

Single-gap Isotropic s -wave Superconductivity in Single Crystals AuSn_4

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Abstract

London, $\lambda_L(T)$, and Campbell, $\lambda_C(T)$, penetration depths were measured in single crystals of a topological superconductor candidate AuSn_4 . At low temperatures, $\lambda_L(T)$ is exponentially attenuated and, if fitted with the power law, $\lambda(T) \sim T^n$, gives exponents $n > 4$, indistinguishable from the isotropic single s -wave gap Bardeen-Cooper-Schrieffer (BCS) asymptotic. The superfluid density fits perfectly in the entire temperature range to the BCS theory. The superconducting transition temperature, $T_c = 2.40 \pm 0.05$ K, does not change after 2.5 MeV electron irradiation, indicating the validity of the Anderson theorem for isotropic s -wave superconductors. Campbell penetration depth before and after electron irradiation shows no hysteresis between the zero-field cooling (ZFC) and field cooling (FC) protocols, consistent with the parabolic pinning potential. Interestingly, the critical current density estimated from the original Campbell theory decreases after irradiation, implying that a more sophisticated theory involving collective effects is needed to describe vortex pinning in this system. In general, our thermodynamic measurements strongly suggest that the bulk response of the AuSn_4 crystals is fully consistent with the isotropic s -wave weak-coupling BCS superconductivity.

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13 **1 Introduction**

14 In recent years, superconductors with topological features in their electronic bandstructure
15 have attracted significant interest for various novel features predicted by a well-developed
16 theory. For example, emerging zero-energy excitations called Majorana fermions [1]. On the
17 material side, the search for topological superconductors (TSCs) is very active but so far has
18 yielded only a few “candidates” whose topological properties have not yet been fully confirmed
19 experimentally, including UTe_2 [2], Sr_2RuO_4 [3–5], UPt_3 [6], 2M-WS_2 [7], and $\text{M}_x\text{Bi}_2\text{Se}_3$
20 with $\text{M}=\text{Cu}$ [8, 9]. The subject of this study, AuSn_4 , is another promising TSC candidate with
21 theoretically predicted non-trivial topological characteristics [10–13].

22 The superconductivity in orthorhombic AuSn_4 with a transition temperature to the super-
23 conducting state, $T_c = 2.4$ K, was discovered in 1962 [14]. This compound is isostructural
24 to PtSn_4 [15] and PdSn_4 [16], which are not superconductors. The first principal study sug-
25 gests semimetallic behavior with type I nodes [12]. The magneto-transport measurements
26 show two-dimensional (2D) superconductivity in AuSn_4 [11, 17]. Recently, ARPES measure-
27 ments supported by DFT calculations [13] revealed nearly degenerate polytypes in AuSn_4
28 crystals, making it a unique case of a three-dimensional (3D) electronic band structure with
29 properties of a low-dimensional layered material. Thermodynamic magnetization and spe-
30 cific heat measurement in AuSn_4 single crystals are consistent with conventional nodeless
31 s -wave Bardeen-Cooper-Schrieffer (BCS) [18, 19] superconductivity [11]. Scanning tunnel-
32 ing microscopy (STM) measurements determined the superconducting gap to T_c ratio close
33 to the s -wave BCS value of $\Delta/T_c = 1.76$ [13]. However, other STM measurements suggest
34 unconventional 2D superconductivity with a mixture of p -wave surface states and s -wave
35 bulk [10]. Clearly, more measurements are required for an objective and conclusive determi-
36 nation of the nature of superconductivity in AuSn_4 .

37 Here, we probe the bulk nature of superconductivity in AuSn_4 single crystals by measur-
38 ing London and Campbell penetration depths using a highly sensitive tunnel-diode resonator
39 (TDR). Furthermore, we examine the response to a controlled non-magnetic point-like disorder
40 induced by 2.5 MeV electron irradiation. We conclude that AuSn_4 is a robust isotropic
41 s -wave superconductor in the bulk. However, we cannot exclude the possibility that it could
42 have a different type of superconductivity in the surface atomic layers, where the STM is most
43 sensitive.

