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POLARIZED NEUTRON REFLECTOMETRY STUDIES OF FLUX PENETRATION IN $YBa_2Cu_3O_7$ SUPERCONDUCTING FILMS

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The technique of Polarized Neutron Reflectometry (PNR) was used for the measurement of the magnetic flux penetration in high–temperature (high–T_c) superconducting thin films. Different superconducting phases of high–T_c films were studied, namely the Meissner state ($H < H_{c1}$) and the mixed state ($H_{c1} < H < H_{c2}$). The experiments on high–T_c films were made in two scattering geometries: reflection from the "vacuum–film" or "substrate–film" interface. For the Meissner state the model fit to the experimental data gave a magnetic penetration depth of 1350±150 Å along the c–axis at a temperature of $T = 2$ K. The c–axis was oriented perpendicular to the film surface. The model included the intrinsic exponential decay of the penetrating flux. In the mixed state the distribution of flux–lines in the high–T_c thin film was determined for the first time by PNR in an external magnetic field of 1.5 kOe applied parallel to the film surface.
Key words: high–T_c superconducting film, magnetic penetration depth, polarized neutron reflection, vortex.

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I. INTRODUCTION

The technique of Polarised Neutron Reflectometry (PNR) is a sensitive probe to determine the *absolute* value and the *shape* of the magnetic penetration depth λ_L, one of the fundamental parameters of superconductors. This technique has already been used to measure the magnetic field penetration depth in classical superconducting films in the Meissner state for niobium [1] $\lambda = 410$ Å, subsequent experiments were carried out on pure lead and lead–bismuth films [2, 3]. Recently, more investigations on Nb system were published [4, 5] $\lambda =$ 900–1450 Å. The high difference in the values obtained for λ for different films can be explained by the fact that it was not the magnetic penetration depth that was measured but the screening length $\lambda > \lambda_L$. The screening length λ takes into account electron scattering at defects in the sample and depends on the coherence length ξ and the electron's mean free path l in the following way [6]: $\lambda = \lambda_L\sqrt{1+\xi/l}$. This effect is discussed in Ref. 7, in which the measured value for l was corrected for electron scattering and = 430 was obtained, which is the best result today for Nb–films. Following the advent of high–T_c superconductors, measurements on $YBa_2Cu_3O_{7-x}$ were also reported [8, 9, 7], but the quality of the obtained results was not sufficient which was explained by an insufficient quality of the samples itself. In our previous publications [10, 11] we optimized both, parameters of the samples and of the PNR method applied to this problem. For this purpose the model calculations, careful selection and the characterization of the samples were made. This allowed us to provide a series of experiments on $YBa_2Cu_3O_7$ thin films with a thickness of 3000 Å epitaxially grown on $SrTiO_3$. The experiments were performed in two scattering geometries (discussed in 7, 10, 11) in an external magnetic field of 500 Oe at 4.8 K with the reflection from the air side and 300 Oe at 2 K with the reflection from the substrate side. Both measurements gave coinciding results for the London penetration depth. One should note that for high–T_c superconductors $\xi \ll l$, therefore in the experiment the penetration depth λ_L is measured.

The application of higher magnetic field parallel to the film surface allows the magnetic field to penetrate into the sample as quantized vortices of magnetic flux. In the limit of a thin film this flux lines are expected to be ordered in one row along the center plane. Increasing the magnitude of the external field the vortex system switches from a line to a staggered array [15]. Until now there were no PNR experiments on thin high–T_c films in the mixed state. With our first experiments we showed the feasibility of measuring vortices arrangement in thin films by means of PNR.

II. METHOD DESCRIPTION

If the external magnetic field H is applied parallel to the surface of a superconducting film and its value does not exceed the first critical field H_{c1}, then the magnetic flux penetrates inside the film from both interfaces with decaying magnitude. In the London limit this decay is intrinsic exponential, then the profile of magnetic flux inside a superconducting film with the thickness d is described by [14]:

$$\mathbf{B}(x) = \mathbf{H}\left(\frac{2x-d}{2\lambda_L}\right)\Big/\cosh\left(\frac{d}{2\lambda_L}\right), \qquad (1)$$

where x is the distance from the center towards the surfaces of the film. The neutron specular reflection depends on the variation of the scattering potential perpendicular to the sample surface. The scattering potential contains a nonmagnetic part, proportional to the nuclear scattering length density and a magnetic term determined by the difference between the magnetic induction inside and outside the sample. Then the neutron scattering length density profile is given by

$$Nb^{\pm} = Nb_n \mp cH\cosh\left(\frac{2x-d}{2\lambda_L}\right)\Big/\cosh\left(\frac{d}{2\lambda_L}\right), \qquad (2)$$

where Nb_n is the nuclear part of the scattering length density (SLD) (N is the number of atoms in a unit volume, b_n is the scattering length density), and $c=2.31\times10^{-8}$ nm^{-2}Oe^{-1}. The resulting SLD Nb is spin dependent and, therefore, in the experiment two reflectivity curves R^+ and R^- will be obtained due to the two spin states of the neutron in a magnetic field.

