$

where $1/T_1$ is the SLR rate and $\beta \in (0, 1]$ is the stretching exponent. While a value of $\beta < 1$ modifies the form of an exponential decay, equivalent to the presence of faster (slower) relaxation at early (late) times, the quantity T_1 captures the characteristic $1/e$ decay time of p_z . We note that this model accurately describes the data over *all* measurement conditions, without over-parameterization.⁸⁷

The SLR data were fit using Eqs. (1)–(3). Note that, for this definition of $A(t)$, the terms A_0 and p_0 are not uniquely defined and instead we used a single “effective” parameter, $\tilde{A}_0 \equiv A_0 p_0$, which is on the order of $\sim 10\%$ for our data. Similarly, to mitigate the known “inflation” of $\tilde{A}_0$ from Eq. (3) [i.e., from the divergence in Eqs. (2) and (3) when $t \rightarrow 0$ s], we constrained our fits such that $\tilde{A}_0$ was shared across measurements at constant E (i.e., in a so-called “global” fit). Empirically, $\tilde{A}_0$ was found to vary with E

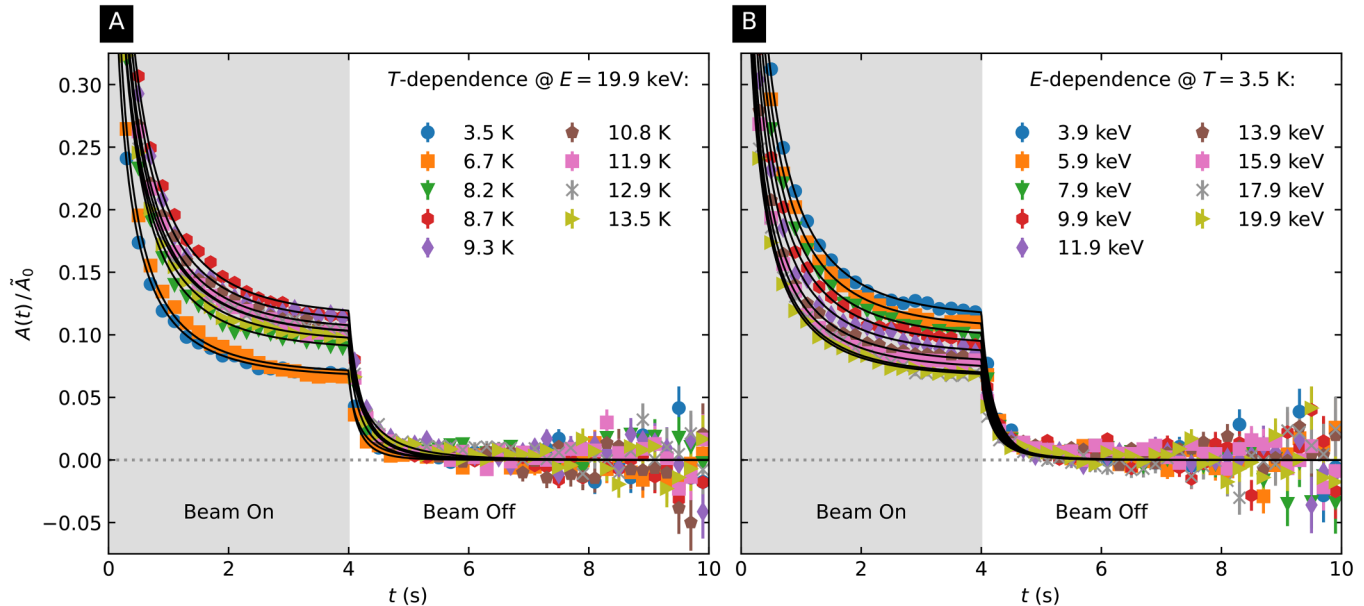


FIG. 2. Typical ^8Li SLR data in Nb(300 nm)/ Al_2O_3 at low temperatures $T < 15$ K and implantation energies $E \leq 20$ keV (where all implanted $^8\text{Li}^{+}$ stops in the film—see Fig. 1) in an applied magnetic field $B_0 = 20$ mT parallel to the sample surface. The data (represented as histograms) have been binned by a factor of 20 for clarity and the solid lines represent fits using a stretched exponential relaxation function (described in Sec. III B). In all cases, the SLR rate is orders of magnitude faster than at higher fields³⁰ with much of the (normalized) asymmetry $A(t)/A_0$ vanishing within the first few milliseconds (the vertical scale has been adjusted to better show the remaining signal). (a): Temperature dependent measurements at a constant implantation energy $E = 19.9$ keV. Upon cooling the sample from the normal state ($T > T_c$) to the Meissner state ($T < T_c$) the SLR rate increases, as evidenced by the increased haste in the decay of the “curves.” (b): Implantation energy dependent measurements at a constant temperature $T = 3.5$ K. In the Meissner state, as the implantation energy increases (equivalent to ^8Li sampling deeper below Nb’s surface) the SLR rate increases. Note that equivalent measurements in the normal state ($T > T_c$) show no such dependence on energy.

according to a piecewise linear function (i.e., with different slopes above and below ~ 4 keV) and we imposed the additional constraint that it must fall along this line.⁸⁸ Lastly, we point out that for our $R(t, t')$, numeric integration is required to evaluate Eq. (2). For this, we used tanh-sinh quadrature,^{89,90} (i.e., a so-called “double-exponential” technique), which provides both accurate and expedient evaluation of our model. The fit quality was good for all the SLR data, with a typical reduced $\chi^2 \approx 1.02$ for each measurement. The fitting was performed by a custom Python script using the mumpy, bdata, and bfit libraries.^{91,92} A subset of the fit results are shown in Fig. 2, with the SLR rates and stretching exponents extracted from the analysis presented in Figs. 3 and 4.

