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Repetition of the disordered pattern in successive solidifications of vortex matter observed by Bitter decoration

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Repeated images of the vortex structure in $YBa_2Cu_3O_{7-\delta}$ single crystals have been obtained, using the Bitter technique. By removing the iron dots between each decoration experiment, it is possible to image the vortices in different cooldown runs done under the same field and temperature conditions. The images show that these different realizations of a disordered vortex state found in twinned crystals are very similar at long range and differ only in small-scale detail. This is unusual behavior in glassy systems, where expectations are that successive configurations will differ over many scales. In contrast, ordered vortex crystals in clean samples can form with different orientations in different runs, therefore differing at long ranges. Double-sided decorations have been performed in twinned samples, and the correlation between images in both sides of the sample is similar in magnitude but slightly different qualitatively than that found in successive decorations.

Glasses form a large class of materials, from window glass to metallic glasses,[1] polymers, and spin glasses.[2,3] They share a disordered nature,[4] and several common features,[5] however, there is no agreement on a general definition of a glass, and several open questions remain. The glass transition is characterized by ergodic symmetry breaking. Upon cooling, the system remains in a restricted region of phase space with respect to the high-temperature phase. In general, it is assumed that many configurations, i.e., points in phase space, have almost the same free energy, so that in each realization of the glass, the system is equally likely to fall in any region (out of many local minima) in phase space. Recently, the vortices in high-T_c superconductors have been shown to possess glassy characteristics. ‘‘Vortex glass,’’[6] ‘‘Bose glass,’’[7] and ‘‘Bragg glass’’[8] phases have been proposed theoretically and in some instances, found experimentally.[9–11]

In general, vortex glasses are defined experimentally with respect to their dynamical (i.e., flux creep, scaling for I-V curves, etc.), with less emphasis on their structural properties. However, an interesting characteristic of vortex glasses is that it is possible to obtain images of the vortex positions in real space, so that static configurations can be known in detail. Since the early work of Essman and Trauble,[12] decorations by the Bitter technique have been extremely useful for obtaining information on vortex matter.[13–17] We have found that in $YBa_2Cu_3O_{7-\delta}$ (YBCO), Bitter decorations can be repeated on the same sample after cleaning the iron particles of the previous experiment.[16] Therefore, in these disordered vortex systems, a unique opportunity is offered for studying different realizations of an amorphous structure by directly imaging the ‘‘particles’’ involved and obtaining information of the type of ergodic symmetry breaking when passing from the high-temperature to the low-temperature phase.

In the present paper, we report successive decorations in twinned single crystals of YBCO, and we find that the vortices fall almost over the same positions in the first and second decorations. This means that different realizations of the vortex glass are very similar at long range and differ mostly in small-scale detail, being, therefore, relatively close in phase space. In contrast, successive decorations in twin-free regions show an ordered hexagonal vortex lattice where crystalline grains may form in different orientations for each realization of the vortex structure.

Our experiments were performed on YBCO single crystals prepared by growth from the melt as described in Ref. 18. They were fully oxygenated with typical values of T_c around 91.5 K and approximate dimensions $(1\times1\times0.01)\ \mathrm{mm}^3$.

Decorations by the Bitter technique were performed at 4 K in a field of 36 Oe, using a field cooling (FC) procedure. Scanning electron microscope images were obtained at room temperature, and, afterwards, the sample was washed in ultrasound, using isopropyl alcohol. This procedure removes all trace of the iron particles and gives a surface clean enough to allow further decoration. The second decoration was performed at the same nominal field and FC procedure, and new images were taken. We have found that the average number of vortices is the same (within 1%) in images (of around 800 vortices each) taken in different regions of the sample, and the density agrees with that obtained from the average magnetic flux seen in magnetization measurements in similar samples.

Figure 1 shows two successive decorations in a heavily twinned region of one of the samples. The images can be aligned by a micron-size irregularity of the surface, seen in the lower left-hand corner. The vortex structure is disordered, with no trace of hexagonal lattice, but there is a clear orientational order along the twin boundary (TB) direction. In the image, the TB’s run parallel to the vertical direction, and vertical ‘‘columns’’ of vortices can be easily identified. Because the positions of the TB’s are disordered, the distance between vortex columns has large fluctuations around a mean value, and furthermore, in each TB the number of vortices pinned is different. The orientational order could be seen also in Fourier transforms of the images, which show bands, indicating a spread in vortex distances and the orientation due to the TB’s.