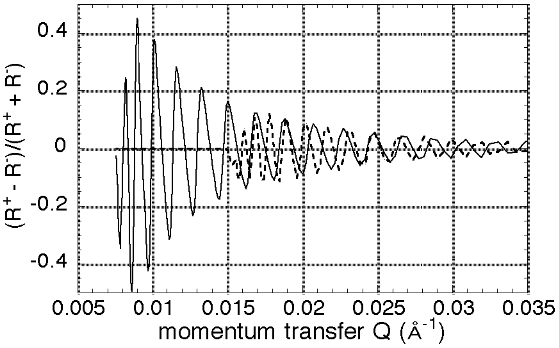

Fig. 1. Model calculation of the spin–asymmetry $SA = (R^+ - R^-)/(R^+ + R^-)$ as a function of momentum transfer for an $YBa_2Cu_3O_7$ thin film with a thickness of 3000 Å on $SrTiO_3$ substrate. Solid line corresponds to the reflection from the substrate side, dashed line — from the vacuum side.

If the magnitude of the external magnetic field exceeds the first critical field H_{c1} , than the magnetic flux penetrates into the film as vortices. The vortex lattice in thin films with H parallel to the surface is different from the Abrikosov lattice in the bulk superconductor. Due to the quasi two–dimensional case there will be a range of fields $H > H_{c1}$ in which the vortices will be placed in one line close to the center of the film. One expects that this "line –vortex structure will be changed to a "zig–zag – like one with increasing external field. The value of this field can be numerically calculated [15]. The presence of the vortex–plane inside the film will give rise to a modification of the neutron SLD distribution with respect to the Meissner state, so from the PNR experiment one can extract the flux–line density averaged along the surface and its distribution perpendicular to the film plane. The presence of the pinning centers deforms the perfect order of vortices and can give rise to a broadened flux distribution around the center plane.

Prior to the experiments the model calculations were performed to study two different possibilities of the scattering geometry: reflection from the air–film interface or substrate–film interface. In fig. 1 the results of the calculation are shown for a $YBa_2Cu_3O_7$ film with a thickness of 3000 Å on $SrTiO_3$ substrate. The magnetic effect shows up in the difference between R^+ and R^- which is demonstrated in the plot of the Spin–Asymmetry $SA = (R^+ - R^-)/(R^+ + R^-)$. The advantage of the scattering through the substrate is obvious due to a better contrast and, therefore, higher amplitude oscillations of the SA–function. However the disadvantage is that an intensity factor of 5 is lost when the neutrons traverse the $SrTiO_3$–substrate.

We performed experiments in both geometries to obtain independent results on the same sample.

III. HIGH–T_c/$SrTiO_3$ EXPERIMENT

A. Meissner state ($H < H_{c1}$)

The high–T_c films of $YBa_2Cu_3O_7$ were epitaxially grown on $SrTiO_3$ single crystal with a CeO_2 seed–layer. The c–axis was perpendicular to the film surface. The quality of the samples was tested in–situ and ex–situ by various methods (RHEED, AFM, STM and X–ray diffraction). The films were c–oriented with a twinned structure of a–b domains of 100–200 nm in size with a small mosaicity. The transition temperature of the high–T_c film was 87 K.

The neutron reflectometry experiments were performed on D17 (ILL, Grenoble) [13] and on the Spectrometer of Polarised Neutrons (SPN, JINR, Dubna) [12]. D17 has a monochromatic beam and SPN is a time–of flight reflectometer. In both cases cryomagnets were used. The cryostat on SPN was specially designed for reflectometry experiments with two couples of sapphire windows for a laser alignment of the samples.

First the experiments were made on D17 from the substrate side. For the sample characterization the reflectivity was measured at room temperature and the structure of the film (thicknesses, density and interface roughnesses) was obtained after a fit to the data (fig. 2a, curve 1). A surface layer consisting presumably of $BaCO_3$ of 25 Å in thickness was found on the film surface. This layer is formed due to the humidity and a contact with atmosphere. The defects and the quality of the films were probed by off–specular scattering. We did not detect any noticeable off–specular scattering in contrast to the rather huge off–specular scattering measured from the film in Ref. 5. The SLD profile is shown in fig. 2 b.

