

# Nonresonant microwave absorption study of intrinsic Josephson coupling in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystals

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The results of nonresonant microwave absorption (NRMA) studies in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  single crystals for the  $H\parallel c$  axis and  $H\parallel ab$  plane are reported. In the former orientation, over a few degrees below  $T_c$ , the NRMA signal shapes evolve continuously as a function of temperature. We interpret these results as indicative of rapid strengthening of Josephson coupling between the pancake vortices in the  $\text{CuO}_2$  bilayers on cooling below  $T_c$ . For  $H\parallel ab$  there is almost no change in the signal shape in the temperature range studied. In this case, no loss component attributable to Josephson coupling is observed.

## I. INTRODUCTION

The layered structure and the extremely short  $c$ -axis coherence length  $\xi_c$  of the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  ( $\text{Bi}_2\text{212}$ ) superconductor suggest<sup>1</sup> that it consists of stacks of superconducting sheets of  $\text{CuO}_2$  bilayers Josephson coupled across an intervening nonsuperconducting medium. With  $\xi_c(0) \leq 1 \text{ \AA}$  (Ref. 2) and the interbilayer separation  $s = 15 \text{ \AA}$ ,<sup>3</sup> for temperatures  $T \lesssim T_c - 1 \text{ K}$ ,  $\xi_c < s/\sqrt{2}$  and therefore a description in terms of the Lawrence-Doniach model<sup>4</sup> is applicable. Indeed, direct evidence for such Josephson coupling in  $\text{Bi}_2\text{212}$  was provided by Kleiner *et al.*<sup>5</sup> through the demonstration of ac and dc Josephson effects in these crystals. In addition, there have been several studies<sup>6-10</sup> providing indirect evidence for the occurrence of interlayer Josephson coupling. For example, Duran *et al.*<sup>6</sup> interpret the results of their mechanical oscillator experiments in terms of two-dimensional Abrikosov (pancake) vortices nucleated in the planes interconnected by Josephson strings between the  $\text{Cu-O}_2$  planes. Grover *et al.*<sup>7</sup> have reported the observation of superposition and interplay between two magnetic responses for  $H\parallel c$  in  $\text{Bi}_2\text{212}$  in the temperature interval  $T^* < T < T_c$ , where  $T^*$  is the magic temperature at which the temperature dependent magnetization curves for  $H > H_{\min} (\approx 0.1 \text{ kOe})$  intersect, i.e.,  $dM/dH = 0$  for  $H > H_{\min}$  at  $T = T^*$ . They attribute the two magnetic responses to the superconducting fluctuations in the individual bilayers and the superconducting network made up of Josephson current flows across weakly coupled  $\text{CuO}_2$  bilayers. However, there have been fewer reports over the years which provide information about the onset and the strengthening of the phase-locking phenomenon across the normal to superconducting crossover region.

The method of nonresonant microwave absorption (NRMA) has emerged as a sensitive and specific technique for the study of absorption of microwave radiation by Josephson junctions in high- $T_c$  superconductors.<sup>11</sup> Since the

viscosity of the Josephson vortices are orders of magnitude lower than that of Abrikosov vortices,<sup>12</sup> the dominant contribution to the microwave loss comes from the former when present. We have, therefore, subjected the  $\text{Bi}_2\text{212}$  system to NRMA studies for  $H\parallel c$  and  $H\parallel ab$  orientations. Many workers<sup>13,14</sup> have reported on the microwave absorption studies of  $\text{Bi}_2\text{212}$  single crystals. Recently, a paper by Baginskii *et al.*<sup>15</sup> describes the electromagnetic absorption studies in these crystals in rf (700 MHz) range. They observe an additional absorption which they attribute to the Josephson component of the vortices. We present here a more elaborate and quantitative analysis of the process of the Josephson coupling and examine our results in the light of viscous motion of vortices in layered superconductors. A preliminary report of this study has been presented earlier.<sup>16</sup> For  $H\parallel c$ , very close to  $T_c (\sim 1 \text{ K})$ , we find a signature of a response attributable to the viscous motion of Abrikosov vortices in the superconducting layers. Over a few degrees below this temperature, a particular component of NRMA rapidly grows, reaches a maximum and then diminishes, which we interpret as due to "thickening" of the Josephson-junction medium to the flow of quantized flux. For  $H\parallel ab$ , there is no change in the shape of the NRMA signals over the same temperature interval, presumably because in this orientation, no nucleation and stacking of pancake vortices is warranted. Instead, the steady field ( $H\parallel ab$ ) is shielded by the so-called coreless Josephson vortices.<sup>17,18</sup>

