# **Evidence of two-dimensional Josephson strings in high-purity**  $YBa_2Cu_3O_{7-\delta}$  **single crystals**

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A strange oscillating magnetization appears in high purity  $YBa_2Cu_3O_{7-\delta}$  single crystals for a parallel to the layers applied field. For these magnetic oscillations few things are yet known. Our results over an extended field and temperature regime show that they are actually evidence of the two-dimensional vortices existing at low enough temperatures. We demonstrate that the oscillations are a reliable and accurate tool for extracting crucial parameters in the vortex matter study. And, surprisingly, our data reveal that for a parallel field, the temperature dependence of the magnetization contradicts the present theoretical expectations.  $[$ S0163-1829(99)13621-X $]$ 

### **I. INTRODUCTION**

The detection of the locked-in vortex state with the theoretically predicted, accompanying, intrinsic pinning has been the focus of a great deal of experimental interest. First an anomaly in torque measurements was observed in an untwinned YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystal by Farrell *et al.*<sup>1</sup> and was attributed to a transition to the locked-in state.<sup>2</sup> A similar anomaly was observed in single crystals of  $Bi_2Sr_2CaCu_2O_8$ by Steinmeyer *et al.*<sup>3</sup> and of  $Tl_2Ba_2CaCu_2O_8$  by Chung, Chaparale, and Naughton.<sup>4</sup> On the other hand, careful transport measurements in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> exhibited an abrupt, impressive drop of resistivity for fields applied in the *ab* plane and flux motion geometry vertical to the layers,  $5.6$  due to intrinsic pinning. Elegant vector magnetization measurements<sup>7,8</sup> proved also to be useful in detecting the lock-in transition, by monitoring the accompanying transverse Meissner effect.

In magnetic hysteresis measurements, however, intrinsic pinning appeared much later, in the pioneering measurements of Oussena *et al.*<sup>9</sup> as a unique sequence of magnetization ''jumps'' or ''oscillations,'' the lock-in oscillations as we have chosen to call them. Nevertheless, these first observations are still far from being fully and detailed analyzed; since then clear cut, sharp oscillations have neither appeared in the literature nor been studied. Demagnetizing effects, orientation problems or, as we shall show, problems of sample quality, conspire against the observation of lock-in oscillations.

As a result, the signature of intrinsic pinning in magnetic hysteresis measurements is still virtually unexplored and many questions remain unanswered. The *exact* mechanism that produces the lock-in oscillations has yet to be fully presented, analyzed, and mapped on the experimentally observed magnetic hysteresis shape. And, although theoretically the problem seems trivial and the answer well documented,<sup>10</sup> experimentally it is still far from certain in *which* way changes in a parameter of the vortex system as critical as temperature is, will affect the magnetic oscillations or even the measured critical currents. Finally and importantly, it remains open for elucidation in what extend we can benefit from the study of lock-in oscillations in exploring the mixed phase and how magnetic hysteresis measurements can actually compensate for intrinsic weaknesses of the other techniques used in the observation of the locked-in state.

Indeed, torque measurements are unable to give any essential information on the dynamics that govern the behavior of locked-in vortices. Even their apparent use in finding out the angular boundaries of the locked-in state can be restricted due to thermal fluctuations and extrinsic pinning effects which can mask or extinguish the transition.<sup>11,12</sup> On the other hand, in the case of magnetotransport studies one has to take into account that for applied fields parallel to the layers the resulting vortices (known as Josephson vortices or Josephson strings) experience a quasi-two-dimensional  $(2D)$  state<sup>10,13</sup> at low enough temperatures, below a threshold value *T*\*  $\sim$  80 K. It is therefore evident that the limitation of transport measurements to high temperatures, above 80 K, makes the study of the quasi-2D regime and consequently the detection of the 2D to 3D transition<sup>10,13</sup> temperature  $T^*$ , impossible. Furthermore, in interpreting a transverse Meissner effect one has to be aware of possible electrodynamic and not thermodynamic (vortex lock-in) origins.<sup>8</sup> In any case, these techniques, for different reasons, are not in a position to enlighten us about the behavior of the 2D Josephson strings system or verify the existing theories for the vortex lattice structure and behavior in the quasi-2D regime.

A major step forward to fill this extended gap of knowledge is made in the present work. After reiterating the unique up to now presented results of lock-in oscillations, we go further by fully and analytically describing, for the first time, the underlying mechanism; in addition we explore the effect of an in-plane rotation of the magnetic field in the observed magnetic oscillations. Subsequently, we illustrate how random point disorder can affect the magnetic oscillations, by altering the effectiveness of intrinsic pinning. We compare

directly magnetic hysteresis measurements in the locked-in state with torque magnetometry measurements, demonstrating that lock-in oscillations can be an accurate, straightforward and powerful tool for extracting parameters vital in vortex matter studies.

But even more surprisingly, we reveal the temperature dependence of the measured magnetization for applied magnetic fields parallel to the layers. We clearly show that in strong contradiction to theoretical expectations the critical current *does not* decrease monotonically with temperature.

