Size and mass of Cooper pairs determined by low-energy $\mu$SR and PNR

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The Pippard coherence length $\xi_0$ (the size of a Cooper pair) in an extreme type-I superconductor was determined directly through high-resolution measurement of the nonlocal electrodynamic effect combining low-energy muon spin rotation spectroscopy and polarized neutron reflectometry. The renormalization factor $Z \equiv m^*_c/m$ is the mass of the Cooper pair and the electron, respectively) resulting from the electron-phonon interaction, and the temperature dependent London penetration depth $\xi_0(T)$ were determined as well. A general expression linking $\xi_0$, $Z$ and $\lambda_0(0)$ is introduced and experimentally verified. This expression allows one to determine experimentally the Pippard coherence length in any superconductor, independent of whether the electrodynamics is local or nonlocal, conventional or unconventional.

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also allows one to determine $Z$. Electrons near the Fermi surface are dressed by a cloud of virtual phonons, leading to an enhancement of their effective mass and consequently to a reduction of the Fermi velocity $v_F$. Below the critical temperature $T_c$, the phonon mediated attraction of electrons exceeding their screened Coulomb repulsion results in formation of the Cooper pairs. In BCS theory electron-electron coupling is weak: $N(0)V \ll 1$, where $V$ is the pairing potential and $N(0)$ is the single-spin electron density of states at the Fermi surface obtained from specific-heat measurements. Eliashberg's strong-coupling theory (SCT) is free from this limitation and agrees better with experiments [1].

In SCT the effective mass of electrons near the Fermi surface is $m^* = Z m$ and consequently the effective mass of the Cooper pairs is $m_{cp}^* = 2 Z m$. Correspondingly, $\lambda_L$ and $\xi_0$ are renormalized with respect to their values in the weak-coupling ($wc$) limit as

$$\lambda_{Lwc} = \sqrt{\frac{Z}{2}} \lambda_{Lwc},$$

$$\xi_0 = \frac{\xi_{0wc}}{Z}.$$  

If $\lambda_L(T \to 0)$ is measured, the factor $Z$ can be inferred from Eq. (1). $\lambda_{Lwc}^0(0)$ can be obtained from the Faber-Pippard formula for $\lambda$ derived for the electrons not interacting with the lattice [10]. This leads to

$$\lambda_{Lwc}^0(0) = \sqrt{\frac{3c^2}{8\pi e^2N(0)v_F^2}} ,$$

where $e$ is the electron charge. On the other hand, if $\xi_0$ is measured, $Z$ can be calculated from Eq. (2) using the BCS definition $\xi_{0wc}^0(0) = \hbar v_F/\pi \Delta(0) = 0.18\hbar v_F/k_BT_c$, where $\Delta(0) \equiv \Delta(T = 0)$ is the energy gap and $\hbar = h/(2\pi)$ and $k_B$ are the Planck and Boltzmann constant, respectively.

Unfortunately neither of these approaches is applicable for quantitative analysis since reliable calculation of $v_F$ is not possible due to the complex topology of the Fermi surface in polyvalent metals [11]. However, if both $\lambda_L(0)$ and $\xi_0$ are known, one can eliminate $v_F$, thus obtaining

$$Z = \frac{c^2k^2}{12.5\pi T^2_1e^2\gamma} \cdot \frac{1}{\lambda_L(0)2\xi_0^2},$$

where it is taken into account that $N(0) = 3\gamma/2\pi^2k_B^2$ ($\gamma$ is the electron heat capacity coefficient [11]). Eq. (1) is a general expression independent of the relationship between the current density and the vector potential and of the specific nature of the electron-electron pairing.