In the technique of NRMA (Ref. 11) the sample is subjected to a steady magnetic field and microwave radiation of known power is made incident transverse to it. A standard continuous-wave nuclear magnetic-resonance (NMR) or electron-paramagnetic-resonance (EPR) spectrometer can be used to study the magnetic-field-dependent absorption of the rf or microwave radiation. The signal is recorded using a magnetic-field modulation in conjunction with phase sensitive detection and therefore the field derivative  $dP/dH$  of the

absorbed power  $P$  is recorded. In single crystals and single crystalline thin-film samples the field-dependent dissipation of microwave power originates in the viscous motion of quantized flux caused by induced microwave currents. The intensity and shape of NRMA signal depend upon various parameters like temperature, dc magnetic field, microwave power, and above all, on the nature of the sample. Exotic phenomena such as paramagnetic Meissner effect (alias Wohleben effect) have characteristic NRMA signals.<sup>19</sup> The technique of NRMA has been very effective in distinguishing between the dissipation originating from intra- and intergranular regions in superconducting samples.<sup>20</sup>

## II. EXPERIMENTAL

We have recorded NRMA signals in single-crystal samples of Bi2212 using a Bruker ER 200D X-band EPR spectrometer equipped with an Oxford ESR 900 continuous-flow cryostat. The rectangular cavity operated in the  $TE_{102}$  mode with the magnetic vector  $H_{rf}$  being maximum at the center of the cavity where the sample is placed and directed along the vertical. The dc magnetic field  $H$  is varied in the horizontal plane and thus  $H \perp H_{rf}$  always. The sample had dimensions 2 mm, 2 mm, and 50  $\mu\text{m}$  along the  $a$ ,  $b$ , and  $c$  directions. Thus, the  $a$  and  $b$  dimensions were much larger than  $\lambda_J$ , the Josephson penetration depth ( $\approx 0.14\text{--}0.7 \mu\text{m}$ ).<sup>5</sup> Therefore, we are in the large junction limit leading to the formation of Josephson vortices. The sample was placed at the center of the cavity mounted on a flattened end of a quartz rod with a small amount of glue. The temperature was crosschecked with a calibrated copper-constantan thermocouple situated close to the sample. The values of magnetic-field modulation and microwave power were 4 G (at 100 kHz frequency) and 20 mW, respectively.  $H$  was swept from  $-50$  to 1050 G and the microwave frequency was 9.45 GHz. The sample temperature was varied from 4.2 to 100 K. However, in this report we shall concentrate on the nominally reversible regime limited to a few degrees below  $T_c$ . It must be mentioned here that conventionally the microwave surface resistance is measured using the cavity perturbation technique. Such measurements give the surface resistance directly and then it is straightforward to fit the results with various theories. A few such studies on single crystals of Bi2212 are listed in Refs. 21 and 22. However, with the combination of field modulation and phase sensitive detection, the EPR technique is highly sensitive and has enabled us to distinguish between different sources of dissipation of microwave power.

The studies being reported here have been made on air-annealed single crystals with nominal composition Bi2212. The crystals were grown by the traveling-zone flux method.<sup>23</sup> Such crystals always admit the possibility of tiny intergrowth of layers of Bi2201<sup>24</sup> and they are also, not free from dislocations.<sup>25</sup> These act as intergranular weak links giving rise to characteristic low-field NRMA signals (see below). The two sample pieces used for NRMA study were well characterized by electrical resistivity and by ac and dc magnetization measurements. The crystals in the as grown form had nominal  $T_c$  of 84 K with  $\Delta T = 0.7$  K. After annealing in air, the two pieces had nominal  $T_c \sim 82.5$  K with  $\Delta T \sim 0.35$  K. The particular sample on which NRMA data

