

Thermodynamic Evidence for Pressure-Driven Evolution towards Weak-Coupling Superconductivity in Pb

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The thermodynamic critical field B_c provides direct access to the superconducting condensation energy, yet its pressure dependence has been studied much less extensively than that of the transition temperature. Here, muon-spin-rotation relaxation measurements of the thermodynamic critical field B_c of elemental Pb under hydrostatic pressure up to ≈ 2.3 GPa are reported. From the magnetic-field distribution in the intermediate state, $B_c(T)$ is determined and $B_c(0)$ is extracted at different pressures, thereby providing direct thermodynamic access to the pressure evolution of the condensation-energy scale. It is shown that, within the experimentally accessible pressure range, the pressure dependence of $B_c(0)$ follows that of the superconducting gap $\Delta(0)$ more closely than that of the transition temperature T_c . The measured pressure derivative of the coupling ratio $\alpha = \Delta(0)/(k_B T_c)$ is furthermore found to agree with the difference between the pressure derivatives of $B_c(0)$ and T_c , providing a direct thermodynamic confirmation of the relation between these quantities. Together, these results show that pressure suppresses strong-coupling effects in Pb. When viewed together with previously reported high-pressure data for B_c and T_c , the present results indicate that this tendency continues toward a regime in which α becomes nearly pressure independent.

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Introduction—Understanding how the characteristic energy scales of a superconductor evolve under external pressure provides important insight into the microscopic mechanisms governing superconductivity [1–4]. In conventional phonon-mediated superconductors, pressure modifies both the electronic structure and the lattice dynamics, typically leading to phonon hardening and a reduction of the electron-phonon coupling strength [3–6]. As a consequence, compression can drive the system gradually from the strong-coupling regime toward the weak-coupling Bardeen-Cooper-Schrieffer (BCS) limit [7]. Experimental signatures of such a crossover are usually inferred from the pressure dependence of the superconducting transition temperature T_c . However, T_c is determined by the linearized gap equation and reflects a delicate balance between competing effects, including phonon hardening and changes in the electron-phonon interaction [5,6,8]. Thermodynamic quantities that probe the condensation energy can therefore provide a more direct view of the superconducting energy scale [7]. In this context, direct pressure measurements of thermodynamic quantities are particularly valuable, since they can reveal how the

condensation-energy scale evolves under compression and thereby provide information that is not contained in T_c alone.

One such quantity is the thermodynamic critical field B_c , which is directly related to the free-energy difference between the superconducting and normal states [9,10]. The corresponding superconducting condensation-energy density is given by

$$U_0 = \frac{B_c^2(0)}{2\mu_0}, \quad (1)$$

where $B_c(0)$ is the zero-temperature thermodynamic critical field. Within weak-coupling BCS theory, the condensation energy can also be written as [9–12]

$$U_0 = \frac{1}{2} N(E_F) \Delta^2(0) = \frac{3}{4\pi^2 k_B^2} \gamma_e \Delta^2(0), \quad (2)$$

where $N(E_F)$ is the electronic density of states at the Fermi level, $\Delta(0)$ is the superconducting energy gap at zero temperature, k_B is the Boltzmann constant, and $\gamma_e = (2\pi^2/3)k_B^2 N(E_F)$ is the Sommerfeld coefficient. For simplicity, Eqs. (1) and (2) are written in terms of a single gap value $\Delta(0)$. More generally, for an anisotropic superconductor this quantity should be understood as an orientation-dependent thermodynamic gap scale in a single crystal, or as the corresponding Fermi-surface average in the polycrystalline case considered here.

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Combining Eqs. (1) and (2) yields the approximate scaling relation

$$B_c(0) \propto \Delta(0)\sqrt{\gamma_e}, \quad (3)$$

showing that the thermodynamic critical field is governed by both the superconducting gap and the electronic density of states. Equation (3) should be understood as an approximate thermodynamic scaling relation: $B_c(0)$ is governed predominantly by the superconducting energy scale $\Delta(0)$, while the contribution from the electronic specific-heat coefficient γ_e represents a weaker correction.

