Superconductivity in the regime of attractive interactions in the Tomonaga-Luttinger liquid

Ž. Gosar,¹ N. Janša,¹ T. Arh,¹ P. Jeglič,¹ M. Klanjšek,¹ H. F. Zhai,² B. Lv,² and D. Arčon¹,³

¹Institute Jožef Stefan, Jamova c. 39, 1000 Ljubljana, Slovenia
²The University of Texas at Dallas, 800 West Campbell Road Richardson, Texas 75080-3021, USA
³Faculty of mathematics and physics, University of Ljubljana, Jadranska c. 19, 1000 Ljubljana, Slovenia

While the vast majority of known physical realizations of the Tomonaga-Luttinger liquid (TLL) have repulsive interactions defined with the dimensionless interaction parameter \( K < 1 \), we here report that Rb₂Mo₃As₃ is in the opposite TLL regime of attractive interactions. This is concluded from a TLL-characteristic power-law temperature dependence of the \(^{87}\text{Rb} \) spin-lattice relaxation rates over broad temperature range yielding the TLL interaction parameter for charge collective modes \( K_c = 1.4 \). The TLL of the one-dimensional band can be traced down to \( T_c = 8 \) K (in 9.39 T), where unusual bulk superconducting state is stabilized by the presence of a 3D band and characterized by the temperature independent Knight shift, the absence of Hebel-Slichter coherence peak in the relaxation rates, and the small, possibly nodal, superconducting gap. The uniqueness of this superconducting state stems from the attractive interactions defining the precursor TLL and the weak disorder effects associated with the Rb positional disorder.

A universal paradigm of the Tomonaga-Luttinger liquid (TLL) describes the physics of interacting fermions in one dimension remarkably well and predicts their most characteristic features that can be directly verified in experiments: collective excitations which generally separate into spin and charge modes and a power-law decay of the spin and charge correlation functions at long distances that lead to power-law dependencies of the corresponding experimental quantities as a function of temperature or frequency. Experimentally, such power-law dependencies were taken as a hallmark of TLL physics in molecular conductors like TTF-TCNQ or Bechgaard salts (TMTSF)₂X³, carbon nanotubes [8, 9] and in one-dimensional antiferromagnetic insulators [10–12].

These physical realizations of the TLL may be very diverse, but their behavior is universal as the extracted power-law exponents are functions of only the TLL parameter \( K \), which characterizes the sign and the strength of interactions between collective excitation modes. While the interactions in the TLLs are generally repulsive with \( K < 1 \), the only example of attractive interactions with \( K > 1 \) has been found in the quantum spin ladder system \((C_7H_{10}N)_2CuBr_4\) [13, 14]. In one-dimensional metals, \( K \) depends on the strength of various scattering processes [15], which also define the competing orders: charge density wave, spin density wave or superconductivity. Remarkably, both the singlet and the triplet superconducting fluctuations are predicted [1 2] in the regime of attractive interactions. However, as most of the TLL candidates are characterized by \( K < 1 \) and since they often undergo a dimensional crossover from one dimension to three dimensions before they enter into the superconducting state [16], the emerging superconductivity in the \( K > 1 \) regime remains experimentally unexplored.