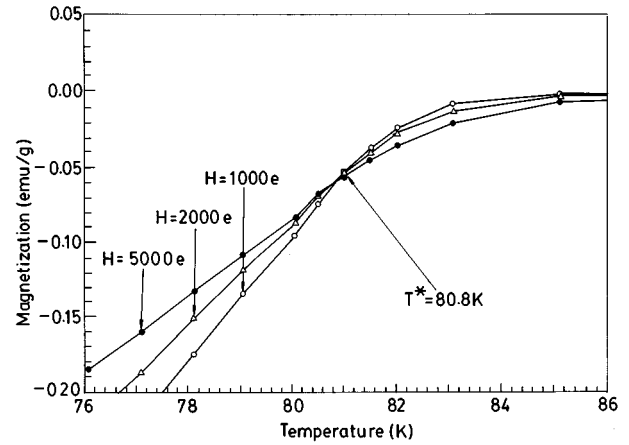


FIG. 1. Magnetization vs temperature for  $H||c$  measured using a superconducting quantum interference device magnetometer (Quantum Design, MPMS<sub>2</sub>). Note that the diamagnetic response extends by a few degrees beyond the nominal  $T_c = 82.5$  K.

are being explicitly shown in this report had  $T^*$  value of 80.8 K (see Fig. 1). It is apparent from the data of Fig. 1 that superconducting fluctuations give significant diamagnetic contribution up to several degrees above its nominal  $T_c$  value of 82.5 K (as determined from zero-field resistivity and in-phase ac susceptibility measurements).

## III. RESULTS

Figure 2 shows NRMA signals recorded at various temperatures (77–86 K) for  $H||c$  orientation. It is apparent that the line shapes evolve continuously as the temperature

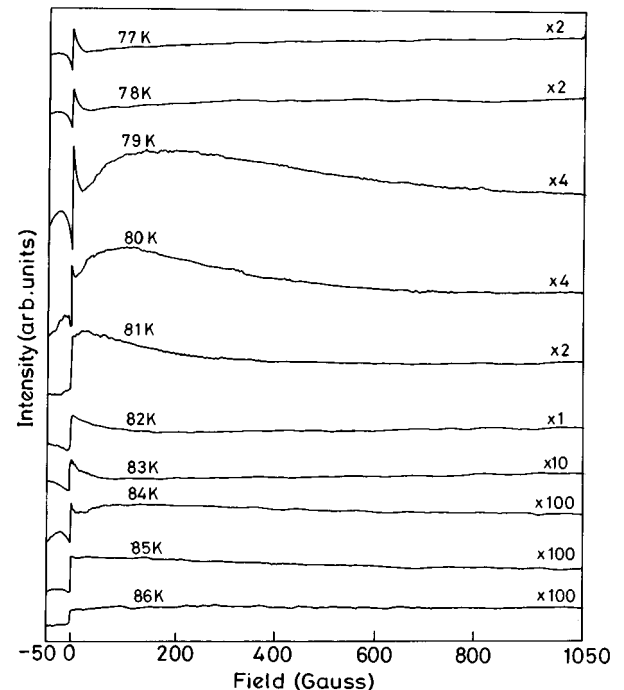


FIG. 2. NRMA signals at various temperatures for  $H||c$ . Especially noteworthy is the temperature-dependent evolution of broad winglike structure in the intervals 78–81 K and 83–86 K.

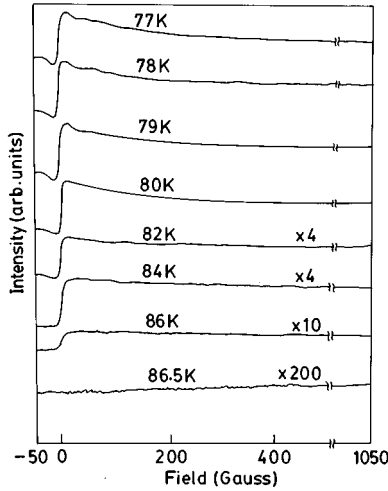


FIG. 3. NRMA signals at various temperatures for  $H||ab$ . The temperature dependent evolution of the line shapes observed for  $H||c$  is absent for this orientation.