It is convenient to express the gap in terms of the dimensionless ratio

$$\alpha = \frac{\Delta(0)}{k_B T_c}. \quad (4)$$

The parameter α is commonly used as a measure of coupling strength via comparison with the weak-coupling BCS value $\alpha_{\text{BCS}} \equiv 1.764$. Values significantly above α_{BCS} indicate enhanced strong-coupling effects, whereas values close to α_{BCS} correspond to the weak-coupling limit. Combining Eqs. (3) and (4) gives the relation between the logarithmic pressure derivatives of B_c , T_c , γ_e , and α ,

$$\frac{d \ln B_c(0)}{dp} = \frac{d \ln T_c}{dp} + \frac{1}{2} \frac{d \ln \gamma_e}{dp} + \frac{d \ln \alpha}{dp}.$$

In simple metals, the pressure dependence of the electronic specific-heat coefficient γ_e is generally weak compared with that of T_c and B_c [2–4,13]; a quantitative estimate for Pb is given in Supplemental Material [14]. Therefore, to a good approximation, the difference between the pressure derivatives of B_c and T_c reflects the pressure evolution of the gap ratio,

$$\frac{d \ln \alpha}{dp} \simeq \frac{d \ln B_c(0)}{dp} - \frac{d \ln T_c}{dp}. \quad (5)$$

This relation shows that the pressure dependences of the thermodynamic critical field and T_c provide direct information on the pressure evolution of the gap ratio α . If, in addition, α is found to decrease with pressure, the pressure evolution of B_c provides thermodynamic evidence for the suppression of strong-coupling effects. When this tendency is considered over a sufficiently broad pressure range, it may be interpreted as an evolution toward the weak-coupling limit.

In this Letter, muon-spin-rotation relaxation (μ SR) is used to determine the thermodynamic critical field of elemental Pb under hydrostatic pressure up to ≈ 2.3 GPa. Unlike transport- or susceptibility-based determinations of field-dependent phase boundaries, μ SR in the intermediate state provides direct access to the equilibrium

thermodynamic critical field B_c through the internal magnetic-field distribution, making it possible to follow the pressure evolution of the condensation-energy scale directly rather than inferring superconducting energy scales from T_c alone. Analysis of the temperature dependence $B_c(T)$ within the phenomenological α model [28,29] yields the pressure evolution of the key superconducting parameters T_c , $B_c(0)$, $\Delta(0)$, and $\alpha = \Delta(0)/(k_B T_c)$. It is shown that, in the investigated pressure range, the pressure dependence of $B_c(0)$ follows that of the superconducting gap more closely than that of the transition temperature, and that the pressure derivative of α is directly linked to the difference between the pressure derivatives of $B_c(0)$ and T_c . These results provide direct thermodynamic evidence that pressure suppresses strong-coupling effects in Pb. The comparison with previously reported high-pressure data [30] is then used to show that this low-pressure trend continues at higher pressures.

Experimental details—Details on sample preparation, transverse-field (TF) μ SR experiments under pressure, the analysis of μ SR data, and the α model used to describe the $B_c(T, p)$ dependence are provided in Supplemental Material [14].

Experimental data—The TF- μ SR measurements were carried out in the intermediate state, i.e., when the Pb sample separates into superconducting (Meissner) and normal-state domains [10,31–40]. In this state, the magnetic induction in the superconducting domains is zero, whereas in the normal-state domains it is equal to the thermodynamic critical field, B_c [9,10]. This makes TF- μ SR a particularly direct probe of $B_c(T)$, since the field distribution measured by the implanted muons contains a distinct contribution from the normal domains at $B = B_c$ [35–46]. It should be noted that, at finite field, the superconducting-to-normal transition in a type-I superconductor is first order along the equilibrium line $B = B_c(T)$. In the present experiment, however, no separate correction is required, since the μ SR signal determines the equilibrium thermodynamic critical field directly from the local field in the normal domains.

