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# Properties and origins of non-resonant microwave absorption in composites of high-temperature superconductors

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**Abstract.** The temperature and magnetic field dependence of the width intensity and position in magnetic field of the derivative of the non-resonant microwave absorption has been measured in a granular sample of $YBa_2Cu_3O_{7-\delta}$. Measurements were made on a single sample in order to keep constant the effect of sample microstructure. They were made with appropriate control of the many parameters known to influence the behaviour of the absorption. The measurements reveal new behaviour. The hysteresis is shown to display a time dependence after the removal of a magnetic field attributed to the decay of the magnetization. The theory of Xia and Stroud for the microwave response of a superconducting loop in a magnetic field formed by Josephson links is extended to the case of a granular composite where there is a distribution of loop areas and orientations with respect to the magnetic field. Assuming that the distribution of loop areas is determined by a Boltzman function of the energy of the current loop in a magnetic field, it is possible to completely account for the line shape and the temperature dependence of the intensity. The temperature and magnetic field dependence of the field position and width of the line are concluded to be a result of the temperature and field dependence of the flux trapping properties of the superconductor. It is shown that the microwave absorption can be used to measure the flux trapping behaviour of the material, such as the decay of the magnetization.

## 1. Introduction

The ceramic composites of the high-temperature copper oxide superconductors show an intense frequency-independent absorption of microwaves at energies less than the superconducting gap [1–12]. The intensity of the absorption increases with the application of a DC magnetic field and therefore the derivative of the absorption can be observed using an electron paramagnetic resonance spectrometer. The absorption has been suggested to be due to the the existence of micron-sized current loops in the granular composite formed by Josephson links between the grains [4, 11, 12, 13]. As the DC $H$ field is increased flux jumps occur in these loops causing large voltage pulses which produce current in excess of the critical current for the loop. The resulting normal current then absorbs microwave energy. However, it has not been demonstrated previously that this model can account for the detailed properties of the absorption such as the temperature and magnetic field dependence of the field position, width and intensity of the absorption.

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The principle purpose of this work is to extend the model of Xia and Stroud [13] for the microwave absorption of a single superconducting loop of a square micron area to the case of a granular composite where there is a distribution of loop areas and orientations with respect to the DC magnetic field, and to examine whether the model can account for the detailed behaviour of the absorption. It is shown that the theory in conjunction with the flux trapping behaviour of the materials can account for the line shape, width, magnetic field and temperature dependence of the derivative of the absorption.

Because the properties of the absorption depend to some extent on the microstructure of the sample, such as the grain size, which may differ from sample to sample, it is difficult to use measurements of different properties from different workers to compare with calculations. Further, in the early studies of the properties of the absorption, many of the parameters which influenced the behaviour were not known and thus not properly controlled in the various measurements. For example, a measurement of the temperature dependence of the magnetic field position of the centre of the line requires that the magnetic field be swept to the same maximum DC magnetic field on each measurement because the maximum field applied to the sample also affects the field position of the line. In this work the important properties of the line have been measured in a single sample thereby making the effect of microstructure a constant in the various measurements and taking care to control all parameters which are known to affect the behaviour of the absorption. While measurements of some of these properties have been previously reported, in different samples these measurements provide a comprehensive consistent set of data on the behaviour of the absorption in a single sample. Also these measurements have revealed new behaviour not previously observed. For example, it has been observed here that the microwave absorption displays a dependence on time elapsed after the removal of a magnetic field.

## 2. Experimental details

The microwave absorption was studied using a Varian E-12 electron paramagnetic resonance spectrometer operating at 9.2 GHz. The sample temperature was controlled using an Air Products closed cycle displex refrigeration system. The temperature was monitored by a thermocouple in contact with the sample. When the temperature dependence of the centre of the derivative was measured the samples were first cooled below $T_c$ in zero magnetic field. At each temperature the magnetic field was swept to the same maximum value of a 100 G. This must be done, because as will be shown below the position and the width of the line also depend on the magnitude of the magnetic field that was applied before the sweep. All measurements used the same modulation of 40 G and sweep rate of 100 G/min.