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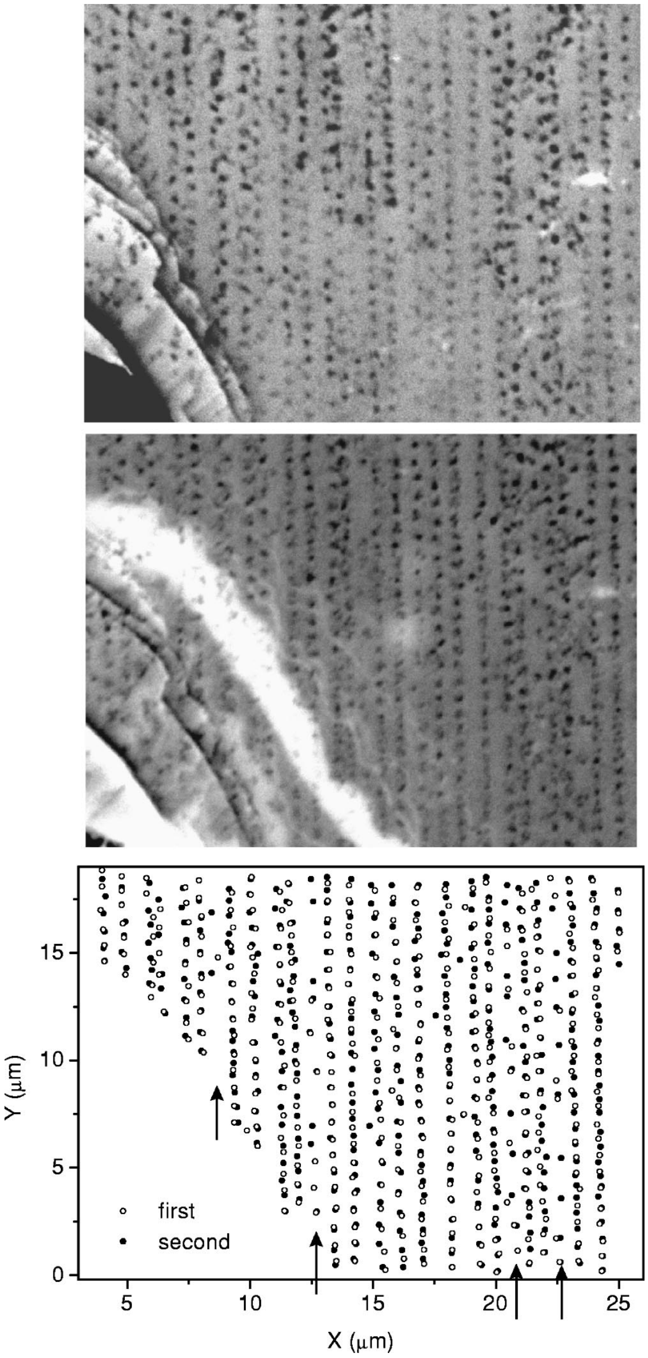

FIG. 1. Upper panel: First decoration, field cooled at 36 Oe. Middle panel: Second decoration, obtained under the same conditions after warming to room temperature and removing the iron particles of the first. The white region is produced by residues left after cleaning. Lower panel: Vortex positions in part of the above images, on the same plot. Arrows show the ''interstitial'' vortices, which do not seem to be pinned to a particular TB.

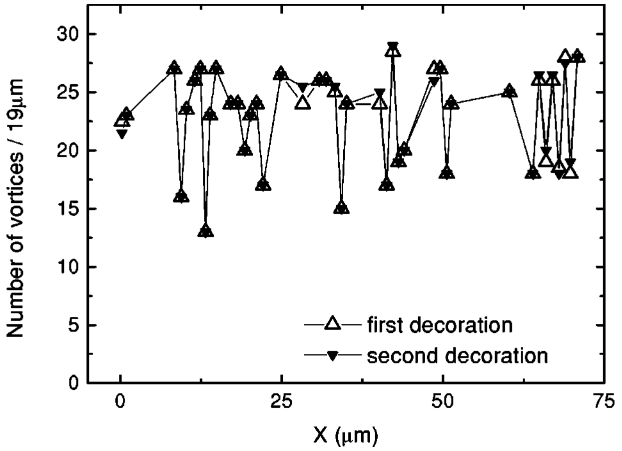

FIG. 2. Number of vortices over a (vertical) distance of 19 μm along a column, vs the (horizontal) column position for two successive images. Up triangles: first decoration. Down triangles: second decoration.