We now consider the results from fitting. In the normal state below ~ 15 K, $1/T_1$ varies approximately linearly with temperature (see Fig. 3); however, its slope is much larger than in measurements at higher fields,⁸⁰ resembling the ^8Li SLR in Au⁸¹ and other elemental metals.³⁷ Similarly, the (extrapolated) intercept of $1/T_1$ at 0 K is also non-zero. Note that these two features are inconsistent with the so-called Korringa response^{39,93} expected for relaxation due to electrons in a metal’s conduction band. We shall return to this point below.

Just below the film’s “bulk” $T_c \approx 8.9$ K, there is a pronounced “kink” in the temperature dependence of $1/T_1$. The discontinuity at ~ 8.8 K is suggestive of a transition temperature,

with the shift to lower- T being consistent with the suppression of T_c by the small (20 mT) applied field. As the temperature is lowered further, $1/T_1$ increases non-linearly, with the sharpest increase occurring within the first few Kelvin below the “kink.” At colder temperatures, the increased SLR ceases, with $1/T_1$ saturating by ~ 4 K. This $1/T_1$ “dome” is consistent with the expected ^8Li SLR signature for Meissner screening.^{57,60} Interestingly, as noted earlier, both field- and zero-field-cooled measurements are in good agreement with each other, suggesting that either flux trapping has little influence on the observed SLR rates or there is little flux trapped in the film.

Interestingly, we find a stretching exponent β close to ~ 0.5 at all measurement conditions, showing only weak variation with temperature or implantation energy (see Fig. 4). Above Nb’s T_c , β is nearly constant and independent of E , showing only a small degree of scatter. Near T_c , a small local maximum is observed, with the exponent decreasing smoothly as the temperature is lowered. At the lowest measured temperature (~ 3.5 K), some E -dependence to β is evident, with β increasing with decreasing E , implying the SLR becomes more homogeneous closer to the sample’s surface. While $\beta = 0.5$ is often ascribed to heterogeneous relaxation resulting from, for example, spatial disorder,⁸² its absence at high field⁸⁰ suggests it must be of a different origin (cf. the field-dependent ^8Li SLR in Au⁸¹). Given the ambiguity of isolating the cause the non-

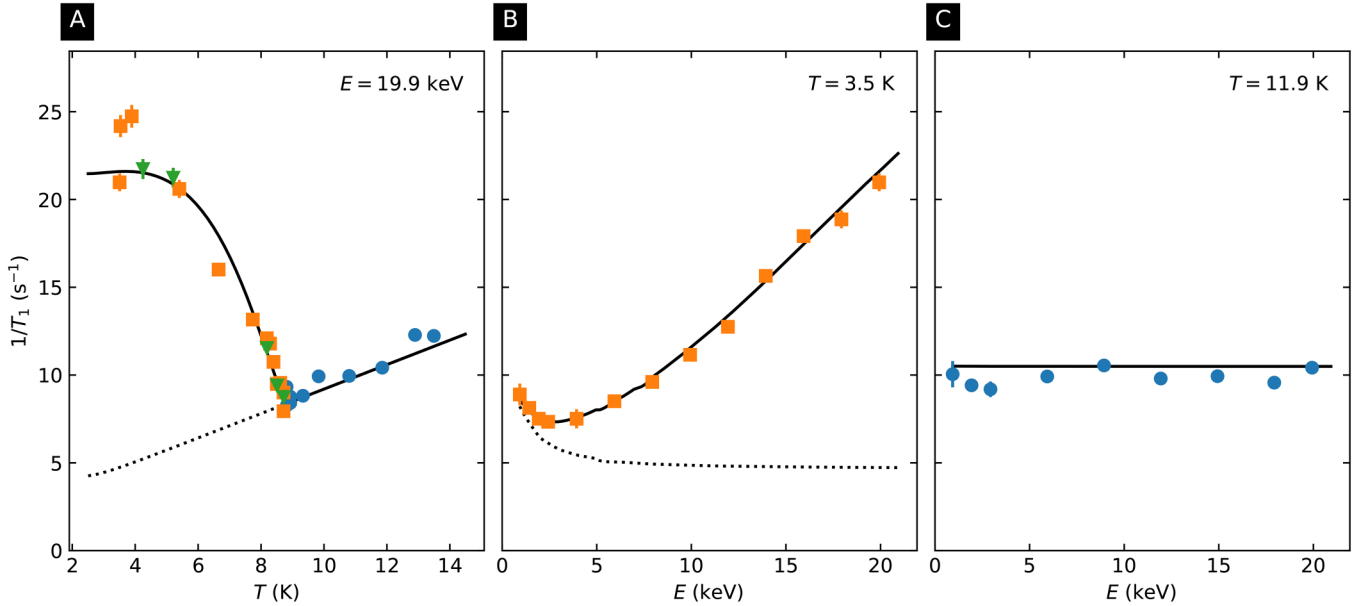


FIG. 3. ^8Li SLR rates $1/T_1$ in Nb(300 nm)/Al₂O₃ at 20 mT parallel to its surface, determined from fits to a stretched exponential model [Eqs. (1)–(3)] described in the text. The symbols distinguish different groups of data [normal state (•); Meissner state, field-cooled (■); Meissner state, zero-field-cooled (▼)], all in good agreement with each other. The solid black line (—) denotes a fit of the $1/T_1(E, T, B)$ model to all the data, clearly capturing all of the main features. For comparison, the dotted black line (···) shows the predicted normal-state response for the SLR rate. (a) Temperature dependence of $1/T_1$ at constant implantation energy ($E = 19.9$ keV), corresponding to a mean stopping depth of ~ 70 nm. In the normal state, the SLR rate varies linearly with temperature, but is orders of magnitude larger than the electronic (i.e., Korringa^{39,93}) relaxation observed at higher applied fields.⁸⁰ At $T_c \approx 8.8$ K, there is a pronounced kink in the temperature dependence and $1/T_1$ increases substantially with decreasing T . This is the manifestation of Meissner screening sensed through the dipole-dipole coupling (i.e., “cross-relaxation”) with the Nb host’s nuclear spins (see, e.g., Refs. 57 and 60). (b) Implantation energy dependence of $1/T_1$ at constant temperature in the Meissner state ($T = 3.5$ K). At energies > 4 keV, the SLR rate increases monotonically with increasing E , consistent with enhanced field screening deeper below the surface. At energies ≤ 4 keV, there is a small upturn in $1/T_1$ with decreasing E , likely a result of a (weak) paramagnetic contribution from the native oxide layer at the Nb surface.^{94,95} (c) Implantation energy dependence of $1/T_1$ at constant temperature in the normal state ($T = 11.9$ K). In contrast to the data in the Meissner state, no meaningful energy (i.e., depth) dependence is observed.