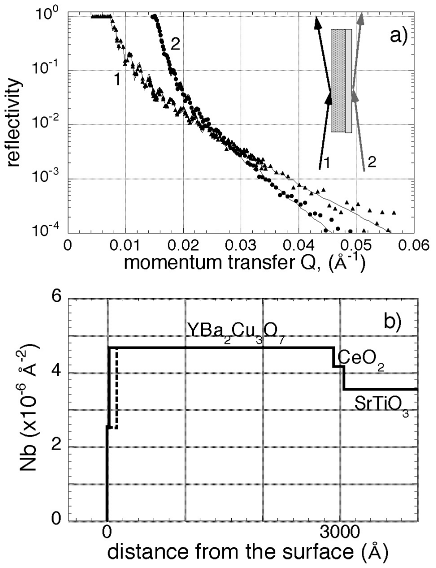

Fig. 2. a) reflectivity curves of the $YBa_2Cu_3O_7$ thin film on $SrTiO_3$ substrate measured at room temperature. Black triangles (curve 1) correspond to the reflection from the substrate side, circles (curve 2) show the reflection from the vacuum side, solid lines correspond to the fit to the data. The scattering geometries are shown in the sketch on the right side. b) the scattering length density profile deduced from the fit to the data.

Then the sample was cooled down to T=2 K , an external magnetic field of 300 Oe was applied parallel to the film surface and the spin dependent reflectivities R^+ and R^- were measured from the substrate–film side. The fit to the experimental R^+ and R^- reflectivities was made simultaneously with the only parameter of the fit — the magnetic part of the SLD, all other parameters (thicknesses, roughness, density) were taken from the room temperature experiment and fixed. The experimental SA is shown in fig. 3a together with a fit to the data. The value of (1350 ± 150) Å was obtained for the penetration depth Eq (2) with an intrinsic exponential profile. Other line– shapes were not yet tested and would require better statistics.

The next experiment was made after half a year on the SPN with the reflection from the vacuum — film side (see fig. 2a, curve 2). The results of the room temperature experiment showed that the thickness of the $BaCO_3$ surface layer increased from 25 to 125 Å. The SLD profile resulting from the fit to the data are depicted by the dashed line in fig. 2 b. The low — temperature experiment was made at T= 4.8 K and magnetic field of 500 Oe. Again for the fit only the magnetic part of the SLD profile was variable and the nuclear scattering part was taken from the fit to the room–temperature data (see fig. 3 b). The value of the penetration depth λ_L=(1400±100) A was obtained for the reflection from the air — film side. The complete result for SLD profile for both experiments is depicted in fig. 3 c. In the case of high–T_c films the penetration depth is measured because the correlation length is very short and thus electron scattering at defects should be negligible.

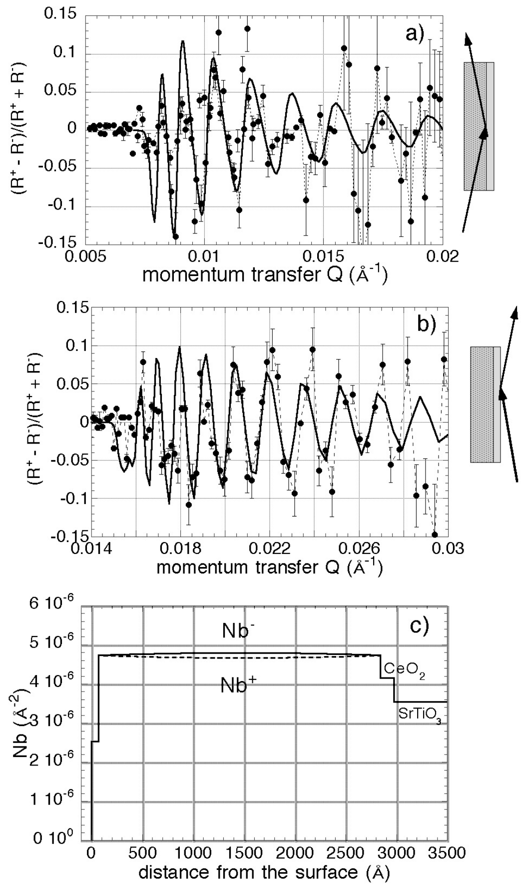

Fig. 3. a) reflection from substrate side at T = 2 K and external magnetic field of 300 Oe; b) reflection from vacuum side at T= 4.8 K and magnetic field of 500 Oe. The deduced SLD profile is shown in figure 3 c.

