Microwave Absorption Peaks: Signatures of Spin Dynamics in Cuprates

S. Sridhar and Z. Zhai

Physics Department, Northeastern University, 360 Huntington Avenue, Boston, MA 02115

A. Erb

DPMC, Université de Genève, CH-1211 Genève 4, Switzerland

Abstract. We show that a common feature of temperature-dependent microwave absorption is the presence of absorption peaks. $ac$ loss peaks can arise when the internal $T$-dependent magnetic relaxation time crosses the measurement frequency. These features are observed in the insulating ($Sr_{x}Ca_{14-x}Cu_{24}O_{41}$, $La_{5/3}Sr_{1/3}NiO_{4}$ and $YBa_{2}Cu_{3}O_{6+δ}$), pseudo-gap ($T > T_{c}$ in underdoped $YBa_{2}Cu_{3}O_{7−δ}$, $Hg : 1223$ and $Hg : 1201$) and superconducting ($T < T_{c}$) states of the oxides. The commonality of these features suggests a magnetic (spin) mechanism, rather than a quasiparticle origin, for the so-called “conductivity” peaks observed in the cuprate superconductors.

During the last few years, there have been extensive and careful experimental [1,2] as well as theoretical [3] studies of the microwave properties of the cuprate superconductors. In parallel, several experiments have been shown to be consistent with a $d$-wave order parameter (OP) [4]. The linear behavior of the penetration depth $\lambda(T)$ at low temperature $T$ is frequently cited as evidence of $d$-wave symmetry. However a consensus is emerging that the totality of the microwave data is not explainable in terms of a pure $d$-wave gap - quasiparticle scenario, and that the (dynamic) microwave response may be decoupled from the (static) OP symmetry. The principal issues are summarized below:

1. $YBCO$ and $Hg : 1223$ are definitely not pure $d$-wave superconductors as deduced from the microwave measurements. This is signaled by the presence of multiple “conductivity” peaks, as shown in Fig. 1(c). Within a conventional gap-quasiparticle scenario, the data are consistent with mixed symmetry, e.g. $d + s$. While mixed symmetry is allowed in orthorhombic $YBa_{2}Cu_{3}O_{7−δ}$, it is not allowed in tetragonal $Hg : 1223$. This suggests that the OP symmetry may be decoupled from the crystal symmetry.

2. The measured microwave absorption is significantly higher than estimates...
based upon $d$-wave calculations, using acceptable estimates of the scattering times. The discrepancies are very large (orders of magnitude) for all superconductors such as $Bi:2212$, with the possible exception of $YBa_2Cu_3O_{7-\delta}$.

3. **Anomalous state above $T_c$:** In materials such as $Hg:1223$, $Hg:1201$ and underdoped $YBa_2Cu_3O_{7-\delta}$ we find that the surface resistance $R_s$ is not equal to the surface reactance $X_s$ ($R_s \neq X_s$). This indicates that the pseudogap state above $T_c$ is not a normal metal with ordinary Ohmic conductivity, and may signify the importance of magnetic contributions to the microwave impedance. Thus the transition to the superconducting state takes place from an unconventional state, and that suggests that sum rules may not hold [5].

4. The measured nonlinear response (which is a major limitation of the use of the cuprate superconductors in microwave applications) is significantly higher than estimates based upon $d$-wave calculations [6].

5. **Internal Josephson effect:** A unique feature of the cuprate superconductors is the strong microwave response at very low field levels well below $H_{c1}$. In ultraclean $YBa_2Cu_3O_{6.95}$ samples, a nonlinear response which can be described as a single Josephson junction in the $ab$-plane, is observed [7]. This can arise from

![FIGURE 1](image-url)
a bulk Josephson effect between two superconducting components. Similar behavior was also seen in Bi: 2212 single crystals [8].

6. Magnetic Recovery Effect: A remarkable effect is the decrease of microwave absorption for small applied dc fields [9]. A compelling interpretation of this effect is that small fields reduce magnetic scattering [10]. It is interesting to note that reduction of magnetic absorption by moment-orienting fields is well-known in magnetic systems [11].

7. 2nd Harmonic generation [12]: Frequently 2nd harmonics are observed in many microwave harmonic generation experiments. This is inconsistent with a time-reversal invariant state such as a pure d-wave OP, and must originate from extrinsic sources. A magnetic origin may well be possible.

To obtain a broad perspective on the microwave response of the cuprates, we have studied single crystal samples of a variety of cuprates, from superconductors such as Y: 123, Hg: 1223, Hg: 1201, insulating or weakly doped members such as YBCO$_6$, and insulating PrBaCuO, and the spin-ladder / chain compounds (Sr, Ca) – Cu – O. The measurements are carried out using very high Q superconducting cavities [13] that enable precision measurements of the microwave susceptibility and impedance.