Representative Fourier transforms of the muon-time spectra, corresponding to the magnetic-field distribution in the pressure-cell sample assembly at $p = 2.34$ GPa, are shown in Fig. 1(a). The field distribution consists of three well-defined peaks. The peaks at $B = 0$ and $B = B_c$ correspond to muons stopping in superconducting (Meissner-state) and normal-state domains of the Pb sample, respectively. The peak at the applied-field position, $B = B_{\text{ap}}$, originates from muons stopping in the pressure-cell walls and precessing in the applied magnetic field. Such a three-peak field distribution is characteristic of TF- μ SR measurements in the intermediate state of a type-I superconductor and directly illustrates how the equilibrium thermodynamic critical field is obtained experimentally [39–42,45,47]. In contrast to conventional magnetic or

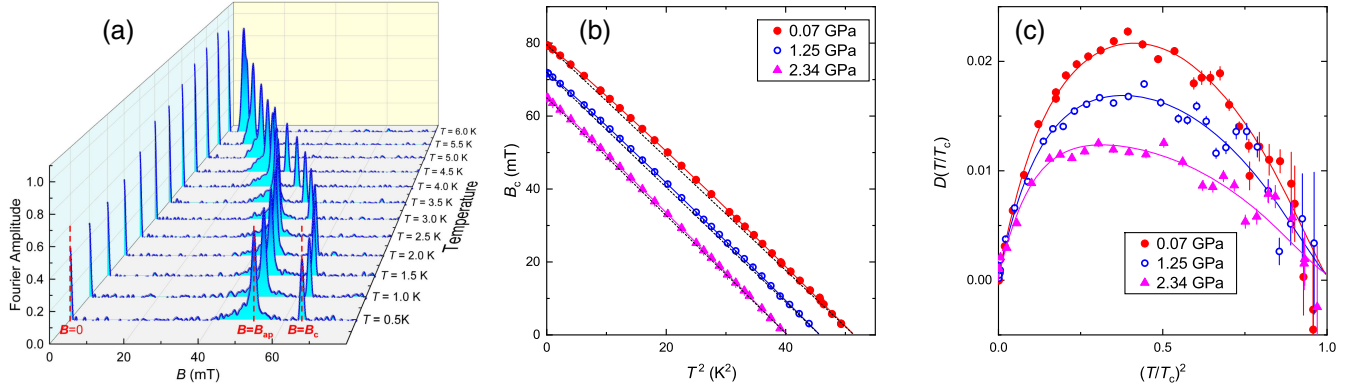


FIG. 1. (a) Fourier transforms of the muon-time spectra at $p = 2.34$ GPa, showing the magnetic-field distribution in the pressure-cell sample assembly. Three distinct peaks are observed: (i) a peak at $B = 0$, corresponding to muons stopping in superconducting domains of the Pb sample (Meissner state); (ii) a peak at the applied field, $B = B_{ap}$, arising from muons stopping in the pressure-cell walls; and (iii) a peak at $B = B_c$, corresponding to muons stopping in normal-state domains of the Pb sample. (b) Temperature dependence of the thermodynamic critical field $B_c(T)$ measured at pressures $p = 0.07, 1.25$, and 2.34 GPa. The solid lines are fits within the α model, while the dashed lines represent parabolic dependences. (c) Temperature dependence of the deviation function $D(T) = B_c(T) - B_c(0)[1 - (T/T_c)^2]$, highlighting the deviation of $B_c(T)$ from parabolic temperature dependence. The solid lines are fits using the α model.

transport measurements under pressure, which usually infer phase boundaries from macroscopic criteria, the present μ SR experiment determines B_c directly from the internal field distribution in the sample. Since the normal domains in the intermediate state are in thermodynamic equilibrium, the field extracted from their corresponding peak yields the equilibrium thermodynamic critical field itself.

The temperature dependence of the thermodynamic critical field obtained at $p = 0.07, 1.25$, and 2.34 GPa is presented in Fig. 1(b). The dashed lines show the parabolic dependence of $B_c(T)$, which is used here as a reference. As discussed in Refs. [28,29], the deviation of $B_c(T)$ from this parabolic form is governed by the value of $\alpha = \Delta(0)/(k_B T_c)$. To illustrate this more clearly, Fig. 1(c) shows the deviation function $D(T) = B_c(T) - B_c(0)[1 - (T/T_c)^2]$. The solid lines in panels (b) and (c) are fits performed within the phenomenological α model, adapted for strong-coupling superconductors such as Pb [28,29,47]. Within this model, the normalized gap function is assumed to follow the BCS form, while α is treated as an adjustable parameter (see Supplemental Material [14] for further details). The progressively smaller deviation of $B_c(T)$ from the parabolic form with increasing pressure therefore directly reflects the decrease of α . Analysis of the $B_c(T)$ data yields the pressure-dependent parameters T_c , $B_c(0)$, $\Delta(0)$, and α which form the basis for the pressure analysis presented below. It should be noted that the superconducting transition temperatures discussed in the following are obtained from fits of the measured thermodynamic critical-field curves $B_c(T)$ within the α model. Since $B_c(T)$ is itself a thermodynamic quantity, the corresponding T_c values should likewise be understood as thermodynamic transition temperatures.