Because hysteresis effects were studied some care was taken to separate possible instrumental hysteresis from behaviour intrinsic to the sample. Previous studies of the hysteresis have not discussed the role of instrumental hysteresis in the data. Instrumental hysteresis is evident when the time to sweep through a line is fast compared with the response time setting of the spectrometer. In this work these effects were separated out by recording a resonance of $Cr^{3+}$ from a ruby crystal also located in the microwave cavity with the superconductor. By appropriate orientation of the ruby crystal, a resonance line of the $Cr^{3+}$ can be arranged to occur at a magnetic field in the region of the microwave absorption allowing the observation of the $Cr^{3+}$ resonance and the microwave absorption

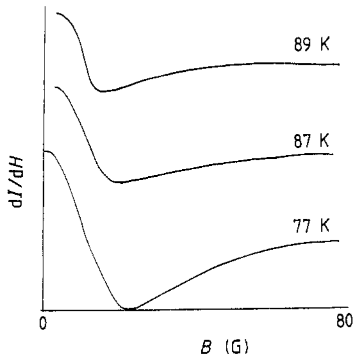

**Figure 1.** Derivative of non-resonant microwave absorption at low magnetic fields at temperatures near $T_c$, showing the narrowing and shifting of field position of line as temperature changes.

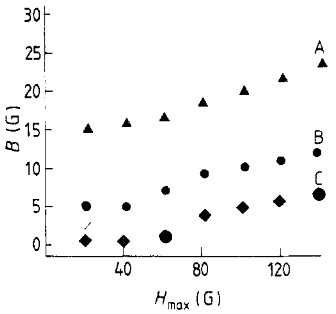

**Figure 2.** Curve A: Field position at the centre of absorption versus previously applied magnetic field. The measurement is made as a function of increasing values of the previously applied magnetic field. Curve B: Effect of previously applied magnetic field on line width. Curve C: Hysteresis of magnetic field at the centre of the line versus maximum DC magnetic field applied to the sample.

during the same sweep. It is generally recommended that the time to sweep a line should be about ten times the spectrometer response time setting. A small hysteresis was, however, observed even when this condition was met.

The hysteresis was observed in this work to be time dependent. This time dependence was measured in the following way. A DC magnetic field of 1000 G was applied to the sample after the sample had been cooled below $T_c$ in zero magnetic field. This field was then rapidly reduced to 250 G and from this field a rapid recording of the derivative was made on the down sweep. The time elapsed from the removal of the 1000 G field was recorded. After this scan the field was rapidly returned to 250 G and another down scan recorded and the elapsed time measured. This procedure was repeated a number of times.

The samples of the 90 K superconductor $YBa_2Cu_2O_{7-\delta}$ were prepared and characterized by methods previously described [14].

## 3. Results

Figure 1 shows the derivative of the absorption versus increasing DC magnetic field, at some different temperatures near $T_c$. Note that in this particular observation the centre of the derivative is not at $H = O$. The data in figure 1 shows that the line width broadens and the centre of the line shifts to higher magnetic field as the temperature is lowered below $T_c$. The position of the centre of the line is the magnetic field half way between the maximum and minimum of the derivative. These effects are continuous down to 20 K, the lowest temperature of our measurement. As observed by many workers, the intensity of the absorption also increases as the temperature is lowered below $T_c$ [1, 2, 4, 8, 11]. Figure 2 (curves A and B) show the effect of the magnitude of the previously applied magnetic field on the magnetic field position of the line and the line width respectively, at 77 K. The measurement is made by sweeping the DC magnetic field to a given strength then returing the field to zero and sweeping to a higher field. A number of workers have observed that the absorption displays a hysteresis in the

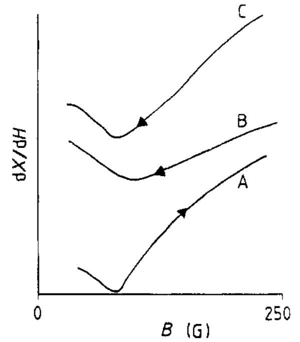

**Figure 3.** Derivative of microwave absorption measured on the up scan (curve A) and on the down scan at $t = 0$ s (curve B) and $t = 29$ (curve C) after removal of a 1000 G field