A common feature that emerges from this wide data set of measurements is the presence of peaks in the temperature dependent microwave absorption, accompanied by transition-like changes in the dispersion. The microwave loss is thus frequently non-monotonic and this behavior is not restricted to YBa$_2$Cu$_3$O$_{7-\delta}$. Remarkably such non-monotonic temperature dependent absorption is observed in insulating (Sr$_x$Ca$_{14-x}$Cu$_{24}$O$_{41}$ and YBa$_2$Cu$_3$O$_{6.0}$), pseudo-gap (above $T_c$ in under-doped YBa$_2$Cu$_3$O$_{7-\delta}$, Hg: 1223 and Hg: 1201) and superconducting (below $T_c$) states of the cuprates (see Fig. 1(c)), and is a signature of spin dynamics in the microwave response.

The presence of non-monotonic temperature-dependence of the microwave absorption, leading to peaks, presents an important clue, since it is not observed in any other type of superconductor. Instead, as discussed later in this paper, peaks in the microwave absorption are characteristic of magnetic dynamics. This raises the possibility that the microwave absorption in the cuprate superconductors is not due to quasiparticle dynamics, and instead is indicative of underlying spin dynamics. Hence the microwave response may be de-coupled from the underlying pairing symmetry of the superconducting OP.

Sr$_{14}$Cu$_{24}$O$_{41}$

To illustrate the underlying physics, we discuss our recent microwave measurements on this spin chain/ladder material [14]. The data shows a microwave absorption peak as shown in Fig. 1(a). This is accompanied by a drop in the microwave dispersion (not shown). The static magnetization does not show these changes, and hence this is a purely dynamic property.
The data in Fig. 1(a) represents a microwave loss peak and can be described as spin freezing at microwave frequencies [14]. The occurrence of loss peaks can be understood by considering a complex susceptibility $\chi = \chi_o/(1 + i\omega\tau) \equiv \chi' + i\chi''$. When the relaxation rate $\tau^{-1}(T)$ varies rapidly with $T$ and crosses the measurement frequency $\omega$, a peak occurs when $\omega\tau = 1$, this is shown in Fig. 2. When $\tau(T)$ increases with decreasing temperature $T$ (Fig. 2 (a)), then $\chi'$ shows a drop with decreasing $T$, as shown in the Fig. 2 (b), and $\chi''$ shows a peak (Fig. 2 (c)). The experimental data for both $\chi'$ and $\chi''$ are in very good agreement with the middle and bottom panels of Fig. 2, as we have shown in ref. 21. Note that the changes in absorption (i.e. $\chi''$) and dispersion ($\chi'$) can easily be mistaken for a superconducting transition without further information from static measurements.

The dominant relaxation mechanism in this material is due to spin-spin relaxation at these temperatures. Relaxation due to mobile holes is rapidly suppressed because of the charge ordering at around $250K - 300K$ seen in synchrotron X-ray scattering [15].

Our experimental data on this material thus provides clear and unambiguous evidence for microwave loss peaks due to spin dynamics in the cuprates, and also shows that spin relaxation occurs at GHz frequencies in these materials.

$\text{YBA}_2\text{Cu}_3\text{O}_{6.0}$

Our measurements on the parent compound $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ further underscores the importance of spin dynamics in microwave measurements of the cuprates. Results for $\chi''$ of $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$ measured at $10GHz$ are shown in Fig. 1(b). The loss term $\chi''$ shows a pronounced peak at around $14K$ with an onset at $50K$ which is accompanied by a corresponding feature in $\delta\chi'(T)$ (not shown). Overall there are close similarities to the results of this material as well as other insulating cuprates such as the ($\text{Sr,Ca}) - \text{Cu} - \text{O}$ family and also $\text{PrBaCuO}$.

A striking feature of the data in Fig. 1 (b) is the close similarity to zero-field $\mu\text{SR}$ $1/T_1$ data shown in Fig. 2 of Niedermayer, et al. [16] on $\text{YBa}_2\text{Cu}_3\text{O}_{6.0}$, in which also a peak is seen around $22K$. The close correspondence provides clear evidence that the $10GHz$ measurements are studying the same spin dynamics seen
in the $\mu SR$ experiment although at a different (shorter) time scale.

Possible magnetic origin to absorption peaks in superconducting cuprates

In the conventional quasiparticle conductivity scenario, the peak is understood from $\sigma_1 = n_{qp}(T)e^2\tau(T)/m$ as a competition between increasing $\tau(T)$ and decreasing $n_{qp}(T)$ with decreasing $T$. In a (non-quasiparticle) magnetic scenario, the peaks in surface resistance $R_s$ should be regarded as microwave absorption or loss peaks, rather than conductivity peaks. The peaks then occur due to the crossing of the magnetic relaxation time $\tau$ with the measurement frequency $\omega$ as temperature $T$ is varied. In a magnetic scenario, the peak is similar to loss peaks observed generally in $ac$ driven relaxation systems, in which for $T > T_p$ ($T_p$ is the temperature at the absorption peak) the system relaxes in phase with the $ac$ drive, while for $T < T_p$ the system cannot follow the drive and the absorption decreases.

Note that the absorption peak, which represents the out-of-phase response, is necessarily accompanied by a decrease of the (in-phase) susceptibility [17]. This is indeed what is observed even in the superconducting state. The peaks $A$, $B$ and $C$ are all accompanied by apparent decreases in the “penetration depth” or the reactive response.