Figure 2 presents the pressure dependences of the parameters extracted from the fits of $B_c(T)$: $T_c(p)$ [panel (a)],

$B_c(0, p)$ [panel (b)], $\Delta(0, p)$ [panel (c)], and $\alpha(p)$ [panel (d)]. All four quantities decrease approximately linearly over the investigated pressure range. The linear fits yield the logarithmic pressure derivatives $d \ln T_c / dp = -5.00(6) \times 10^{-2} \text{ GPa}^{-1}$, $d \ln B_c(0) / dp = -7.92(9) \times 10^{-2} \text{ GPa}^{-1}$, $d \ln \Delta(0) / dp = -7.3(2) \times 10^{-2} \text{ GPa}^{-1}$, and $d \ln \alpha / dp = -2.6(3) \times 10^{-2} \text{ GPa}^{-1}$. Remarkably, the logarithmic pressure coefficient of α is nearly equal to the difference between those of $B_c(0)$ and T_c , consistent with Eq. (5).

Discussion—The central idea of this Letter is that the thermodynamic critical field provides access to the superconducting condensation energy and therefore probes the superconducting energy scale in a way that is complementary to the transition temperature alone. The present μ SR measurements provide a direct realization of this concept for elemental Pb.

The low-pressure results summarized in Figs. 1 and 2 establish a consistent picture. First, the temperature dependence $B_c(T)$ is well described by the α model, and the deviation from a simple parabolic form decreases systematically with increasing pressure. Second, the pressure dependences extracted from these fits show that $B_c(0)$ and $\Delta(0)$ evolve in a very similar manner, whereas T_c decreases more slowly. Third, the pressure dependence of the gap ratio $\alpha = \Delta(0)/(k_B T_c)$ is consistent with the thermodynamic relation introduced in Eq. (5).

These observations follow naturally from the framework developed above. Since in simple metals the pressure dependence of the Sommerfeld coefficient γ_e is relatively weak [2–4,13], and for Pb can be estimated to provide only a secondary correction (see Supplemental Material [14]), the nearly equal logarithmic pressure derivatives of $B_c(0)$ and $\Delta(0)$ reported in Figs. 2(b) and 2(c) represent the