44 **2 Samples and Methods**

45 Single crystals of AuSn_4 were grown with excess Sn flux [13, 20, 21]. High-purity Au and Sn
46 were mixed in a 12:88 ratio in a fritted crucible and sealed in a quartz ampoule under an Ar
47 gas atmosphere. The ampoule was heated to 1100 °C over 12 hours, then cooled to 250 °C
48 in 12 hours, and significantly slower to 230 °C over 90 hours. The ampoule was held at this
49 temperature for 48 hours prior to removal from the furnace.

50 The London penetration depth, $\lambda(T)$, was measured using a sensitive frequency-domain
51 self-oscillating tunnel-diode resonator (TDR) operating at a frequency of around 14 MHz.
52 Measurements were performed in a ^3He cryostat with a base temperature of ≈ 400 mK, which

53 is $0.17T_c$, allowing us to examine the low-temperature limit, which starts below approximately
 54 $T_c/3$, where the superconducting gap is approximately constant [19]. The experimental setup,
 55 measurement protocols, and calibration are described in detail elsewhere [22–26].

56 Briefly, the sample placed inside the inductor of the LC - tank circuit affects the total in-
 57 ductance, L , leading to a shift of its resonant frequency, $2\pi f = 1/\sqrt{LC}$ by the amount propor-
 58 tional to the magnetic susceptibility of the sample, $\Delta f = G\chi(T)$, where G is the calibration
 59 constant [23,25]. A small excitation magnetic field of the setup, $\sim 20\text{mOe}$, ensures the regime
 60 of a small-amplitude linear magnetic response where $\chi = \lambda_m/R \tanh(R/\lambda_m) - 1$. Here R is
 61 the effective dimension of the sample and λ_m is the total measured magnetic penetration
 62 depth [25]. The sample of this study had dimensions $0.6 \times 0.4 \times 0.1 \text{ mm}^3$, which gives the
 63 effective dimension $R = 84.1 \mu\text{m}$, calculated using the procedure described in Ref. [25]. With
 64 a penetration depth smaller than a few micrometers for most of the temperature range, we
 65 can simplify the relation for magnetic susceptibility to $\chi \approx \lambda_m/R - 1$. Therefore, the change in
 66 $\lambda_m(T)$ with respect to a reference point at the lowest temperature, $\lambda_m(T_{min})$ is proportional
 67 to the relative frequency shift, $\Delta\lambda_m(T) = \lambda_m(T) - \lambda_m(0.4 \text{ K}) = (R/G)\Delta f$. For all practi-
 68 cal purposes, $\lambda_m(0) \approx \lambda_m(0.4 \text{ K})$. Although the calibration constant G can be calculated, in
 69 our setup it is measured directly by mechanically extracting the sample from the coil at low
 70 temperature, thus providing a robust calibration specific for each sample studied.

71 In a zero magnetic field, there are no vortices and the measured penetration depth is the
 72 London penetration depth, $\lambda_L(T)$. Therefore, by measuring the change, $\Delta\lambda_L(T) = \lambda_m(T, B=0)$,
 73 and adding the absolute value, $\lambda(0)$ determined from other measurements, such as fitting
 74 $\Delta\lambda(T)$ in Fig.2, a full London length is obtained, $\lambda_L(T) = \lambda_L(0) + \Delta\lambda_L(T)$. In the presence
 75 of a magnetic field, in a small-amplitude linear AC response, the vortex-mediated Campbell
 76 length adds to the London length, $\lambda_m^2 = \lambda_C^2 + \lambda_L^2$ [27–29]. Upon approaching the transition
 77 temperature, T_c , the penetration depth can only increase up to a normal-metal skin depth.
 78 Since the magnetoresistance at T_c is known and in this case is negligible, the value at T_c
 79 is used as a fixed reference point [13]. Measuring $\lambda_m(T, B)$ in finite magnetic fields, the
 80 curves are shifted vertically, so that the saturation flat parts above T_c match. Once such a
 81 vertical shift procedure is performed, the full Campbell penetration depth is extracted from
 82 $\lambda_C = \sqrt{\lambda_m^2 - \lambda_L^2}$.

