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Spin fluctuations in the exotic metallic state of Sr_2RuO_4 studied with β-NMR

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A β-NMR study was performed on a Sr_2RuO_4 crystal in the metallic state using a beam of spin-polarized $^8Li^+$ implanted at a mean depth of 90 nm. The $^8Li^+$ spin-lattice relaxation rate is strongly influenced by the onset of incommensurate spin fluctuations. The nuclear relaxation rate can be explained using previously published bulk ^{17}O NMR and inelastic neutron spectroscopy measurements of the dynamic magnetic susceptibility to model the hyperfine coupling. A well-resolved quadrupolar-split NMR for $^8Li^+$ implies a static stopping position in an interstitial site. The $^8Li^+$ Knight shift is highly sensitive to the anisotropic static susceptibility.

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Sr_2RuO_4 is a key compound in the study of correlated electrons in metals [1]. It is stoichiometric and can be prepared as large high-purity single crystals, nearly free of disorder and amenable to many experimental methods. Sr_2RuO_4 is isostructural with the cuprate parent compound La_2CuO_4. The combination of this crystalline point-group symmetry with the spin-orbit coupling in Sr_2RuO_4 has led to recent predictions that it may host a topological superconducting phase which would be a strong candidate to support Majorana fermions [2,3]. In contrast to La_2CuO_4, Sr_2RuO_4 is a good two-dimensional conductor in the *ab* plane below room temperature, and below ~130 K it shows three-dimensional (3D) metallic transport [4]. Quantum oscillations at low temperature indicate that it is a good metal with a three-sheet Fermi surface that is well studied theoretically [5]. The resistivity scales as T^2 below 25 K so that the machinery of Fermi-liquid theory is applicable [6]; however, despite this, the exotic low-temperature superconductive state still has many competing explanations [7]. NMR has played a key role in elucidating the magnetic properties of both the normal [8–11] and superconducting states [12]. Initial interpretation of the NMR emphasized a $\mathbf{q} = 0$ ferromagnetic (FM) response [13,14], but subsequent inelastic neutron scattering (INS) showed a much more substantial antiferromagnetic (AFM) contribution at four incommensurate in-plane wave vectors $\mathbf{q_0} = (\pm 0.3, \pm 0.3)$ [15,16] due to Fermi surface nesting of the α and β sheets in the band structure. The dynamic susceptibility $\chi''(\mathbf{q},\omega)$ determined from INS admitted a quantitative model of the NMR spin-lattice relaxation rate (SLR) $1/T_1$ via the Moriya relation [15],

$$\frac{1}{T_1} = \frac{\hbar k_B T \gamma^2}{(g\mu_B)^2} \sum_q [A(\mathbf{q})]^2 \frac{\chi''_\perp(\mathbf{q},\omega_0,T)}{\omega_0}, \qquad (1)$$

where γ_N is the nuclear gyromagnetic ratio; $A(\mathbf{q})$ is the hyperfine form factor, determined by the site and hyperfine coupling; $\omega_0/\hbar = \gamma_N H_0$ is the NMR frequency, which corresponds to $\omega \to 0$ on the energy scale of the INS; and $\chi''_\perp$ is the imaginary part of the dynamic susceptibility.

NMR Knight shifts K also yielded local probe measurements of the static response $\chi'(0,0)$ but were found to be coupled differently to the various band contributions to the susceptibility χ. The ^{17}O and ^{101}Ru shifts are found to be T dependent, and the former is significantly more anisotropic than the bulk χ. Detailed modeling of the hyperfine couplings was then used to deduce the spin susceptibility χ_{spin} [8]. The complete absence of a change in K through the superconducting transition is the main evidence for spin-triplet superconducting order [12]. This and some other features of K remain puzzling [17], such as the substantial isotropic contribution to the ^{17}O shift in the metallic state [8].