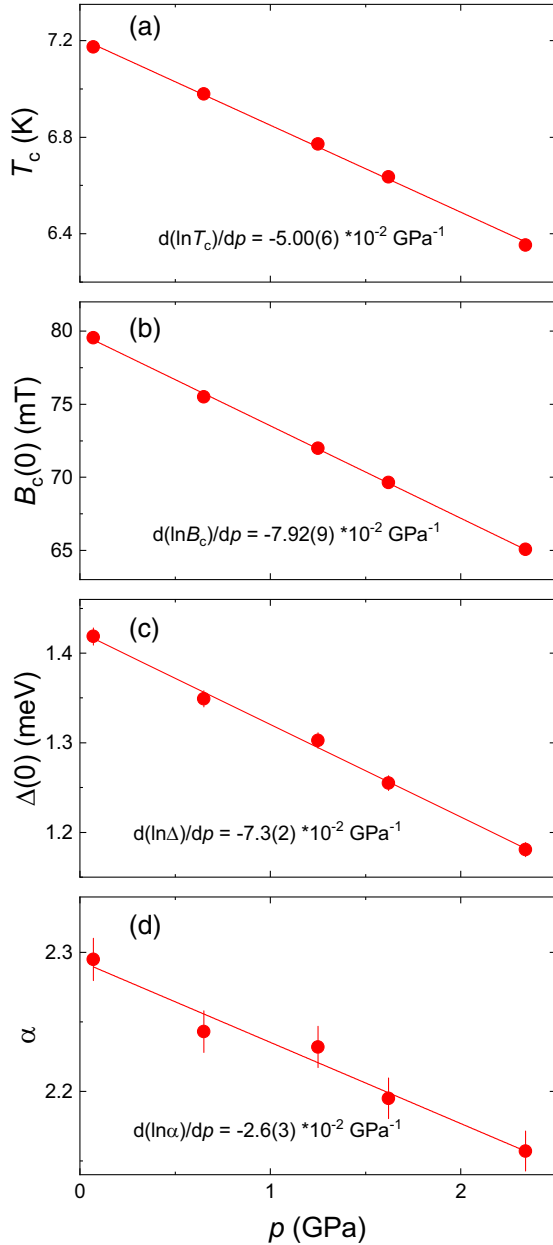


FIG. 2. Pressure dependencies of the superconducting parameters obtained from fits of $B_c(T)$ data using the α model: (a) superconducting transition temperature $T_c(p)$, (b) thermodynamic critical field at zero temperature $B_c(0, p)$, (c) superconducting energy gap $\Delta(0, p)$, and (d) the coupling parameter $\alpha(p)$. Solid lines represent linear fits to the data. The corresponding logarithmic pressure derivatives are indicated.

expected thermodynamic signature of a condensation energy governed primarily by the superconducting gap; see Eqs. (1)–(3). At the same time, according to Eq. (5), the weaker pressure dependence of T_c implies that the gap ratio α must evolve with pressure. This is seen directly in Fig. 2(d), where α decreases approximately linearly over the measured pressure range. Physically, this behavior is consistent with the known strong-coupling character of Pb

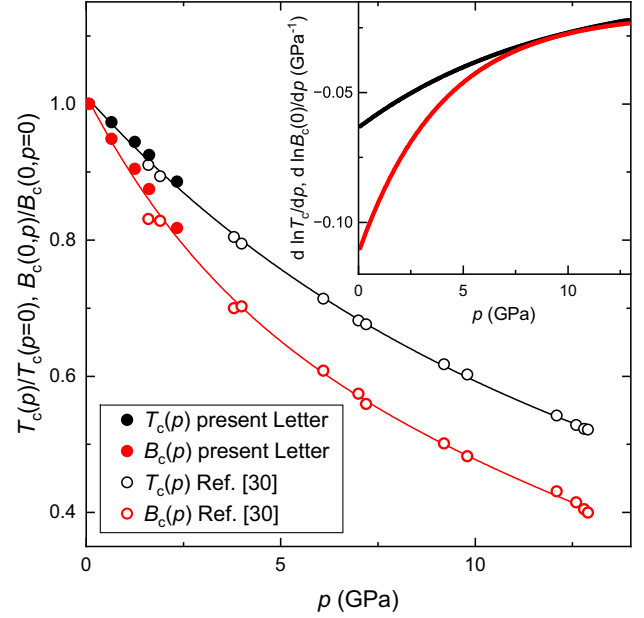


FIG. 3. Normalized pressure dependencies of the superconducting transition temperature $T_c(p)/T_c(p=0)$ and the thermodynamic critical field $B_c(0, p)/B_c(0, p=0)$ for Pb. Closed symbols represent the present μ SR data, while open symbols correspond to literature data from Brandt *et al.* [30]. The solid lines are empirical fits using a double-exponential form. Inset: pressure dependencies of the logarithmic derivatives $d \ln T_c/dp$ and $d \ln B_c(0)/dp$ obtained from the fit functions. The slopes approach each other and become nearly equal above $p \sim 8$ GPa, indicating convergence of the pressure responses of T_c and B_c .

at ambient pressure [7,47–49]: compression hardens the phonon spectrum and reduces the electron-phonon interaction, thereby driving the system toward weaker coupling.

It should be emphasized that the α model used here is phenomenological. For a strong-coupling superconductor such as Pb, deviations from simple BCS behavior are expected, and the quantities $\Delta(0)$ and α extracted from the fits should therefore be regarded as effective thermodynamic parameters. The role of the model in the present Letter is not to provide a fully microscopic Eliashberg description, but rather to quantify the pressure evolution of the measured $B_c(T)$ curves in a standard and internally consistent way. In this sense, the model dependence affects mainly the precise absolute values of the extracted parameters, whereas the central pressure trends—in particular the stronger pressure dependence of $B_c(0)$ compared with T_c , the decrease of α with pressure, and the relation between their logarithmic pressure derivatives—remain robust consequences of the thermodynamic data themselves.