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exponential SLR transients, we suggest that β of ~ 0.5 represents an effective average over all relaxing ^8Li , whose SLR time-constants are too similar to isolate here.

Finally, we note that the implantation energy dependence in the Meissner state (3.5 K) appears more complicated than anticipated. As E increases, $1/T_1$ decreases slightly, reaching a minimum around ~ 2.5 keV (i.e., $\langle z \rangle \approx 10$ nm); however, for all higher E , $1/T_1$ increases monotonically, qualitatively consistent with the expected SLR response from Meissner screening.^{57,60} This rich behavior in the Meissner state is greatly contrasted by that in the normal state above T_c , where any (meaningful) depth-dependence to $1/T_1$ is absent. This fact is strong confirmation that the observed effects originate from Nb’s superconductivity.

With the essential features of the $1/T_1$ data described above, we now consider a model that captures these details, including the magnetic penetration depth λ .

C. The $1/T_1$ model

First, we remark that for a given $^8\text{Li}^+$ implantation energy E , the measured $1/T_1$ represents an average over the probe’s stopping

profile $\rho(z; E)$,

$$\frac{1}{T_1} \equiv \int_0^\infty \frac{1}{T_1(z)} \rho(z; E) dz, \quad (4)$$

where the term $1/T_1(z)$ encapsulates any depth dependence of the SLR rate.⁹⁸ While $\rho(z; E)$ can be reliably determined from Monte Carlo simulations (see Sec. II and Fig. 1), we must postulate a realistic form for $1/T_1(z)$. In general, there can be several contributions to the *observed* SLR rate $1/T_1$, which can be decomposed into distinct mechanisms,⁹⁹

$$\frac{1}{T_1} = \frac{1}{T_1^e} + \frac{1}{T_1^d} + \frac{1}{T_1^q} + \dots$$

Here, $1/T_1^i$ s denote contributions from: conduction electrons ($i = e$), dipole-dipole interactions ($i = d$), and quadrupolar interactions ($i = q$). While other contributions are possible,^{37–40} we restrict ourselves to the most common ones listed above.

We begin by considering $1/T_1^e$. In metals, the SLR is often dominated by the so-called Korringa mechanism,^{39,93} wherein

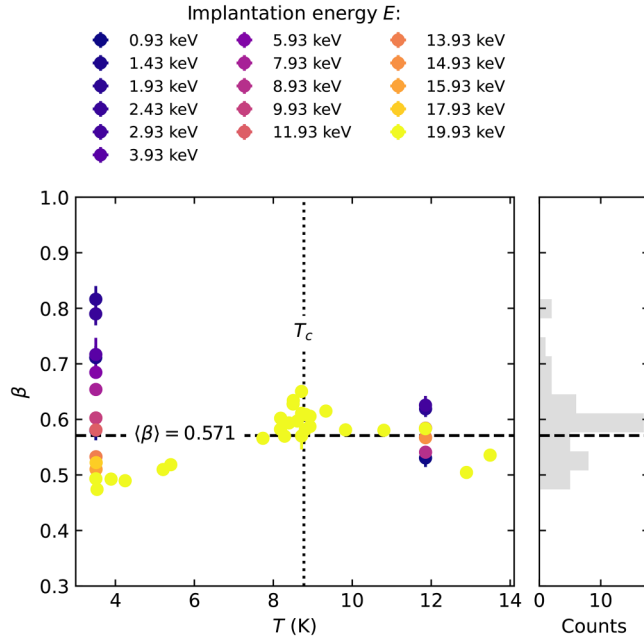


FIG. 4. The temperature (T) and implantation energy (E) dependence of the stretching exponent β , determined from fits of the ^8Li SLR data (Fig. 2) to a stretched exponential relaxation function [Eq. (3)]. β is found to be weakly dependent on both T and E , but remains close to ~ 0.5 for the majority of our measurement conditions. Above Nb's critical temperature T_c (indicated by the vertical dotted line), β is nearly constant and independent of implantation energy E , showing only a small degree of scatter. At T_c , a small local maximum is observed, with the exponent decreasing smoothly as the temperature is lowered. At ~ 3.5 K (i.e., well into the Meissner state), the E -dependence is most pronounced, with the (depth-averaged) relaxation becoming increasingly homogeneous with decreasing E . The exponent's average value ($\langle\beta\rangle \approx 0.571$) is indicated by the horizontal dashed line, with the value included in the inset. The distribution of β values is also shown as a histogram.

relaxation is induced by spin-flip scattering of conduction electrons coupled to the nuclear spin via their hyperfine interaction. From a derivation based on Fermi's golden rule, the sum over electron momenta, when converted to an integral over energy, yields

$$\frac{1}{T_1^e} = \frac{2\pi}{\hbar} A_{\text{hf}}^2 \int_{-\infty}^{+\infty} \rho_e^2(E) f(E) [1 - f(E)] dE, \quad (5)$$

where A_{hf} is the hyperfine coupling, $\rho_e(E)$ is the electronic density of states (DOS), and $f(E)$ is the Fermi-Dirac distribution,

$$f(E) = \left[\exp\left(\frac{E - E_F}{k_B T}\right) + 1 \right]^{-1},$$

with E_F and k_B denoting the Fermi energy and Boltzmann constant, respectively. Note that the above integral is over all electron energies in the conduction band. In a broad band degenerate metal, where $E_F \gg k_B T$, $\rho_e(E)$ is practically constant over the range where the Fermi factor $f(E)[1 - f(E)]$ is non-zero, and the integral