Here we report the results of NMR experiments in the normal state of Sr_2RuO_4 via beta-detected NMR (β-NMR) of $^8Li^+$ ions implanted into a *c*-axis-oriented crystal at 20 keV. Similar to the better-known μSR [18,19], the radioactive spin probe stops at a well-defined crystallographic site in the host lattice and senses its surroundings by a hyperfine coupling to surrounding electrons. The anisotropic β decay is used to monitor the polarization of the radioactive nuclear spin. At the implantation energy used here, the $^8Li^+$ ions stop at an average depth of 90 nm, far enough from the interface that the local response should be characteristic of the bulk, rather than the surface, where Rashba effects and reconstruction may play a role [20]. The main result of this work is that the $^8Li^+$ nuclear spin relaxation rate ($1/T_1$) is found to be very similar to the bulk planar ^{17}O data, with the temperature dependence explained by the incommensurate magnetic fluctuations seen in INS. Second, we found that the Knight shift $K(T)$ of $^8Li^+$ is more strongly temperature dependent than the ^{17}O or ^{101}Ru shifts, suggesting that a large orbital component may contribute to previously observed shifts. These results are an important demonstration that $^8Li^+$ β-NMR is a sensitive probe of the intrinsic metallic properties of a correlated-electron complex oxide.

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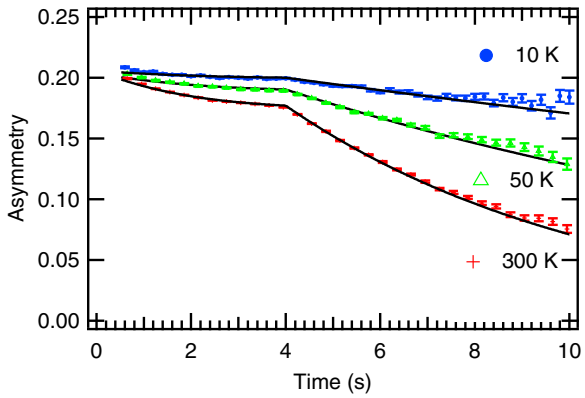

FIG. 1. (Color online) Measured $^8Li^+$ spin lattice relaxation in Sr_2RuO_4 at several temperatures. The pronounced kink is due to the end of the 4-s beam pulse. The fits shown are to a single exponential relaxation convoluted with the square beam pulse shape. A small fast-relaxing background signal at early times is neglected in the analysis.

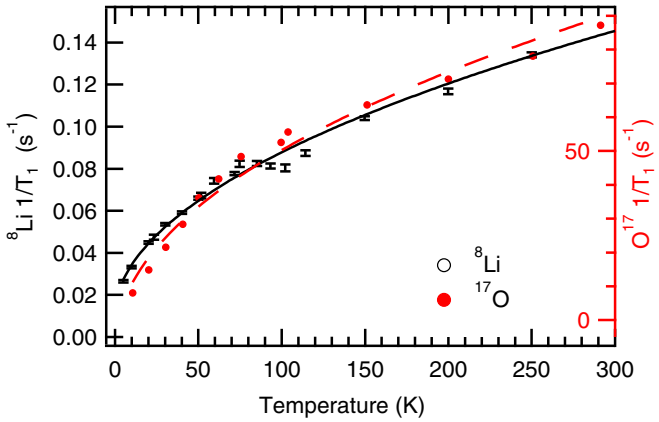

FIG. 2. (Color online) Comparison of the measured $1/T_1$ obtained using $^8Li^+$ β-NMR with the $1/T_1$ obtained from conventional NMR signal of the ^{17}O isotope for $H \parallel c$. The points are the experimental data, and the solid and dashed lines are fits to the temperature dependence expected for a nearly antiferromagnetic metal as described by Eq. (2).