### **II. EXPERIMENTAL**

The single crystals investigated were grown by a conventional self-flux method, using Y-stabilized zirconia crucibles, $14-16$  a method known to give crystals of extremely high purity.<sup>16–18</sup> The high quality of the crystals investigated is further demonstrated by the first observation of a new transition line in the vortex phase diagram of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, a result of the existence of an extremely weakly pinned, quasiperiodical, vortex lattice phase.<sup>19</sup> Samples are oxygenated for 10 days at 500 °C in flowing oxygen and quenched at room temperature. The procedure yields crystals with an oxygen content  $7-\delta=6.91$ , according to the existing diffusivity studies,<sup>20</sup> and a high critical temperature  $T_c = 93.6 \text{ K}.$ The samples have a sharp superconducting transition, with magnetic width  $\Delta T_c$ <0.3 K; this is defined as the temperature range over which the zero field cooled magnetization, in a field of 0.1 mT, varies from 10 to 90%. Crystal DT0 has dimensions  $1.45\times1.2\times0.125$  mm<sup>3</sup> and a mass of 1479  $\mu$ g, while crystal DT3 is  $1.69 \times 1.08 \times 0.091$  mm<sup>3</sup> and weights 1129  $\mu$ g. Crystal thickness is estimated using the mass and the theoretical density of 6.8  $g/cm<sup>3</sup>$ . The crystals are detwinned by applying about 50 Mpa of uniaxial stress at 550 °C in air for 30 min and then reoxygenating for one day at 500 °C in flowing oxygen.<sup>21</sup> The procedure is highly successful and gives detwinned samples for which polarized light microscopy revealed a surface fraction of misaligned phase of much less than  $1\%$ .<sup>22</sup>

Magnetic hysteresis measurements were carried out on a 12 T vibrating sample magnetometer in the University of Southampton. Samples were first zero field cooled at the desired temperature and then subjected to an applied magnetic field  $H_a$ . Unless otherwise specified, in our measurements the direction of  $H_a$  is in the plane defined by either the *a* axis or the *b* axis and the normal to the *ab* plane. The angle that  $H_a$  makes with the basal plane is measured with the accuracy of our setup,  $\theta$ <1°. Measurements were performed with a constant sweep rate of 5 mT/sec. Torque magnetometry measurements were performed in a homemade 4 T torque magnetometer in the University of Paris-Sud in Orsay.23–25 The torque on the sample was measured by rotating the magnetic field in respect to one of the major crystalline axes. A capacitive method was used, in which the deflection of the capacitance electrode gives the corresponding torque on the sample. $^{23,25}$ 

## **III. RESULTS AND DISCUSSION**

In the locked-in state the fishtail shape of the magnetic hysteresis changes fundamentally.<sup>9</sup> Shown in Fig. 1 are the



FIG. 1. The measured magnetization for an applied field  $H_a$  in the *ab* plane, increasing up to 12 T and at the indicated temperatures, for sample DT0. (a)  $H_a||a$  axis, (b)  $H_a||b$  axis.

magnetization curves at different temperatures up to *T*  $=60$  K for crystal DT0, in an increasing applied field up to  $H_a$ =12 T. The applied field is parallel to one of the principal axis of the *ab* plane (each separate case is indicated on Fig. 1); in order to see the effects clearly we shifted the curves along the vertical axis, a procedure that does not qualitatively affect the signal. It is clear that the magnetic isotherms exhibit a strong oscillatory behavior, with oscillations whose periodicity increases with field. As seen in Fig. 1, and below we will illustrate in much more detail, the maxima of these oscillations are temperature independent.

Here we give a full account of the mechanism, pictured in Fig. 2, comparing the theoretical predictions to the experi-



FIG. 2. Schematic representation of the transition between two successive commensurate states,  $n=4$  and  $n=3$ , which produces the observed lock-in oscillations: (a) Commensurate state  $n=4$ , (b) vortices between planes are only weakly pinned, (c) formation of kinks and antikinks: domains with  $n=4$  and  $n=3$ , and (d) commensurate state  $n=3$ .



FIG. 3. Structure of the equilibrium, compressed, Josephson vortex lattice with ratio  $a/l = 2\gamma/\sqrt{3}$ , i.e., the magnetically observed case for a parallel to the layers applied field. Dark areas represent the cross section of the Josephson cores.

mental results as these are mapped in Fig. 1. The anisotropic London model predicts for nearly parallel fields a hexagonal vortex lattice compressed along the *c* axis and expanded in the plane direction of the crystal, $13,26$  with a field independent ratio  $a/l = 2 \gamma/\sqrt{3}$  [Fig. 2(a)], where *a* and *l* stand for the average vortex distance parallel and perpendicular to the *ab* plane, respectively, see also Fig. 3. The vortex lattice with the *a*/*l* ratio predicted by the anisotropic London model is a state of minimum vortex-vortex interaction energy. This state in the layered cuprate superconductors will be commensurate with the order parameter modulation along the *c* axis, for certain only values of the applied field  $H_n$ . For  $H_a$  $H_n$ , it is  $l = nd$ , where  $n = 1,2,3,...$ , and the cores of the Josephson strings (Josephson cores or Josephson nuclei), members of the compressed hexagonal lattice, lie in between the layers. This is the case in the dips of the magnetic hysteresis  $(e.g., points A and E in Fig. 1).$ 