(to an excellent approximation) evaluates to

$$\frac{1}{T_1^e} \approx \frac{2\pi}{\hbar} A_{\text{hf}}^2 \rho_e^2(E_F) k_B T. \quad (6)$$

The salient feature of Eq. (6) is its linear temperature dependence, which is distinct from most other SLR mechanisms. This also produces an SLR rate that is *independent* of the applied magnetic field. This electronic relaxation has been reported previously for ^8Li implanted in this Nb film,⁸⁰ where below ~ 20 K,

$$\frac{1}{T_1^e T} \approx 1.271(39) \times 10^{-2} \text{ s}^{-1} \text{ K}^{-1}.$$

Importantly, these rates are at least an order of magnitude smaller than our observed $1/T_1$ s, leading us to conclude that the importance of $1/T_1^e$ is negligible at our measurement conditions.¹⁰⁰

Next, we consider $1/T_1^q$. The ^8Li resonance in Nb at low temperatures shows unambiguously that some probes stop in sites with non-cubic symmetry,⁸⁰ where a static (i.e., time-averaged) electric field gradient (EFG) remains finite. Considering Nb's body-centered cubic (BCC) structure,¹⁰¹ there is some difficulty in precisely assigning a stopping site. Earlier work suggested the possibility of an interstitial dumbbell-like ^8Li -Nb defect complex at low- T ,⁸⁰ which is consistent with preliminary results from density functional theory (DFT) calculations;⁸⁶ however, no firm assignment has yet been made. We note though that the presence of a non-zero *static* EFG does not necessitate a dynamic component sufficient for causing SLR (cf. ^8Li β -NMR in Bi¹⁰²). Moreover, $1/T_1^q$ is expected to follow a power-law that is strongly dependent on temperature,¹⁰³ but not applied field, which is inconsistent with our data and measurements at higher fields.⁸⁰ At our measurement conditions, such a phonon-mediated mechanism should be negligible, given that T is far below Nb's Debye temperature (~ 270 K) and there are very few thermally populated phonons. On that basis, we rule it out as a significant source for our $1/T_1$.¹⁰⁴

With the above two mechanisms ruled out as meaningful contributors, it is curious that a T -linear component of the SLR rate persists; however, this is not a novel observation and similar behavior has been observed in the ^8Li β -NMR of materials whose NMR response is metallic (see, e.g., Refs. 81, 105 and 106). As the exact origin remains unclear, we adopted an empirical term in our model,

$$\frac{1}{T_1^l} \approx c \times T, \quad (7)$$

where c is the slope of the linear relation. This does not, however, account for the finite intercept at $T = 0$ K, which must find another origin such as the $1/T_1^d$, considered below.

The SLR term $1/T_1^d$ accounts for relaxation resulting from magnetic fluctuations due to dipole-like fields in the vicinity of the probe. While the interaction is sensitive to the local field, it is sufficiently decoupled by typical NMR fields (>1 T) such that its contribution to SLR is often minor. Instead, the static (i.e., time-averaged) component of the dipole-dipole interaction is often the main observable, dictating the intrinsic width of the NMR line.¹⁰⁷

Outside this “high field” limit, however, both this quantity¹⁰⁸ and the SLR rate become field-dependent, the latter often becoming dominant in ^8Li β -NMR (see, e.g., Refs. 57, 59, 60, 80, 81 and 109). This contribution to the SLR can be considered as a form of “cross-relaxation” with the host ^{93}Nb nuclear spins ($I = 9/2$; $\gamma/(2\pi) = 10.4523(5) \text{ MHz T}^{-1}$; 100 % abundance),¹¹⁰ whose dynamic behavior is stochastic with characteristic time-constants T_1 and T_2 for their longitudinal and transverse components, respectively (the latter being of the greatest importance here). In essence, the polarization of the ^8Li probes “leaks” into the ^{93}Nb spin “bath” at a rate $1/T_1^d$. This process is strongly suppressed by large magnetic fields, except near (avoided) level-crossings in the eigenstates of the coupled spin system (see, e.g., Refs. 111 and 59) or at low magnetic fields where it dominates.¹¹² In fact, cross-relaxation between ^8Li and ^{93}Nb nuclei has been studied previously using (neutron activated) ^8Li β -NMR of LiNbO_3 .¹¹¹

Qualitatively, the SLR behavior of this cross-relaxation process can be described by a Lorentzian-like expression, with $1/T_1^d \propto B^{-2}$ (see, e.g., Refs. 57, 59, 60, 80, 81 and 109). Recognizing that our ^8Li probe is unlike any of the “host” ^{93}Nb spins, it is possible to describe their interaction more rigorously. Explicitly, for a probe spin I coupled to a lattice spin S , the heteronuclear dipole-dipole contribution to the SLR rate can be written as³⁸

$$\frac{1}{T_1^d} = \frac{1}{3} S(S+1) \langle \delta B_d^2 \rangle \left\{ \frac{1}{3} J_0(\omega_S - \omega_I) + J_1(\omega_I) + 2J_2(\omega_S + \omega_I) \right\}, \quad (8)$$

where $\langle \delta B_d^2 \rangle$ is the mean-squared fluctuating field¹¹³ and

$$J_n(\omega_i) = \frac{\tau_c}{1 + \omega_i^2 \tau_c^2}, \quad (9)$$

which is the n -quantum spectral density function (i.e., the Fourier transform of the (auto)correlation function describing stochastic fluctuations in the local electromagnetic field that “relax” the probe’s spin) characterized by an (exponential)¹¹⁴ correlation time τ_c .¹¹⁵ Note that while Eq. (9) is among the simplest forms for J_n , most reasonable choices for the (auto)correlation function yield similar results (see, e.g., Ref. 116). In fact, the validity of this formulation of heteronuclear dipole-dipole SLR has been confirmed quantitatively in a previous β -NMR experiment on dilute ^8Li in (isotopically pure) ^7Li metal.¹¹⁷