Here we focus on an intriguing class of solids, which are derived from the self-assembly of chains (or nanotubes) and for which coupling between chain/nanotube units is inherently weak. Molybdenum-chalcogenide (Li₂MoS₂, Na₂₋₄Mo₆Se₈) and molybdenum-oxide (Li₀₉₉Mo₅O₁₇) nanotubes/chains [17, 20] display many unusual properties associated with TLLs, but at the same time they also show a high degree of disorder due to the intercalation of metallic atoms between the nanotubes, which mask the TLL and superconducting properties. The disorder effects seem to be less important in Cr-pnictide quasi-one-dimensional metals, e.g., \( A₂Cr₃As₃ \) (\( A = K, Rb, Cs \)) with Cr₃As₃ chains as main building units, which remarkably also show low-temperature superconductivity with the critical temperature \( T_c \approx 5 \) K [21, 29]. There seems to be a growing consensus for the unconventional singlet superconducting states in this family of materials based on the large specific-heat jump at \( T_c \) and large upper critical fields exceeding Pauli limit [21, 31], absence of the Hebel-Slichter coherence peak and the power-law dependence of nuclear spin-lattice relaxation rates [24, 29] and the penetration depth proportional to temperature [25] that implies the nodal-type of a superconducting gap. Moreover, a possible ferromagnetic quantum critical point has been proposed based on the tuning of the nuclear spin-lattice relaxation rates by the Cr-As-Cr angle [31]. However, the complicated electronic structure comprising two quasi-one-dimensional (1D) and one three-dimensional (3D) Fermi surface [32] and the presence of a Korringa component in the nuclear spin-lattice relaxation cast some doubts on the direct applicability of the TLL model for \( A₂Cr₃As₃ \) [24].

Recently, superconductivity has been discovered in \( K₂Mo₂As₃ \) made of assembled Mo₂As₃ chains (Fig. 1D) with a relatively high zero-field \( T_c(0) = 10.4 \) K [33]. Although \( A₂Mo₃As₃ \) are isostuctural to their Cr counterparts \( A₂Cr₃As₃ \) and the first principle calculations suggest that they share similar electronic structure [34], the \( 4d \) character of electrons and stronger spin-orbit coupling...
and 130 samples of Rb state, and measured sample show phase pure sample with the A powder diffraction measurements on the polycrystalline type structure – hexagonal crystal lattice with a space used to synthesize A field (blue), 5 T (green) and 9 T (red).

superconductivity develops, but the insensitivity of the Quantum Design PPMS system. The Rb the reduced superconducting gap may separate this family of superconductors into its own class. Here we report on the normal and superconducting state in Rb$_2$Mo$_3$As$_3$ and find some fundamental differences with respect to the family of A$_2$Cr$_3$As$_3$. Strikingly, TLL of the 1D component of the Fermi surface in which effective attractive interactions between collective charge excitations prevail can be followed in $^{87}$Rb nuclear magnetic resonance (NMR) experiments over an extremely broad temperature range between 200 K and $T_c$. On the other hand, $^{75}$As nuclei couple more strongly to the 3D band. Directly from such a multi-orbital state, superconductivity develops, but the insensitivity of the $^{87}$Rb Knight shift to the onset of the superconducting state, the absence of Hебel-Slichter coherence peak and the reduced superconducting gap even in a moderate field imply its unconventional character.

Powder samples of Rb$_2$Mo$_3$As$_3$ were prepared via standard high-temperature solid state method previously used to synthesize A$_2$Cr$_3$As$_3$ samples [22, 23]. X-ray powder diffraction measurements on the polycrystalline sample show phase pure sample with the A$_2$Cr$_3$As$_3$-type structure - hexagonal crystal lattice with a space group of $P6_3/m2$ [21]. The sample was further investigated by dc magnetic susceptibility measurements in the zero-field-cooled (ZFC) and field-cooled (FC) modes at $\mu_0 H = 1$ mT (Fig. 1b) using VSM option of a Quantum Design PPMS system. The Rb$_2$Mo$_3$As$_3$ compound indeed shows a bulk superconductivity below zero-field $T_c(0) \approx 10$ K with the large diamagnetic shielding $\Delta T \chi_{ZFC}$ of 97% at 2 K demonstrating sample’s high quality. Next, the temperature dependence of the zero-field resistivity, $\rho(T)$, was measured (Fig. 1b) using Quantum Design PPMS system. Initially, the resistivity at high temperatures decreases with decreasing temperature, but then it shows a minimum at $T_{min} \sim 210$ K. Below $T_{min}$, $\rho(T)$ starts to increase with decreasing temperature. Just above $T_c(0) = 10.4$ K, $\rho(T)$ first almost flattens as a function of temperature and then it sharply drops to zero where the superconducting state sets in. The normal-state increase in $\rho(T)$ with decreasing temperature below $T_{min}$ may imply the weak localization effects due to the presence of disorder and Coulomb repulsion [2, 15]. Measurements of $\rho(T)$ in the magnetic field show a suppression of superconductivity and the corresponding decrease of $T_c$ with increasing magnetic field (Fig. 1c). In the field of 9 T, the critical temperature decreases to $T_c \approx 8$ K. The magnetization and resistivity data thus confirm the high quality of the sample and the emerging superconducting state, but could not decide alone on the quasi-one-dimensional electronic structure and the potential TLL physics in this material nor on the symmetry of the superconducting state.