moves across nominal  $T^*$  (80.8 K) and  $T_c$  (82.5 K) values. The signal at 77 K shows that NRMA is dominated by a narrow signal. Such narrow signals are usually observed in ceramic samples of high-temperature superconductors,<sup>20</sup> where they originate from intergranular weak links. We believe that in case of the single-crystal sample of Bi2212, the narrow signal at 77 K and the narrow, low-field component at other temperatures, arise from inevitably present weak links of interunit-cell variety in this system. As the temperature moves towards  $T^*$  value, we start to see another component of absorption as a broad winglike signal (see NRMA signal at 79 K). This component grows in intensity across the 80–81 K region and it appears to diminish as the temperature approaches nominal  $T_c$  value. We associate this particular component with the evolving Josephson coupling between the  $\text{CuO}_2$  bilayers in this temperature interval. Further, the microwave power was varied and the signals were recorded as a function of the field at an intermediate temperature of 80 K. It was observed that the intensity of the “Josephson” component is proportional to the square root of the power, indicating that we are in the Ohmic region. Forward and reverse sweeps of the magnetic field showed no measurable hysteresis. The signals in the interval 84–86 K are about 25 times weaker than those at lower temperatures. The signal shape at 84 K once again appears to reveal the existence of absorption from two different sources which could be identified with superconducting fluctuations persisting above  $T_c$  and/or a part of it could be attributed to small regions of the sample insufficiently air annealed. An important feature of the signal at 86 K is the characteristic quasi-linear dependence of dissipation on field, which usually<sup>20</sup> is associated with the motion of Abrikosov flux lines in bulk of the material. If we choose to follow the evolution of NRMA signals from 86 to 77 K, the signals in the interval 86 to 82 K would appear to serve as precursors of signals which follow below 82 K.

Figure 3 shows the NRMA signals recorded for  $H||ab$  at various temperatures. The significant difference from the signals for  $H||c$  is apparent. The dramatic evolution of the signal shape as a function of temperature observed earlier is

absent now. Instead, only a marginal increase in the intensity of the signal between 86–80 K is seen. However, we do observe a kink at low fields in the temperature range 77–80 K suggestive of a coupling signature, which may be due to slight misorientation of the  $ab$  plane of the crystal with respect to applied steady field.

#### IV. DISCUSSION

Coffey and Clem<sup>26</sup> have presented a detailed phenomenological treatment of microwave surface impedance in terms of complex penetration depth  $\lambda$  as function of frequency  $\omega$ , magnetic-flux density  $B$  and temperature  $T$ . However, we present here an analysis relevant to our data using the simpler and somewhat more transparent method due to Portis *et al.*<sup>27</sup> particularly applicable to NRMA experiments. It describes the complex surface impedance of a superconductor subjected to a static magnetic field  $H$  and incident upon by microwave radiation. The surface impedance  $Z = R_s - iX_s$  where the real part

$$R_s = X_o \{ [-1 + (1 + 4f^2 H^2 / B_o^2)^{1/2}] / 2 \}^{1/2} \quad (1)$$

and imaginary part

$$X_s = X_o \{ [1 + (1 + 4f^2 H^2 / B_o^2)^{1/2}] / 2 \}^{1/2}$$

with  $X_o = 4\pi\omega\lambda\mu^{3/2}/c^2$  and  $B_o = 8\pi\omega\mu\lambda^2\eta/\phi_o$  where  $\lambda$  is the London penetration depth,  $\omega$  is the microwave frequency,  $\eta$  is the coefficient of viscosity experienced by the moving fluxons,  $\mu$  is the magnetic permeability, and  $\phi_o$  is the unit flux.  $f$  stands for the fraction of free or weakly pinned fluxons. In the NRMA context, the induced microwave currents cause dissipation by driving the fluxons and real part is the lossy part. We have to therefore compute the derivative  $(dR_s/dH)$  of the real part of surface impedance as a function of static field  $H$ . It is a usual practice to do this computation for a given value of  $B_o$  which incorporates all other important parameters like  $\eta$  and  $\lambda$ . To do a meaningful comparison between calculated and observed signals, we need to first identify the different sources of dissipation.

As mentioned earlier the central narrow signal at 77 K (Fig. 2) and other temperatures comes from the Josephson junctions in the intergranular region. Such signals can be aptly described by a model due to Dulic *et al.*<sup>28</sup> The loss in such junctions occurs due to reduction of critical currents as the magnetic field easily penetrates. The expression for field derivative of the loss is given by

$$S_M = \frac{((1/2)I_{mw}^2 R)^{1/2}}{((1/R)(\hbar/2e)\omega_{mw})^2 (1+N)^{3/2}} \times \left( \frac{-dI_c}{dH} H_M + I_M \sin(\Phi_o) \right) \cos(\omega_M t), \quad (2)$$

where  $I_{mw}$  is the induced microwave current,  $I_c$  is the junction critical current,  $R$  is junction resistance,  $H_M$  is the modulation field,  $\omega_M$  is the modulation frequency,  $\Phi_o$  is the phase, and  $N = I_c^2 F^2(H)$  ( $F(H)$  is the envelope of the diffraction relation  $[\sin(\pi H/\phi_o)]/(\pi H/\phi_o)$ ).