The first question we wish to address is whether the vortices will pin to the same TB for successive decorations. We have counted the number of vortices N (for a fixed length of 19 μm along the column) in each vortex column for approximately 40 TB's, and in Fig. 2, we plot N against the coordinate of the TB for two successive decorations. The vortex column positions coincide within experimental resolution and the number of vortices in some of the columns has small variations when comparing both realizations of the glassy state. The fact that the vortex columns occupy the same random positions (that is, they are pinned to the same twin boundary) and reproduce almost the same density along the TB is one of the main results of this experiment. Although we show in detail only a few images, the same result is obtained in all heavily twinned regions we have checked. It is also found in samples where three successive decorations were made.

On samples that have been detwinned, however, the behavior is different. Figure 3 shows the Delaunay triangulation corresponding to succesive FC decorations at 52 Oe in a twin-free zone, where a surface imperfection can be used to pinpoint the same region of the sample in both images. The hexagonal lattice is seen in both, and Fourier transforms show the hexagonal order, but the orientation of the vortex crystals is not repeated exactly. This can be appreciated in the lower left-hand corner of the images, where the first decoration has a grain boundary, absent in the second. This is not surprising, because it is expected that the energy difference for growth in a different direction is small so that, for example, pinning fluctuations produce different orientations in $Bi_2Sr_2CaCu_2O_8$.[19] Grains that grow in unrelated directions have also been seen in double decorations (where the iron particles are not removed between one decoration and the next) in $NbSe_2$.[20] However, our observation of similar behavior in the hexagonal vortex lattice of YBCO emphasizes the fact that orientational freedom is absent in the disordered vortex phase at the TB's.

We can now look in more detail into the individual vortex positions. The small differences in N seen in Fig. 2 for some TB's is not due to experimental uncertainties, and is small but significant, as can be seen in the lower part of Fig. 1 where an expanded image is shown. The positions of the vortices have been determined by looking for the maximum density in each image, by means of a computer program. Both pictures can be aligned by use of the surface irregularities without further adjustment, and we plot the resultant vortex positions in the same graph using different symbols. It is seen that the coincidence is not perfect, although for a majority of the vortices in the first decoration, it is possible to find a vortex in the second that is closer to it than the

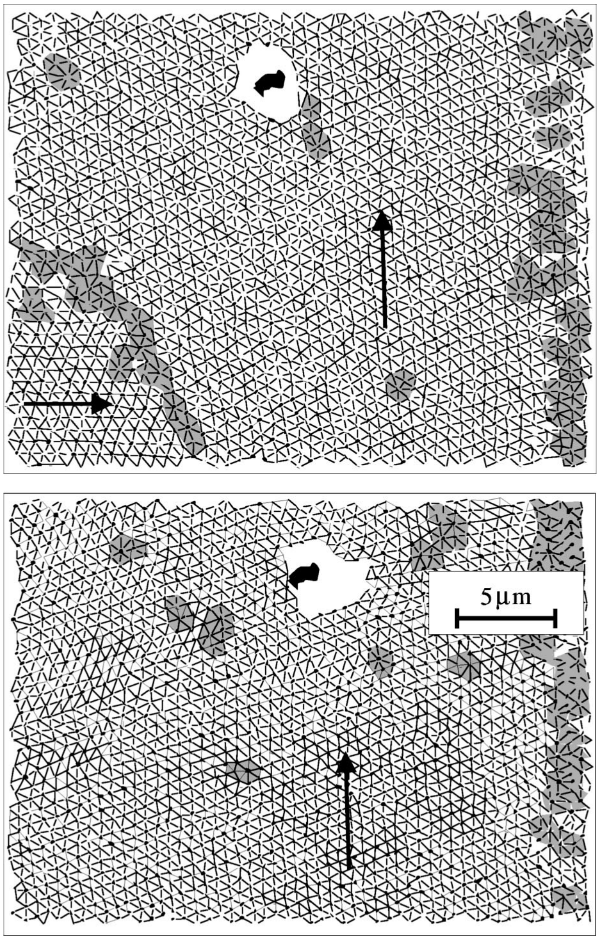

FIG. 3. Delaunay triangulations of two successive FC decorations at 52 Oe in an untwinned sample. Arrows indicate grain orientation. In the upper picture, a grain boundary is apparent in the lower left-hand corner, which is absent in the second decoration (lower picture).

average vortex spacing. Therefore, there is short-range decorrelation between the different realizations of the disordered structure.