We thus turn to $^{87}$Rb (nuclear spin $I = 3/2$) NMR, which was measured in magnetic fields of 4.7 T and 9.39 T with the corresponding reference Larmor frequencies of $\nu_L = 65.442$ MHz and 130.871 MHz, respectively. We note that Rb atoms are intercalated into the voids between the Mo$_3$As$_3$ chains (Fig. 1a) and thus couple to the intrinsic intra-chain low-energy dynamics through the contribution of $xy$ dispersive Mo 4$d$ bands. As shown in Fig. 2a, the $^{87}$Rb NMR spectrum measured in 9.39 T is composed of a narrow peak corresponding to the central 1/2 $\leftrightarrow$ −1/2 transition, which is shifted for $\sim 56$ kHz with respect to the reference frequency, and the two broadened satellite peaks that symmetrically flank the central one and which yield the quadrupole frequency $\nu_Q \approx 280$ kHz. The second-order quadrupole shift of the central transition line, $\delta \nu \propto \nu_Q^2/\nu_L$, is thus small. The measured line shift must therefore come almost entirely from the hyperfine coupling to the itinerant electrons, i.e., the Knight shift $K_{87}$. In addition to this $^{87}$Rb resonance, we find also a broader component with a very small shift and whose satellite transitions cannot be resolved (Fig. 2a). The two-component NMR spectrum reflects two Rb sites (at the 3k and 1c positions [21]. Fig. 1a) in the structure, where only one of them (most probably Rb1 at the 3k position) strongly couples to the Mo$_3$As$_3$ chains. We note that the analogous two-component NMR spectrum has been observed also in $^{133}$Cs NMR of Cs$_2$Cr$_3$As$_3$ [24].

The central transition Rb1 peak strongly shifts with temperature between room temperature and $T_c$. More specifically, on cooling the shift decreases from the room
FIG. 2. (a) $^{87}$Rb powder NMR spectra in Rb$_2$Mo$_3$As$_3$. In the field of 9.39 T the reference Larmor frequency is $\nu_L = 130.871$ MHz. Top: the full spectrum measured at $T = 200$ K. Arrows indicate the two satellite transitions at $\pm \nu_Q$ relative to the Rb1-site central 1/2 ↔ 1/2 transition. The region of the non-shifted Rb2 component is also indicated. Bottom: the full temperature dependence of the central transition line of Rb1. (b) Temperature dependence of the $^{87}$Rb NMR Knight shift for Rb1 site. Temperature dependencies close to $T_c$ for the $^{87}$Rb NMR Knight shift (c) and for the integrated signal intensity multiplied by temperature (d). Please note, that $T_c \approx 8$ K (marked by dashed vertical lines) in the NMR field of 9.39 T.