Increasing  $H_a$  but staying near  $H_n$ , any changes in the vortex density can be accommodated with motion of the cores parallel to the layers  $[Fig. 2(b)].$  There are only two factors opposing such a guided motion: firstly the repulsive vortex-vortex interaction which initially, for the commensurate values of *a*, is expected to be minimum and secondly the pinning of Josephson vortices by extrinsic defects. Taking into account the absence of a normal core, one expects Josephson strings to be much less effectively pinned by extrinsic defects. This was explicitly shown by Blatter et al.,<sup>10</sup> who demonstrated the reduction of the elementary pinning force acting on a Josephson vortex, compared with the force acting on an Abrikosov vortex, by a factor of approximately  $(\xi_{ab}/\lambda_i)^3$ , for both  $\delta T_c$  and  $\delta$ l pinning;  $\xi_{ab}$  is the in-plane coherence length and  $\lambda_j$  the Josephson length,<sup>10,13</sup> the core's length along the layers—see also Fig. 3. We should also consider that for the case of vortices laying parallel to the layers there is a dramatic softening of the component of the shear modulus parallel to the basal plane,  $c_{66}^{\parallel}$ . Using the anisotropic scaling approach, $10$  we have

$$
c_{66}^{\parallel} = c_{66} \varepsilon^3, \tag{1}
$$

where  $c_{66}$  is the shear modulus in the isotropic case while  $\epsilon = 1/\gamma$ ,  $\gamma$  being the anisotropy parameter.

Due to the aforementioned reasons, the motion of the Josephson strings is greatly facilitated for directions parallel to the layers. This explains the extremely weak irreversibility measured for the field lying in the *ab* plane: the scaled to size width of the magnetic hysteresis for transverse geometry  $(H_a||c$  axis) increases up to approximately 30 times when compared to this at the longitudinal geometry.<sup>27</sup> Thus, from all the above, it is evident that any increase of  $H_a$  will lead to a decrease of the vortex spacing *a* and an accompanying enhancement of the mutual repulsion of the strings (points of upward curvature such as  $B$  in Fig. 1).

Nevertheless, in order to proceed from one commensurate state,  $H_{n+1}$  with  $l=(n+1)d$ , to the next,  $H_n$  with  $l=nd$ , changes in *l* have to occur for field values in the intermediate region of  $H_{n+1}$  and  $H_n$ . This is illustrated in Fig. 2(c) and has been a theoretical suggestion long before the observation of lock-in oscillations.<sup>13,28</sup> The increase of  $H_a$  leads to a point where the parallel motion of the cores is prohibited due to strong repulsion applied by the other cores in the same ''channel;'' now it is favorable for the nuclei to actually *cross* the CuO<sub>2</sub> layers by the creation and motion of pairs of kinks and antikinks. A fast reorganization of vortices from the  $n+1$  to the *n* commensurate state begins. In the quasi-2D regime, which is the case for the temperature regime discussed here, the kink-antikink pair is a 2D Abrikosov vortexantivortex pair in the  $ab$  plane.<sup>13</sup> In this incommensurate state, the pair has to face the enhanced pinning interactions present in the  $CuO<sub>2</sub>$  plane. Thus, one expects the effective pinning force to maximize resulting to a peak in the magnetic hysteresis. This is the case in our lock-in oscillations for points as C in Fig. 1.

As it is obvious from the geometry of the problem, $^{13}$  the kink-antikink pair faces a force parallel to the *ab* plane and once it is nucleated, it also moves in the direction parallel to the layers (points of downward curvature such as D in Fig. 1). In this way, the whole vortex line is "transported" to the neighboring interlayer spacing. Finally the vortex system arrives at the new commensurate state and the circle is then repeated  $|Fig. 2(d)|$ .

Note that the above description should be valid only up to a threshold value of the magnetic field

$$
H_0 = \Phi_0 / \gamma d^2, \qquad (2)
$$

where  $H_0 \sim 200 \text{ T}$  in the case of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>. Above  $H_0$ , all the interlayer spaces are occupied and any further increase of  $H_a$  results in a mere further stacking of nuclei in each of the interlayer spaces.<sup>13,29</sup>

Figure 4 illustrates the competitive nature of point disorder and layered structure, critical for the appearance of the lock-in transition. Pictured are the magnetic hysteresis loops for crystal DT0, for two different temperatures 5 and 60 K.  $H_a$  is parallel to the *b* axis. In one case the misalignment from the *ab* plane is  $\theta_1 < 1$ ° while in the other we intentionally created a misalignment of  $\theta_2 \approx 2^{\circ}$ . For  $\theta_1$  lock-in oscillations appear at 60 K, indicating that we are below the critical angle for lock-in  $\theta^*$ . On the other hand, for  $\theta = \theta_2$  at 60 K the absence of oscillations in the magnetic hysteresis verifies that this second angle is above the critical one. As Blatter *et al.*<sup>10</sup> have shown,  $\theta^*$  can be considered either temperature independent, or even weakly increasing with decreasing temperature. Therefore, we expect to observe lock-in oscillations also at 5 K, for  $\theta = \theta_1$ . However, as Fig. 4(b) shows, no oscillations appear at this temperature and the magnetic hysteresis is essentially the same as for the second, misaligned, orientation. At these very low temperatures, even for