83 Point-like disorder was introduced at the SIRIUS facility in the Laboratoire des Solides
 84 Irradiés at École Polytechnique in Palaiseau, France. Electrons, accelerated in a pelletron-
 85 type linear accelerator to 2.5 MeV, knock out ions, creating vacancy-interstitial Frenkel pairs
 86 [30,31]. During irradiation, the sample is immersed in liquid hydrogen at around 20 K. This
 87 ensures efficient heat removal upon impact and prevents immediate recombination and mi-
 88 gration of the produced atomic defects. The acquired irradiation dose is determined by mea-
 89 suring the total charge collected by a Faraday cage located behind the sample. As such, the
 90 acquired dose is measured in the “natural” units of C/cm^2 , which is equal to $1 \text{ C}/\text{cm}^2 \equiv 1/e \approx$
 91 6.24×10^{18} electrons per cm^2 . Upon warming to room temperature, some defects recom-
 92 bine, and some migrate to various sinks (dislocations, surfaces, etc.). This leaves a metastable
 93 population, about 70%, of point-like defects [32,33]. Importantly, the same sample has been
 94 measured before and after electron irradiation.

95 3 Results

96 3.1 London penetration depth

97 Figure 1 shows the low-temperature dependence of the change in the London penetration
 98 depth, $\Delta\lambda(T) = \lambda(T) - \lambda(T_{min} = 0.4 \text{ K})$ before (blue circles) and after $2.5 \text{ C}/\text{cm}^2$ electron

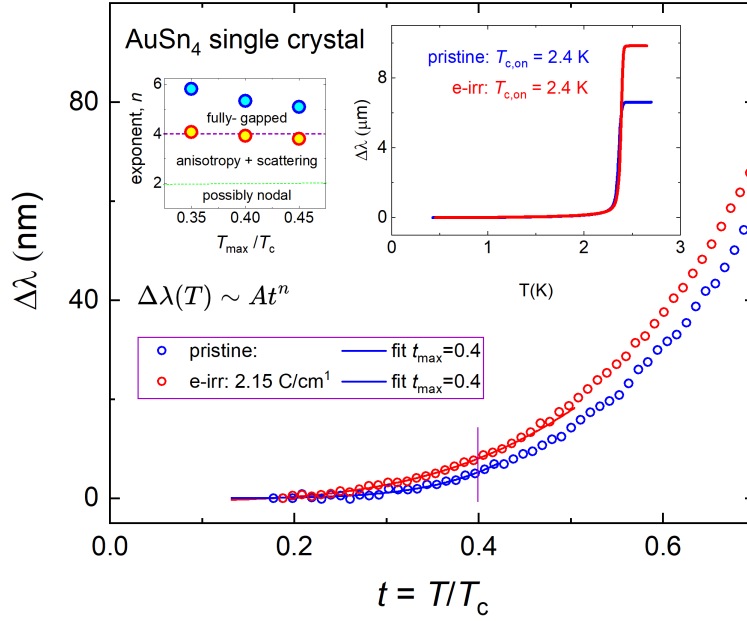


Figure 1: Main Panel: Low-temperature temperature variation of the London penetration depth $\Delta\lambda(T) = \lambda(T) - \lambda(0.4 \text{ K})$ as a function of normalized temperature, $t = T/T_c$, for pristine (blue circles) and irradiated at 2.5 C/cm^2 (red circles) single crystal of AuSn_4 . Lines show fits to the power law, $\Delta\lambda(T) \sim At^n$, with the upper range of $t_{\text{max}} = 0.4$. The top right inset shows the $\Delta\lambda(T)$ in the whole temperature range, showing sharp superconducting transition with onset $T_c = 2.4 \text{ K}$ for both pristine and electron irradiated state. The top left inset shows the exponent n versus the upper limit of the power-law fitting, $t_{\text{max}} = T_{\text{max}}/T_c$, indicating robustness of the power law, experimentally indistinguishable from exponential.

99 irradiation (red circles). The upper left inset shows the exponent n determined from the
 100 power-law fitting, $\Delta\lambda(T) \sim At^n$, as a function of the upper fitting limit, $t_{\text{max}} = T_{\text{max}}/T_c$. The
 101 solid lines in the main frame show an example of such a fitting with $t_{\text{max}} = 0.4$. The results
 102 show a robust and consistent behavior with $n \geq 4$, indicating experimentally indistinguishable
 103 from the exponential temperature dependence. The exponent, n , decreased after irradiation
 104 as it should be in an s -wave superconductor [34, 35].

