Evidence for nonmonotonic magnetic field penetration in a type-I superconductor

Citation for published version (APA):
Kozhevnikov, V. F., Giuraniuc, C. V., Bael, van, M. J., Temst, K., Haesendonck, van, C., Mishonov, T. M., Charlton, T., Dalgliesh, R. M., Khaidukov, Y. N., Nikitenko, Y. V., Aksenov, V. L., Gladilin, V. N., Fomin, V. M., Devreese, J. T., & Indekeu, J. O. (2008). Evidence for nonmonotonic magnetic field penetration in a type-I superconductor. Physical Review B, 78(1), 012502-1/4. Article 012502. https://doi.org/10.1103/PhysRevB.78.012502

DOI:
10.1103/PhysRevB.78.012502

Document status and date:
Published: 01/01/2008

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.

Download date: 22. Sep. 2023
Evidence for nonmonotonic magnetic field penetration in a type-I superconductor

V. F. Kozhevnikov,1,2 C. V. Giuraniuc,1,4 M. J. Van Bael,1 K. Temst,3 C. Van Haesendonck,1 T. M. Mishonov,4 T. Charlton,5 R. M. Dalgliesh,5 Yu. N. Khaidukov,6 Yu. V. Nikitenko,5 V. L. Aksenov,5 V. N. Gladilin,1,7,8 V. F. Kozhevnikov,1,2 C. V. Giuraniuc,1,* M. J. Van Bael,1 K. Temst,3 C. Van Haesendonck,1 T. M. Mishonov,4 T. Charlton,5 R. M. Dalgliesh,5 Yu. N. Khaidukov,6 Yu. V. Nikitenko,5 V. L. Aksenov,5 V. N. Gladilin,1,7,8 V. F. Kozhevnikov,1,2 C. V. Giuraniuc,1,* M. J. Van Bael,1 K. Temst,3 C. Van Haesendonck,1 T. M. Mishonov,4

1Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, 3001 Leuven, Belgium
2Science and Mathematics Division, Tulsa Community College, Tulsa, Oklahoma 74119, USA
3Instituut voor Kern- en Stralingsfysica, Katholieke Universiteit Leuven, 3001 Leuven, Belgium
4Department of Theoretical Physics, St. Clement of Ohrad University of Sofia, 1164 Sofia, Bulgaria
5ISIS Science Division, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, United Kingdom
6Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, 141980 Dubna, Moscow Region, Russia
7Theoretische Fysica van de Vaste Stoffen, Universiteit Antwerpen, 2020 Antwerpen, Belgium
8Physics of Multilayer Structures, State University of Moldova, 2009 Chisinau, Moldova
9Instituut voor Theoretische Fysica, Katholieke Universiteit Leuven, 3001 Leuven, Belgium

(Received 2 June 2008; published 10 July 2008)

Polarized neutron reflectometry (PNR) provides evidence that nonlocal electrodynamics governs the magnetic field penetration in a low-κ superconductor. The sample is an In film with a large elastic mean-free path. It is shown that PNR can resolve the difference between the reflected neutron spin asymmetries predicted by the local and nonlocal theories of superconductivity. The experimental data support the nonlocal theory, which predicts a nonmonotonic decay of the magnetic field.

DOI: 10.1103/PhysRevB.78.012502

PACS number(s): 74.20.–z, 74.25.Ha, 78.70.Nx

In this Brief Report we pose and answer experimentally the following fundamental questions: Are nonlocal electrodynamics effects measurable in superconductors? Can the nonmonotonic decay of magnetic field penetration predicted by the nonlocal theory be observed? To what extent can polarized neutron reflectometry (PNR) resolve the difference between local and nonlocal diamagnetic responses expected for type-I superconductors?

Nonlocality is a key concept of superconductivity theory, but its experimental verification is still not established. In the Meissner state, a magnetic field applied parallel to the surface located at \(z=0\) causes the magnetic induction \(B(z)\) to penetrate over a depth \(\lambda = B(0)^{-1}B(z)dz\). In the London (local) limit, \(B(z) = \exp(-z/\lambda_L)\), where \(\lambda_L\) is the London penetration depth. In 1953, to explain the variation of \(\lambda\) in Sn due to a change of the mean-free-path \(\ell\), Pippard proposed that the current density is related to the average of the vector potential over a region of size \(\xi_0\) (the Pippard coherence length). More recently the concept of nonlocality was applied to high-\(T_c\) cuprates.

