Critical Currents and Order-Disorder Phase Transition in the Vortex States of YBa$_2$Cu$_4$O$_8$ with Chemically Introduced Disorder

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A series of YBa$_{2-x}$Sr$_x$Cu$_4$O$_8$ single crystals was measured to study the influence of site disorder on the transition line $H_{ss}(T)$ between quasi-ordered vortex lattice and highly disordered vortex glass, as well as on the maximum critical current density within the glass phase, $j_{c}^{\text{max}}$. When 32% of Ba is replaced by Sr, $j_{c}^{\text{max}}$ is an order of magnitude higher than in the unsubstituted compound. In contrast, the transition field $H_{ss}$ first drops by a factor of about five with a substitution of just 10% of Sr for Ba, and then remains approximately constant for higher Sr contents. Our results indicate that in very clean systems the order-disorder transition is affected very strongly by any crystallographic disorder, while above a certain threshold it is relatively robust with respect to additional disorder. In all substituted crystals $H_{ss}$ monotonically decreases with an increase of temperature.

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I. INTRODUCTION

The vortex matter of cuprate superconductors has a rich phase diagram in the $H$-$T$ plane (see e.g. Ref. 1). Not long after the discovery of the cuprate superconductors it has been established that there are at least two phases, a fluid (liquid or gas) phase at high temperatures and fields and a solid (lattice or glass) phase at low temperatures and fields. It was shown that the transition between these two phases is of first order, at least in low fields near some critical point. 

Recently, it has been shown1,2,4,5,6,7 that there are two distinct solid phases. In low fields the vortices form a quasi-ordered lattice in a so-called Bragg glass,8,9,10,11,12 which is stable against the formation of dislocations. In higher fields they form a highly disordered, entangled solid. The nature of the high field phase is not well understood. It could be anything from a highly viscous fluid to a vortex glass with unbounded barriers against vortex movement. We will simply term it “glass” henceforth.

The high-field glass phase is caused by weak, random and uncorrelated disorder due to point-like defects, such as oxygen vacancies. This type of disorder is present even in the most pure crystals of cuprate superconductors. Strong, correlated disorder due to extended defects, such as twin boundaries or columnar irradiation defects, has got a different influence on the vortex matter and can lead to additional phases, e.g. a “Bose glass” phase, located at low fields. Correlated disorder may not be present in all crystals. In the following, when we simply speak of “disorder”, we are referring to weak random point-like disorder.

The pinning induced by the disorder has two effects on the vortex lattice. Firstly, it localizes vortices by trapping them in low-lying metastable states, hardening the solid. Secondly, it promotes transverse wandering of vortices from their ideal lattice positions. The mechanism of the destruction of the lattice, triggering the transition to the glass phase, is very similar to the mechanism of melting at higher temperature. Similar to the way the lattice looses translational order and melts into a fluid, when thermal fluctuations become of the order of the lattice spacing, it looses translational order and “melts” into a glass, when the disorder-induced line wandering becomes of the order of the lattice spacing. In both cases the stability against the formation of topological defects, such as dislocations, is lost. The massive proliferation of dislocations accompanying the destruction of translational order leads to an entangled configuration of vortices, i.e. topological order is lost as well. In highly anisotropic superconductors, the lattice-glass transition may fall together with a decoupling between the layers, of which there is some evidence in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO). This is not expected in less anisotropic systems, like YBa$_2$Cu$_3$O$_{7-\delta}$ (Y123).

There is considerable evidence that the phase transition between lattice and glass, like the melting transition between lattice and fluid, is of first order and is even a continuation of the melting line.

Since the solid-solid transition is an order to disorder transition with the disordered phase located at high fields, the transition line $H_{ss}(T)$ can be expected to shift to lower fields with the introduction of additional disorder in the crystal structure. Calculations of $H_{ss}$, based on a Lindemann criterion, confirm this expectation. Indeed, there seems to be a correlation between the position of $H_{ss}$ and the purity of a sample, as can be seen by comparing different measurements on Y123 crystals. Also, studies of the effect of electron irradiation on the transition between lattice and glass have found a systematic decrease of $H_{ss}$ with increasing irradiation.
An alternative way to introduce structural disorder into a system is to partially substitute an element of the compound. However, the generally varying level of oxygen within samples, which is difficult to measure and control, constitutes a serious problem. YBa$_2$Cu$_4$O$_8$ (Y124) is a compound very well suited for studying the dependence of $H_{ss}$ on chemically introduced disorder, because in this compound the oxygen content is fixed to 8 per unit cell. Another advantage of Y124 is that twinning, which leads to strong correlated disorder, does not exist in this compound. Finally, Y124 has got an intermediate anisotropy $\gamma \approx 12$, which, as compared to Y123, shifts the glass phase down to fields more easily attainable experimentally. To exclude or at least to minimize concurrent sources of disorder in the system it is advisable to use single crystals for any measurements.