105 The upper right inset of Fig.1 shows $\Delta\lambda(T)$ of the same sample in its pristine state and
 106 after 2.15 C/cm^2 electron irradiation as a function of absolute temperature T . One might
 107 think that for some reason (e.g., defect annealing and recombination), there was no increase in
 108 disorder after irradiation. It is straightforward to prove that this is not the case. The saturation
 109 of the measured $\lambda(T)$ above T_c occurs when it reaches the skin depth of the normal state,
 110 $\delta_{\text{skin}} = \sqrt{\rho/\mu_0\pi f}$, where $\mu_0 = 4\pi \times 10^{-7} \text{ H/m}$ is the vacuum permeability, and ρ is the
 111 resistance. More precisely, $\delta_{\text{skin}}(T_c) = 2\lambda(T_c)$ [36]. We did not measure resistivity in this
 112 particular AuSn_4 sample, but we directly compared resistivity from transport measurements
 113 and extracted from the skin depth on the same samples of other compounds and always found
 114 good quantitative agreement [37, 38]. Furthermore, the upper critical fields are small, $H_{c2}^{\parallel ab} =$
 115 130 Oe and $H_{c2}^{\parallel c} = 90 \text{ Oe}$ [11]. Consulting with published magnetoresistance [13], we find
 116 that the expected variation of δ_{skin} just above T_c is negligible. On the other hand, the top right
 117 inset in Fig.1 shows a substantial increase in saturation value after electron irradiation. This
 118 proves a substantial increase of resistivity, which can only be due to added disorder scattering.
 119 Therefore, the fact that the superconducting transition temperature T_c remains unchanged is
 120 consistent with the Anderson theorem for isotropic s -wave superconductors [39, 40]. We have

121 observed similar robust superconductivity in another low- T_c superconductor with non-trivial
122 topology, LaNiGa₂ [41].

123 The exponential temperature dependence of $\lambda(T)$ can be fitted with the well-known low-
124 temperature asymptotic BCS, $\Delta\lambda(T) = \lambda(0)\sqrt{\frac{\pi\delta}{2t}}e^{-\frac{\delta}{t}}$ [19], where the ratio $\delta = \Delta(0)/T_c$
125 was fixed at $\delta \approx 1.764$, leaving only one free parameter $\lambda(0)$. The fitting is shown in the
126 top panel of Fig.2. It produces $\lambda(0) = 150$ nm in the pristine state (blue fitting curve and
127 blue data symbols) and $\lambda(0) = 258$ nm after 2.15 C/cm² electron irradiation (red curve and
128 symbols). With these numbers, we can calculate the superfluid density in the full tempera-
129 ture range using $\rho_s(T) \equiv (\lambda(0)/\lambda(T))^2 = (1 + \Delta\lambda(T)/\lambda(0))^{-2}$. The bottom panel of Fig.2
130 shows $\rho_s(T)$ by blue and red circles for the pristine and irradiated states of the same sam-
131 ple, respectively. The theoretical lines of the clean (blue) and dirty (red) limits were calcu-
132 lated self-consistently using the Eilenberger formalism [43]. The analytical dirty limit formula,
133 $\rho_s = (\Delta(T)/\Delta(0)) \tanh(\Delta(T)/2T)$ reproduces the numerical calculation precisely [42]. We
134 note that due to a limited number of data points, good fits of $\lambda(T)$ can also be obtained with
135 slightly different ratios of $\Delta(0)/T_c$. However, then the full-range superfluid density curve does
136 not fit. It fits only with the weak-coupling isotropic BCS value of 1.764. In summary, Fig.2
137 shows that the classical BCS theory describes the experimental data well.

138 To summarize our findings from measurements of the London penetration depth, $\lambda(T)$,
139 several independent parameters: (1) low-temperature behavior of $\lambda(T)$; (2) full temperature
140 range behavior of ρ_s ; (3) disorder-independent T_c before and after electron irradiation, fully
141 agree with the BCS theory for the isotropic s – w ave gap with the ratio $\delta = \Delta(0)/T_c \approx 1.76$.
142 This is the nature of superconductivity in the bulk of AuSn₄ crystals. However, our measure-
143 ments would not pick up a tiny signal coming from the surface atomic layers, so unconventional
144 topological features are still possible.

145 3.2 Campbell penetration depth

146 The temperature variation of the magnetic penetration depth before (top panel) and after (bot-
147 tom panel) electron irradiation, measured in various dc magnetic fields applied along the c –
148 axis, is shown in Fig.3. The field values are shown next to each curve. Solid lines correspond
149 to zero-field cooling (ZFC) in all curves, and dotted lines correspond to field cooling (FC) pro-
150 tocols. For one curve, this is shown by arrows. The ZFC and FC curves are indistinguishable,
151 implying that the process is totally reversible, which indicates a parabolic shape of the pinning
152 potential.