The present μ SR experiment is restricted to the low-pressure regime, but it already establishes the relevant thermodynamic trend directly. To illustrate how this tendency develops further at higher pressure, Fig. 3 compares the present data with the high-pressure results of Brandt *et al.* [30] for $B_c(0, p)$ and $T_c(p)$. In the main panel, the

normalized pressure dependences of T_c and $B_c(0)$ differ most strongly at low pressure, where the pressure evolution of α is largest. With increasing pressure, however, the slopes of the two curves gradually approach each other. This becomes especially clear in the inset of Fig. 3, which shows the logarithmic derivatives $d(\ln T_c)/dp$ and $d(\ln B_c)/dp$ obtained from the empirical fit functions.

The result of this extended comparison is that the logarithmic pressure derivatives of T_c and $B_c(0)$ become nearly identical for $p \gtrsim 8$ GPa; see the inset of Fig. 3. Within Eq. (5), this means that $d \ln \alpha / dp$ becomes very small in this pressure range. In other words, the strong-coupling corrections to the gap ratio cease to evolve significantly, and Pb approaches a regime in which α is nearly pressure independent. Since the low-pressure μ SR data show that α decreases with increasing pressure, the subsequent convergence of $d \ln B_c(0)/dp$ and $d \ln T_c/dp$ at higher pressures is naturally interpreted as thermodynamic evidence that Pb evolves from the strong-coupling regime toward the weak-coupling limit.

More broadly, the present analysis clarifies why the thermodynamic critical field is a particularly informative quantity in pressure studies of conventional superconductors. Whereas T_c reflects the onset of superconductivity through the linearized gap equation, B_c probes the fully developed superconducting state through the condensation energy. The combined analysis of $B_c(T)$, the extracted pressure dependences of $B_c(0)$, T_c , $\Delta(0)$, and α , together with the comparison to higher-pressure data, therefore yields a coherent thermodynamic picture of how the superconducting energy scales of Pb evolve under compression.

A further important aspect of the present Letter is methodological. The qualitative statement that pressure weakens electron-phonon coupling in Pb is not, by itself, new. What is new here is that this evolution is followed directly through the thermodynamic critical field and hence through the condensation energy. In the intermediate state of a type-I superconductor, TF- μ SR provides direct access to the equilibrium value of B_c from the internal field distribution, avoiding criterion-dependent definitions of superconducting phase boundaries. The present Letter therefore demonstrates that pressure measurements of B_c can reveal information complementary to that obtained from T_c alone and can disentangle the evolution of the condensation-energy scale from that of the transition temperature.

Conclusions—The thermodynamic critical field of elemental Pb under hydrostatic pressure up to 2.3 GPa has been determined by means of muon-spin-rotation relaxation measurements in the intermediate state. Analysis of the resulting $B_c(T)$ curves within the α model has provided the pressure dependences of the zero-temperature critical field $B_c(0)$, the transition temperature T_c , the superconducting energy gap $\Delta(0)$, and the gap ratio α in the

low-pressure regime. It has been found that, within the experimentally accessible pressure range, $B_c(0)$ follows $\Delta(0)$ much more closely than it follows T_c . This is the central thermodynamic result of the present Letter and follows naturally from the relation between B_c and the condensation energy. At the same time, the weaker pressure dependence of T_c implies a decrease of the gap ratio $\alpha = \Delta(0)/(k_B T_c)$. The experimentally determined difference between the logarithmic pressure derivatives of $B_c(0)$ and T_c is found to agree, within the uncertainty of the fit, with the measured pressure derivative of α , thereby providing a direct thermodynamic confirmation of the relation between these quantities.

When combined with previously reported high-pressure measurements, the present low-pressure results further indicate that the pressure-induced weakening of coupling continues toward a regime in which the pressure evolution of α becomes very small. In this sense, the present μ SR data establish the low-pressure thermodynamic trend directly, while the literature comparison places this trend into a broader pressure context.

More generally, this Letter demonstrates that measurements of the thermodynamic critical field under pressure provide information that is complementary to that obtained from the transition temperature alone. Because B_c probes the condensation energy of the superconducting state, it provides direct access to the evolution of the superconducting energy scale and coupling strength under compression. In this respect, μ SR offers a unique advantage for type-I superconductors, since it allows the equilibrium thermodynamic critical field to be determined directly from the internal field distribution in the intermediate state.

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Data availability—There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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