Microwave Electrodynamics of low T_C and high T_C Systems with Coexisting Superconductivity and Magnetism

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We investigate the microwave electrodynamics of both artificial low T_C superconducting / magnetic (S/M) layered structures and high T_C cuprates. In particular we focus on Nb/CuMn (superconducting / spin-glass) bilayers, and on GdBa_2Cu_3O_{7−δ} (GBCO) thin films, which show coexistence of superconductivity and long range ordered antiferromagnetism below T_N=2.2 K. In both cases we are interested in the influence of magnetism on superconductivity. Moreover, in the GBCO case, we want to shed light on the problem of the determination of the pairing symmetry in the cuprates. First we show surface impedance data at 10 GHz on Nb/CuMn bilayers. We extract information about the induced order parameter in the magnetic layer and we compare it with the exotic behavior predicted for S/M proximity systems. Then we present microwave surface impedance data at three different frequencies on GBCO c-axis oriented epitaxial thin films. Both the resistance and the reactance data on GBCO show an unusual low temperature behavior, mainly due to change in magnetic permeability. This result indicates that the paramagnetism of the rare-earth ions has to be taken into account when extracting the superconducting penetration depth as a function of temperature, and thus determining the pairing state symmetry of the cuprates.

1. Introduction

The temperature dependence of the microwave surface impedance is a powerful tool in the investigation of microscopic properties of superconductors. It gives valuable information about the inhomogeneous superconducting properties of layered systems, both natural and artificial, and it is used to study the pairing symmetry and other properties of the cuprates.

We investigate the microwave electrodynamics of both artificial low T_C superconducting / magnetic (S/M) layered structures and high T_C cuprates, with the basic idea of using artificial layered conventional superconductors as model systems

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for the cuprates. In particular we focus on Nb/CuMn (superconducting/spin-glass) bilayers, and on GdBa2Cu3O7−δ (GBCO) thin films, where the Gd3+ ions carry magnetic moments which align parallel to the c-axis and order antiferromagnetically below $T_N \simeq 2.2$ K in the three crystallographic directions. In both cases we are interested in the influence of magnetism on superconductivity.

Exotic phenomena are predicted for superconducting/ferromagnetic (S/M) coupled layered structures, such as critical temperature oscillations versus the M layer thickness and spontaneous persistent currents in rings interrupted by an S/M/S junction. All of these properties depend on the presence of a spatially dependent phase for the order parameter in the M layer that, for suitable thicknesses, gives rise to a π shift between adjacent superconducting layers. From the experimental point of view, observations of $T_c$ oscillations, seen as an indirect proof of the π phase theories, have been reported. More insights into this problem may result from microwave surface impedance measurements on Nb/CuMn bilayers.

On another matter, the intriguing coexistence of long-range ordered antiferromagnetism and superconductivity appears in rare-earth element (RE) based compounds, such as (RE)Rh$_4$B$_4$ and (RE)Mn$_6$Sb$_6$, (RE)Ba$_2$Cu$_3$O$_7$ cuprates, and quaternary borocarbides (RE)Ni$_2$B$_2$C. In general the magnetic behavior is associated with localized moments of the RE ions, and superconductivity exists in a separate electronic subsystem in the material. The interplay of superconductivity (SC) and antiferromagnetism (AF) in these systems can be quite complicated, and varies dramatically upon doping with different RE elements. Another important issue is the effect of paramagnetism of the AF subsystem (above the Neel temperature $T_N$) on the determination of the magnetic penetration depth temperature dependence in cuprate superconductors, such as the electron-doped Nd$_{2−x}$Ce$_x$CuO$_4$. It has been claimed that paramagnetism of the Nd ions above $T_N = 1.2$ K masks the true behavior of the screening length in this system, leading to an incorrect conclusion about the pairing state symmetry of the electron-doped cuprates. To address these issues we have focused on the electrodynamic properties of GBCO.

We will discuss the measurements and results in the following section.