In SCT $Z = 1 + \lambda_m$, where $\lambda_m$ is a mass-enhancement parameter calculated in Ref. [1] using tunneling spectroscopy experimental data. $\lambda_m$ can also be obtained from McMillan’s equation for $T_c$ [12], but this approach is less reliable [1]. Therefore, for nonlocal superconductors Eq. (1) furnishes a bridge between the $B(z)$ and the tunneling data, hence providing an independent test for the validity of $\lambda_L$ and $\xi_0$ inferred from the $B(z)$ data. On the other hand, the vast majority of superconductors are local. For those Eq. (1) allows one to determine $\xi_0$ from the measured values of $\lambda_L(0)$ and $Z$ using, e.g., LE-$\mu$SR and tunneling experiments, respectively.

In this work $B(z)$ is measured using the LE-$\mu$SR [13, 14] and PNR [15] techniques. Combining these complementary techniques enables an independent verification of the inferred values of $\xi_0$ and $\lambda_L(T)$ and an estimate of their total uncertainties. The samples were two indium films whose parameters are listed in table 1. The films were deposited on an oxidized silicon wafer (IN-1) and on a sapphire crystal (IN-2) held at room temperature via thermal evaporation of indium shots (99.9999% purity) at a base pressure $\leq 5 \times 10^{-9}$ mbar. The IN-1 sample was used for PNR measurements and for LE-$\mu$SR measurements down to 2.9 K. The IN-2 sample was used for LE-$\mu$SR measurements at lower temperatures.

The samples’ DC magnetization exhibits a clear-cut first-order phase transition with deep supercooling. The

TABLE I: Parameters of the samples, $d$ is the thickness, $o$ is the oxide layer thickness (see Ref. [2]), $RRR$ is the residual resistivity ratio; $\ell$ is the elastic mean free path.

| Sample | $T_c(\text{K})$ | $d$ (\text{cm}) | $o$ (\text{nm}) | $RRR$ | $\ell$ (\text{\mu m}) |
|--------|----------------|----------------|----------------|-------|----------------------|
| IN-1   | 3.415          | 2.3            | 2.5            | $\leq 1$ | 560                 |
| IN-2   | 3.415          | 6              | 3.3            | $\leq 1$ | 730                 |

FIG. 1: Time spectra of muons with energy of 22 keV at an applied field of 47 Oe in the normal state (a) and in the Meissner state (b). The curves are fits to the Gaussian (a) and the time-domain (b) models. The inserts present the field spectra $P(B)$. In the insert in (b) $bg$ is the background contribution, while the curve represents two Gaussian peaks to approximate the experimental spectrum. $A_0$ is the maximum observable asymmetry of the muon decay.
FIG. 2: Reduced field $B/B_0$ obtained from the LE-μSR measurements using the Gaussian model. The depth is the average stopping distance of the implanted muons calculated from the Monte Carlo TRIM.SP code. The solid (dashed) line is the field profile calculated from the nonlocal (local) theory at $T = 2.92$ K with $\xi_0 = 380$ nm, $\lambda_L(0) = 30$ nm and the dead layer $z_0 = 4$ nm, inferred from the $\chi^2$ and the time-domain analysis. The insert presents the same field profiles on a linear scale.

measured critical field $H_c(T)$ and $T_c$ perfectly agree with values reported in literature [16, 17]. The surface of the films consists of nearly atomically flat terraces (root-mean-square roughness of the terrace areas is $<2$ Å) with a typical size around 5 μm with voids in between. This size is much larger than $\xi_0$ and the total area of the voids does not exceed 3% of the sample surface. Therefore the terrace surface structure should not affect the electrodynamic properties of the films. For both samples the elastic mean free path $\ell \gg \xi_0$, therefore the samples are type-I superconductors in the pure limit [2].

The LE-μSR experiments were performed at the μE4 beamline of the Swiss Muon Source at the Paul Scherrer Institute [18]. Typical spectra of muon polarization $P$ are presented in Fig. 1. In the normal state $B$ is uniform and the $P(B)$ spectrum (insert in Fig. 1a) has Gaussian shape due to nuclear dipole broadening. In the Meissner state $B$ is not uniform and $P(B)$ is not Gaussian. However, Gaussian spectra (curve in the insert of Fig. 1b) provide a good first approximation.

