10 Muon-Spin Rotation (μSR) and Magnetization Studies of Superconducting Materials

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10.1 Introduction

In the work carried out over the past year we have continued our detailed study of the magnetic properties of high-temperature superconductors and related magnetic systems. In order to achieve this goal we have used a combination of muon spin rotation (μSR), electron paramagnetic resonance (EPR), together with bulk SQUID and torque magnetometry measurements, and thermal expansion measurements.

The μSR technique probes the local internal field distribution in the mixed state of a superconductor. Over the past years this technique has been applied, among other things, to understanding the microscopic arrangements of flux vortices and associated transitions (flux-lattice melting) in highly anisotropic superconductors, where significant progress has been achieved. In the last year we have continued our μSR studies of $Bi_2Sr_2CaCu_2O_8$ (BSCCO), in which we intend to investigate the exotic vortex behavior at a microscopic level, and extended these studies to highly anisotropic organic superconductors and BSCCO containing columnar defects induced by irradiation with heavy ions (see Sec. 10.2).

The nature of the symmetry of the superconducting pair wave function is a fundamental question of current interest. In order to test the pairing symmetry we have carried out precise measurements of the ab-anisotropy of $Tl_2Ba_2Cu_3O_{6+x}$ films by means of a specially designed torque sensor based on a silicon lever (see Sec. 10.3.1). In addition, we have improved the design and the performance of our recently constructed nano-torque magnetometer (see Annual Report 95/96). For this purpose we have designed a new type of piezoresitive cantilever, which can simultaneously operate in the flexion and torsion mode and allows calibration measurements with an integrated calibration coil (see Sec. 10.3.2).

EPR is a powerful method to study crystal electric fields and static and dynamic correlations on a microscopic level. Last year this technique has successfully been used the first time to demonstrate an oxygen isotope effect of the ferromagnetic exchange energy in colossal magnetoresistive manganites (see Sec. 10.5.1), providing microscopic evidence for the formation of Jahn-Teller (JT) polarons in doped manganites (see Sec. 10.4). Furthermore, we performed EPR measurements in order to investigate the peculiar magnetic properties of the spin-ladder compound $Sr_{0.72}CuO_2$ at a microscopic level.

We have continued our oxygen-isotope effect studies in the giant magnetoresistance ferromagnetic Jahn-Teller (JT) perovskite $La_{1-x}Ca_xMnO_3$ by means of magnetization and EPR measurements, as well as measurements of the thermal expansion coefficient (see Annual Report 95/96). Our results provide strong evidence for the formation of JT polarons in these materials (see Sec. 10.5.1). Furthermore, we have performed magnetization and thermal expansion measurements on ^{16}O and ^{18}O samples of the cuprate superconductor $La_{2-x}Sr_xCuO_4$ for investigating the isotope effect on the in-plane penetration depth (see Sec. 10.5.2). Our results show that polaronic charge carriers exist and condense into Cooper pairs in the cuprate superconductors, as originally suggested by Bednorz and Müller.

This work was done in collaboration with scientists from ETH Zürich (G. Blatter, K. Conder, and J. Karpinski), IBM Rüschlikon Laboratory (C. Rossel), University of Birmingham (E.M. Forgan and S.H. Lloyd), University of St. Andrews (S.L. Lee and C. Ager), Institut Max von Laue-Paul Langevin, Grenoble (R. Cubitt), University of Oxford (S.J. Blundell), RIKEN-RAL (F.L. Pratt), Tohoku University (T. Sasaki), and Free University of Amsterdam (B. Stäuble-Pümpin).

10.2 μSR measurements of highly anisotropic superconductors

The high-temperature superconducting cuprates show anisotropic superconducting and magnetic behavior. This is largely due to their layered structure, where the currents preferentially flow in the CuO_2 planes. This anisotropy is parameterised by $\gamma = \lambda_c/\lambda_{ab}$, where λ_c and λ_{ab} are the penetration depths for currents flowing perpendicular and parallel to the superconducting planes, respectively. The high-temperature superconductor $Bi_{2.15}Sr_{1.85}CaCu_2O_{8+\delta}$ (BSCCO) shows an extreme anisotropy with γ as high as 200 for optimally doped samples. Therefore in this material a vortex line penetrating the sample is better described by a stack of 'pancake' vortices, each confined to a CuO_2 plane, rather then an extended, three dimensional (3D) object. These 'pancake' vortices interact both within the planes as well as between different adjacent planes. The interactions within the planes originate always from the supercurrents flowing in the CuO_2 planes. Between the planes the interactions can arise from Josephson tunneling currents flowing between the planes or also from screening currents in the adjacent layers. These two cases are referred to as Josephson coupling and electromagnetic coupling, respectively. Usually, Josephson coupling is taken to make up the main part of the inter-layer coupling in high temperature superconductors. However, for extremely anisotropic superconductors, obeying the relation $\gamma s > \lambda_{ab}$, where s is the inter-layer separation, Blatter *et al.* [1] have shown, that the vortex behavior may well be dominated by electromagnetic coupling.