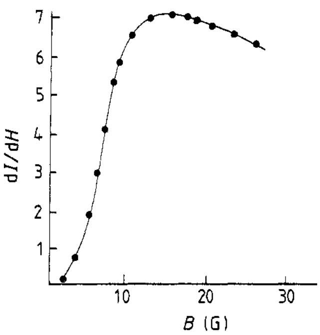

**Figure 4.** Measured line shape at 77 K in $YBa_2Cu_3O_{7-\delta}$ (full curve) and calculated line shape based on flux jump model (full circles).

magnetic field position and intensity of the line [1, 11, 10, 14]. The field position of the centre of the derivative is different when the magnetic field is increased to a given value compared with when the field is decreased from that value. Curve C of figure 2 shows a plot of the difference of the centre of the derivative for the up and down sweep versus the maximum DC magnetic field applied to the sample at 77 K. The magnitude of the hysteresis versus the maximum field applied is considerably smaller than previously reported measurements because many of the observations report the hysteresis as the separation between the peaks of the derivative rather than the centre of the derivative and because this data is corrected for instrumental hystersis. The separation of the peaks of the derivative on the up and down scan is not a measure of the hysteresis because the line width on the up and down scan is also changing with the maximum field applied. The hysteresis is a result of flux trapped in the sample. When the applied magnetic field is returned to zero some field remains trapped in the sample as long as it is kept below $T_c$ and this can be measured from the difference of the centres of the derivative on the up and down scan. Note also that there is a hysteresis in the peak height of the absorption in that the peak height is smaller on the down sweep of the magnetic field. The ratio of the peak heights on the down sweep to the up sweep correlates inversely to the magnitude of the hysteresis and therefore to the trapped flux. The magnitude of the hysteresis also depends on whether the sample is cooled below $T_c$ in a magnetic field or in zero field. The hysteresis is larger when the field is applied before the sample is cooled below $T_c$ compared with when the field is applied after cooling. It was found that the hysteresis depends on the orientation of the DC magnetic field with respect to the geometry of the pellet. For the pellet in the form of a flat platelet, the hysteresis is larger, for a given applied field, when the field is parallel to the large face compared to when the field is perpendicular to the face. This behaviour is consistent with the known effects of sample geometry and orientation on the properties of flux trapping. The hysteresis is also temperature dependent: for a given maximum applied DC magnetic field, it increases with decreasing temperature, reflecting the increase in the flux trapping at lower temperatures.

The hysteresis displays a dependence on time elapsed after the removal of a DC magnetic field. Figure 3 is an example result of the time dependence measured, as described in the experimental section, showing the absorption on the up scan and two successive times on the down scan after the removal of the 1000 G field. Clear changes in the absorption with time are evident. Both the field position of the maximum and the intensity of the absorption recorded on the down scan have changed with time. It is important to note that at the rapid rate of the down scan, the difference in field position of the absorption on the up and down scan does not represent the intrinsic hysteresis of the magnetization of the sample because at rapid scan rates there is considerable instrumental hysteresis. However, this can be separated out using the calibration resonance of the $Cr^{3+}$ as described above. The instrumental hysteresis for a given scan rate does not depend on time elapsed and therefore the time dependence shown in figure 3 is associated with intrinsic behaviour of the sample. Both down scans in the figure were recorded at the same instrumental conditions. The magnitude of the hysteresis of the field position of the absorption is changing with elapsed time from removal of the 1000 G field. The intensity of the $Cr^{3+}$ calibration resonance does not depend on the direction of sweep and elapsed time meaning there is no instrumental hysteresis in the resonance intensity. The time dependent intensity changes observed in figure 3 are also intrinsic to the superconducting sample.