In the experiment, a beam of radioactive $^8Li^+$ from the Isotope Separation and Acceleration (ISAC) facility at TRIUMF [21] was spin polarized in flight using collinear optical pumping [22] and implanted into a cleaved c-axis Sr_2RuO_4 platelet crystal, part of the one used in Ref. [23], measuring $\sim 3 \times 4 \times 0.5$ mm. The 20 keV polarized ion beam with an intensity of $\sim 10^6$ ions/s was focused onto a spot ~ 2 mm in diameter centered on the sample mounted on a sapphire plate in a cold-finger cryostat contained within a high-homogeneity superconducting solenoid at $B_0 = 6.55$ T $\parallel c$ [24]. The implantation profile was modeled with the Stopping and Range of Ions in Matter (SRIM) software [25,26]. As the nuclear polarization at implantation is already far from thermal equilibrium, unlike conventional NMR, no rf magnetic field is required for a SLR measurement. Instead, the β decay asymmetry is monitored as a function of time both during and after a 4-s beam pulse. The normalized asymmetry from a pair of detectors is proportional to the average spin polarization of the ensemble of implanted $^8Li^+$. Pulses of $^8Li^+$ with alternating polarization are collected for a measurement time of ~ 20 min to obtain sufficient statistics.

Typical SLR data are shown in Fig. 1. The relaxation is relatively slow, about half as fast as $^8Li^+$ in Pt at 300 K [27]. Upon cooling, it slows further but remains measurable at 5 K. The data are fit to a single exponential relaxation $[\exp(-t/T_1)]$ convoluted with the beam pulse shape [28] to extract the relaxation rate $1/T_1$ plotted in Fig. 2 as a function of temperature together with similar data for the planar oxygen ^{17}O site in a c-axis field from Ref. [8]. It is immediately apparent that, aside from the $\sim$500-fold reduced rate for $^8Li^+$, the temperature dependence is nearly identical, demonstrating the extrinsically implanted $^8Li^+$ probe senses the same excitations as the intrinsic ^{17}O. This is noteworthy given $^8Li^+$ represents a point defect which causes at least some local lattice strain and modified electron density. Therefore our result implies that the dynamic susceptibility and spin fluctuations in Sr_2RuO_4 are robust against such local perturbations.

The form of $1/T_1$ in Fig. 2 does not follow the linear Korringa law for a conventional metal, e.g., for $^8Li^+$ β-NMR [27,29–31]. Instead, we expect that spin fluctuations, reflecting strong spin correlations, make the dominant contribution to $1/T_1$; however, the character of these fluctuations is not known *a priori*. It is well established that Sr_2RuO_4 does not possess long-range magnetic order at any temperature, but the Stoner-enhanced susceptibility is close to a magnetic instability [15,32]. Although the system remains paramagnetic, it can order under a moderate degree of chemical substitution, resulting in either antiferromagnetism or ferromagnetism [32–36]. The delicate balance between competing forms of order is implied by the behavior of closely related compounds: the isostructural Ca_2RuO_4 is antiferromagnetic, whereas $SrRuO_3$ is a ferromagnetic metal. Early INS experiments showed that incommensurate AFM fluctuations produce the largest q-localized contribution to the dynamic susceptibility of Sr_2RuO_4 [15]. Subsequent studies revealed the existence of a broad background of low-q fluctuations [11,37]. As a magnetic sensor, a nuclear $^8Li^+$ spin relaxes according to the weighted average of both FM and AFM fluctuations [9], so it is informative to determine the dominant source of relaxation. Within a self-consistent renormalization picture, Moriya [38] has treated the effects of spin fluctuations on nuclear spin relaxation in nearly magnetic metals, finding $1/T_1$ has a differing temperature dependence for FM and AFM fluctuations. In the high-T limit, for the AFM case, $1/T_1$ is approximately proportional to $\sqrt{T}$, while in the FM case it is linear. The relaxation rates shown in Fig. 2 are fitted to

$$\frac{1}{T_1} = c\frac{T}{\sqrt{T - T_N}} \tag{2}$$

for $T > T_N$, where T_N is the Néel ordering temperature. While there is no magnetic transition, the fits describe the data well over a wide temperature range with the value of $T_N \to 0$ K (within 5 K) for both ^{17}O and $^8Li^+$. Past work demonstrated that Sr_2RuO_4 is close to a quantum critical point resulting in a characteristic ω/T scaling of χ'' [11]. At higher temperatures, the high-energy magnetic excitations detected by neutrons are described with an invariant ω/T scaling function; however,