Using μSR we can microscopically probe the vortex distribution in the mixed state of type II superconductors. This is done over the field probability distribution $p(B)$, which is intimately related to the spatial distribution $B(r)$. This has allowed us in the past to observe such interesting phenomena as vortex lattice melting and the dimensionality of the vortex system [2, 3]. This year we have investigated these phenomena further to obtain information on the nature of the inter-layer coupling in the extremely anisotropic BSCCO. Following the analysis of Blatter *et al.* [1], we investigated the field dependence of the melting temperature in detail, as differences are expected for the different types of inter-layer coupling. Especially, there should be a crossover in the shape of the melting line, at the temperature T^{em}, where the criterion for the dominance of electromagnetic coupling breaks down, $\lambda(T^{em}) = \gamma s$. This behavior was indeed observed [4] and allowed us to determine γ to be $\simeq 160$.

The nature of the inter-layer coupling also has interesting consequences on the dimensional crossover field. At this field, there is a strong change in the field distribution p(B) [5] to a more symmetric shape. This has been interpreted as arising from the improved adjustment of pancake vortices to pinning sites, when the inter-layer coupling becomes weaker than the intra-layer coupling. For a system consisting of Josephson coupled pancake vortices a calculation of these properties gives $B_{2D} = \Phi_0/(\gamma s)^2$ [6], where $\Phi_0 = h/2e$ is the flux-quantum. This means that the Josephson coupling is dramatically weakened as the cores of the Josephson vortices associated with the Josephson currents start to overlap. For superconductors such as BSCCO however, where $\gamma s > \lambda_{ab}$ the vortices are still further apart then the typical length scale of their interaction (λ_{ab}), such that they interact very weakly. Hence the electromagnetic inter-layer coupling is still stronger than the intra-plane interactions, delaying the crossover up to a field of $B_\lambda = \Phi_0/\lambda_{ab}^2$, where the vortices are only λ_{ab} apart and therefore interact more strongly. We have investigated the dimensional crossover for a number of samples having different oxygen contents and therefore different penetration depths using μSR. The result is shown in Fig. 10.1, where we have plotted the crossover field as a function of the inverse square of the penetration depth. It can be clearly seen that the crossover field in the samples studied follows the behavior expected from the above discussion. To study the dependence of this dimensional crossover on the anisotropy we have also carried out measurements on BSCCO samples containing columnar defects induced by irradiation with 17.3 GeV U ions. The tracks of these heavy ions act as very strong pinning sites, such that a stack of pancake vortices will be ordered along them effectively increasing the dimensionality of the vortex system. In our μSR and small angle neutron scattering (SANS) measurements we have observed this effect. The field distribution $p(B)$ shows a tail at high fields and there is scattering from extended vortex lines in SANS [9] at fields much higher than the crossover field in the pristine material. Furthermore we have shown that the 2D thermal fluctuations present in the unirradiated samples [3] are strongly reduced in those containing columnar defects [9]. This is due to the fact that the ionic tracks strongly pin the vortices along their length, which is of importance to applications since thermal fluctuations of vortices may lead to resistive behavior. Moreover, we have started an extensive study of the vortex behavior in a superconducting charge transfer salt of the organic

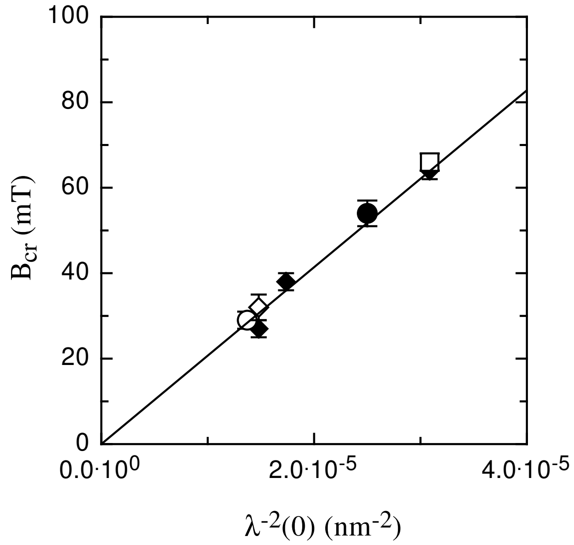

Figure 10.1: The crossover field B_{cr} as a function of $\lambda_{ab}^{-2}(0)$ for different samples of BSCCO. Open symbols correspond to μSR measurements on samples studied in ref. [5]: circle is sample S_1; square S_2; diamond S_3. Filled diamonds are taken from Ref. [7] and are obtained from magnetization measurements using Hall bar arrays. Filled circles are from a Tl-2212 sample taken from Ref. [8], where the crossover field is determined by magnetization measurements. The solid line corresponds to the relation $B_{cr} = \Phi_0/\lambda_{ab}^2$, which describes the data without any adjustable parameter.

molecule bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET). We studied the superconducting material κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ (ET-SCN), with a transition temperature of approximately 10.4 K, that shows extremely anisotropic behavior as BSCCO. We therefore expect to observe a dimensional crossover similar to that in BSCCO. Due to the very long penetration depths (600 nm) in these samples however, there might be some differences with respect to the inter-layer coupling. Our results on ET-SCN, where we observe a dimensional crossover at a field of $\simeq 5$ mT [10], however are consistent with a mechanism as in BSCCO, where the dominating inter-layer coupling is of electromagnetic nature.