Temperature value of $\sim 56$ kHz to $33$ kHz at $T = 50$ K and then becomes almost temperature independent between 50 K and $T_c$. Since the Knight shift, $K_{87} = \frac{a_{\text{Rb}}}{N_A \mu_B} \chi(T)$ (here $a_{\text{Rb}}$, $N_A$ and $\mu_B$ are the $^{87}$Rb hyperfine coupling constant, the Avogadro number and the Bohr magneton, respectively), is directly proportional to the contribution to the static spin susceptibility from a band that dominantly couples to $^{87}$Rb, we conclude that associated intrinsic susceptibility $\chi(T)$ is also significantly temperature dependent. This would be highly unusual for the Pauli susceptibility of the Fermi liquid in two- and three-dimensions, as there the temperature dependence shows up only on the scale of Fermi energy. On the other hand, large and temperature dependent $\chi(T)$ is not uncommon to one-dimensional metals [17]. Calculations for the Hubbard model of interacting electrons in one dimension in fact predict a maximum in $\chi(T)$ at $T \sim 0.1t$ (t is the hopping matrix element) followed by an inflection at $T \sim 0.1t$ [22]. While the comparison between the experimental and theoretical data can be at this stage only at a qualitative level, the temperature dependent $K_{87}$ implies the dominant coupling of $^{87}$Rb to the 1D bands and explains why $K_{87}$ does not follow bulk susceptibility (Fig. S1 in [30]) with contributions from both 1D and 3D bands [34].

In order to quantitatively discuss the possible TLL physics, we switch to the $^{87}$Rb spin-lattice relaxation rate (Fig. 3), $1/T_1$, which is proportional to the sum of the imaginary part of the electron spin susceptibility $\chi''(q, \omega_L)$ over the wave vector $q$ and calculated at $\omega_L = 2\pi \nu_L$: $1/T_1 \propto \sum_q |a_{\text{Rb}}(q)|^2 \chi''(q, \omega_L)$. This makes $1/T_1$ an extremely sensitive probe of the low-energy dynamics. Upon cooling from the room temperature, $1/T_1$ has a weakly pronounced field-dependent maximum at $T \sim 250$ K (Fig. S2 in [30]), which coincides with the minimum in $\rho(T)$ and indicates the freezing out of the Rb$^+$ ion dynamics, most probably of Rb at the 1c position where the alkali metal vacancies are primarily located [32]. However, once such dynamics is completely frozen on the time-scale given by $\omega_L$ below 200 K, a clear power-law temperature dependence of $1/T_1 \propto T^p$ is observed in 4.7 T and 9.39 T over more than a decade in temperature between $T_c$ and 200 K. This is a signal that the low-energy dynamics is described by the critical phenomena of collective modes and is thus a hallmark of the TLL. $1/T_1$ shows no contribution from the ferromagnetic spin fluctuations similar to those recently reported for the superconducting $\text{A}_2\text{Cr}_3\text{As}_3$ [31]. Finally, $1/T_1$ data does not follow the Fermi-liquid-like Korringa relaxation behavior 1/T1T $\propto K_{87}^2$ either, which implies that the 3D part of the Fermi surface has a negligible contribution to the $^{87}$Rb spin-lattice relaxation in Rb$_2$Mo$_3$As$_3$. The spin-lattice relaxation is thus completely dominated by the two 1D bands, which are responsible for the TLL behavior. On the other hand, complementary $^{75}$As $1/T_1$ zero-field nuclear quadrupole resonance (NQR) measurements (Figs. S3 and S4 in [30]) indeed show 1/T1 $\propto T$ (Fig. 3) and suggest that $^{75}$As sitting in the Mo$_3$As$_3$ chains predominantly couple to the part of the Fermi surface with a 3D character. Combined $^{87}$Rb and $^{75}$As spin-lattice relaxation rates thus corroborate multi-band physics [34] and the interplay of 1D and 3D bands in the studied system.