2. Measurements and results

We measure the effective (due to the finite film thickness) surface impedance

$$Z_{Seff}(T, \omega) = R_{Seff} + iX_{Seff} = \sqrt{i \omega \mu(T, \omega) / \sigma(T)} \coth \left[ t \sqrt{i \omega \mu(T, \omega) / \sigma(T)} \right],$$

where $\mu = \mu_0 \mu_r$ and $\sigma$ are respectively the complex magnetic permeability and complex conductivity and $t$ is the film thickness. The first factor on the right hand side is the bulk surface impedance $Z_S$, where $\mu = \mu_0$ for non magnetic superconductors, and the second factor is the finite thickness correction. In the Nb/CuMn bilayer case $\mu = \mu_0$, and the measured surface impedance (or the penetration depth) and the surface resistance are some overall quantities which do not correspond to the individual impedance of the constituents, because of the non-uniform nature of the superconducting proximity-coupled systems.

We perform the measurements with the parallel plate resonator (PPR) technique
with the rf field in the film plane (the ab plane in the GBCO case) down to 1.4 K. The PPR resonance frequency \( f(T) \) and quality factor \( Q(T) \) data are converted to changes in the effective surface reactance and the effective surface resistance and to complex conductivity.

2.1. Nb/CuMn bilayers

We have focused on Nb/CuMn bilayers, deposited by dc sputtering, with a Mn concentration of 2.7%. We have analyzed seven samples characterized by the same Nb layer thickness (\( d_S = 1500 \) Å) and different CuMn layer thicknesses (\( d_M = 30, 60, 90, 120, 150, 180, 240 \) Å), where the Nb is the first layer on the substrate. After the surface impedance measurements, the top CuMn layer of one pair of samples was removed by using a dilute HNO\(_3\) solution, in order to characterize the underlying Nb layer.

Surface impedance measurements have been performed on the Nb/CuMn bilayers and on the underlying Nb film at 10 GHz from 1.7 K to \( T_C \).

Figure 1 shows the change in the effective penetration depth \( \Delta \lambda_{eff}(T) = \Delta X_{Seff}/\mu_0\omega \) for the Nb film and the Nb/CuMn (S/M) bilayers, compared to previous results on Nb/Cu (S/N) bilayers. Here we are not removing the geometric correction due to the finite thickness of the sample. The shape of the \( \Delta \lambda_{eff}(T) \) curves for the S/M bilayers is not very different from the temperature dependence for the Nb, but shows only an enhancement, while a strong linear-in-temperature character was evident in \( \Delta \lambda_{eff}(T) \) for Nb/Cu. Moreover there is no systematic
dependence of $\Delta \lambda_{\text{eff}}(T)$ on the CuMn layer thickness, in striking contrast to the case of Nb/Cu bilayers, where a strong dependence of $\Delta \lambda_{\text{eff}}(T)$ on the normal layer thickness has been observed.

By developing proximity effect models suitable for the S/M case,\textsuperscript{15,16} we find a temperature independent proximity effect correlation length (as expected on the basis of the Radovic et al. picture predicting the $\pi$-phase shift for S/M multilayers,$^3$) and we learn that the screening is dominated by the S layer with a strongly suppressed order parameter near the interface. These results confirm that the CuMn layer is acting as a ferromagnet in the screening process.

Similar conclusions can be extracted from the surface resistance data, which again are in contrast to the Nb/Cu case in the same manner.

2.2. \textit{GBCO thin films}

The samples we have investigated are pairs of identical c-axis oriented GBCO epitaxial films, laser ablated on (100)-cut LaAlO$_3$ single crystal substrates. The film thickness is 300 nm, the superconducting critical temperature measured by AC susceptibility is 92.5 K and the transition width is 0.3 K.