B. Mixed state ($H_{c1} < H < H_{c2}$)

With the application of higher values of the external magnetic field parallel to the surface of the superconducting film magnetic flux penetrates inside the film. This effect will cause a change in the scattering potential profile and, thus, changes the R^+ and R^- reflectivities.

A quantized flux–line can be described with a simple model of a core of a normal material with the radius of the order of the coherence length ξ surrounded by a region with decaying magnitude of the magnetic field of the size of the penetration depth λ_L. Taking into account that magnetic flux penetrates also from both interfaces one can expect that in case of a thin superconducting film of a finite thickness d the lower critical field H_{c1} will be always larger than in the bulk material. It means that if the sample is thinner than or about equal to the penetration length, the field inside the film is only slightly lower than the external field. In this case the energy cost of this partial field expulsion is smaller then the vortex nucleation. The vortex nucleation becomes favorable only for higher value of the applied external field. This was calculated by Abrikosov [16, 17] and can be expressed by the following formula:

$$H_{c1}^{d} = H_{c1}^{\infty} \frac{1 + \frac{2}{K_0(1/k)} \sum_{n=1}^{\infty} (-1)^n K_0(nd/\lambda)}{1 - \cosh^{-1}(d/2\lambda)}, \qquad (3)$$

where H_{c1}^{∞} is the lower critical field of bulk material, $k = \lambda/\xi$, and K_0 is the Hankel function of zero order and imaginary argument. This equation was extended by Kogan for the case of an anisotropic superconductor [18]. The result is similar to Eq. (3) with appropriate anisotropic parameters $H_{c1//ab}$, λ_{ab}, k_{ab}.

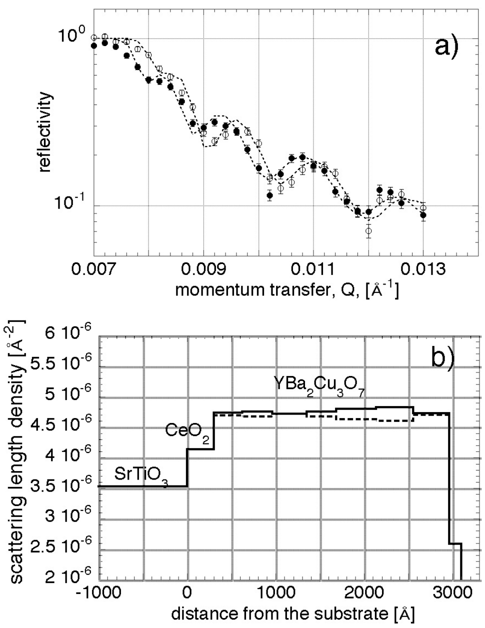

Fig. 4. a) Spin–dependent R^+ and R^- reflectivities as functions of momentum transfer measured from $YBa_2Cu_3O_7$ thin film at the temperature $T = 2$ K in the external magnetic field of 1.5 kOe. Dashed lines correspond to the fit to the data. The scattering length density profile is shown in b). The absence of the splitting between SLD for the spin–up and spin–down neutrons in the central part of the film indicates the penetration of the magnetic flux into the film.

Fig. 4 a shows the results of our experiment in the external magnetic field of 1.5 kOe together with the fit to the data. For the fit we used a model in which the film was divided into 7 sublayers with fixed nuclear part of the SLD. Thicknesses of each sublayer i and magnetic part of SLD Nb_m^i were parameters of fit. For the starting parameters no assumption was made about the distribution of the magnetic flux inside the film, so that the initial values were set to zero: Nb_m^i=0. The resulting SLD profile is presented in fig. 4 b. In contrast to the Meissner state, the absence of the splitting between SLD for the spin–up and spin–down neutrons in the central part of the film indicates the penetration of the magnetic flux into the film.

IV. CONCLUSIONS

A study of the magnetic field penetration depth in a high temperature (high–T_c) superconducting film was performed with polarised neutron reflectometry in two scattering geometries. The fit to the reflectivity curves yielded the neutron scattering length density profile and thus a precise image of the composition of the film. The film had a good quality seen by an unnoticeable off–specular scattering. The fit to the spin–asymmetry gave a magnetic–penetration depth of 1350±150 A along the c–axis at the temperature of T=2 K. The model included an intrinsic exponential decay of the penetration depth. For the first time the spin–asymmetry was determined with high resolution over an extended Q–range for a high–T_c film.

The results presented in this article show the feasibility of measuring the magnetic–flux penetration into the high–temperature superconducting films in the mixed state using the technique of Polarized Neutron Reflectometry.

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