The power-law fit of $^{87}$Rb $1/T_1$ yields $p = 1.4$ for both 4.7 T and 9.39 T measurements. This is dramatically different from $p \approx 0.75$, determined in K$_2$Cr$_3$As$_3$ [23] or $p = 0.34$ in single-wall carbon nanotubes [3] and immediately suggests that 1D bands of Rb$_2$Mo$_3$As$_3$ are in a different TLL regime. In general, TLL is described by separate spin and charge collective modes, each of these described by the corresponding TLL interaction parameters $K_s$ and $K_c$, respectively. Assuming the spin invariance in low fields, which gives $K_s = 1$, we focus only on the charge mode, which is affected by the interactions [33]. For the single-band TLL case, $1/T_1T \propto T^{\eta/2-2}$ is predicted, where the power law exponent $\eta = 2K_c + 2$ holds the information about the TLL parameter for the kF spin fluctuations. From the determined power-law exponent we get $K_c = p = 1.4$, which puts the studied system into the regime with dominant attractive interactions. This is in striking contrast to K$_2$Cr$_3$As$_3$, where the same analysis gives $K_c = 0.75$ and thus implies more conven-
the temperature dependence of $^{87}\text{Rb}\ 1/T_1$, the onset of superconductivity is seen as a sudden suppression of $1/T_1$ (Fig. 1b) without any Hebel-Slichter coherence peak, which would be a hallmark of the conventional isotropic Bardeen-Cooper-Schrieffer (BCS) s-wave superconductivity. The absence of the Hebel-Slichter coherence peak is common to unconventional superconductors, including related iron-pnictide superconductors [11] and $\text{A}_2\text{Cr}_3\text{As}_3$ [28, 31]. In $\text{Rb}_2\text{Mo}_3\text{As}_3$, $1/T_1$ apparently follows an activated temperature dependence, $1/T_1 \propto \exp(-\Delta_0/k_\text{B}T)$ down to $T_c/T = 2.7$ (Fig. 1b). Attempts to fit $1/T_1$ to the power law $T^n$ for $T_c > T > T_c/2.7$ with $n \sim 3 - 5$ result in a worse fit – for example, at the lowest temperature the measured value of $1/T_1$ is larger by a factor of $\sim 5$ compared to what is expected for such a power-law dependence. The final verdict on the form of $^{87}\text{Rb}\ 1/T_1$ temperature dependence in the superconducting state can be only reached by extending measurements to lower temperatures. Nevertheless, we use the fitting of $1/T_1$ to the activated temperature dependence to estimate the superconducting gap $\Delta_0 \approx 0.9$ meV (Fig. 1d and Table I in [29]). Using $T_c \approx 8$ K deduced from the onset of superconductivity in $\rho(T)$ (Fig. 1d), we can then calculate the energy gap $T_c$ ratio $2\Delta_0/k_\text{B}T_c = 2.4$. Equivalent analysis and conclusions hold also for measurements in $4.7$ T. This is inconsistent with the standard BCS prediction in the weak coupling limit for which $2\Delta_0/k_\text{B}T_c \geq 3.52$. Remarkably, the temperature dependence of the zero-field $1/T_1$ for the $^{75}\text{As}$, which couples to the 3D part of the Fermi surface, can still be described by the thermally activated dependence, but with a larger $\Delta_0 = 1.6$ meV and $2\Delta_0/k_\text{B}T_c = 3.7$. No Hebel-Slichter coherence peak has been found in this case, too.

The smallness of the extracted $\Delta_0$ for the $^{87}\text{Rb}$ coupled to the 1D bands either indicates (i) the presence of nodes in the superconducting gap, which would comply also with the spin-triplet superconductivity or (ii) the quenched disorder introduced near the superconductor-to-insulator transition when the length scale of the inhomogeneity is comparable to or larger than the coherence length $\xi_\text{c}$. In the former case, $\text{Rb}_2\text{Mo}_3\text{As}_3$ would be the best of our knowledge the first example of a triplet superconductor developed from the TLL characterized by $K_c > 1$. In the latter case, the source of disorder could be connected to the $\text{Rb}^+\text{1c}$ sites, whereas the effective attractive interactions within the TLL place this system very close to the metal(superconductor)-to-insulator critical point. The increase in $\rho(T)$ with decreasing temperature below $T_{\min}$ (Fig. 1c) seems to corroborate this suggestion. Although further experiments are needed to reveal the true nature of the superconducting state, the temperature independent Knight shift, the absence of the Hebel-Slichter coherence peak and the smallness of the extracted superconducting gap signify unconventional character of the bulk superconducting state that develops from the TLL in the regime of attractive interactions.