## 4. Mechanism of the microwave absorption

Xia and Stroud [13] recently analysed the electromagnetic response of a 1 $\mu m^2$ superconducting loop of wire formed by weak links of Josephson junctions in an increasing magnetic field. They showed that when such a loop, oriented perpendicular to an applied magnetic field, was subjected to microwave radiation it displayed a series of equally spaced sharp absorptions of microwaves as a function of increasing magnetic field. The current flowing through a junction depends on the relative phase of the wave functions of the Cooper pairs in each superconducting part of the junction and can be represented as

$$I = I_0 \sin(\theta_1 - \theta_2) \qquad (1)$$

where $\theta$ is the phase. The application of a DC magnetic field perpendicular to the junction changes the relative phase of the current in each part of the junction by,

$$\Delta\theta = 2\pi\phi/\phi_0 \qquad (2)$$

where $\varphi$ is the flux threading the loop and $\varphi_0$ the quantum of flux. Because flux is quantized in the superconducting loop, there are phase jumps as the applied field increases. These occur when the field in the loop changes by an integral multiple of the unit quantum of flux. These flux jumps occur in a very short time of the order of $10^{-12}$ s and therefore produce large voltage pulses given by:

$$V(t) = (h/2e)\, \mathrm{d}\theta/\mathrm{d}t. \qquad (3)$$

When this occurs the critical current of the superconductor is exceeded and normal current flows. It is this pulse of normal current that absorbs microwave energy. Thus the microwave absorption spectrum of this loop as a function of increasing DC magnetic field will be a series of equally spaced sharp absorptions, which are separated from each other by $\varphi_0/A$ where $A$ is the area of the loop. Microwave energy is also absorbed but less intensely in zero magnetic field. This is a result of the microwaves themselves producing normal current in some loops in which the critical current is exceeded. This normal current is a result of flux jumps associated with the time-dependent $H$ component of the microwaves.

Both of these processes are believed to be the cause of the intense non-resonant microwave absorption in the granular composites of the copper oxide superconductors [1–4, 11]. The current loops in the granular composite are formed by weak links between or on the grains. There is a distribution of loop areas and orientations of the loops with respect to the DC magnetic field. The sharp line structure is therefore broadened out and the observed broad absorption reflects the distribution of loop areas and orientations in the composite. If this mechanism is valid, it should be possible to account for the detailed properties of the absorption such as the shape of the line and its field and temperature dependence.

### *4.1. Line shape*

In order to determine if this model can account for the shape of the absorption line, it is necessary to know the distribution of loop areas and orientations. It will be assumed that the distribution of loop areas is determined by a Boltzman distribution for the energy of a current loop in a magnetic field. This energy is $\frac{1}{2}\boldsymbol{B}\cdot A\boldsymbol{I}$. Thus for a given current $\boldsymbol{I}$ and magnetic field $\boldsymbol{B}$, the fraction of all loops having area $A$ will be given by:

$$f = \exp(-\boldsymbol{B}\cdot A\boldsymbol{I}/kT). \tag{4}$$

Note that as the field is changed, the distribution changes. It can be shown that the distribution of loop areas over angle with respect to the direction of the applied magnetic field is constant [15]. The line shape in the granular composite is computer simulated in the following way. At each magnetic field $\boldsymbol{B}$, a sharp microwave absorption will occur for those loops in the distribution that meet the flux jump condition at that field, i.e., $BA = N\varphi_0$. A narrow Gaussian derivative is centred at each $\boldsymbol{B}$ for each $A$ of the distribution that meets this condition. The height of the derivative is weighted by $f$ which determines the fraction of loops of a given area. The contribution of the lines for all areas at a given $\boldsymbol{B}$ are then added. The calculation is done for all $\boldsymbol{B}$ of the sweep. The distribution function $f$ is arbitrarily truncated at some maximum $A$. It is assumed that there is a finite maximum allowed loop area. In order to fit the experimental line shape in the composite the maximum value of $A$, the current $\boldsymbol{I}$ and the width of the individual derivatives are treated as adjustable parameters. Figure 4 shows the fit of the measured line shape in $YBa_2Cu_3O_{7-\delta}$ at 77 K. The full curve is the measured absorption; the points are the calculated simulation. The parameters of the fit are $I = 0.2\ \mu$A, $A_{\max} = 7\ \mu\text{m}^2$ and the intrinsic line width is 3.5 G. The computer simulation of the line shape based on the flux jump model is in very good agreement with the measured line shape, lending support to the proposed mechanism for the absorption.