In the nonlocal theory \(B(z)\) deviates from a simple exponential decay; it is nonmonotonic and, moreover, changes sign at a specific depth. In the pure limit \((\xi_0 \ll \ell)\) \(B(z)\) is a function of the intrinsic parameters \(\lambda_L(T=0)\) and \(\xi_0\), and the temperature \(T\). The magnitude of this nonlocal effect is determined by the ratio \(\xi_0^2/\lambda_L(0) \approx 1/k_\lambda\); the smaller the Ginzburg-Landau parameter \(k\), the bigger the nonlocal effect. It is most significant in “extreme” type-I superconductors, such as Al (\(k \approx 0.01\)) and In (0.06). For these the results of the Pippard theory are identical to those of the Bardeen-Cooper-Schrieffer theory, in which nonlocality follows from the spatial separation of electrons in Cooper pairs. Thus, if confirmed, the nonlocal effect allows one to measure the size of the Cooper pairs (\(\xi_0\)) and \(\lambda_L(0)\), which are currently calculated using the theory.

\(B(z)\) in In, calculated in local and nonlocal approaches, with \(\xi_0=0.38 \mu m\) and \(\lambda_L(0)=0.025 \mu m\), for \(T=1.8\) K, is shown in Fig. 1. Details on the formalism can be found in Ref. 5. In the nonlocal approach \(B(0)/e=k_B=2(\lambda_L)\). The sign reversal is expected at \(z=5.5\lambda_L\), and the amplitude of the reversed field is about 0.03\(B(0)\). Indium is chosen due to its convenience for experiment.

An observation of sign reversal was reported in Ref. 6. An external ac magnetic field \(H\) with amplitude up to 30 Oe was applied parallel to a cylindrical Sn film, and a strongly attenuated (\(10^8\) times) signal with reversed phase was detected inside the cylinder at 2.88 K and 25 Oe. It was interpreted as a sign change in the penetrating field. However, this interpretation is questionable because the phase difference drops back to zero at a larger (30 Oe) field, whereas the critical field \(H_c\) at 2.9 K is 115 Oe.

![FIG. 1. Magnetic-induction profiles B(z) in a semi-infinite In sample. The dashed (solid) line corresponds to the local (nonlocal) relation between current density and vector potential.](image-url)
Nowadays $B(z)$ can be measured directly using PNR (Ref. 8) and low-energy muon spin rotation (LE-μSR) (Ref. 9) techniques. We comment briefly on the latter before focusing on the former.

In the LE-μSR technique polarized $\mu^+$ (lifetime $2.2 \mu$s) are implanted in a sample over a distance determined by the muon energy. $B(z)$ is obtained from the precession frequency of the muon spins at stopping distance. However, in practice the muon precession is strongly damped due to a broad distribution of stopping distances.\(^{10}\) This is the main difficulty in applying the LE-μSR technique to fields with a sharp profile.

Recently the LE-μSR technique was used to measure $B(z)$ in Pb, Nb, and Ta.\(^{10}\) Most interesting is the reported nonexponential shape of $B(z)$ for all these metals. The nonlinearity of $\log B(z)$ plots is marginal, which is coherent with the theory in view of the fairly high $\kappa$ of the studied samples. For example, $\kappa$ of pure Nb (residual resistivity ratio $RRR = 1600$) is 1.3 at 3 K and 1.0 at 7 K.\(^{11}\) However, in Ref. 10 $\kappa$ of less pure Nb ($RRR = 133$) is reported to be 0.7(2). This inconsistency with established literature data suggests that the muon probing results may contain some hidden uncertainties. Therefore, additional experiments would be worthwhile, in particular to verify the reliability of these results.

The PNR technique is based on the change of the neutron index of refraction in a magnetized medium. When a polarized neutron beam is incident on a laterally uniform sample under a grazing angle, its specular reflectivity $R$ is determined by the profile of the neutron-scattering potential below the surface. $R$ is measured versus momentum transfer $Q = 4\pi \sin \theta / \lambda_n$, where $\theta$ is the angle of incidence and $\lambda_n$ is the neutron wavelength. The scattering potential consists of a nuclear and a magnetic part, which results in different reflectivities $R^+$ and $R^-$ for neutrons with spins parallel (up) and antiparallel (down) to the applied field, respectively. The sample magnetization is obtained from the spin asymmetry $s = (R^+ - R^-)/(R^+ + R^-)$ by fitting $s(Q)$ data with $s(Q)$ simulations based on theoretical models for $B(z)$. The neutron reflectivity also depends on some other spin-independent parameters such as beam divergence $d\theta / \theta$, these parameters are determined independently and fine tuned using $R$-data for a nonmagnetically sample. Then $s(Q)$ is solely determined by the sample magnetization. PNR has been applied for measuring superconducting properties of Nb,\(^{12,13}\) high-$T_c$ cuprates,\(^{14-16}\) and Pb.\(^{17,18}\)