The onset of superconductivity below $T_c \approx 8$ K is barely seen in the line shift of the $^{87}\text{Rb}$ NMR spectra measured in $9.39$ T (Fig. 2a), but can be deduced from the wipeout effect (Fig. 2b). The absence of a noticeable shift upon cooling through the superconducting transition may imply the elusive spin-triplet superconductivity [9]. If this is the case, then the superconductivity should persist at fields exceeding the paramagnetic limit, as it was indeed previously observed also in $\text{A}_2\text{Cr}_3\text{As}_3$ samples [21, 30]. However, in triplet superconductors, the spin-wave contribution leads to yet another power-law scaling of $1/T_1$ with temperature below $T_c$ [40].

The zero-field $2\Delta_0/k_\text{B}T_c \approx 8$ K is $\approx 3.5$ in $\text{Rb}_2\text{Mo}_3\text{As}_3$. In $\text{Rbg}_{2/3}\text{Si}_{1/3}$, the onset of superconductivity is seen as a sudden suppression of $1/T_1$ (Fig. 1b) without any Hebel-Slichter coherence peak, which would be a hallmark of the conventional isotropic Bardeen-Cooper-Schrieffer (BCS) s-wave superconductivity. The absence of the Hebel-Slichter coherence peak is common to unconventional superconductors, including related iron-pnictide superconductors [11] and $\text{A}_2\text{Cr}_3\text{As}_3$ [28, 31]. In $\text{Rb}_2\text{Mo}_3\text{As}_3$, $1/T_1$ apparently follows an activated temperature dependence, $1/T_1 \propto \exp(-\Delta_0/k_\text{B}T)$ down to $T_c/T = 2.7$ (Fig. 1b). Attempts to fit $1/T_1$ to the power law $T^n$ for $T_c > T > T_c/2.7$ with $n \sim 3 - 5$ result in a worse fit – for example, at the lowest temperature the measured value of $1/T_1$ is larger by a factor of $\sim 5$ compared to what is expected for such a power-law dependence. The final verdict on the form of $^{87}\text{Rb}\ 1/T_1$ temperature dependence in the superconducting state can be only reached by extending measurements to lower temperatures. Nevertheless, we use the fitting of $1/T_1$ to the activated temperature dependence to estimate the superconducting gap $\Delta_0 \approx 0.9$ meV (Fig. 1d and Table I in [29]). Using $T_c \approx 8$ K deduced from the onset of superconductivity in $\rho(T)$ (Fig. 1d), we can then calculate the energy gap $T_c$ ratio $2\Delta_0/k_\text{B}T_c = 2.4$. Equivalent analysis and conclusions hold also for measurements in $4.7$ T. This is inconsistent with the standard BCS prediction in the weak coupling limit for which $2\Delta_0/k_\text{B}T_c \geq 3.52$. Remarkably, the temperature dependence of the zero-field $1/T_1$ for the $^{75}\text{As}$, which couples to the 3D part of the Fermi surface, can still be described by the thermally activated dependence, but with a larger $\Delta_0 = 1.6$ meV and $2\Delta_0/k_\text{B}T_c = 3.7$. No Hebel-Slichter coherence peak has been found in this case, too.

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![FIG. 3](image-url). Temperature dependence of the zero-field $^{75}\text{As}$ (blue triangles) and $^{87}\text{Rb}$ spin-lattice relaxation rates $1/T_1$ measured in $4.7$ T (green squares) and $9.39$ T (red circles) for the $\text{Rb}_2\text{Mo}_3\text{As}_3$ powder. The power-law dependence $1/T_1 \propto T^p$ with $p = 1.4$ (solid line) for $^{87}\text{Rb}$ data demonstrates the TLL physics of 1D bands. For comparison, linear and quadratic dependencies are denoted with dotted lines, respectively. $^{75}\text{As}$ nuclei, which are predominantly coupled to the 3D electronic band, show the conventional $1/T_1 \sim T$ behavior (dashed line).
and is stabilized by the presence of the 3D band.