In Eqs. (8) and (9), the connection to the local magnetic field is through the Larmor relation,

$$\omega_i = \gamma_i B(z), \quad (10)$$

where γ_i is the gyromagnetic ratio of the i^{th} nuclear spin, and $B(z)$ is the local field, written suggestively to emphasize its spatial (i.e., depth) dependence. With Eq. (10) providing the connection between the SLR rate [via Eqs. (8) and (9)] and the local field, we now consider the details of the latter.

In the normal state (i.e., when $T > T_c$), there is no meaningful depth-dependence and $B(z) = B_0$, the applied magnetic field. In the limit that τ_c is temperature-independent, Eq. (8) reduces to a constant, equivalent to the non-zero $1/T_1$ at 0 K (i.e., a finite

intercept) that we infer from extrapolating our data. By contrast, in the Meissner state, the local field is expected to be a strong function of z and we ascribe the E -dependence of our data to this fact. A salient feature of a superconductor in the Meissner state is the complete expulsion of magnetic flux from its interior. This is achieved by supercurrents within the material counter-acting the external field, causing it to “decay” over very short distances below the material’s surface (on the order of ~ 10 to $\sim 100 \text{ nm}$). Quite generally, the screening of the magnetic field can be written as^{16,17,62}

$$B(z) = B_0 \left(\frac{2}{\pi} \right) \int_0^\infty \frac{q}{q^2 + K(q)} \sin(qz) dq, \quad (11)$$

where B_0 is the applied field, z is the depth below the surface, and $K(q)$ is the Fourier transform of the integrand kernel relating the current density $\mathbf{j}$ to the vector potential $\mathbf{A}$ inside a superconductor (see, e.g., Ref. 19). In the absence of any appreciable dependence on the wavevector q , $K(q) \approx 1/\lambda^2$ and Eq. (11) reduces to

$$B(z) = B_0 \exp\left(-\frac{z}{\lambda}\right),$$

which is the famous (phenomenological) result of the London brothers.⁶¹ A similar calculation, accounting for the boundary conditions specific to a film with thickness ζ , as well as a non-superconducting “dead layer” d at the material’s surface (see, e.g., Ref. 22), yields⁶²

$$B(z) = B_0 \times \begin{cases} 1, & z \leq d, \\ \frac{\cosh[(\zeta/2 - z - d)/\lambda]}{\cosh[\zeta/(2\lambda)]}, & d < z \leq \zeta. \end{cases} \quad (12)$$

We note that Eq. (12) assumes no contribution from non-local electrodynamics.^{16,17,19} Though non-local effects are expected to be present in “clean” Nb,¹⁹ they are known to be weak and, as is shown below, they can be safely neglected.

While Eqs. (8)–(10), and (12) define the connection between the SLR rate and the spatial dependence of B , these alone cannot account for the temperature dependence in $1/T_1$ that we observe. To reconcile this deficiency, it is necessary to also consider the temperature dependence of the penetration depth λ . Typically, λ is nearly temperature-independent when $T \ll T_c$, but diverges such that $\lim_{T \rightarrow T_c} \lambda(T) = +\infty$. While this behavior arises naturally from Eq. (11) in the Bardeen-Cooper-Schrieffer (BCS) formulation of $K(q)$,¹⁷ the expressions are computationally cumbersome,¹⁹ and we instead adopt the empirical “two-fluid” expression to describe the temperature dependence,

$$\lambda(T) = \begin{cases} \frac{\lambda_0}{\sqrt{1 - (T/T_c)^4}}, & T < T_c, \\ +\infty, & T \geq T_c, \end{cases} \quad (13)$$

where λ_0 is the penetration depth at 0 K. We note that Eq. (13) has been shown previously to accurately capture the temperature dependence of $\lambda(T)$ in Nb.¹⁹

While these details can correctly explain the majority of our observations about the data, a remaining feature requiring explanation is the upturn in $1/T_1$ observed at low- T and low- E . At these E

(<2.5 keV), we expect a substantial fraction of the implanted $^8\text{Li}^+$ stop within the first ~ 5 nm of the surface (see Fig. 1), where a native oxide layer (e.g., Nb_2O_5) resides.⁷⁵ In the bulk, these oxides are known to be paramagnetic (e.g., due to oxygen sub-stoichiometry),⁹⁴ and this magnetism has been observed at the surface of Nb.^{95,118,119} This leads us to postulate that the upturn is due to fluctuations in these electronic moments localized in the surface oxide layer. Noting that the upturn is appreciable at 3.5 K, but negligible at 11.9 K (see Fig. 3), this SLR mechanism must be highly temperature dependent, with the fluctuation spectrum “freezing” into the vicinity of ^8Li ’s Larmor frequency within a narrow temperature range. While this process can be described, in general, by a formalism analogous to that in Eqs. (8) and (9), our data are too limited for such a treatment and we instead adopt an additional power law term for stopping depths less than ~ 5 nm,

$$\frac{1}{T_1^p} \approx \frac{C}{T^b}, \quad (14)$$

where b is the exponent and C is a constant. As is shown below, this gives a reasonable description of our data.

Based on the above discussion, we propose the following hierarchy to the SLR rates:

$$\frac{1}{T_1^q} < \frac{1}{T_1^e} \ll \frac{1}{T_1^l} \sim \frac{1}{T_1^p} \sim \frac{1}{T_1^d}.$$

We stress that only $1/T_1^d$ can account for the SLR response observed upon transition from the normal to the Meissner state.^{57,60} With these pieces in place, we now turn our attention to the $^8\text{Li}^+$ stopping profile $\rho(z; E)$ —the remaining “ingredient” required to evaluate Eq. (4).