10.3 Magnetization measurements of high-T_c and classical superconductors by torque magnetometry

10.3.1 Pairing symmetry in single-layer tetragonal $Tl_2Ba_2CuO_{6+x}$ from in-plane torque anisotropy

Since the discovery of high-T_c superconductivity several experiments for testing the pairing symmetry in these novel superconductors were performed, but the situation is still controversial. One part of the experiments, such as the phase-sensitive experiments of Tsuei et al. [11] performed with $Tl_2Ba_2CuO_{6+x}$ (Tl2201) and $YBa_2Cu_3O_{7-\delta}$ tricrystals yield evidence for a $d_{x^2-y^2}$ symmetry [12, 13]. On the other hand, a number of experiments, such as the Josephson tunneling experiment along the c-axis of optimally doped $YBa_2Cu_3O_{7-\delta}$ by Sun et al. [14] point to a s-wave symmetry. Recently, Müller [15] has proposed, that in these short-coherence length superconductors two condensates with different symmetry (s- and d-wave), but the same T_c, may be present, in agreement with a large number of experimental results.

In our experiment we studied the in-plane torque anisotropy in a Tl2201 film grown on $SrTiO_3$ (001) by rf magnetron sputtering [16]. Films of two different thicknesses were used. The tetragonal structure of Tl2201, checked at room temperature with x-rays, is certainly an advantage for this kind of experiment, compared to orthorhombic compounds like $YBa_2Cu_3O_{7-x}$. However, we cannot totally exclude a small orthorhombic distortion at low temperature. The films were patterned into disks of 2 mm diameter to avoid geometrical effects. A capacitance torque meter

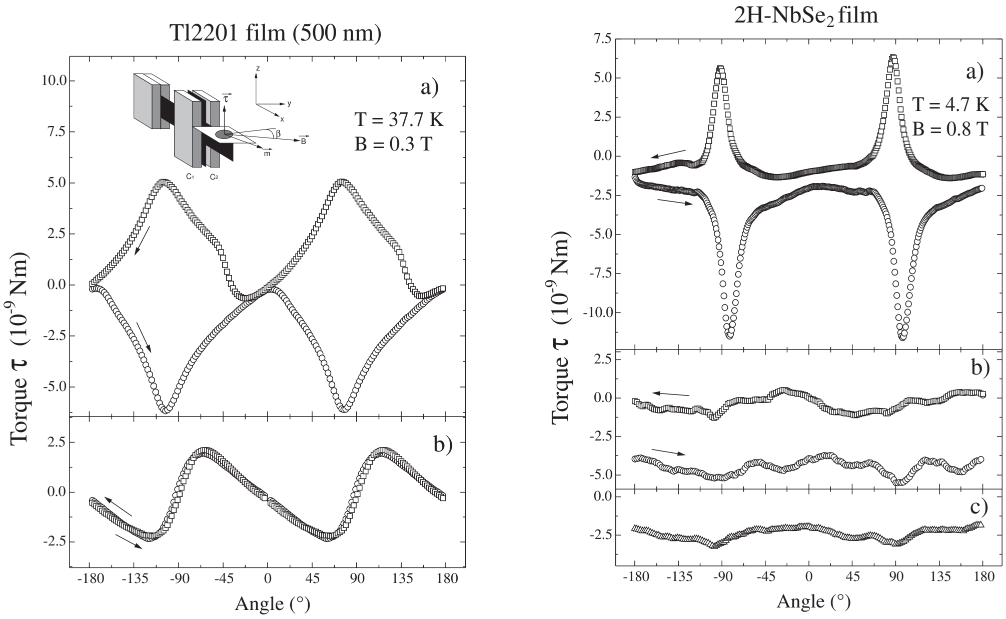

Figure 10.2: Left : a) Out-of-plane torque of a 500 nm-thick Tl2201 film. In this case the c-axis magnetization dominates and the large irreversibility is due to pinning. b) In-plane torque, almost perfectly reversible. Insert: Thin film mounted on the capacitance torque meter. Right : Angular dependent torque of a 2H-$NbSe_2$ film. a) Before proper in-plane alignement of B. b) After alignement corrections. c) Reversible part of the torque.

based on a silicon lever allows measurements on thin films grown on substrates with a very good resolution ($\sim 10^{-11}$ Nm) (see insert of Fig. 10.2a, left). The intrinsic background signal induced by the lever is negligibly small. For these measurements the magnetic field has to be applied exactly in the *ab*-plane of the sample to avoid an unwanted contribution from the *c*-axis magnetization (Fig. 10.2a, left). A misalignment less than $0.07°$ was achieved with a small magnetic field perpendicular to the main external field.

The comparison of the in-plane torque of the Tl2201 (Fig. 10.2b, left) and the 2H-$NbSe_2$ (Figs. 10.2b and c, right) shows two totally different behaviors. For the conventional superconductor also a pinning contribution in the *ab*-plane was ob-