In conclusion, we have studied Rb$_2$Mo$_3$As$_3$ composed of the assembly of weakly coupled Mo$_3$As$_3$ chains and showed that the 1D bands adopt TLL behavior over a strikingly broad temperature range between 200 K and 8 K. A power-law dependence of $^{87}$Rb $1/T_1$ to an activated temperature dependence yielding $2\Delta_0/k_B T_c \approx 2.4$, while solid line is a fit of $^{75}$As $1/T_1$ to the same model with $2\Delta_0/k_B T_c \approx 3.7$ (Table I in [36]). The power-law dependence, $1/T_1 \propto T^4$, anticipated for nodal-type superconductivity is shown as a dotted line.

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Superconductivity in the regime of attractive interactions in the Tomonaga-Luttinger liquid
Supplemental Material

ž. Gosar,¹ N. Janša,¹ T. Arh,¹ P. Jeglič,¹ M. Klanjšek,¹ H. F. Zhai,² B. Lv,² and D. Arčon¹,³,

¹Institute Jožef Stefan, Jamova c. 39, 1000 Ljubljana, Slovenia
²The University of Texas at Dallas, 800 West Campbell Road Richardson, Texas 75080-3021, USA
³Faculty of mathematics and physics, University of Ljubljana, Jadranska c. 19, 1000 Ljubljana, Slovenia
TABLE I. Summary of fitting parameters for the spin-lattice relaxation rates in the superconducting state of $\text{Rb}_2\text{Mo}_3\text{As}_3$. The critical temperature $T_c$ has been taken as an onset of the superconducting transition in the resistivity measurements (Fig. 1d in the main text), while the superconducting gap $\Delta_0$ has been determined from the fits of the spin-lattice relaxation rates to the thermally activated behavior (Fig. 4b in the main text).

| Nuclei | Magnetic field (T) | $T_c$ (K) | $\Delta_0$ (meV) | $2\Delta_0/k_B T_c$ |
|--------|--------------------|-----------|------------------|-------------------|
| $^{75}\text{As}$ | 0 | 10 | 1.62 | 3.7 |
| $^{87}\text{Rb}$ | 4.7 | 9 | 0.89 | 2.4 |
| $^{87}\text{Rb}$ | 9.39 | 8 | 0.83 | 2.4 |

FIG. 1. Molar susceptibility of $\text{Rb}_2\text{Mo}_3\text{As}_3$ powder sample measured with the zero-field-cooling protocol and in the 0.1 T magnetic field. Quantum Design MPMS magnetometer has been used in these experiments.
FIG. 2. $^{87}$Rb $1/T_1$ measured in Rb$_2$Mo$_3$As$_3$ powder sample shows enhancement in a lower magnetic field around the maximum, which is expected for the Bloembergen-Purcell-Pound-type relaxation.
FIG. 3. $^{75}$As NQR spectrum of $\text{Rb}_2\text{Mo}_3\text{As}_3$ powder measured at $T = 20$ K. Thin red, green and magenta lines are individual Lorentzian components used to deconvolute $^{75}$As NQR spectra. Solid thick blue line is their sum. Arrow indicates the frequency at which spin-lattice relaxation rate measurements were conducted.
FIG. 4. The $^{75}$As NQR signal intensity as a function of delay time $\tau$ in the spin-lattice relaxation rate measurements. Standard inversion-recovery pulse sequence $\pi - \tau - \pi/2 - \tau_1 - \pi - \tau_1 - \text{echo}$ (pulse lengths were typically $\pi/2 = 3.1$ $\mu$s and the delay time $\tau_1 = 100$ $\mu$s) was used. The $^{75}$As magnetization recovery curve was fitted to the expression for $^{75}$As $I = 3/2$: $M_z(\tau) = M_0 (1 - (1 + s) \exp[-(3\tau/T_1)^\alpha])$. Here $M_0$ is the signal amplitude, $s$ measures the efficiency of the nuclear magnetization inversion, and $\alpha \approx 0.9$ is the stretching exponent.