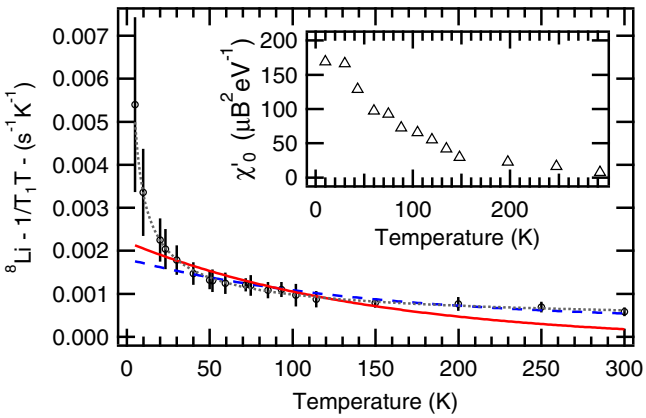

FIG. 3. (Color online) A plot of $1/T_1T$ vs temperature for $^8Li^+$ in Sr_2RuO_4 with $B_0 = 6.55$ T $\parallel c$ compared with the predicted temperature dependence (solid line) based on inelastic neutron scattering. The dashed line includes the addition of a Korringa-like term to the model susceptibility. The gray dotted line assumes that the relation $1/T_1 \propto T^\alpha$ survives below the Fermi liquid crossover. The inset shows the temperature dependency of the amplitude parameter taken from INS in Ref. [15] used to calculate the imaginary susceptibility via Eq. (3).

this relation is altered below the Fermi-liquid crossover (25 K) [11]. In contrast our result demonstrates the Fermi-liquid crossover does not modify the temperature scaling of the low-energy fluctuation spectrum detected by $^8Li^+$ and the intrinsic ^{17}O spin. Instead, the nuclear spins relax according to the fluctuation spectrum predicted by Moriya's theory for a nearly antiferromagnetic metal, thus motivating a more detailed comparison with the neutron result.

Following Sidis *et al.* [15], we use Eq. (1) and the AFM χ'' from INS to *calculate* $1/T_1$ for $^8Li^+$ using $\gamma_N = 6.301$ MHz/T for $^8Li^+$. From INS [15,16] the dynamic susceptibility is peaked at $\mathbf{q_0}$ and in energy is well described by a Lorentzian,

$$\chi''(\mathbf{q},\omega,T) = \chi_0'(\mathbf{q_0},T)\frac{\Gamma\omega e^{b(\mathbf{q}-\mathbf{q_0})^2/(\Delta_q)^2}}{(\omega^2+\Gamma^2)}, \qquad (3)$$