We measured the effective surface impedance of the GBCO thin films from 1.4 K to $T_c$ at three different resonance frequencies: 10.4, 14.7, 17.9 GHz. The changes in surface reactance and surface resistance have been converted to absolute values using $X_{\text{Seff}}(77\text{K})=49 \text{ m}\Omega$ and $R_{\text{Seff}}(77\text{K})=0.48 \text{ m}\Omega$ measured at 10 GHz by the variable spacing parallel plate resonator technique.\textsuperscript{17}

In figure 2 we show $R_{\text{Seff}}(T)$ and $X_{\text{Seff}}(T)$ at 10.4 GHz over the entire measurement temperature range. The high temperature behavior is consistent with a $d$-wave temperature dependence for the surface impedance.\textsuperscript{17} The deviations from this behavior start below 30 K, where the magnetic effects due to $\mu(T)$ come into play.\textsuperscript{18} Both $R_{\text{Seff}}(T)$ and $X_{\text{Seff}}(T)$ show a minimum at two different temperatures, $T \simeq 25 \text{ K}$ and $\simeq 7 \text{ K}$, respectively. Then $R_{\text{Seff}}(T)$ and $X_{\text{Seff}}(T)$ increase upon reducing the temperature, and a strong peak is observed in $R_{\text{Seff}}(T)$.

![Graph](image)

Fig. 2. Effective surface resistance $R_{\text{Seff}}(T)$ and reactance $X_{\text{Seff}}(T)$ of GBCO at 10.4 GHz.
Fig. 3. Real part of the modified conductivity $\sigma_{1m}$ at different frequencies (a), and rescaled imaginary part $\lambda_m^{-2}$ (same symbols as for the real part) (b).

The same behavior is found at the other two frequencies with some extra frequency dependence other than the trivial $X_S \sim \omega$ and $R_S \sim \omega^2$ observed for superconductors. This is clearly seen in figure 3, where we show the data at the three frequencies as modified complex conductivity $\sigma_m(T, \omega) = \sigma(T, \omega)/\mu_r(T, \omega)$, defined through $Z_S(T, \omega) = \sqrt{i\omega\mu_0/\sigma_m(T, \omega)}$. The real part, $\sigma_{1m} = 2R_S\omega\mu_0/X_S^2$, presents frequency dependent peaks around $T_N$ (fig. 3(a)). In figure 3(b) we show a rescaled imaginary part $\lambda_m^{-2} = \sigma_{2m}\omega\mu_0 = (\omega\mu_0/X_S)^2$, where the frequency dependence is less pronounced. This behavior, mainly due to change in magnetic permeability, indicates that the paramagnetism of the rare-earth ions has to be taken into account when extracting the superconducting penetration depth as a function of temperature, and thus determining the pairing state symmetry.

3. Conclusions

In conclusion, microwave surface impedance measurements have been performed on Nb/CuMn (superconducting/spin-glass) proximity coupled bilayers and on GBCO thin films. The presence of magnetic contributions in these systems gives rise to unusual surface impedance and conductivity behaviors at microwave frequencies.

In the Nb/CuMn case both $\Delta\lambda_{eff}(T)$ and $R_S(T)$ are very different from the Nb/Cu case, showing that the superconducting properties of the Nb layer are strongly suppressed near the interface and the CuMn layer does not participate too much in the screening of the applied rf magnetic field, as expected for a proximity effect between a superconductor and a ferromagnet. Moreover, these measurements can be described with a proximity effect correlation length which does not depend on temperature, consistent with previous data on $T_c$ oscillations vs. CuMn layer thickness, and the Radovic et al. theoretical picture predicting a $\pi$-phase shift in S/M systems.

In the GBCO case strong unusual features are observed in the temperature dependence of surface impedance and conductivity. The effects of paramagnetism and antiferromagnetism are shown to have a significant influence on $\lambda(T)$ and $R_S(T)$, that may lead to incorrect interpretation on the pairing state symmetry of the
cuprates. This conclusion indirectly supports some recent results showing predominant d-wave symmetry also in the electron-doped cuprates.¹⁹

Acknowledgements

The authors want to acknowledge J. Claassen, M. Coffey, P. Fournier H. Harshevarden, M. Pambianchi, A. Pique, A. Porch and A. Schwartz.

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