The simulation has been performed by assuming the current is independent of the loop area and applied field. It is known, however, that the current in a junction decreases with increasing DC magnetic field. In the approximation of the line shape calculation described above, the fitting parameter $I$ may be thought of as the average current over all loop areas and magnetic fields. The distribution of loops will also be determined by the physical microstructure of the sample, particularly the particle size distribution. This has not been explicitly written into the distribution. However, it will manifest itself by a different set of fitting parameters in a composite having a different distribution of particle sizes.

The value of the field at the peak of the derivative has been related to the average loop area [4]. In this analysis, the average value of the loop area, for a given sweep maximum $B_{\max}$ will be:

$$\langle A\rangle = \int_0^{B_{\max}}\int_0^{A_{\max}} A\exp(-\boldsymbol{B}\cdot A\boldsymbol{I}/kT)\,\mathrm{d}B\,\mathrm{d}A \Bigg/ \int_0^{B_{\max}}\int_0^{A_{\max}} \exp(-\boldsymbol{B}\cdot A\boldsymbol{I}/kT)\,\mathrm{d}B\,\mathrm{d}A. \tag{5}$$

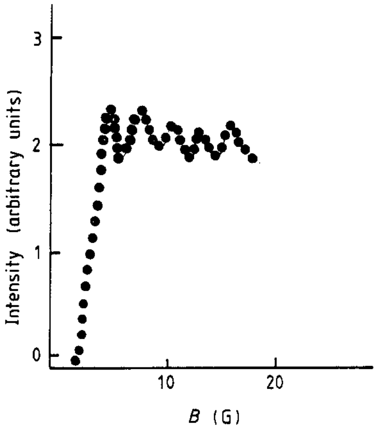

**Figure 5.** A calculated absorption line showing structure obtained by narrowing the intrinsic line width.

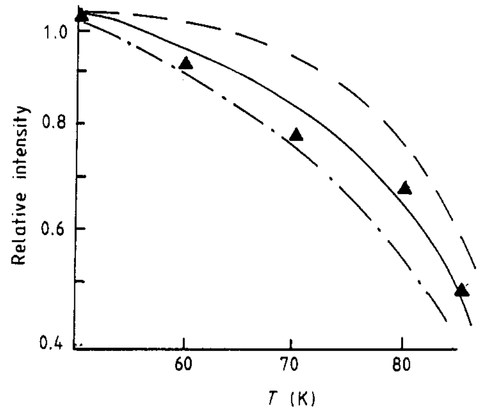

**Figure 6.** Calculated temperature dependence of intensity of absorption at three different magnetic fields: — — —, 5 G; —·—, 20 G; ——, $H_{\max}$. Points (▲) are experimental values at maximum of derivative.

Since the distribution function changes with the applied field the average loop area will depend on the maximum field applied and will therefore not be simply related to the peak of the derivative. Numerical evaluation of equation (5) for the absorption shown in figure 4 using the fitting parameters cited above yields $\langle A\rangle = 3.37\ \mu\mathrm{m}$ compared with $1.38\ \mu\mathrm{m}$ obtained by using $\varphi_0/H_{\max}$ for the average loop area.

There have been some reports of fine structure superimposed on the broad absorption line [4, 16]. It has been found here that when the intrinsic line width of the absorption at each flux jump is narrowed, the simulation shows the possibility of fine structure on the absorption.

An example of the structure that can be produced by the simulation is shown in figure 5. This simulation was performed using the same parameters as those used for figure 4 except that the intrinsic line width was narrowed to 1.0 G. Structure can also be produced on the line by narrowing the distribution of loop areas. The simulation does support the possible observation of structure on the absorption.