where the T dependence of the average response at $\mathbf{q_0}$, $\chi_0'(\mathbf{q_0},T)$, was determined from a higher-energy measurement (6.5 meV) extrapolated to zero energy [15] and is shown in the inset of Fig. 3. The spin susceptibility of Sr_2RuO_4 is significantly anisotropic. The in-plane fluctuations have a magnitude of $\chi_0'(\mathbf{q_0},0) = 140\mu_B$, whereas the out-of-plane fluctuations have a magnitude of $220\mu_B$. For $H \parallel c$ we expect $^8Li^+$ couples to the weaker in-plane fluctuations transverse to its spin via a weak isotropic hyperfine coupling. For this reason the values in the inset of Fig. 3, which represent the average of the two components measured with unpolarized neutrons [15], are reduced by the ratio $\frac{140}{180}$ in the calculation. The temperature dependency of the width of the excitation Δ_q and the damping energy Γ and the constant b are reported in Ref. [11]. The form factor $A(q)$ in Eq. (1) depends on the $^8Li^+$ site and its local hyperfine couplings; however, it is important to point out that regardless of the microscopic site, no site will systematically have $A(\mathbf{q_0}) = 0$ as $\mathbf{q_0}$ is incommensurate, so they should all sense the AFM fluctuations. Calculations show that the q-dependent form factor would only rescale the calculated rate, and therefore we adopt a simple effective pointlike coupling, $A(\mathbf{q}) = A$, and note that in a more accurate picture, A would represent the $A(\mathbf{q_0})$ tensor with contributions from both dipolar and hyperfine contact coupling. Under the assumption of a pointlike coupling, $A(\mathbf{q})$ factors out of the sum in Eq. (1), and to compute $1/T_1$ we obtain $\frac{\chi''}{\omega_0}$ using the parameters from the INS data [16]. For ^{17}O, the hyperfine form factor $A \approx 33$ kG/μ_B. The magnitude of the hyperfine coupling is not well known for $^8Li^+$, but if we assume a simple dipolar coupling with local Ru moments directed along the c axis, this provides a lower limit for the coupling of $^8Li^+$ in the interstitial Wyckoff c site (d site) $A \approx 1$ (2) kG/μ_B, which would likely be enhanced by a hyperfine contact interaction of similar magnitude [17]. Treating the magnitude of A as an adjustable parameter for $^8Li^+$, we infer that $A = 2.5 \pm 0.5$ kG/μ_B to account for the absolute relaxation rate, which is reasonable given the previous estimate. We plot $1/T_1T$ for $^8Li^+$ in Fig. 3 as a function of temperature together with the calculated value from Eq. (1) as outlined above. It is clear that sensitivity to the T-dependent incommensurate fluctuation accounts for the upturn in $1/T_1T$ observable below 100 K. An additional mechanism attributed to the background low-q fluctuations [9,11,37] is required to explain the finite relaxation at higher temperatures. To account for this second contribution, not explicitly included in Eq. (3), an additional constant term of $4.0 \pm 0.7 \times 10^{-4}$ s^{-1}K^{-1} is added to $1/T_1T$, corresponding to metallic Korringa relaxation from the γ band. Although we use this widely accepted model, it does not fully explain the sharp increase in the $^8Li^+$ $1/T_1T$ below the Fermi-liquid crossover (25 K). On the other hand, if the $\sqrt{T}$ scaling were preserved, this would predict the upturn shown by the gray dotted line in Fig. 3. Although the low-temperature relaxation is near the intrinsic limit imposed by the radioactive $^8Li^+$ lifetime, the $\sqrt{T}$ scaling is similar to that of the intrinsic ^{17}O data in Fig. 2. We note that the frequencies probed by thermal neutron spectroscopy are four to five orders of magnitude higher than those causing nuclear $1/T_1$, and although ω/T scaling is broken in the high-energy regime, a weaker form of scaling may survive in the low-frequency limit.

In order to better understand the $^8Li^+$ site, resonance measurements were conducted at several temperatures. Unlike the pulsed SLR measurements mentioned above, the following resonance measurements used a continuous $^8Li^+$ beam where the time-integrated polarization was destroyed using a transverse sinusoidal rf signal stepped through a frequency range, as in continuous wave (cw) NMR. Figure 4 shows typical resonance data. At 300 K, the resonance is split into a well-resolved multiplet of four small quadrupole satellites, corresponding to the $|\Delta m| = 1$ transitions for the spin $I = 2$ nucleus, interlaced with three sharp double-quantum resonances observable in cw mode [39–42]. The extra double-quantum resonances originate from the large time-dependent rf field which drives higher-order transitions [39,40] with an intensity that depends on the strength of the rf field. As with solid-state NMR, the electric quadrupole moment of the nucleus couples to the electric field gradient (EFG) at the site of the nucleus. The spectrum was fitted to a sum of Lorentzians with the splittings determined by a single parameter [43], the quadrupole frequency $\nu_Q = 7.539 \pm 0.019$ kHz at 300 K, which implies a single well-defined site is responsible for the resonance, as confirmed by recent complementary zero-field

