Observation of enhanced nuclear spin-lattice relaxation by superconducting fluctuations in thin films by β-NMR

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Abstract

We report β-NMR investigations of polarized ^{8}Li nuclei implanted in a thin Pb film and in Ag/Nb bilayers. We observe a pronounced singular peak in the longitudinal spin relaxation at the superconducting temperature, which we ascribe to superconducting fluctuations. Such an enhancement of the $1/T_1$ relaxation at T_c has never been observed. The magnitude of the effect cannot be solely explained by the enhancement of the dynamic electron spin susceptibility as calculated by diagrammatic techniques.

Introduction

Near the superconducting critical temperature T_c,thermodynamic fluctuations of the order parameter occur, leading to the appearance of pairing correlations with critically increasing life time when approaching T_c from above [1]. The effects of pairing fluctuations are reported above T_c in the excess conductivity (paraconductivity), tunnel conductivity, enhanced diamagnetism, and in many other electronic properties [2].

The importance of fluctuations increases in systems of lower dimensionality as thin films, wires and small particles. Unconventional superconductors such as cuprates, with their quasi 2D electronic structure, low superfluid density and short coherence length, are even more susceptible to fluctuations [3]. In these systems the question about the importance of fluctuations is closely related to the nature of the transition from the pseudogap regime into the superconducting phase, which is still not understood and probably related to the close proximity of a quantum critical point [4].

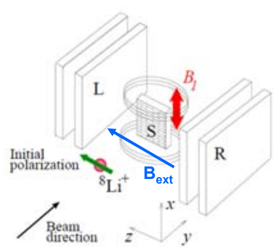
NMR can give detailed information of the normal and superconducting electronic state. It has played a fundamental role in elucidating the nature of low and high-T_c superconductivity [5].
The contribution of superconducting fluctuations to T_1 has been calculated at different levels of approximation (see [1] for an overview). Unless strong pair-breaking mechanisms are introduced, a singular peak in $1/T_1$ is expected in the case of s-wave pairing.
However, until now no singular behavior of the spin-lattice relaxation at T_c has been reported, either in conventional or in unconventional superconductors. In conventional superconductors the observation of superconducting fluctuations is believed to be difficult due to the smallness of the effect [6]. In case of d-wave pairing the dominant contribution, the so-called Maki-Thomson term, is suppressed [7,8].

Experiment

We measured resonance curves and the longitudinal spin relaxation rate $1/T_1$ of polarized ^{8}Li ions implanted in a Pb film and in the Ag overlayer of Ag/Nb bilayers.

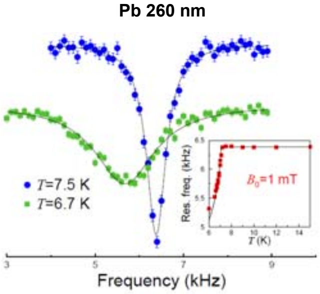
Samples: Pb : 260 nm, T_c= 7.12 K, $\Delta T_c \lesssim$ 8 mK
Ag/Nb : Ag 30-120 nm on Nb 300nm,
(due to the proximity effect Ag becomes superconducting at T_{cNS} = 9.1K ($\Delta T_{cNS} \lesssim$ 40 mK)

β-NMR Seup at ISAC



Results

Pb 260 nm



Resonance curves of ^{8}Li ions implanted at 5 keV in Pb with a magnetic field of 1 mT applied parallel to the surface of the film.

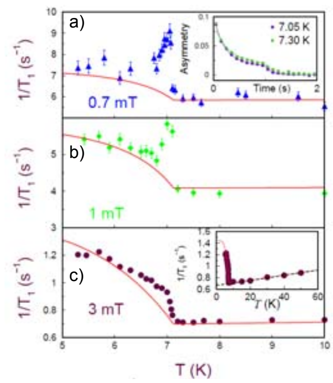
At this energy the implantation range of Li extends to ~ 100 nm with a mean implantation depth and rms of 34.05 nm and 19.22 nm, respectively.
The insert shows the temperature dependence of the resonance frequency.

The temperature dependence of the shift reflects the appearance of the Meissner state. The data can be well fitted by a solution of the London equation taking into account the implantation profile and yielding an effective penetration depth

$$\lambda_{eff}(T) = \frac{\lambda_L(0)}{\sqrt{1-(\frac{T}{T_c})^4}}\sqrt{1+\frac{\xi_0}{\ell}}$$

with $\lambda_{eff}(0) = 62.2 \pm 0.3$ nm, $\xi_0 = 83$ nm and $\ell = 48$ nm

Pb 260 nm



Spin lattice relaxation rate $1/T_1$ versus temperature from single exponential fits to relaxation data in the Pb film (260 nm) with implantation energy 5 keV. a) B_{ext} =0.7 mT, b) B_{ext} =1$ mT, c) B_{ext} =3 mT.

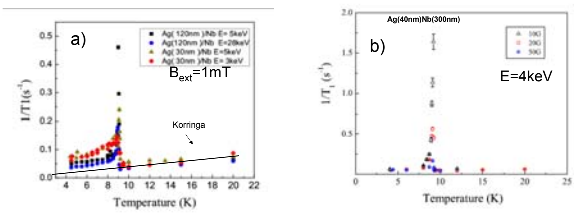
The solid curves show the contribution from the host nuclear dipole dynamics including the increase in relaxation due to the field screening below T_c and a small Hebel-Slichter contribution [9].