At sufficiently low temperatures, for  $H||c$ , straight stacks of pancake vortices are formed.<sup>29</sup> Thermal motion of the pancakes sets in at higher temperatures disrupting the

straight stacks. When the pancakes move away from the core axis, a phase change between the pancake vortices associated with the Josephson current across the layers is created, giving rise to Josephson vortices connecting the pancakes. The formation of such Josephson vortices is energetically favorable for intervortex separation larger than the phase screening length.<sup>30</sup> Then, since the vortices are mobile, the coupling of the pancakes through the Josephson strings can be understood to be in a state of dynamic equilibrium<sup>10</sup> for a given field and temperature. This is similar to the static case of a tilted vortex (away from  $c$  axis) where the pancakes are not stacked straight but are displaced leading to the formation of Josephson strings connecting the pancakes.<sup>30</sup>

We postulate that the dominant contribution to the microwave loss comes from the viscous motion of these Josephson vortices which terminate at either end on pancakes in the planes driven by microwave currents. This state of flux flow is established after crossing the irreversibility line (IL) which is not only a function of the magnetic field and temperature but is also dependent on the measuring frequency  $\omega$ .<sup>13</sup> Our operating frequency of 10 GHz is close to but lower than the estimated depinning frequency of  $\approx 40$  GHz for Bi2212.<sup>13</sup> It is therefore expected that part of the loss occurs due to thermally activated flux flow (TAFF). Further, some fluxons also will be unpinning due to the proximity of the operating frequency to the depinning frequency. So, to account for the loss below the IL we add a fraction of the thermally activated fluxons in the expression for the real part of the surface impedance. Since most of the fluxons are unpinning due to the combined effect of thermal activation and the proximity of the operating frequency to the depinning frequency, not much hysteresis is expected as is experimentally observed. As we increase the magnetic field isothermally we cross the IL to enter a complete flux-flow state where all the fluxons are free to contribute to the microwave power loss.

If  $f$  is the fraction of completely free fluxons at  $H < H_{irr}$  (irreversibility field), the number of pinned fluxons is  $(1-f)$ . Let  $U(H, T, \omega)$  be the activation energy of pinned fluxons, then the fraction of thermally activated fluxons  $F$  can be written as  $F = (1-f) \exp\{-[U(H, T, \omega)/KT]\}$ , where  $U = U_o[(1 - T/T_c)^{3/2}/H] \ln(\omega_p/\omega)$ .<sup>13</sup>

Therefore the modified expression for the real part of the surface impedance is

$$R_s = X_o \left[ -1 + \left( 1 + 4(f+F)^2 \frac{H^2}{B_o^2} \right)^{1/2} \right]^{1/2} \quad \text{for } H < H_{irr} \quad (3)$$

and at  $H \geq H_{irr}$ ,  $f = 1$ , hence  $F = 0$ , so we get back the earlier expression (1) with  $f = 1$  which describes a complete flux-flow state.

Kleiner *et al.*<sup>5</sup> have shown evidence for series arrays of Josephson junctions consisting of different numbers of phase locked coupled layers of Bi2212. This would lead to a distribution of coupling strengths of such arrays and in turn a range of field values required to decouple them. To describe this aspect we use a treatment analogous to the one used by Silva *et al.*<sup>22</sup> to take into account the decoupling of Josephson junctions. The power loss due to decoupling can be written as

$$P_J = \Delta P_o (1 - e^{-(H/H_d)}), \quad (4)$$

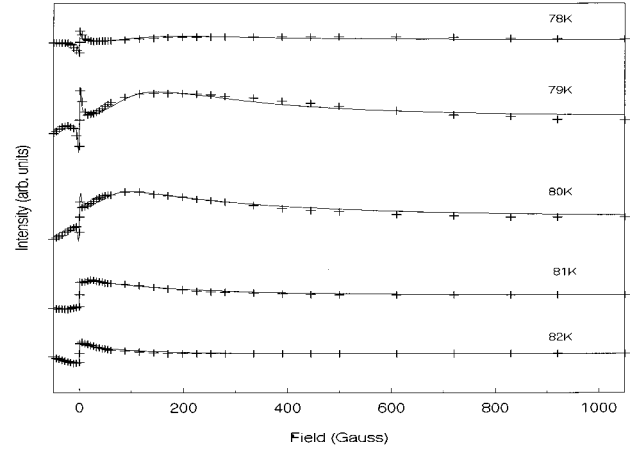


FIG. 4. Calculated derivatives (continuous lines) of the power loss vs field  $H$  at various temperatures (see text), (+) marks represent the observed signals.