As mentioned in Sec. II, $^8\text{Li}^+$ stopping profiles can be simulated reliably using Monte Carlo codes (e.g., SRIM^{72,73}), producing histograms to represent $\rho(z; E)$ (see Fig. 1). For our analysis, it was convenient to have the ability to describe these profiles at *arbitrary* E , which can be accomplished by fitting the simulated profiles and interpolating their “shape” parameters. Empirically, we found that $\rho(z; E)$ at a given E can be described, in general, by

$$\rho(z; E) = \sum_i^n f_i p_i(z), \quad (15)$$

where $p_i(z)$ is a probability density function, $f_i \in [0, 1]$ is the i^{th} stopping fraction, constrained such that

$$\sum_i^n f_i \equiv 1,$$

and z is the depth below the surface. For our $\text{Nb}_2\text{O}_5(5\text{ nm})/\text{Nb}$ target (see, e.g., Ref. 75), the stopping data are well-described using $n = 2$ and a $p(z)$ given by a modified beta distribution.¹²⁰

Explicitly,

$$p(z) = \begin{cases} 0, & z < 0 \\ \frac{(z/z_0)^{\alpha-1}(1-z/z_0)^{\beta-1}}{z_0 B(\alpha, \beta)}, & 0 \leq z \leq z_0, \\ 0, & z > z_0, \end{cases} \quad (16)$$

where $z \in [0, z_0]$ is the depth below the surface and $B(\alpha, \beta)$ is the beta function,

$$B(\alpha, \beta) \equiv \frac{\Gamma(\alpha)\Gamma(\beta)}{\Gamma(\alpha + \beta)},$$

with $\Gamma(s)$ denoting the gamma function

$$\Gamma(s) \equiv \int_0^\infty x^{s-1} \exp(-x) dx.$$

Note that the “extra” z_0 in the denominator of Eq. (16) ensures proper normalization of $p(z)$. This (phenomenological) parameterization of the $^8\text{Li}^+$ stopping profiles was found to work well for $E \leq 30$ keV and a subset of the fit results are shown in Fig. 1.

With all the pieces now in hand, we briefly summarize the most important details of our $1/T_1$ model. The major SLR term $1/T_{1d}$ [Eqs. (8) and (9)] has a field dependence of the form

$$1/T_{1d} \propto 1/(1 + \varsigma B^2),$$

where ς is a scalar value. Above T_c , B is equal to the external field and is independent of ^8Li ’s position z below Nb’s surface, resulting in a constant $1/T_{1d}$. Below T_c , the local magnetic field in Nb decreases as z approaches the film’s center [Eq. (12)] due to the Meissner effect, causing the spatially averaged $1/T_{1d}$ to increase with decreasing temperature. This change is abrupt, occurring immediately below T_c , where the penetration depth λ changes rapidly [Eq. (13)]. At lower temperatures, λ approaches a constant value and so too does $1/T_{1d}$. Similarly, as the implantation energy E is increased, the implanted ^8Li sample depths closer to the film’s center (see Fig. 1) where the local field is lowest, causing the spatially averaged $1/T_{1d}$ to increase with increasing implantation energy (up to the maximum E of 20 keV used here).

Finally, with these details in mind, it is straightforward to construct a model to describe the $1/T_1$ data for Nb in Fig. 3. Explicitly, a full 3D fit (i.e., $1/T_1$ vs E , T , and B) was performed by: combining Eqs. (7)–(10) and (12)–(14) into an expression for $1/T_1(z)$; parameterizing $\rho(z; E)$ using Eqs. (15) and (16) and interpolating their “shape” parameters; plugging these expressions into Eq. (4) and evaluating the integral numerically.¹²¹ The fit included *all* experimental data below 15 K,¹²² and the result is shown in Fig. 3. A summary of the main fit parameters determined from this procedure is given in Table I.

IV. DISCUSSION

It is clear from Fig. 3 that our model for the ^8Li $1/T_1$ (described in Sec. III C) is able to accurately reproduce our SLR data in the Nb film. Such good agreement is a strong confirmation

TABLE I. Summary of the main fit parameters and their uncertainties (rounded according to the Particle Data Group's rules⁹⁶), determined from fits of the $1/T_1(E, B, T)$ model to the data in Fig. 3 (solid black line). For convenience, a brief description of each parameter and a list of equations where they appear are also included. Further details about the model and fitting procedure are given in Sec. III C.

Symbol	Value	Units	Description	Appearances
d	5.0 ± 0.9	nm	Thickness of the non-superconducting “dead layer” at the film’s surface	Eq. (12)
λ_0	51.5 ± 2.2	nm	The film’s magnetic penetration depth	Eqs. (12–13)
B_d	$(41.1 \pm 2.1) \times 10^{-6}$	T	Magnitude of the dipolar field at the ^8Li stopping sites	Eq. (8)
τ_c	$(6.1 \pm 0.5) \times 10^{-6}$	s	NMR correlation time for the fluctuations causing SLR	Eq. (9)
T_c	8.775 ± 0.014	K	The film’s superconducting transition temperature	Eq. (13)
c	0.697 ± 0.011	$\text{s}^{-1} \text{K}^{-1}$	Slope of the T -linear contribution to the total SLR rate	Eq. (7)
C	$(2.03 \pm 0.19) \times 10^3$	$\text{s}^{-1} \text{K}^b$	Constant in the near-surface paramagnetic SLR contribution	Eq. (14)
b	4.5		Exponent in the near-surface surface paramagnetic SLR contribution	Eq. (14)

of the choices made throughout our analysis. To further contextualize the results, we now consider the extracted parameters explicitly.