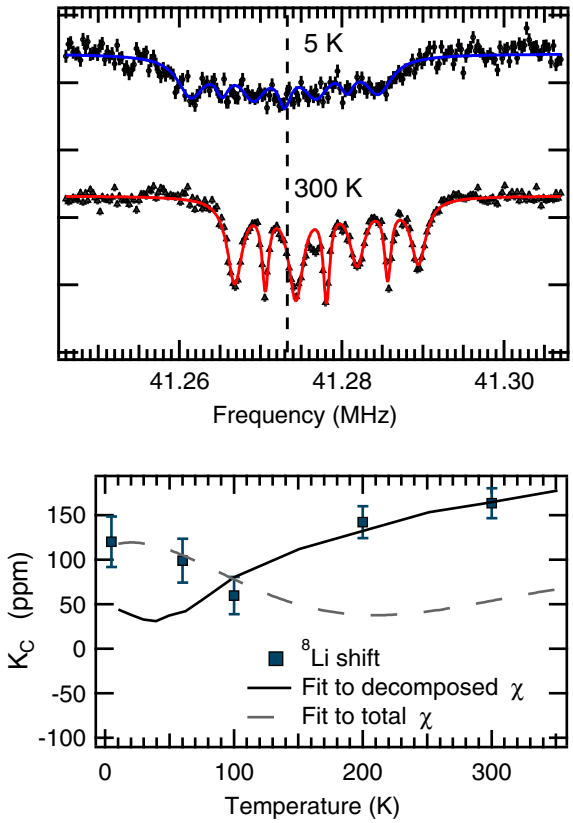

FIG. 4. (Color online) (a) Quadrupolar split resonance at 300 and 5 K in Sr_2RuO_4 (offset vertically for clarity). The solid curve is a fit to a sum of Lorentzians. The vertical line is the frequency of the Larmor frequency. (b) Summary of frequency shift as a function of temperature compared with the calculation based on hyperfine coupling to the three-component bulk susceptibility (solid line) and total susceptibility (dashed line).

measurements that showed only a single resonance line [44]. The observed quadrupole resonance shows a small electric field gradient with axial symmetry. Calculations using a point-charge model and density-functional theory show that the site that best fulfills the criteria is the Wyckoff d site for the space group $I4/mmm$ positioned at $(0, \frac{1}{2}, \frac{1}{4})$ between two SrO planes. At lower temperature, the component lines broaden slightly, reducing the intensity; however, the overall line shape has comparable width, suggesting that the site does not change. From the fits, we extract the resonance frequency ν from the center of the multiplet pattern and define the relative shift:

$$K = \frac{\nu - \nu_0}{\nu_0}, \tag{4}$$

where ν_0 is the reference resonance for $^8Li^+$ in MgO at 300 K in the same H_0. For light nuclei like $^8Li^+$ with generally small hyperfine couplings, the demagnetizing fields can contribute a significant fraction of the shift [45]. The correction for a thin plate sample is $(8\pi/3)\chi$ [46]. For Sr_2RuO_4 $\chi \approx 1 \times 10^{-3}$ emu/mol and varies only weakly in this temperature regime, giving a demagnetizing correction of about +115 ppm. We subtract this contribution using $\chi(T)$ from Ref. [6]. The resulting shift K_c expressed in parts per million is shown as a function of temperature in Fig. 4. In a simple metal, the shift K_c is related to the $q = 0$ hyperfine coupling A and the Pauli spin susceptibility χ_{Pauli}:

$$K_c = A\chi_{\text{Pauli}} + K_{\text{orb}}, \tag{5}$$

where χ_{Pauli} is independent of temperature, as found for $^8Li^+$ in simple elemental metals [29–31]. K_{orb} is a T-independent orbital shift that is small for $^8Li^+$. The magnetic susceptibility in Sr_2RuO_4 is only weakly temperature dependent, changing by ∼10% in the relevant temperature range [6], and thus coupling to the total $\chi(0,0)$ cannot explain the large changes observed in the $^8Li^+$ shifts. The Knight shifts for ^{17}O and ^{101}Ru show a substantial T-independent component added to the lesser T-dependent contribution with varying magnitudes and signs [8]. It is widely thought that a large fraction of this nearly isotropic χ corresponds to χ_{Pauli} [47] and the variation is driven by coupling to the anisotropic susceptibilities decomposed into the α and β bands. Recently, it was pointed out, however, that orbital contributions have likely been underestimated [17]. The $^8Li^+$ shift in Fig. 4 shows a much larger relative change than χ of any of the ^{17}O or ^{101}Ru shifts. A likely explanation for this is that χ_{Pauli} is only a small fraction of χ and K_{orb} is small for $^8Li^+$. The dashed line in Fig. 4(b) shows a fit to the data using Eq. (5) assuming a small hyperfine coupling of ≈ 1 kG/μ_B along with a negative T-constant term. This roughly describes the data below 100 K but fails at higher temperature. To investigate further we considered the possibility that $^8Li^+$ experiences different hyperfine couplings to each of the temperature-dependent susceptibility components associated with the different Sr_2RuO_4 bands:

$$K_c = A_{xz}\chi_{xz}(T) - A_{xy}\chi_{xy}(T) + K', \tag{6}$$

where $\chi_{xz}(T)$ is the susceptibility from the $4d_{xz/yz}$ orbitals responsible for the α/β bands and $\chi_{xy}(T)$ originates from the $4d_{xy}$ orbital responsible for the γ band. The choice of sign of the second term is arbitrary since it depends on electronic details of the transferred and dipolar hyperfine field [8]. Using values from Ref. [8] for χ_{xz} and χ_{xy} determined from ^{17}O shifts, we find that anisotropic changes in χ may explain the $^8Li^+$ shift above 75 K, as shown by the solid line in Fig. 4(b) with fitted hyperfine parameters $A_{xz} \approx 4$ kG/μ_B, $A_{xy} \approx 4$kG/μ_B, and $K' = 470$ ppm. The last term contains the effect of a large temperature-independent contribution including the contribution from the orbital part. No choice of hyperfine parameters, even with unconstrained signs, allows this model to describe the shifts below 75 K, perhaps because it relies on past decomposition of χ based on ^{17}O shift measurements which may be sensitive to isotope-specific orbital effects. Although a quantitative treatment of the Knight shift remains elusive, the same qualitative statement holds for both scenarios discussed: while the $^8Li^+$ relaxation rate is dominated by hyperfine coupling to the α/β bands, the Knight shift has a large contribution from the bulk γ band.

In summary, the application of the β-NMR technique shows that polarized $^8Li^+$ implanted in Sr_2RuO_4 senses the spin fluctuations of the host and gives analogous relaxation to the NMR of the intrinsic ^{17}O nuclei. This implies $^8Li^+$ β-NMR is a sensitive probe of low-frequency magnetic excitations and has the capability to detect both near-surface and bulk phenomena. The temperature dependence of the SLR at 90 nm from the Sr_2RuO_4 surface is driven by the incommensurate AFM fluctuations well known from the bulk material, similar

to the Moriya theory for a weak itinerant antiferromagnet and a quantitative model based on the experimental neutron spectroscopy. The $^{8}Li^{+}$ Knight shift shows a much stronger relative T dependence than the intrinsic ^{17}O NMR shifts, likely due to the modification of the large T-independent component, which may be orbital in origin.

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