where  $\Delta P_o$  is the power absorbed when all the junctions are decoupled at a given temperature.  $H_d$  is the mean decoupling field.

We add this expression to that describing complete flux-flow state to account for the total power loss after crossing the IL. Therefore, the field derivative of the power loss for  $H < H_{irr}$  is given by combining Eqs. (2) and (3)

$$\frac{dP}{dH} = S_M + \frac{dR_s}{dH}.$$

For  $H \geq H_{irr}$ , we combine Eqs. (1), (3), and (4):

$$\frac{dP}{dH} = S_M + \frac{dR_s}{dH} + \frac{dP_J}{dH}.$$

Figure 4 shows an evolution of calculated NRMA signals following this approach along with the experimental results in the temperature range 78–82 K. The two are seen to match quite satisfactorily. For these calculations we took both  $\eta$  and  $\lambda$  to be temperature dependent,  $\eta(T) = \phi_o H_{c2}(T) / \rho_n c^2$ , where the upper critical field  $H_{c2}(t) \propto (1 - t^2) / (1 + t^2)$  (Ref. 26) ( $t = T/T_c$ ), and  $\lambda(t) = \lambda(0) / (1 - t^4)^{0.5}$ .<sup>26</sup> Therefore,  $B_o(t) = 8\pi\omega\mu\lambda^2(t)\eta(t) / \phi_o = B_o(0) / (1 + t^2)^2$  such that  $B_o(0) = 8\pi\omega\mu\lambda^2(0)\eta(0) / \phi_o$ . For  $\lambda(0)$  we substituted 3000 Å (Ref. 31) and obtained two sets of  $B_o(0)$  values as 100 and 1000 G. These values imply  $\eta(0)$  values to be  $10^{-7}$  and  $10^{-6}$  cgs units, respectively. Such values for  $\eta(0)$  are usually considered reasonable for the two known sources of dissipation due to viscous motion of fluxons namely, the intragrain weak links and the bulk in ceramic samples. At an order of magnitude level, therefore, the identification of above  $\eta$  values with the corresponding two components for  $H \parallel c$  in Bi2212 single crystals appears reasonable. For the relative proportions of the two components of dissipation at different temperatures in Fig. 4, we have been guided by the relative intensities of the experimentally observed signals at the equivalent temperatures in Fig. 2. We, therefore, labeled the calculated curves in Fig. 4 by the respective temperature values.

In the flux-flow model by Portis *et al.*<sup>27</sup> the dissipation due to flux flow starts from near zero fields. Also, the peak in

the field derivative of the surface impedance occurs close to zero field.<sup>20</sup> In the experimentally observed signals, the broad part identified with the appearance of Josephson coupling has peaks at high fields. We believe that the onset of the complete flux-flow state can be associated with these peaks, so that the field value at the peak can be termed as the irreversibility field  $H_{irr}$  which defines the crossover from TAFF to free flux-flow state.

## V. SUMMARY

To conclude and summarize, we have studied the temperature dependence of magnetic-field-dependent microwave absorption in Bi2212 single crystals. For  $H\parallel c$ , the absorption is seen to consist of three different components. One of these components can be correlated with the presence of Josephson coupling between the  $\text{CuO}_2$  planes. The key result is that

coherence between pancakes located across nearest-neighbor  $\text{CuO}_2$  bilayers evolves over a narrow range of temperature interval across the nominal  $T^*$  value. In contrast, for  $H$  in the  $ab$  plane, no loss component attributable to Josephson coupling is observed pointing out that it is necessary to have some  $c$ -axis component of the magnetic field. Only then, pancakes displaced from the  $c$  axis can arise and to compensate for the phase difference between these pancakes, the Josephson strings would form.

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