153 In the presence of an external DC magnetic field, Abrikosov vortices penetrate the sample
154 and form a vortex lattice. Then the measured penetration depth, λ_m , has two contributions,
155 the usual London penetration depth that in this section we explicitly denote as λ_L , and the
156 Campbell penetration depth λ_C , which is a characteristic length scale over which a small ac
157 perturbation is transmitted elastically by a vortex lattice into the sample [44–47]. More specifi-
158 cally, the amplitude of the ac perturbation must be small enough so that the vortices remain
159 in their potential well, and their motion is described by the reversible linear elastic response.
160 In this case, $\lambda_m^2 = \lambda_L^2 + \lambda_C^2$ [27, 48]. This requirement of a very small amplitude makes most
161 conventional ac susceptibility techniques inapplicable for the measurements of the Campbell
162 length. Specialized frequency domain resonators with sufficient sensitivity to a small excita-
163 tion ac magnetic field are needed [49, 50]. Until now, only a few experimental studies have
164 been published [29, 49–52].

165 Figure 3 shows the temperature-dependent variation of the magnetic penetration depth,
166 $\lambda_m(T) = \lambda_L(0) + \Delta\lambda_m(T)$, for different values of the dc magnetic field applied parallel to
167 the sample c –axis. For $\lambda_L(0)$, we have used the values obtained from the BCS fit; see the
168 upper panel of Fig. 2. Then, we assumed that, above T_c , the resistivity is field independent,

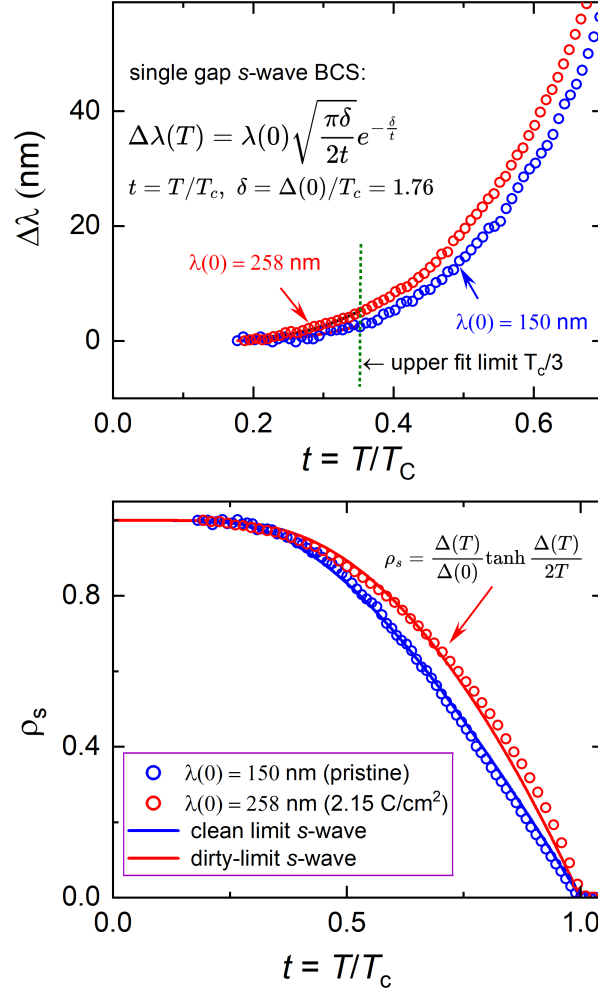


Figure 2: Top panel. Fit to the BCS low-temperature asymptotic, $\Delta\lambda(T) = \lambda(0)\sqrt{\frac{\pi\delta}{2t}}e^{-\frac{\delta}{t}}$ with a fixed ratio $\delta = \Delta(0)/T_c \approx 1.76$ leaving only one free parameter, $\lambda(0) = 150 \text{ nm}$ in the pristine sample (blue fitting curve and blue data symbols) and $\lambda(0) = 258 \text{ nm}$ after 2.15 C/cm^2 electron irradiation (red curve and symbols). Bottom panel: Superfluid density calculated from the data, $\rho_s(T) = (1 + \Delta\lambda(T)/\lambda(0))^{-2}$. Solid lines show self-consistent full temperature range calculations using Eilenberger formalism for pristine (blue line) and irradiated (red line) states. The known analytical expression for the s-wave dirty limit is shown in [42].