The nonlocal effect in $B(z)$ measured with PNR was discussed in Refs. 12, 13, 17, and 18. Although some deviation from exponential decay was noticed in Refs. 13 and 17, no confirmation of the nonlocal theory was obtained. The authors of Ref. 18 correctly pointed out that experiments with lower-$\kappa$ materials are desirable to verify nonlocality, but their overall conclusion was that PNR is incapable of detecting nonlocality in any superconductor.

Figure 2 shows $s(Q)$ calculated for an In layer with the “local” and “nonlocal” field profiles; details on the formalism are available in Ref. 19. The simulations indicate that the difference between spin asymmetries for the local and nonlocal approaches can be of the order of 10%, which is feasible for state-of-the-art PNR facilities. Therefore it is interesting to reassess the problem of nonlocality with PNR applied to a low-$\kappa$ material.

![FIG. 2. Simulations of $s(Q)$ for a semi-infinite In layer based on the local (dashed lines) and on the nonlocal (solid lines) approaches. The instrumental resolution $\Delta Q/Q$ is 0.01, 0.03 and 0.1 for the curves 1, 2 and 3, respectively.](image-url)
BRIEF REPORTS

PHYSICAL REVIEW B

78

acterized by the measured dc magnetization detected.

resistivity. The shape of the tent with the negative result of Rutherford backscattering reflectometer measured on our sample: No oxide film has been detected.

The electromagnetic properties of the sample were characterized by the measured dc magnetization and electrical resistivity. The shape of the $M(H)$ curves is typical for type-I superconductors. The obtained phase diagram $H_c(T)$ agrees well with the literature data.\(^7\) $T_c$ of our sample (3.415 K) matches the tabulated value of 3.4145 K.\(^21\) and RRR=540. Correspondingly, $\ell \approx 11$ $\mu$m is much larger than $\xi_0$. Therefore, our sample is a type-I superconductor in the pure limit.

PNR experiments were performed on the REMUR reflectometer at the Joint Institute for Nuclear Research (Dubna) and on the CRISP instrument at ISIS (Oxford). Both sets of measurements confirm that splitting of the $R^*(Q)$ and $R^-(Q)$ curves is achievable for our sample. The ISIS data, which are the most detailed, allow a quantitative analysis to which we now turn.

CRISP operates with a spin-polarized polychromatic pulsed neutron beam. The angle $\theta$ and $\Delta Q/Q$ were set to 0.24 degrees and 3%, respectively.

The reflectivity in the Meissner state was measured at $T = 1.8$ K and $H=77$, 140, 166, and 194 Oe [$H_c(1.8$ K) = 205 Oe]. The obtained data sets are shown in Fig. 3. The $R(Q)$ dependencies exhibit a hill caused by total reflection from the substrate. The splitting between $R^*$ and $R^-$ is clearly visible near $Q_c$, different magnitudes of the error bars are due to different times of exposure. The data obtained at 77 and 166 Oe have the smallest statistical error and will be used for further discussion.

The data obtained in the normal state ($T=4.6$ K) are shown in Fig. 4. Solid curves are simulations, in which the sample is a pure In film on a SiO$_2$ substrate. In the simulations $\Delta Q/Q$ was allowed to vary due to the unknown uncertainties of the set value and of the geometrical factor (as only part of the beam covers the sample).

The simulation curve near $Q_c$ is mostly controlled by the resolution (see Fig. 4 and, e.g., Ref. 17). The next segment, down to the foothill, is determined by the roughness of the sample surface. The location of the ascending part $[0.011 < Q(\text{Å}^{-1}) < 0.014]$ is governed by the film thickness. The segment following the hill is determined by the substrate scattering properties. No attempts were made to achieve a better fit for that segment, because there the spin asymmetry is indistinguishable from zero.

The best fit (curve 1 in Fig. 4) was obtained for the model sample with $\sigma=14$ nm and $\Delta Q/Q=2.5\%$. Fitting the ascending part enables one to determine $d$ in situ. The statistical error of the reflectivity data in this region being $\pm 5\%$, the thickness was found to be $2400 \pm 30$ nm, in agreement with the nominal thickness of $2.5 \mu$m. These parameters were further used for simulating the spin asymmetry. Attempts to introduce an indium oxide layer on top of the sample yielded no reasonable fit for any appreciable thickness ($>1$ nm) of the oxide layer. This is consistent with our expectation that the oxide layer does not affect the neutron reflectivity.