$B(z)$ points obtained using a Gaussian model [8] for the depolarization of the precessing muons are presented in Fig. 2 along with the $B(z)$ curve calculated from the local and nonlocal theories. The sample temperature was determined in situ based on the $H_c(T)$ phase diagram obtained from magnetization measurements. As can be seen in Fig. 2, the “Gaussian” $B$-points exhibit a pronounced non-exponential depth dependence, consistent with the nonlocal effect. Discrepancies between the points and the “nonlocal” theoretical curve are mainly caused by incomplete adequacy of the Gaussian model. On the other hand, the qualitative consistency of the “Gaussian” points with the nonlocal theory justifies the application of a time-domain model [19], directly assuming the “nonlocal” shape of $B(z)$ with $\lambda_L$ as an adjustable parameter.

In nonlocal superconductors the effective penetration depth $\lambda_{\text{eff}} \approx (\lambda^2_0/\xi_0)^{3/2}$ [2]. Therefore either $\lambda_L$ or $\xi_0$ has to be determined using an additional criterium. Since $\lambda_{\text{eff}}$ is mainly sensitive to $\lambda_L$ and $\xi_0$ weakly depends on temperature, this parameter is $\xi_0$. Its optimal value was found from $\chi^2$ analysis of the global fits (simultaneous fitting of all implantation energy data sets at each temperature). The fits were performed with different $\xi_0$ chosen around a theoretical value of 377 nm [12]. The graphs for $\chi^2$/NDF (NDF stands for number of degrees of freedom) versus $\xi_0$ are presented in Fig. 3. Values of $\xi_0$ at the minima and the best estimate for $\xi_0$ are given in the insert. The values of $\lambda_L$ obtained from the global fits with $\xi_0 = 380$ nm are presented in a summary graph in Fig. 5.

The PNR measurements were performed on the D3 reflectometer at the NRU reactor in Chalk River. The experimental data and simulations for the reflectivity of neutrons polarized parallel ($R^+$) and antiparallel ($R^-$) to the magnetic field and for the spin asymmetry $(R^+ - R^-)/(R^+ + R^-)$ are presented in Fig. 4. In agreement with our LE-μSR results and previous PNR results [6], the neutron spin asymmetry (Fig. 4, insert (a)) simulated with $B(z)$ calculated from the nonlocal theory matches the experimental data significantly better than the simulation based on the London field profile. The “nonlocal” $B(z)$ was calculated with $\xi_0 = 380$ nm obtained from the LE-μSR data. The best match of the simulated spin asymmetry with the experimental data was achieved for $\lambda_L(T = 0.3$ K) = 28.0 ± 2.5 nm.

Results for $\lambda_L$ inferred from the LE-μSR and PNR measurements are shown in Fig. 5. The values of $\lambda_L(T)$ are consistent with each other and agree with the two-fluid formula [2]. The best estimate of $\lambda_L(0)$ is 30 ± 2 nm.

Having determined $\lambda_L(0)$ and $\xi_0$ we calculated $Z$ from Eq. (3). The results are summarized in the table inserted in Fig. 5, from which it can be concluded that the value of
mine the Pippard coherence length of the Cooper pairs, and the $\lambda$ one to infer $Z$ obtained in this work agrees with the value obtained from electron tunneling data [1] and from McMillan’s equation for $T_c$ [12].

In conclusion, high resolution measurements of the magnetic field profiles $B(z, T)$ were performed, for the first time, in an extreme type-I superconductor (indium) combining LE-$\mu$SR and PNR measurements. The $B(z)$ profiles quantitatively confirm the Pippard/BCS nonlocal electrodynamic effect. The $B(z)$ data were used to determine the Pippard coherence length $\xi_0$, the renormalization factor $Z$ for the electron-phonon mass enhancement of the Cooper pairs, and the London penetration depth $\lambda_L(T)$. The experimentally verified equation (4) allows one to infer $\xi_0$ in any superconductor from the results for $\lambda_L(0)$ and $Z$.

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