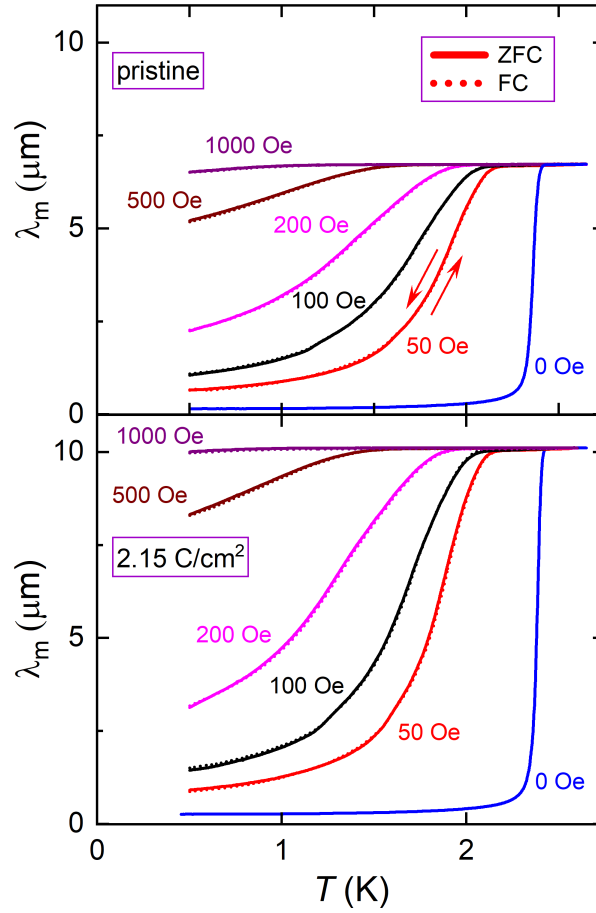


Figure 3: Temperature variation of the measured magnetic penetration depth, λ_m , before (top panel) and after (bottom panel) electron irradiation, measured with the various dc magnetic fields applied along the c -axis. The field values are shown. Solid lines correspond to zero-field cooling (ZFC), and dotted lines correspond to field cooling (FC) protocols. For one curve, this is shown by arrows. The ZFC and FC curves are indistinguishable, implying that the process is completely reversible, indicating the pinning potential's parabolic shape. Note that the axes scales are the same in the top and bottom panels, aiding in a visual comparison of the effect of irradiation.

169 so we adjusted other curves to match that value. The top panel shows a pristine state, and the
 170 bottom panel shows the same sample after electron irradiation.

171 Generally speaking, the Campbell penetration depth can exhibit a hysteresis upon warm-
 172 ing and cooling, indicating an anharmonic (non-parabolic) pinning potential and/or strong
 173 pinning [47, 49, 52, 53]. Therefore, there are two types of measurement protocols: zero-field
 174 cooling (ZFC) and field cooling (FC). In the ZFC protocol, the Campbell length is measured
 175 on warming after the sample was cooled in a zero magnetic field and the target field was ap-
 176 plied at the base temperature (solid lines in Fig. 3). In the FC protocol, measurements are
 177 performed on cooling in a target magnetic field applied above T_c (dotted lines in Fig. 3). For
 178 both pristine and irradiated states, $\lambda_m(T)$ shows a monotonic increase with temperature, and
 179 there is no hysteresis between the ZFC and FC protocols. To aid in visualizing the effect of
 180 irradiation, the scales of the axes in Fig. 3 are the same in the top and bottom panels. It is
 181 clear that the measured penetration depth has increased after electron irradiation.