### *4.2. Effect of magnetic field on line position and width*

The difference in field position of the centre of the line on the up and down sweeps is a result of the trapping of flux by the superconductor. When the centre of the line occurs at a higher field on the down sweep compared to the up sweep, the trapped field is opposite in direction to the applied field and its magnitude is the difference between the centres of the two lines. This also means that on the next up sweep after the application of a given magnetic field, the centre of the line should be shifted up by an amount equal to the trapped field. A comparison of curves A and C in figure 2 shows this effect in that the magnitude of the hysteresis for a given maximum of sweep corresponds well with the shift of the line on the next upward sweep of the field. In fact the microwave absorption can be used to measure $H_{c1}$, the lowest magnetic field at which flux begins to penetrate the sample. The value of $H_{c1}$ is the field at which the absorption begins to show an upward shift of field position at the centre of the derivative and the field at which a hysteresis is first evident. From this measurement, the data in figure 2 yields a value of

60 G for $H_{c1}$ at 77 K, which is in reasonable agreement with measurements in composites of this material at this temperature.

The increase in the width of the absorption line with the magnitude of the previously applied DC magnetic field may also be due to flux trapping. It is noted that the onset of broadening with the maximum applied field corresponds to the field at which the upward shift and hysteresis first occurs. This suggests that the line broadening is also associated with flux trapping. It is not entirely clear how increased trapped flux can lead to a broadening of the line. One possibility is that the trapped field may be altering the distribution of loop areas. In a loop of current formed by a Josephson junction there is a maximum allowable current which decreases with an increasing DC magnetic field. In terms of the analysis of the line shape in section 4.1, this means that the fitting parameter $I$ is decreasing as the magnetic field is increased. It is found that reducing this parameter in the simulation does result in a broadening of the line. This indicates that the line broadening with increasing magnetic field may be due to a reduction of the average Josephson current in the loops because of increased trapping of flux.

### 4.3. Temperature dependence of the line

While the procedure in section 4.1 is appropriate for simulating the line at one temperature, some refinements are necessary to examine the temperature dependence of the non-resonant absorption. At a given magnetic field the intensity of the absorption, besides being proportional to the number of loop areas meeting the flux jump condition, will also be proportional to the current flowing in the loop. Thus the intensity at a given magnetic field will be proportional to

$$I \exp(-\boldsymbol{B}\cdot AI/kT). \qquad (6)$$

The Josephson current $I$ is temperature dependent. For $T > T_c/2$ the current will be proportional to the magnitude of the superconducting gap, which will be assumed to follow a mean field temperature dependence $(T_c - T)^{1/2}$. Thus the temperature dependence of the line will determined by:

$$C_1(T_c - T)^{1/2} \exp[-BAC_2(T_c - T)^{1/2}/kT] \qquad (7)$$

where $C_1$ and $C_2$ are fitting parameters. The temperature dependence will depend on the magnetic field at which it is measured because at each field there is a different distribution function. It is more meaningful, therefore, to plot the height of the derivative at a given magnetic field versus temperature because this is proportional to the microwave absorption at this field. The intensity determined by integrating the derivative is not unique in that it depends on the maximum field of the sweep. Figure 6 shows a plot of the height of the derivative of simulation versus temperature at three different magnetic fields. The points are the experimental values measured at the maximum of the derivative and agree well with the predictions of the simulation at the maximum.

The temperature-dependent simulation does not predict a large shift of the line to higher fields or a broadening of the line as the temperature is lowered. The temperature dependence of the line position and width therefore must reflect the temperature dependence of flux trapping. Curves A and B in figure 2 show that the line broadens and shifts to higher magnetic fields with increasing applied field, reflecting the fact that more flux is trapped as the applied field increases. Similarly for a given applied field the amount of trapped flux will increase as the temperature is lowered. The temperature dependence of the magnetization of the sample has been shown to be [17]:

$$M = (-J_c kT/E) \ln(t) \qquad (8)$$

where $t$ is the time, $J_c$ the critical current and $E$ the flux pinning barrier. The critical

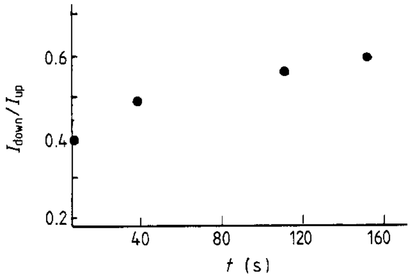

**Figure 7.** Plot of ratio of peak height on down scan to up scan to a constant magnetic field of 250 G as a function of time elapsed after removal of a 1000 G field.