We shall start our discussion with the film’s critical temperature $T_c = 8.775(14)$ K, which is slightly below its “bulk” value of ~ 8.9 K determined from magnetometry. This difference, however, is consistent with the well-known suppression of T_c by an applied field and its small uncertainty emphasizes that it is well-defined by our model. This has bearing on $\lambda(T)$, whose temperature dependence is defined by Eq. (13). This expression’s other term of consequence, the magnetic penetration depth at 0 K λ_0 , was determined to be $51.5(22)$ nm. The magnitude of this value is common for Nb films (see, e.g., Refs. 123–129) and larger than the so-called London penetration depth λ_L often quoted for bulk Nb (see below). Noting that our film is thick compared to Nb’s BCS coherence length $\xi_0 \approx 40$ nm (derived from an average of literature values^{1,123,124,126,130–136}), this “discrepancy” can be understood not in terms of a finite-size effect,¹²⁹ but rather in terms of the “dirty”^{62,137} character of our film. It can be shown that, at 0 K, one obtains the simple relationship⁶²

$$\lambda_0 = \lambda_L \sqrt{1 + \frac{\xi_0}{\ell}}, \quad (17)$$

defining the influence of the carrier mean-free-path ℓ on λ_0 . Taking $\lambda_L \approx 29$ nm (based on an average of literature values^{1,123–126,130–133,135,138–140}), and re-arranging Eq. (17), we obtain $\ell = 18.7(29)$ nm, in good agreement with the range of values found in similarly prepared films (see, e.g., Refs. 129 and 141–143). Within the Pippard¹⁶ or BCS¹⁷ theories, such a short ℓ corresponds to an “effective” coherence length $\xi \approx 13$ nm, placing the results firmly within the “local” regime (i.e., where $\lambda > \xi > \ell$ —see, e.g., Ref. 137). Note that, in this limit, accounting for non-local electrodynamics in the calculation of $B(z)$ ^{16,17} is unnecessary, in contrast to that expected for “clean” Nb.¹⁹

We now turn our attention to the correlation time $\tau_c = 6.1(5) \times 10^{-6}$ s of the fluctuations causing the ^8Li SLR. We remark that this value is larger by a factor of ~ 10 than the quantity derived from the ~ 0.2 T “width” of the Lorentzian field-dependence quoted for earlier measurements at 10 K and higher fields.⁸⁰ We emphasize that, though not explicitly discussed in Ref. 80, the field-dependence is a manifestation of the cross-relaxation discussed in

Sec. III C. It should be remarked, however, that differences in the two quantities are expected, for example, due to the orientation dependence of spectral densities (see, e.g., Refs. 37–39). While the earlier estimate was made using a limited set of measurements,⁸⁰ our value is derived using both a more comprehensive dataset and a more accurate model for the field dependence,^{38,117} suggesting that is the more reliable quantity. This is confirmed by its close proximity to the ^{93}Nb spin-spin relaxation (SSR) time $T_2 \approx 24 \mu\text{s}$,¹⁴⁴ differing from τ_c by only a factor of ~ 4 . Such a level of agreement is remarkable given the (indirect) manner in which τ_c is identified. We also note that similar behavior has been observed for the low-field ^8Li SLR in $\text{Bi}_2\text{Te}_2\text{Se}$.¹⁰⁵ Interestingly, the τ_c in the Nb film is substantially shorter than in NbSe_2 ,⁵⁷ presumably due to the reduced ^{93}Nb spin density and ^8Li ’s weaker coupling to their fluctuations.

Next, we consider the “coupling” term in Eq. (8): the dipolar field $B_d = 41.1(21) \mu\text{T}$. Interestingly, this value is a factor of ~ 10 smaller than estimates obtained from lattice sums¹⁰⁷ for several plausible interstitial sites in Nb’s BCC lattice,¹⁰¹ as well as the ^8Li resonance linewidths, both reported previously.⁸⁰ This level of difference isn’t unexpected, as the B_d in Eq. (8) is a *dynamic* (i.e., fluctuating) quantity, which need not be similar in magnitude to its *static* (i.e., time-averaged) counterpart. In fact, this level difference is comparable to the case of ^8Li in Cu, where estimates of B_d from detailed measurements of $1/T_1$ in the vicinity of avoided level crossings (ALCs)⁵⁹ are an order of magnitude smaller than those inferred from lattice sums¹⁴⁵ and resonance linewidths.^{145,146} We stress that $1/T_1$ ALC measurements should provide the most accurate determination of B_d ; however, much like the use of lattice sums^{107,108} they require a structural model of the $^8\text{Li}^+$ site in the host material. While we do not have an independent experimental verification of our B_d at this time, we suggest that its value is reasonable. Key to verifying this hypothesis will be developing a detailed understanding of the $^8\text{Li}^+$ defect site using quantum chemical models. Current information from high-field β -NMR experiments⁸⁰ and preliminary DFT calculations⁸⁶ suggest that a Li–Nb dumbbell-like defect complex is formed at the low temperatures employed in these experiments.