182 Figure 4 shows the Campbell penetration depth as a function of an applied magnetic field,

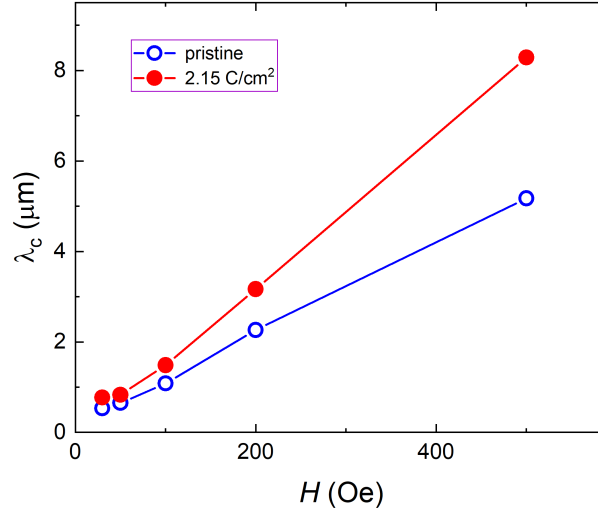


Figure 4: Campbell penetration depth, $\lambda_C^2 = \sqrt{\lambda_m^2 - \lambda_L^2}$ as a function of an applied magnetic field, H , evaluated from the data shown in Fig. 3 at a fixed temperature of $T = 0.5$ K. for a FC protocol comparing pristine (blue symbols) and irradiated (red symbols) states of the same sample.

183 H , evaluated from the data shown in Fig. 3 at a fixed temperature of $T = 0.5$ K for a FC
 184 protocol comparing pristine (blue symbols) and irradiated (red symbols) states of the same
 185 sample. The Campbell length λ_C increases after electron irradiation. In the simple Campbell
 186 model [44, 45], $\lambda_C^2 = \phi_0 H / \alpha$, where ϕ_0 is the magnetic flux quantum and α is the curvature
 187 of the pinning potential, $\alpha = d^2 U / dr^2$. The critical current density $j_c = \alpha r_p / \phi_0 = H r_p / \lambda_C^2$,
 188 where r_p is the radius of the pinning potential, usually assumed to be of the order of the
 189 coherence length, ξ . We note that this critical current is not the same as the persistent current
 190 obtained in conventional magnetization measurements, which is based on the Bean model
 191 that assumes a constant vortex density gradient [54, 55]. In the present measurements, the
 192 critical current is a parameter of the model defining the equilibrium Campbell length without
 193 persistent Bean currents present. It represents a theoretical current density supported by a
 194 specific pinning potential, $U(\mathbf{r})$. The conventional measured current density is lower due to
 195 magnetic relaxation, which is very fast on short time scales and later slows down to become
 196 time-logarithmic [56].

197 In a more general picture, α is determined by the elementary pinning forces [46, 47, 57].
 198 In the original model with a fixed r_p , the Campbell length is expected to scale as $\lambda_C \sim \sqrt{H}$,
 199 but Fig. 4 shows a practically linear temperature dependence, especially after irradiation. This
 200 indicates that vortex pinning in AuSn₄ is more complicated with a field-dependent radius of
 201 the pinning potential, which is possible, for example, in a collective pinning theory when the
 202 vortex lattice evolves from the single-vortex pinning regime to the vortex bundle regime [56].
 203 In addition, it is known that the coherence length increases with the magnetic field [58].
 204 Therefore, if $\xi \sim H$, then λ_C will be a linear function of the applied field. As for the difference
 205 between pristine and irradiated states, it is possible that the collective pinning in the pristine
 206 state is replaced by the disordered vortex phase after electron irradiation, and one cannot
 207 directly compare the critical current densities using the same formula. In any case, the nature
 208 of pinning in AuSn₄ requires further investigation.

4 Conclusions

We report measurements of London, $\lambda_L(T)$, and Campbell, $\lambda_C(T)$, penetration depths in single crystals of the topological superconductor candidate AuSn_4 to elucidate the nature of superconductivity in the bulk. Several independent parameters studied before and after 2.5 MeV electron irradiation unambiguously point to isotropic single s -wave gap weak coupling BCS superconductor. Specifically, the superfluid density before and after electron irradiation overlaps almost perfectly with the parameter-free theoretical BCS curves in the full temperature range for clean and dirty limits, respectively. The Campbell penetration depth before and after electron irradiation does not show hysteresis between the ZFC and FC data, indicating a parabolic shape of the pinning potential. However, the H -linear behavior of λ_C implies either the field-dependent Labusch parameter, α , or the radius of the pinning potential, r_p , or both. Considering the low pinning in AuSn_4 single crystals and the point-like nature of the induced defects, such a field dependence may be expected in the vortex bundle regimes within the collective pinning theory [56].

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