To quantify this decorrelation, we have used the rms fluctuation between successive images, defined as

$$\langle D_{1,2}\rangle = \frac{1}{N}\sum_{i=1}^{N}\sqrt{(x1_i - x2_i)^2+(y1_i - y2_i)^2}, \qquad (1)$$

where $x1, y1, x2, y2$ are the coordinates of the vortices in the first and second images, respectively. We find a value of $\langle D_{1,2}\rangle = 0.2\,\mu\text{m}$ averaging over 380 vortices. However, the structure observed is anisotropic, and it is meaningful to ask for the deviation along the twin boundary direction, and perpendicular to it. Defining an rms fluctuation in the same way as in Eq. (1) but considering only the distances along $\langle D_{1,2}\rangle_{\parallel}$ the TB or perpendicular to it $\langle D_{1,2}\rangle_{\perp}$, we find $\langle D_{1,2}\rangle_{\parallel} = 0.18\,\mu\text{m}$ and $\langle D_{1,2}\rangle_{\perp} = 0.065\,\mu\text{m}$. The average vortex spacing along the TB direction is $a_{0\parallel} \approx 0.9\,\mu\text{m}$, so that the difference between successive decorations is, on average, only $\approx 20\%$ of the vortex spacing although $a_{0\parallel}$ has typical fluctuations of around $0.1\,\mu\text{m}$. The fact that $\langle D_{1,2}\rangle_{\perp} < \langle D_{1,2}\rangle_{\parallel}$ shows the importance of the alignment introduced by the TB's.

The average vortex separation observed is close to the value needed to conserve magnetic flux, so it seems that despite the constraints imposed by pinning at the TB's, the vortices can usually find a TB at a convenient distance to fulfill the flux conservation condition, and, furthermore, they pin to the same TB's when the decoration is repeated. The average vortex density is governed by the compression modulus C_{11} of the lattice, which is much stronger than the shear modulus C_{66}, so this would indicate that pinning by the TB's is strong enough to overcome the shear modulus of the vortex lattice, but not the compression modulus. It also seems to indicate that in some regions, the TB's are close enough so that the vortices find a convenient pinning site within the average lattice parameter $a_0 = \sqrt{\phi_0/H} \approx 0.8\,\mu\text{m}$. In some cases, TB's seem to be located farther apart, and we find vortices in what we have called ''intersitial'' positions (see Fig. 1). In the above calculations, we have taken into account only the vortices pinned to the TB's, and disregarded those found in ''interstitial'' positions. In this case, they are only 8% of the total number of vortices averaged, so the quantitative calculation is not significantly altered; however, the qualitative behavior of these few vortices is different. While the number of interstitial vortices is the same (as required by flux conservation), their positions do not coincide in the succesive decorations.

We have also performed a two-sided decoration on another twinned YBCO crystal from the same batch. It was possible to decorate the two sides without heating the sample significantly by using an appropriate geometry. The images of both sides can be seen in Fig. 4, which shows a region near the sample border, where it is possible to identify the region by characteristics of the pattern. It is seen that the TB's go through from one face of the crystal to the other, producing a very similar vortex pattern in both sides. The correlation between two-sided decorations has been studied in $Bi_2Sr_2CaCu_2O_8$ (Ref. 15) and Nb Se_2.[21] A comparison of the different materials is beyond the scope of this paper. We evaluate the rms deviation between side A and side B and find $\langle D_{A,B}\rangle_{\perp} = 0.1\,\mu\text{m}$ and $\langle D_{A,B}\rangle_{\parallel} = 0.14\,\mu\text{m}$. The thickness of the sample is $12\,\mu\text{m}$.

