MEASUREMENTS OF THE MEISSNER-OCHSENFELD EFFECT OF HIGH-TEMPERATURE SUPERCONDUCTORS BY MEANS OF FREE-RADICAL EPR

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Abstract. – A method for the measurement of the Meissner-Ochsenfeld and induced fields of superconductors by means of EPR of solid free radicals is presented and applied for the study of fields of Y-Ba-Cu-O high-temperature superconductors.

1. Introduction

The use of the electron paramagnetic resonance (EPR) in materials with narrow EPR linewidth enables static magnetic field intensity (H) measurements with high accuracy (about 10^{-6} for $H > 0.5$ kG) in small volumes (down to 10^{-8} cm^3) [1]. For that purpose, mostly samples of solid-state free radicals (FR) are applied (e.g. DPPH with an EPR linewidth $\Delta H = 1$ G [2]). Another, secondary application of this method is the determination of certain magnetic parameters of magnetic materials by FR EPR measurements of local field intensities at the surfaces (or in the vicinity) of samples [3]; it has been used for the investigation of demagnetizing fields and magnetization (e.g. [3-7]), of stray fields at ferromagnetic surfaces (e.g. [3, 8]), etc. Recently, we have applied the FR EPR method for the study of superconductors (SC) [9]. In this contribution we wish to report on the local measurements of the Meissner-Ochsenfeld (MO) and induced (IN) fields at the surface, and in the presurface layer of a ceramic YBaCuO material, at external field intensity of about 13 kG, at temperatures 4 to 300 K.

2. Experimental

In our work the MO field is defined as a field intensity caused by the (partial) expulsion of the magnetic induction out of the SC sample (i.e. by the Meissner-Ochsenfeld effect), the specimen being kept at a constant value of the bias external magnetic field. By the IN field we understand a more general case of the field intensity (caused by the SC sample magnetization) for varying bias fields.

The MO (or IN) field intensity of a superconductor (SC) is measured as a difference between the local field values measured in the presence, and without the SC sample, respectively. The stability and the accuracy of measurements must meet high standards [10]. The results reported here were obtained by an Q-band (around 36 GHz) equipment. The magnetic field (an 15" electromagnet) was electronically stabilized (short-time stability of the order of 10^{-7}) and measured with NMR (relative accuracy of 10^{-7}). The microwave frequency stability was of the same order of magnitude, the absolute accuracy of frequency measurements ammounted to 10^{-9}. The samples were mounted in a continuous-flow helium cryostat, the temperature of which was electronically stabilized and measured within 0.1 K [4].

The SC material (chemical composition $YBa_2Cu_3O_{6.65}$, simple orthorhombic structure, porosity about 15 % , critical temperature T_c = 95 K) was prepared by a two-step heating in oxygen [9]. Selected single crystals of the solid state free radical DPPH [2, 10] (grown from the H_2S solution) in the form of platelets (with dimensions of about 0.03 × 0.2 × 0.4 mm) were used as FR EPR probes. Because their g-factor is anisotropic, the frequency/field ratio of the probes and its temperature dependence must be calibrated in the same geometry as it is used in the MO (or IN) field measurements. The EPR inflection linewidth of these probes ammounts to 0.9 (0.5) G at 300 K and at 36 GHz for fields laying in (pointing normally to) the plane of the platelets, respectively; at 25 K ΔH values begin to broaden with decreasing temperature up to 2.8 (2.5) G at 4 K. In some cases (see Sect. 3) we let to suck a DPPH solution in benzene into the SC samples in order to introduce small DPPH crystals into the microscopic cavities of the porous SC material.

3. Results and discussion

First let us report on the measurements of the MO and IN fields, which were performed on a sample in the shape of rectangular parallelepiped (dimensions 0.7 × 0.9 × 1/4 mm) magnetized along the long axis. One DPPH platelet (with its plane parallel to the field direction) was localized in the middle of the larger parallel side of the SC sample (parallel configuration of the experiment); the other one (its plane is normal to the field direction) was situated in the middle of the SC surface oriented normally to field (normal configuration).