A particularly clear example of the consequences of magnetism is our measurements on the anti-ferromagnetic superconductor $DyNi_2B_2C$, where $T_N = 10.5K$ is greater than $T_c = 6K$ [18]. In the metallic AFM state above $T_c$, we found that $R_s \neq X_s$. Both in the superconducting and AFM states, the data were analyzed in terms of a dynamic magnetic permeability contribution $\mu(\omega, T)$ to the surface impedance since $Z_s = [i\mu(\omega, T)\omega/\sigma]^{1/2}$. However the high frequency measurements do not distinguish between dynamic permeability and a dynamic conductivity contribution such as arising from a narrow Drude peak. In this material, the magnetic contribution is clearly identifiable as arising from the 3-D AFM ordered state, whose influence extends into the superconducting state also, perhaps in the form of strong pairbreaking.

Although there are no 3-D ordered magnetic states in the cuprates, there are lower dimensional structures such as stripes, arising from inhomogeneous hole doping of the parent AFM insulator. Recently, the presence of stripes in the cuprates [19] and nickelates [20] has been established experimentally and theoretically [21,22].

The presence of stripes provides a plausible mechanism for spin contributions to the microwave response in the metallic pseudo-gap and superconducting states. The intrinsic inhomogeneity of the striped state leads naturally to unconventional (non-quasiparticle) transport [23]. Since the region between the charge stripes is a disordered anti-ferromagnet, spin contributions to the microwave response could necessarily arise which are also similar to that in the pure spin chain/ladder compounds.

We have recently shown in the nickelate $La_{5/3}Sr_{1/3}NiO_4$ that stripe formation does lead to a microwave absorption peak [24]. We have also confirmed the glassy dynamics of stripes in this material. Thus we have shown that stripe formation can
lead to non-monotonic microwave absorption and hence stripes can potentially be the magnetic structures responsible for the absorption peaks seen in the cuprates. There is a close similarity of the data of $La_{5/3}Sr_{1/3}NiO_4$ and the $Sr – Cu – O$ compounds discussed earlier, which shows that the microwave absorption peaks are typically observed below a charge ordering transition. This may well be happening in the cuprate superconductors also.

In summary, an extensive analysis of microwave data in the cuprate and related compounds suggests that the presence of microwave absorption peaks is a common signature of spin dynamics. The presence of these peaks in the superconducting state strongly suggests that conventional quasiparticle dynamics is not operative, but that these may be overwhelmed by strong contributions from spin dynamics, possibly due to the presence of stripes.

Discussions with K. Scharnberg are gratefully acknowledged. This research was supported by NSF-9711910 and AFOSR-5710000349.

REFERENCES

1. H. Srikanth, et al., Phys. Rev. B 55, R14 733 (1997).
2. H. Srikanth, et al., Phys. Rev. B 57, 7986 (1998).
3. S. Hensen, G. Müller, C. T. Rieck and K. Scharnberg, Phys. Rev. B 56, 6237 (1997).
4. D.J. van Harlingen, Rev. Mod. Phys., 67, 515 (1995).
5. Z. Zhai, S. Sridhar, et. al, (to be published).
6. T. Dahm, et al., J. of App. Phys. 84, 5662 (1998).
7. Z. Zhai, et. al., Physica C, 282-287, 1601 (1997).
8. T. Jacobs, et al., Rev. Sci. Instr. 67, 3757 (1996).
9. D. P. Choudhury, et al., IEEE TAS, 7, No. 2, p. 1260-1263 (1997).
10. M.A. Hein, Ch.Bauer, G.Muller, J. of Sup., 10, 485 (1997).
11. C. Poole and H. Farach, “Relaxation in Magnetic Resonance”, Academic Press, (New York, 1971).
12. D. P. Choudhury, J. S. Derov and S. Sridhar, (submitted to Appl. Phys. Lett.)
13. S. Sridhar and W. L. Kennedy, Rev. Sci. Instrum. 59, 531 (1988).
14. Z. Zhai, et. al., cond-mat/9903198.
15. D. E. Cox, et al., Phys. Rev. B 57, 10750 (1998).
16. Ch. Niedermayer, et al., Phys. Rev. Lett. 80, 3843 (1998).
17. This is true only if the response is purely relaxational and $\tau$ increases with decreasing $T$. The in-phase susceptibility change can even be “ferromagnetic-like”, i.e. it can increase across $T_p$ in the presence of a restoring force.
18. D. P. Choudhury, H. Srikanth, S. Sridhar and P. C. Canfield, Phys. Rev. B 58, 14490 (1998).
19. J. M. Tranquada, et al., Phys. Rev. Lett. 73, 1003 (1994).
20. S.-H. Lee and S.-W. Cheong, Phys. Rev. Lett. 79, 2514 (1997).
21. V. J. Emery and S. A. Kivelson, Nature (London) 374, 434 (1995).
22. S. R. White and D. Scalapino, Phys. Rev. Lett. 80, 1272 (1998).
23. C. C. Tsuei, et al., (preprint).
24. N. Hakim, et. al., BAPS 44, 1658 (1999).