FIG. 4. Magnetic hysteresis loops for  $H_a \parallel b$  axis and two orientations  $\theta_1$ ,  $\theta_2$  (see text) for (a)  $T = 60$  K and (b)  $T = 5$  K. FIG. 5. Lock-in oscillations for crystal DT0 and for *H<sub>a</sub>* (a) along

our high purity samples, the pinning energy of the random point disorder background is quite large and, as Tachiki<sup>30</sup> predicted, kinks can be nucleated at the defects. The creation of such kinks destroys the alignment of vortices and spoils the lock-in transition between the layers $13,30$  and hence the absence of oscillations. The purity of the samples is therefore an important factor in the determination of the lock-in transition and the observation of lock-in oscillations.

One has to note that the theoretical prediction of jumps in the magnetization existed long before our observations. They were one of the possible consequences considered of commensurability effects between the interlayer distance and the vortex lattice spacing along the  $c$  axis.<sup>13,28,31</sup> Bulaevskii and Clem<sup>28</sup> estimated, using the anisotropic London model, the field dependence of *l* to be

$$
l = (\Phi_0 \sqrt{3}/2\gamma)^{1/2} B_a^{-1/2}
$$
 (3)

with  $B_a = \mu_0 H_a$ . Taking into account, next, the existence of the layered structure and incorporating their results in the Lawrence-Doniach (LD) model, they predicted that the transition from one commensurate to another commensurate state by jumps in *l* (and consequently<sup>32</sup> in *a*), is accompanied by jumps in the measured magnetization.

The periodicity that characterizes the layered structure and the succession of the commensurate states, together with the field dependence of  $l$  as given by Eq.  $(3)$ , are the reasons for the periodicity of the magnetization in the  $H_a^{-1/2}$  space, as depicted in Fig. 5. Data are for DT0, at temperatures of 30, 40, 50, and 60 K with  $H_a$  applied parallel to the *a* and *b* axes and increasing (the data corresponds to these of Fig. 1). Again the curves have been shifted vertically for clarity. We observe a remarkable periodicity of the lock-in oscillations with their period being the same for all temperatures. The temperature independence of the periodicity of the lock-in



the *a* axis, (b) along the *b* axis.

oscillations and of the positions of the magnetization maxima and minima is in agreement with the case of pinning caused by matching of the perpendicular vortex lattice spacing *l* with the layered structure.

We should mention that the magnetic loops for  $T \ge 60$  K display some additional characteristics, namely, a slight shift of lock-in oscillations towards lower fields, which we discuss in detail below, and the appearance of additional peaks at high fields (e.g., between 0.345 and 0.414  $T^{-1/2}$  for  $H_a||a$ axis). These additional peaks are observed at field values comparable to the VSM's accessible range, i.e., around 10 T  $(Fig. 1)$ , which makes their study problematic. We now know that they are not a side effect due to the interaction of the Josephson nuclei with the chains, since they are observed for any orientation of the applied field within the *ab* plane. Though one could speculate that at this point other regular vortex structures which do not have minimal free energy (with  $a/l$  ratio other than  $2\gamma/\sqrt{3}$ ) become energetically favorable, in the field range we can access these extra peaks appear only when increasing the magnetic field.

Since the observed oscillations are a direct consequence of the interaction of the locked-in lattice of Josephson vortices<sup>10,13</sup> with the periodic pinning structure that the layers represent, it is natural to expect that their measurement would allows us to probe the nature of the lattice and the mechanism of vortex motion at the low-temperature quasi-2D regime.

Indeed, based on the lock-in oscillations we can monitor and map the position of the Josephson cores along the *c* axis of the sample in a very simple and accurate way, for every value of the applied magnetic field. From Eqs.  $(2)$  and  $(3)$  it is straightforward to show that

$$
\left(\frac{\sqrt{3}H_0}{2H_n}\right)^{1/2} = n.
$$
\n<sup>(4)</sup>

Taking, for example, the data for crystal DT0 (pictured in Fig. 1), we find one of the commensurate states at  $T$ = 50 K, for  $H_a$  parallel to the *a* axis, at  $H_n$ = 3.45 T. For this value, Eq.  $(4)$  gives  $n=7.94$  that is, by taking the integer part of the result,  $n=8$ . This means the realised Josephson vortex lattice has a spacing perpendicular to the *ab* plane of *l*  $=8d$ ; thus, at this certain value of the applied field, a buffer zone of seven interlayer spaces separates every occupied one by the Josephson cores. Knowing *l*, it is trivial to find the cores separation distance *a* within the same interlayer spacing.<sup>32</sup> In other words, we can *completely* control the vortex entry and exit in the sample, over an extended magnetic field range, fixing both the number and the mutual separation of vortices, a procedure that can be essential for future applications.