The MO fields were investigated in an external magnetic field of about 12.7 kG and plotted as open circles in figure 1. The $H(T)$ values do not depend on the course of the temperature (cooling or warming). The absolute values of MO fields are about two orders of magnitude smaller than values for an ideal type of superconductor. Because the external bias field is much larger than the first critical field (H_{c1}) of our SC material ($H_{c1} < 0.5$ kG) the incomplete exclusion of magnetic induction leads to small MO fields. The IN fields were investigated in two ways: The sample was cooled to 4 K in the external field of 5 G (22 kG), then the field was adjusted to the FR EPR value and the $H(T)$ measurements were performed; the results are plotted in figure 1 as crosses (full circles) for the low (high) field cooling, respectively.

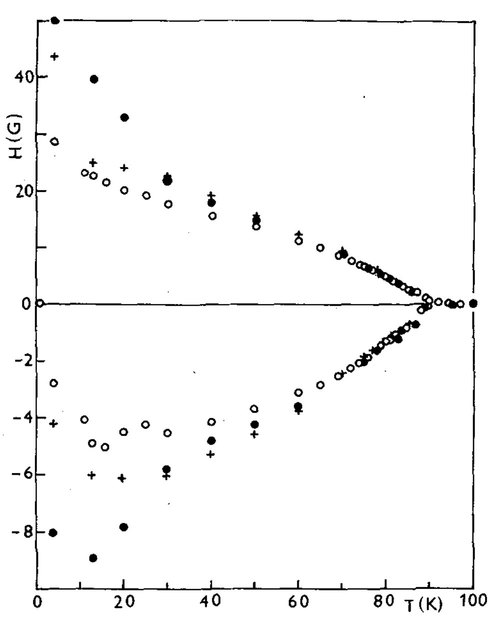

Fig. 1. – The temperature dependence of MO(o) and IN (•, +) fields (H), measured at a bias field intensity of 12.7 kG on a SC sample of a parallepiped shape. The upper (lower) part of the figure, i.e. positive (negative) H values, represents data for the DPPH probe situated at the SC sample surface oriented normally (parallel) to the bias field direction, respectively; for details see text.

In both parallel and perpendicular configurations the signs of the MO and IN fields correspond to the diamagnetic nature of the superconductor, the difference of the field values between the parallel and normal configuration are caused mainly by local demagnetizing effects. For each temperature between 72 K and the transition temperature (T_c) the MO and IN values are equal (and linearly dependent on $(T_c - T)$) within the accuracy of the measurement (about 0.1 K, 0.1 G). It is worth mentioning that the absolute values of MO fields are in all cases smaller than the appropriate values of IN fields at T smaller than 72 K. The general increase of MO and IN fields towards the lower temperatures can be explained by the decreasing penetration of the magnetic field (larger than H_{c1}) into the non-ideal SC sample. For a quantitative description of field dependences one should consider both the macroscopic (sample demagnetization effects) and the microscopic (influence of the structure of the ceramic material, of paramagnetic impurities, e.t.c.) nonhomogeneities of the internal field. Reliable experiments should be performed on single crystals of SC.

In another type of experiment we have investigated the behaviour of MO fields in a presurface layer of a SC sample by means of the "suck-in" DPPH solution. The results [9] point to a large penetration depth (~ 0.1 mm) into the SC sample and to large inhomogeneities of the internal MO fields near the surface of the SC.

Previous experiments show that the FR EPR method is useful for obtaining additional information about the magnetization in SC materials. The use of the common EPR techniques (at frequencies above 8 GHz) enables studies of SC materials mostly in fields above H_{c1}. However, for physically more interesting case, i.e. low fields, one can make use of the FR EPR method (mainly of its local resolution) by applying the low-field EPR technique [11], e.g. by using the highly developed nuclear resonance detection techniques in a broad frequency interval.

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