Simulations of the reflectivity in the Meissner state were performed assuming on both sides of the sample the field profiles shown in Fig. 1. The $s(Q)$ data for fields 77 Oe and 166 Oe, along with simulations for the local and nonlocal field distributions, are shown in Fig. 5.

For field 77 Oe [Fig. 5(a)], the results of the “nonlocal” simulation fit the experimental data somewhat better, but no clear discrimination between the local and nonlocal approaches is possible due to insufficient accuracy of these
FIG. 5. Spin asymmetry at $T=1.8$ K and $H=77$ Oe (a) and 166 Oe (b). The curves are simulations performed within the local (dashed line) and nonlocal (solid line) approaches.

---

*Present address: Interdisciplinary Research Institute, CNRS USR 3078, 59021 Lille, France.

1A. B. Pippard, Proc. R. Soc. London, Ser. A 216, 547 (1953).

2I. Kosztin and A. J. Leggett, Phys. Rev. Lett. 79, 135 (1997).

3M. Tinkham, Introduction to Superconductivity (McGraw-Hill, New York, 1996).

4P. Valko, M. R. Gomes, and T. A. Girard, Phys. Rev. B 75, 140504(R) (2007); K. S. Wood and D. Van Vechten, Nucl. Instrum. Methods Phys. Res. A 314, 86 (1992).

5J. Halbritter, Z. Phys. 243, 201 (1971).

6K. E. Drangeid and R. Sommerhalder, Phys. Rev. Lett. 8, 467 (1962).

7D. K. Finnemore and D. E. Mapother, Phys. Rev. 140, A507 (1965).

8G. P. Felcher, Physica B (Amsterdam) 192, 137 (1993).

9E. Morenzoni, F. Kottmann, D. Maden, B. Matthias, M. Meyberg, Th. Prokscha, Th. Wutzke, and U. Zimmermann, Phys. Rev. Lett. 72, 2793 (1994).

10A. Suter, E. Morenzoni, N. Garifianov, R. Khasanov, E. Kirk, H. Luetskens, T. Prokscha, and M. Horisberger, Phys. Rev. B 72, 024506 (2005).

11D. K. Finnemore, T. F. Stromberg, and C. A. Swenson, Phys. Rev. 149, 231 (1966).

12G. P. Felcher, R. T. Kampwirth, K. E. Gray, and R. Felici, Phys. Rev. Lett. 52, 1539 (1984).

13H. Zhang, J. W. Lynn, C. F. Majkrzak, S. K. Satija, J. H. Kang, and X. D. Wu, Phys. Rev. B 52, 10395 (1995).

14R. Felici, J. Penfold, R. C. Ward, E. Ols, and C. Mataka, Nature (London) 329, 523 (1987).

15A. Mansour, R. O. Hillege, G. P. Felcher, R. B. Laibowitz, P. Chaudhari, and S. S. P. Parkin, Physica B (Amsterdam) 156-157, 867 (1989).

16V. Lauter-Pasyuk, H. J. Lauter, V. L. Aksenov, E. I. Kornilov, A. V. Petrenko, and P. Leiderer, Physica B (Amsterdam) 248, 166 (1998).

17K. E. Gray, G. P. Felcher, R. T. Kampwirth, and R. Hillege, Phys. Rev. B 42, 3971 (1990).

18M. P. Nutley, A. T. Boothroyd, C. R. Staddon, D. McK. Paul, and J. Penfold, Phys. Rev. B 49, 15789 (1994).

19S. J. Blundell and J. A. C. Bland, Phys. Rev. B 46, 3391 (1992).

20K. Temst, M. J. Van Bael, and H. Fritzsche, Appl. Phys. Lett. 79, 991 (2001).

21Handbook of Physical Quantities, edited by I. S. Grigoriev and E. Z. Meilikhov (CRC, New York, 1997).

22V. L. Aksenov, K. N. Jernenkov, S. V. Kochezhnikov, H. Lauter, V. Lauter-Pasyuk, Yu. V. Nikitenko, and A. V. Petrenko (www.jinr.ru/publish/Preprints/2004/047/D13-2004-47-e.pdf).

23D. G. Bucknall, S. Langridge, and R. M. Dalgliesh (www.isis.rl.ac.uk/largescale/crisp/).