Importantly, as it was first shown in Ref. 9, lock-in oscillations can be used for measuring essential physical parameters of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> as is the out-of-plane and in-plane anisotropy. Indeed, from Eq. (3) and the periodicity of the lock-in oscillations in the  $H_a^{-1/2}$  space, it is straightforward to extract  $\gamma$  as

$$
\gamma = \frac{\sqrt{3}}{2} \Phi_0 \left( \frac{\Delta [B_a^{-1/2}]}{d} \right)^2.
$$
 (5)

In Eq.  $(5)$ , one has to take into account that in each period of the magnetization oscillations  $\Delta[B_a^{-1/2}]$ , *l* is changing by *d*. Using the reported value at  $T=120$  K of  $d=1.164$  $\times 10^{-9}$  m (Ref. 33),  $\Phi_0 = 2.067 \times 10^{-15}$  Vs and that for crystal DT0  $\Delta[B_a^{-1/2}]$  is equal to 0.069 and 0.065  $T^{-1/2}$  for the direction of the applied field parallel to the *a* and *b* axes, respectively, we end up with

$$
\gamma_{cb} = \sqrt{\frac{m_c}{m_b}} = 6.3 \pm 0.1
$$
,  $H_a || a \text{ axis},$  (6)

$$
\gamma_{ca} = \sqrt{\frac{m_c}{m_a}} = 5.6 \pm 0.1, H_a || b \text{ axis.}
$$
 (7)

The difference of the anisotropy factors  $\gamma_{cb}$  and  $\gamma_{ca}$  is expected from the observed difference of the periodicity of the lock-in oscillations for  $H_a||a$  axis and  $H_a||b$  axis (Fig. 5) and verifies previous measurements of in-plane anisotropy in  $YBa_2Cu_3O_{7-\delta}$ . Indeed, Dolan *et al.*<sup>34</sup> using the Bitter decoration technique obtained a factor  $\gamma_{ab}$  ranging from 1.11 to 1.15. In our case  $\gamma_{ab}$  is estimated as the ratio of  $\gamma_{cb}/\gamma_{ca}$  and is equal to  $1.13 \pm 0.04$  (Ref. 35).

On the other hand, the values of the out-of-plane anisotropy parameter reported in the literature until now for  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$ , by means of various techniques, are scattered between  $3-10$  (Ref. 36–40). One of the techniques for extracting  $\gamma$ , suffering by fewer ambiguities, has proved to be measurements of torque magnetization.<sup>41</sup> Based on torque measurements, reports in the literature<sup>40,41</sup> give for  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$  single crystals of similar critical temperature with the ones presented here  $(T_c > 90 \text{ K}$ , near the optimum doped regime) values of  $\gamma$  between 4–10.

While our out-of-plane and in-plane  $\gamma$  values, extracted from lock-in oscillations, are in good agreement with all the aforementioned cases, we went further and directly compared the anisotropy parameters extracted from lock-in oscil-



FIG. 6. The measured torque for sample DT3, for the indicated temperatures and  $H_a$  rotated in (a) the *ca* plane and (b) the *cb* plane. Angles are measured from the *c* axis. Symbols represent the experimental data and lines the theoretical fits, using Eq.  $(9)$  (see text).

lations with those extracted with a conventional method, namely, torque magnetometry. For this reason we carried out torque measurements on one of the crystals which exhibited lock-in oscillations, crystal DT3.

As Kogan<sup>42</sup> has pointed out, in high- $T_c$  oxides the tendency of the screening current loops to flow preferentially close to the *ab* plane has an important consequence: the magnetization has a perpendicular to the applied field component. The result is the existence of an anisotropy torque  $\tau$ given by

$$
\frac{\tau(\phi)}{V} = |\mathbf{M} \times \mathbf{H}_a|.
$$
 (8)

*V* is the volume of the sample and  $\phi$  is the angle between the external field  $H_a$  and the *c* axis. For temperatures close to  $T_c$ , by an anisotropic London treatment, Eq.  $(8)$  can be translated to a more practical relation, yielding the angular dependence of the reversible torque<sup>42</sup>

$$
\frac{\tau(\phi)}{V} = \frac{\Phi_0 H_a \sin(2\phi) (\gamma^2 - 1)}{64\pi^2 \lambda_{ab}^2(T)\epsilon(\phi) \gamma} \ln \left( \frac{\beta H_{c2}^{\perp}(T)\gamma}{H_a \epsilon(\phi)} \right), \quad (9)
$$

where  $\beta$  is a constant of order unity,<sup>42</sup>  $H_{c2}^{\perp}(T)$  the upper critical field measured along the *c* axis, perpendicularly to the layers,  $\lambda_{ab}$  the in-plane penetration depth, and  $\varepsilon(\phi)$  $= \sqrt{\sin^2 \phi + \gamma^2 \cos^2 \phi}$ . As Farrell has demonstrated,<sup>1</sup> in  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$  the above equation is valid for temperatures  $T \ge 80$  K; there is also a field requirement,<sup>42</sup> namely,  $H_a$  $\gg H_{c1}$ . In our measurements we worked in temperatures *T*  $>87$  K with an applied field  $H_a=1$  T, satisfying both conditions.