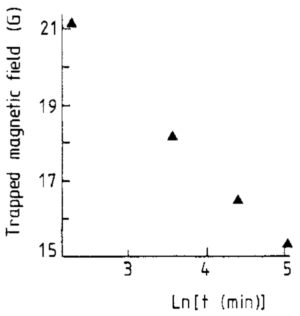

**Figure 8.** Plot of trapped magnetic field versus the log of the time elapsed after removal of 1000 G field obtained from the time dependence of the hysteresis of microwave absorption.

current increases as the temperature is lowered below $T_c$. Thus the trapped flux will increase with lowering temperature. For a given maximum of sweep the line will then shift to a higher field position with lowering temperature, the magnitude of the hysteresis will increase, and the line will broaden. In fact this can be used to measure the temperature dependence of $H_{c1}$ and the amount of flux trapped.

### *4.4. Time dependence of the absorption*

As curve C of figure 2 shows, the hysteresis of the field position of the centre of the line increases with increasing maximum of the sweep field, reflecting the fact that more flux is trapped when a larger field is applied. Also the ratio of the height of the peak on the downward scan to the upward scan decreases with increasing applied magnetic field and thus correlates inversely with the amount of trapped flux, as measured by the hysteresis. The ratio of the peak heights decreases linearly as the magnitude of the hysteresis increases. This would suggest that the time-dependent increase in the peak of the derivative, shown in figure 4, after the removal of the 1000 G field is due to a decay of trapped flux. Figure 7 shows a plot of the ratio of the intensities on the down scan to the up scan versus time elapsed after removal of the 1000 G field. The time to reach a constant peak height is in reasonable agreement with direct measurements of the decay of magnetization in composites of $YBa_2Cu_2O_{7-\delta}$ at 77 K [18]. Using the correlation between the ratio of the peak heights and the magnitude of the hysteresis the time dependence of the trapped flux can be obtained. Figure 8 shows a plot of the trapped flux versus the log of time, showing a straight line as predicted by equation (8). This further supports the conclusion that the observed time dependence is due to the decay of magnetic field trapped in the sample.

## 5. Conclusion

The effect of temperature and magnetic field on the properties of the derivative of the low field microwave absorption has been measured in a granular composite of $YBa_2Cu_3O_{7-\delta}$ with optimum control of parameters which are known to affect the properties. The centre of the derivative shifts to higher magnetic field and the width of the line broadens as the temperature is lowered below $T_c$. The line also broadens and shifts to higher magnetic field as the value of the DC magnetic field applied before the sweep is increased. It is shown that the field position, width and intensity depend on the direction of the field sweep. This hysteresis is also observed to be dependent on the time elapsed after the removal of a large magnetic field.

The derivative of the non-resonant microwave absorption observed in a granular composite of $YBa_2Cu_3O_{7-\delta}$ has been simulated by extending the theory of Xia and Stroud for the electromagnetic response of a single loop of superconducting current formed by Josephson links in a DC magnetic field to the case where there is a distribution of loop areas and orientations. The distribution of loop areas is assumed to be determined by a Boltzmann distribution of the energy of a current loop in a magnetic field. The simulation can account for the detailed shape of the derivative of the absorption and the temperature dependence of the intensity. However, the simulation does not predict the observed broadening and shifts of the line with temperature and magnetic field. These effects are demonstrated to be a result of the field and temperature dependence of the flux trapping behaviour of the composite. It is shown that the microwave absorption can be used to measure the flux trapping properties of the material, including the decay of magnetization.

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