The next quantity to discuss is the slope of the linear term $c = 0.697(11) \text{ s}^{-1} \text{K}^{-1}$ in Eq. (7). There is some difficulty interpreting this value. First, we reiterate that this is *not* due to a Korringa

mechanism,^{39,93} whose slope is orders of magnitude smaller⁸⁰ (see also Sec. III C). The stark difference in these quantities implies that c is highly dependent on B_0 , though the precise mechanism is unclear (e.g., fluctuations in the *longitudinal* component of the Nb spin-system). At the level of empiricism, this behavior has also been observed in the ^8Li β -NMR of other metallic systems, including: Au,^{81,109} Cu,⁵⁹ NbSe₂,⁵⁷ and Bi₂Ch₃ (Ch = Se, Te).^{105,106} For the latter materials, it was suggested that the change in c may be related to charge-carrier-freezing in an applied field (see, e.g., Refs. 147–149); however, its applicability to our elemental metal seems minimal. Alternatively, the behavior in NbSe₂ was thought to be due to fluctuations in the ^{93}Nb T_1 ,⁵⁷ based on reasonable agreement between the estimated τ_c and the $1/e$ decay time for the SLR in the nuclear quadrupole resonance (NQR) regime.¹⁵⁰ While this remains the most plausible explanation for the behavior in Nb, making an equivalent comparison is hampered by the unavailability of ^{93}Nb SLR measurements at comparable fields (see, e.g., Refs. 144, 151, and 152). To be more conclusive, a systematic study of $c(B)$ seems necessary. Such an investigation may be possible in the near future thanks to a newly developed β -NMR instrument designed to operate from 0 to 200 mT.¹⁵³

Finally, we consider the parameterization of the near-surface behavior of $1/T_1$, namely, the upturn in SLR in the Meissner state at $E < 2.5$ keV and the extent of the (non-superconducting) “dead layer.” We note that there is some ambiguity in quantifying these features, owing to the rather limited data at these implantation energies. Empirically, the low- E increase in $1/T_1$ can be described by the power law in Eq. (14) with a fixed exponent $b = 4.5$ and $C = 2.03(19) \times 10^3 \text{ s}^{-1} \text{ K}^{4.5}$. Note that it was not possible to identify unique solutions for C , b , and d without fixing one of the parameters, due to their high degree of correlation. We are not aware of any physical significance for the value of our exponent b used in Eq. (14), making the connection of C to the magnetic properties of Nb₂O_{5-x}⁹⁴ unclear. While paramagnetism in Nb’s surface oxide layer^{95,118,119} remains the likely source, further measurements (e.g., in a Nb₂O₅ film) are necessary to be conclusive. Despite these ambiguities, our model determined the superconducting “dead layer” d to be 5.0(9) nm for our film, in excellent agreement with the typical thickness of the oxide layer that forms natively at the surface of Nb.⁷⁵ While this value also compares well with LE- μSR measurements on a similar film,¹⁹ it is much smaller than typical values found in Nb prepared for SRF applications.^{20–22} This difference is likely due to greater surface roughness in the SRF samples,¹⁵⁴ which is known to influence the extent of the “dead layer” (see, e.g., Refs. 155–157). We emphasize though that this quantity is inherent to a given *sample*, rather than being intrinsic to the material. Further SLR measurements at low implantation energies, including their temperature dependence, are required for deeper insight.

V. CONCLUSIONS

We used ^8Li β -NMR to measure the Meissner screening profile in a Nb(300 nm)/Al₂O₃ thin film. Expanding on an earlier characterization performed at high magnetic fields,⁸⁰ we measured the SLR rate $1/T_1$ ’s dependence on temperature and $^8\text{Li}^+$ implantation energy (i.e., depth below the surface), in an applied field of

20 mT (parallel to the film’s surface). While measurements at constant temperature in the normal state showed no depth-dependence, the SLR rate was found to vary strongly with implantation energy, increasing deeper below the surface. These observations were complemented by temperature dependent measurements at constant implantation energy, showing a linear variation in the normal state ($1/T_1 = 0.697(11) \text{ s}^{-1} \text{ K}^{-1} \times T$), but with a pronounced “kink” at $T_c = 8.775(14) \text{ K}$ and a non-linear increase at lower temperatures where $1/T_1 \propto B(z)^{-2}$, consistent with the ^8Li SLR in other superconductors.^{57,60} From a fit of the data to a model accounting for these details and the $^8\text{Li}^+$ stopping profile, it was found that the Meissner screening is well-described by a simple London model with a magnetic penetration depth $\lambda_0 = 51.5(22) \text{ nm}$ (extrapolated to 0 K). The large λ_0 compared to Nb’s intrinsic London penetration depth $\lambda_L \approx 29 \text{ nm}$ is consistent with a relatively short carrier mean-free-path $\ell = 18.7(29) \text{ nm}$ often found in similarly prepared films.^{129,141–143}

This work constitutes an important advance in the use of ^8Li β -NMR to study materials rich in (stable) nuclear spins at low magnetic fields. Similarly, it provides a basis for understanding and modeling the SLR response in other superconductors, where measurements in this field regime are likely required (i.e., to remain in the Meissner state). For the specific case of studying Nb, this work provides a foundation for analyzing and interpreting results in samples with *engineered* surfaces (e.g., from mild baking^{4,5} or doping^{6,7}), where λ may be spatially *inhomogeneous* (see, e.g., Refs. 158–160). There is even further interest in extending this study to fields on the order of Nb’s superheating field $B_{sh} \sim 200 \text{ mT}$,^{161,162} where SRF cavities typically operate. A new beamline and instrument have been developed for this purpose¹⁵³ and the measurement program is already under way.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Ryan M. L. McFadden: Formal analysis (lead); Investigation (equal); Software (lead); Visualization (lead); Writing – original draft (lead); Writing – review & editing (lead). **Md Asaduzzaman:** Formal analysis (supporting). **Terry J. Buck:** Investigation (equal). **David L. Cortie:** Investigation (equal). **Martin H. Dehn:** Investigation (equal). **Sarah R. Dunsiger:** Investigation (equal). **Robert F. Kiefl:** Conceptualization (equal); Funding acquisition (equal); Investigation (equal); Writing – review & editing (supporting). **Robert E. Laxdal:** Formal analysis (supporting); Writing – review & editing (supporting). **C. D. Philip Levy:** Investigation (supporting); Resources (equal). **W. Andrew MacFarlane:**

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DATA AVAILABILITY

Raw data from the β -NMR experiments were generated at TRIUMF's Centre for Molecular and Materials Science (CMMS) facility and are available for download from: <https://cmms.triumf.ca/>. Derived data supporting the findings of this study are available from the corresponding authors upon reasonable request.

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