Figure 6 shows torque data for sample DT3. In the case of



FIG. 7. The values for the out-of-plane anisotropy parameters  $\gamma_{cb}$  and  $\gamma_{ca}$  as extracted by fitting the experimental data of Fig. 6 with Eq.  $(9)$  (see text).

Fig. 6(a)  $H_a$  was rotated from the *c* axis to the *a* axis, while in Fig. 6(b) to the *b* axis. For each orientation  $\tau(\phi)$  was measured at a number of different temperatures, in the  $[0^{\circ},]$  $90^\circ$  interval. The measurement regime was completely reversible. To eliminate the contribution arising from both the sampleholder and the sample above  $T_c$  we subtracted from the raw signal the measured torque in the same applied field but in the normal state, at  $T=99$  K.

To deduce the anisotropy, we followed the standard procedure and fitted each of the experimental  $\tau(\phi)$  curves pictured in Fig. 6 with the expression of Eq.  $(9)$ , using the usual fitting parameters  $\gamma$ ,  $\lambda_{ab}(0)$ , and  $\beta H_{c2}^{\perp}(T)$ . As clearly seen, Eq.  $(9)$  provides a very satisfactory fit. The resulting values of  $\gamma$ , for both orientations, are shown in Fig. 7. For rotation of  $H_a$  in the *ca* plane, we find<sup>42</sup> an average anisotropy factor  $\gamma_{cb}$ =7.56±0.14 while for rotation in the *cb* plane the average out-of-plane anisotropy is  $\gamma_{ca} = 6.51 \pm 0.22$ . This leads to an in-plane anisotropy  $\gamma_{ab} = 1.16 \pm 0.05$ . On the other hand, for the same crystal, we find the periodicity of the lock-in oscillations to be 0.071 and 0.063  $T^{-1/2}$  for  $H_a||a$  axis and  $H_a \parallel b$  axis, respectively (see also Fig. 8). From Eq. (5) we estimate the anisotropy factors to be correspondingly,  $\gamma_{cb}$  $=6.57\pm0.20$  and  $\gamma_{ca}=5.24\pm0.20$ , i.e., an in-plane anisotropy of  $\gamma_{ab} = 1.22 \pm 0.06$ . The results are in reasonable agreement with those obtained by torque with the maximum deviation from them being in the case of  $\gamma_{ca}$ , approximately 14%. It is interesting to note that this small deviation may point towards a possible temperature dependence of  $\gamma$ . The slightly increased  $\gamma$  values come from torque, which was employed in the temperature regime around 90 K, whereas we extract  $\gamma$  from lock-in oscillations at much lower temperatures. In previous theoretical reports<sup>43</sup> a temperaturedependent anisotropy has been already proposed; it can be directly attributed to coupling of the superconducting layers due to proximity effects. A similar temperature dependence of  $\gamma$  for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals with  $T_c$ =91 K was found by Pugnat et al.,<sup>44</sup> from vector magnetization measurements. However, further systematic studies are needed in order to carefully examine this possibility.45

Thus, lock-in oscillations provide an alternative, reliable and, especially, direct way of estimating the in-plane and out-of-plane anisotropy factors for  $YBa_2Cu_3O_{7-\delta}$ . Due to the



FIG. 8. Lock-in oscillations for crystal DT3, with *Ha* in the *ab* plane and  $(a)$  parallel to the  $a$  axis,  $(b)$  parallel to the  $b$  axis, and  $(c)$ in an angle  $\vartheta$  ~41° with the *b* axis.

great impact of the anisotropy parameters on vortex dynamics (for example, their devastating influence on the elastic modulii), it is obvious that lock-in oscillations can be extremely useful in extracting valuable information on a variety of other research problems  $[e.g.$  the  $(Abrikosov)$  vortex matter phase diagram]; this is particularly true in  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$ , where the additional complication of the inplane anisotropy can introduce effects, leading to a rescaling of the vortex-vortex elasticity.46

In Fig. 8 we show lock-in oscillations for an applied field oriented away from one of the major axes in the basal plane. Data are for sample DT3, at 60 K and  $H_a||ab$  plane, at three different orientations: for the applied field along the two principal axes  $(a \text{ axis and } b \text{ axis})$  of the basal plane and at an angle of  $41^{\circ} \pm 2^{\circ}$  with the *b* axis. For the last orientation of the applied field, tilted at an angle  $\vartheta$  (in our case  $\vartheta$ =41°) from the *b* axis within the *ab* plane, and the geometry of the flowing supercurrents, the relevant effective mass will be an average of  $m_a$  and  $m_b$ :<sup>42</sup>

$$
m(\vartheta) \equiv m(41^{\circ}) = m_a \cos^2 \vartheta + m_b \sin^2 \vartheta. \tag{10}
$$