Insert in (a) typical polarization spectrum, in (c) extended temperature range showing the Korringa contribution, due to the interaction of Li with the spin of the conduction electrons. Fitting this region to the Korringa law we obtain a slope 0.0044 (sK)$^{-1}$, a value very close to that found in other elemental metals [10].

The temperature dependence of $1/T_1$ shows a sharp increase within less than ~ 50 mK at T_c. The peak is more pronounced at 0.07 mT than at 1mT and broadens at 3 mT. We attribute this peak to superconducting fluctuations.

Similar behavior and field dependence with even more pronounced singular peak is observed at T_{cNS} in Ag/Nb when Li is stopped in a proximated Ag overalyer on Nb.

Ag/Nb



Spin lattice relaxation rate 1/T1 versus temperature from single exponential fits to relaxation data in Ag/Nb bilayers. Li is stopped in Ag. Nb does not contribute to the signal due to very fast relaxation. a) Thickness dependence, b) Field dependence.

The detailed structure of the superconducting fluctuations in the Gaussian regime has been calculated by the diagrammatic technique in a microscopic theory involving the hyperfine contact coupling.
The anomalous Maki-Thomson (MT) [1,8] term is the only positive and dominating contribution to the relaxation rate. It is due to fluctuating correlations formed by electrons moving along self-intersecting trajectories. The corresponding enhancement of the NMR relaxation rate over the normal Korringa value can be analytically calculated [7,11]:

$$\frac{\frac{1}{T_1}^{SF}}{\frac{1}{T_1}^{n}} = \frac{\pi\hbar}{8E_F\tau}\frac{1}{\varepsilon-\gamma_\Phi}\ln\left(\frac{\varepsilon}{\gamma_\Phi}\right) \text{ where } \varepsilon = \frac{T-T_c}{T_c},\ \tau = \frac{\ell}{v_F},\ \gamma_\Phi \text{ pair breaking parameter}$$

However, this expression representing the modification of the dynamic spin susceptibility by superconducting fluctuations underestimates the observed jump in spin-lattice relaxation by more than an order of magnitude. Moreover, it does not reproduce its temperature and field dependence.

Conclusions

We have observed a pronounced rise of the longitudinal spin-lattice relaxation in a narrow region close to the critical temperature of thin films reflecting an unexpected strong coupling of the ^{8}Li spin to low frequency fluctuations. Although more quantitative estimates must await further investigations, the results indicate that the singular contribution to the dynamic spin susceptibility as calculated in lower order cannot predict the magnitude and frequency spectrum of the effect. Other mechanisms, that may add to direct contact interaction have to be considered. Since the order parameter is complex phase fluctuations and associated supercurrents can create fluctuating magnetic fields relaxing the spin probe even in cases where the contact term is small or zero [12]. Local fluctuating fields can be also created by diamagnetic fluctuations.

The suppression of $1/T_1$ as a function of magnetic field indicates there is a decrease in the size (and corresponding correlation time) associated with the fluctuating domains with increasing applied field [2]. Our observation also demonstrates the sensitivity of β-NMR to low frequency dynamics in superconductors. Contrary to conventional NMR, depth dependent β-NMR is not hampered by shielding effects of high frequency fields. The implantation range of the polarized nuclei makes them suitable to application in low dimensional systems where fluctuation effects are enhanced e.g. heterostructures containing insulating and superconducting layers, where it can be stopped in the insulating environment to act as a sensitive probe of fluctuations in the nearby superconducting layer .

References

[1] A. Larkin and A. Vaslamov, *Theory of fluctuations in superconductors* OUP, (2009)
[2] W. J. Skocpol and M. Tinkham, Rep. Prog. Phys. 38, 1049 (1975)
[3] V. J. Emery, S. A. Kivelson, Nature 374, 434-437 (1995).
[4] J. Orenstein and A. J. Millis, Science 288, 468 (2000)
[5] A. Rigamonti, F. Borsa and P. Carretta, Rep. Prog. Phys. 61, 1367 (1998)
[6] D. MacLaughlin, *Solid State Physics*, p. 1, Academic Press (1976)
[7] K. Kuboki, H. Fukuyama, J. Phys. Soc. Jpn, 58,376 (1989).
[8] K. Maki, Progr. Theor. Phys. 40, 193 (1968), R. S. Thompson, Phys. Rev. B1, 327 (1970).
[9] M. D. Hossain, Z. Salman, D. Wang, K. H. Chow, S. Kreitzman, T.A. Keeler, C. D. P. Levy, W. A. MacFarlane, R. I. Miller, G. D. Morris, T. J.Parolin, M. Pearson, H. Saadaoui, R. F. Kiefl, Phys. Rev. B 79 , 144518 (2009)
[10] M.D. Hossain et al., Physica B404, 914 (2009)
[11] M. Randeria, A. Varlamov, Phys. Rev. B50, 10401 (1994).
[12] M. Cyrot, Solid State Communications, 11, 747 (1972) 801 (1986).