Low rms displacement between images in the two sides is not unexpected, because of the correlated character of the vortex lines,[18] and furthermore, it indicates that vortex positions must be determined by pinning averaged over the bulk of the sample. The elastic constants of the vortex structure and the correlated TB pinning potentials also favor correlation, in competition with thermal fluctuations and possible differences in pinning over the vortex length. Thermal fluctuations should be comparable in the two-sided decoration and successive decorations, but the effect of the elastic constants is absent in the latter, where the rms displacement along the TB $\langle D_{1,2}\rangle_{\parallel} > \langle D_{A,B}\rangle_{\parallel}$. The differences in pinning potential along the vortex length are absent in the former, and the perpendicular displacement $\langle D_{1,2}\rangle_{\perp} < \langle D_{A,B}\rangle_{\perp}$. The fact that disordered structures related by different mechanisms show different types of correlation is to be expected, but it is remarkable that the correlation has similar absolute magnitude in both cases.

The fact that the sample has correlated pinning potentials probably means we are observing a Bose glass phase[7] and the work of Grigera *et al.*[11] in twinned YBCO seems to con-

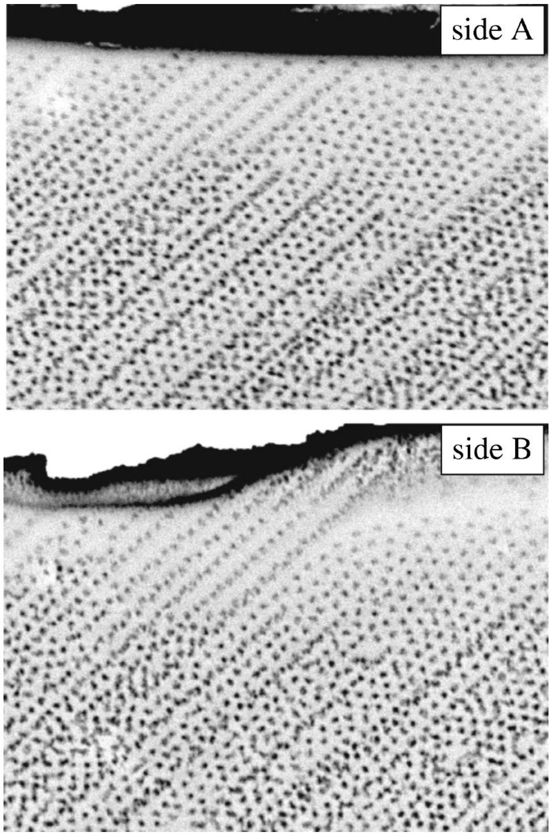

FIG. 4. Two-sided decoration. One of the images has been flipped so that it can be superposed to the other. We can identify the same region of the sample in both images by the characteristic pattern of vortices pinned to the TB's, and using marks in the border as a first rough guide.

firm this. However, we will not discuss in detail which of the vortex glass proposals[6–8] best fits our observations. In any case, the reproducibility of the disordered structure was not explicitly treated in them. It should also be remembered that decorations are done at much lower fields than the transport studies of Grigera *et al.*,[11] and some caution is necessary when extrapolating these results. It is also worth remarking that although the decorations were performed at 4 K, the vortex structure was quenched at a higher temperature.[16,21] We estimate that the quenching temperature is around 86 K from the average flux found in our decorations.[16]

When comparing our results with the expectations in other glasses, the vortex system shows a surprising reproducibility at long range. Numerical simulations of metallic or insulating glasses find different configurations in each realization of the structure.[1,4] For spin glasses, which have quenched disorder as in the vortex system, the Parisi ansatz[1,3] proposes a hierarchical, ultrametric distribution of accessible regions in phase space. Presumably, it would produce real-space distributions differing over a wide range of distance scales. This is not found in our observations. The reproducibility found could be explained if the pinning were so strong that each vortex position is determined much more by the pinning potential than by interaction with its neighbors. Then there could exist a unique configuration that minimizes pinning energy and basically ignores energy increase from vortex interactions, so that the importance of frustration in the system is greatly reduced. In support of this, the images show that the hexagonal order is completely destroyed although the vortex density, i.e., the average lattice parameter, is preserved. An interesting related question is what configuration entropy should be assigned to this sort of glass phase.

In conclusion, we have shown that successive vortex decorations provide unique insight into the reproducibility of the disordered vortex structure in twinned YBCO, and also offer a model glass where structural information can be obtained repeatedly and in detail.

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