This, in conjunction with Eqs.  $(6)$  and  $(7)$ , leads to an anisotropy factor  $\gamma_{41}$ :

$$
\gamma_{41} \cong \sqrt{\frac{m_c}{m(41^\circ)}} = \sqrt{\frac{\gamma_{ca}^2 \gamma_{cb}^2}{\gamma_{cb}^2 \cos^2 41 + \gamma_{ca}^2 \sin^2 41}}.
$$
 (11)

Taking into account that, as mentioned before, the values of the anisotropy parameters for sample DT3 as extracted from the oscillations are  $\gamma_{cb} = 6.57 \pm 0.20$  and  $\gamma_{ca} = 5.24 \pm 0.20$ , from Eq. (11) one finds that  $\gamma_{41} = 5.71 \pm 0.15$ . On the other



FIG. 9. Lock-in oscillations for three different angles between  $H_a$  and the *ab* plane, at 60 K, for DT0 and  $H_a \parallel b$  axis. Angles  $\theta_1$ and  $\theta_2$  are below the critical angle for vortex lock-in  $\theta^*$ ; angle  $\theta_3$  is above it.

hand, directly from lock-in oscillations now  $(Fig. 8)$ , we find the periodicity for the angle of  $(H_a, b \text{ axis})=41^\circ \pm 2^\circ$  to be 0.067  $T^{-1/2}$ , i.e., using Eq. (5)  $\gamma_{41} = 5.95 \pm 0.20$ , in good agreement with the expected result.

Since, according to what we mentioned before, the periodicity of the lock-in oscillations depends only on the characteristics of the layered structure, it should be unaffected by the angle of the applied field with the *ab* plane, as long as this remains below the critical angle  $\theta^*$ . Indeed, Fig. 9 illustrates magnetic hysteresis loops at 60 K, for crystal DT0, with  $H_a$  parallel to the *b* axis and at three different angles with the *ab* plane. Angles  $\theta_1$  and  $\theta_2$  are all less than 1°. In all these measurements lock-in oscillations are present. We also created a misalignment between the sample and the layered structure of  $\theta_3 = 2^\circ \pm 0.5^\circ$ ; as seen in Fig. 9, for this angle the oscillations disappear, indicating the existence of a critical angle of the order of 1°. Furthermore, as Fig. 9 clearly shows, in the locked-in state the position of the maxima and minima of the oscillations are independent from the orientation of the applied field.

An astonishing oddity of the magnetic lock-in oscillations is revealed in Fig. 10. Figure 10 exhibits the temperature development of the lock-in oscillations for crystal DT3, with  $H_a$  along the *a* axis, at an angle  $\theta$  smaller than 1° from the *ab* plane. Measurements for temperatures up to 80 K are pictured.

Between 30 and 40 K the width of the hysteresis  $\Delta m$  is decreasing with temperature; the oscillations are weak. They appear, with a small amplitude, at high magnetic fields larger than approximately 6 T. As demonstrated before, at lower temperatures the pinning energies of the random point disorder prevent vortices from being locked between the  $CuO<sub>2</sub>$ planes; kinks consisting of 2D vortex pancakes with normal cores are created in the *ab* planes and pinned by point defects. $30$  As a result, the oscillations are expected in this case to be either absent or weak.

In the temperature interval  $42-52$  K two are the major findings. First, we observe an enhancement in the amplitude of the oscillations. This is expected: having moved away from low temperatures, in this temperature regime, the creation of kinks at the point defects is less favorable due to the



FIG. 10. Temperature development of the lock-in oscillations in the range 30–80 K, for crystal DT3 and  $H_a||a$  axis.

depression of the elementary pinning energies. Therefore, an increased number of vortices becomes now locked-in. This increased number of locked-in vortices can account for the increase in the amplitude of the oscillations.

But the most unexpected finding is that, simultaneously, the total width  $\Delta m$  of the magnetic hysteresis actually *increases* with temperature. This experimental result comes in straight contradiction with the existing theoretical reports<sup>10</sup> which predict for  $\Delta m$ , in the case of a parallel to the layers applied field, the usual decrease with temperature. Indeed, Fig. 11 depicts the temperature dependence of the width of the magnetic hysteresis  $\Delta m$  (proportional to the critical current) for two constant applied fields of 3.3 and 8.4 T. We verified this result for all crystals that give lock-in oscillations and for both orientations of the applied field within the *ab* plane, i.e., parallel to both principal axes *a* and *b*.

For the increase of the total width of the hysteresis, one has to consider another possible effect. For a parallel to the layers applied field  $H_a$ , due to the easy entrance of vortex cores between the layers, the irreversibility measured by magnetization is weak and reflects the extrinsic pinning, i.e., pinning of the Josephson cores from the extrinsic defects.<sup>13</sup> As mentioned before, in the absence of a normal core, this pinning source for Josephson vortices is also weak. Feinberg theoretically predicts the possibility that raising the temperature this source of pinning can actually *strengthen*, due to the



FIG. 11. Temperature dependence of the width of the magnetization  $\Delta m$  at two different applied fields  $H_a = 3.3$  and 8.4 T, as derived from the raw data of Fig. 10. FIG. 12. A slight shift of the period of the lock-in oscillations at

increased suppression of the order parameter in the Josephson core, leading to an increase of the measured magnetic hysteresis width. Indeed, the suppression of the order parameter in the core has been calculated by Blatter *et al.*,<sup>10</sup> who found that it increases with temperature proportionally to  $\xi_{ab}^2$ . Thus, our results are actually the first experimental evidence in single crystals of the  $J_c$  anomaly hinted by Feinberg.<sup>13</sup> In addition, one has to take into account that the layered structure offers now to an increased number of locked-in strings an enhanced screening of thermal fluctuations and effectively makes these fluctuations 2D (vortex waving between two adjacent layers), restricting dissipation.

Increasing the temperature further,  $\Delta m$  is reduced. At these elevated temperatures, the energy to create kinks is reduced.<sup>10,13</sup> Thermal energy is now sufficiently strong to spontaneously activate vortex kink-antikink pairs, introducing another important mechanism of dissipation.<sup>13,47</sup> Experimentally, the oscillations are no longer observed above 80 K. Taking this temperature<sup>1,8</sup> as the temperature  $T^*$  at which vortices enter the quasi-3D regime and the lock-in transition is not any more realizable due to the increase of the coherence length across the layers  $\xi_c$  above the  $d/\sqrt{2}$  limit, one can extract  $\xi_c(T=0)$  from the temperature dependence of the transverse coherence length $10,13,47$ 

$$
\xi_c(T) = \xi_c(0) \left( 1 - \frac{T}{T_c} \right)^{-1/2}.
$$
 (12)

For  $T=T^*$  we have  $\xi_c(T^*)=d/\sqrt{2}$ . In YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> with the spacing between the superconducting layers being equal to  $d=11.64$  Å (Ref. 33), we estimate from Eq.  $(12)$  that  $\xi_c(0)$ =3.14 Å. The result agrees with estimations of  $\xi_c(0)$ by other methods. For example, estimating  $\xi_c(0)$  from the thermodynamic critical field  $H_c$  derived from specific heat measurements, results in a value of  $\xi_c(0) = 3.2$  Å (Refs. 48). Welp *et al.*<sup>49</sup> using *dc* magnetization measurements have found  $\xi_c(0)=3$  Å.

Finally, Fig. 12 offers a closer look at the data presented in Fig. 10 for crystal DT3 and  $H_3||a$  axis, at elevated temperatures. As mentioned before the magnetization isotherms at higher temperatures,  $T > 50$  K, display a slight shift of the



elevated temperatures. Data are for sample DT3, with  $H_a||a$  axis.

lock-in oscillations at lower fields. This is because at elevated temperatures and as the temperature rises, the periodic pinning potential of the layered structure weakens<sup>10,31</sup> and the nucleation of pairs of kinks, necessary condition for the crossing of the layers, is now favorable for the Josephson cores at slightly lower fields. Note that in a recent numerical study of the lock-in oscillations, Ichioka<sup>50</sup> verified this slight lowering of the critical fields  $H_n$  with the increase of temperature.

#### **IV. CONCLUSIONS**

In conclusion, we have presented an extended study of the magnetization for a large range of magnetic fields and temperatures in the longitudinal geometry, i.e., with the external field in the basal plane of the samples. We have demonstrated that for fields applied parallel to the layers, the observation of lock-in oscillations in the magnetic hysteresis loops of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> can be a powerful tool for the study of the low-temperature quasi-2D regime.

The method offers considerable advantages over other techniques, especially since it covers extensively the lowtemperature regime giving us access to valuable information in the most straightforward, direct and uncomplicated way. We have showed that lock-in oscillations give us a rare insight of the existing Josephson strings: we can accurately map the position of vortices in the sample and picture the realized vortex lattice.

But more importantly, we can reliably extract fundamental parameters, such as the out-of-plane and in-plane anisotropy, the in-plane coherence length and the temperature *T*\* where the Josephson vortex dimensional crossover from the quasi-2D to the 3D region occurs.

We have also illustrated the competing nature of point disorder and layered structure. The first tends to destroy the lock-in transition and quench lock-in oscillations, by offering to vortices favorable locations for the creation of kinks and spoiling their perfect alignment between the layers, even for an external field applied at an angle below the lock-in critical angle. Thus, the need for high purity samples is essential in order to observe the magnetization oscillations.

Finally, we have presented the temperature dependence of

lock-in oscillations up to a temperature of 80 K. Our results show a surprising nonmonotonic temperature dependence of the width of the magnetization curve and consequently of the measured critical current: these two—proportional quantities increase with temperature, at intermediate temperatures. Our results can be explained by considering the interaction of Josephson vortices with extrinsic pinning centres, as random point defects, together with the reduced thermal dimensionality of the locked-in vortices.

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