If vortices are not ordered in a lattice, they can better adapt to the local pinning potential. Therefore, critical current densities can be expected to be higher in the glass phase. High critical currents in high magnetic fields are desirable for many applications. Investigations of the disordered solid state and its boundaries are thus very important in view of practical applications. Many irradiation studies found that critical current densities can increase by orders of magnitude upon irradiating a sample. Studies on compounds with chemical disorder have also found increases of $j_c$ and even the development of a second peak.

In this work, we study the influence of a substitution of isovalent Sr for Ba in YBa$_{2-x}$Sr$_x$Cu$_4$O$_8$ on the lattice to glass transition line $H_{ss}(T)$, the irreversibility line $H_{irr}(T)$ and the critical current densities $j_c$. In the paper, we are going to give evidence of an enhancement of the maximum critical current density in the glass phase $j_{c}^{max}$ by more than an order of magnitude due to structural disorder introduced by Sr substitution. The change of the vortex matter phase diagram due to the influence of relatively strong quenched disorder will be presented.

### II. EXPERIMENTAL

Single crystals of Y124, with and without Sr substitution, were grown using a high-pressure technique, the details of crystal growth are reported elsewhere. Single crystals of YBa$_{2-x}$Sr$_x$Cu$_4$O$_8$ with Sr content up to $x = 0.64$ or 32% were obtained. The crystals were checked with EDX and their structure analyzed with Single Crystal x-ray analysis. The transition temperatures $T_c$ of the crystals were determined with a SQUID magnetometer by temperature sweeps (both zero field and field cooled) in an applied field of 1 Oe. All samples chosen for further magnetic measurements had a well-resolved transition with a transition width (10% to 90%) smaller than 2 K (see Fig. 1).

Measurements of the dc magnetization were performed with a Quantum Design MPMS5 SQUID magnetometer, additional measurements were performed with a non-commercial SQUID magnetometer with a sensitivity better than 10$^{-6}$ emu. All measurements were done with the magnetic field applied parallel to the $c$-axis of the crystal and generally the magnetization was measured at fixed temperatures as a function of the external field being swept up and down. After each change of the field, we waited for a time of 5 min before measuring the magnetic moment four times using a scan length of 2 cm. The relatively short scan length was chosen to ensure maximum homogeneity of the applied field and was necessary to ensure a sufficient resolution of all features of interest discussed below. The wait time of 5 min was chosen to avoid effects of the initial relaxation of the field of the superconducting magnet without extending the time of measurements too much. Throughout the paper $H$ denotes the applied field, i.e. no correction of the demagnetization was made. This is not a problem, however, since the magnetization is always much smaller than the external field except in the low field range, which is not the particular interest in this paper.

The critical current density $j_c$ was calculated from the width of the hysteresis loop using an extended Bean model. The irreversibility field $H_{irr}$ was determined as the field where increasing and decreasing field branches of the $M(H)$ hysteresis loop meet, or equivalently as the field where the critical current density vanishes. A uniform criterion of $j_c = 50$ A cm$^{-2}$ was used. For the crystals studied it corresponds to a criterion of in between 4.4 $\mu$emu and 26 $\mu$emu, which is not far from the accuracy limit of our experimental setup.

### III. RESULTS AND DISCUSSION

In the following we will discuss the influence of Sr substitution on disorder and structure, critical current densities, the irreversibility line, and the transition from lattice to glass.
A. Disorder and Structure

Sr substitution increases the disorder in the structure, evidenced for example by a dramatically increased NQR line width. No evidence for an introduction of correlated disorder was found and since twin boundaries are absent in $YBa_{2-x}Sr_xCu_4O_8$ the assumption of the disorder in our crystals to be purely random and point-like seems justified.

The Sr substitution also induces a variation of structural parameters, causing a charge redistribution. Bond valence sum calculations and nuclear quadrupole resonance measurements indicate a transfer of holes from oxygen to copper atoms in the $CuO_2$ planes and from copper to oxygen in the chains. The charge redistribution can explain the increase of $T_c$ upon substituting more than 10% Sr for Ba. The initial decrease of $T_c$ may be due to the increased site disorder.

An important structural modification due to Sr substitution is a decrease of the thickness of the blocking layer $d_b$ separating the superconducting $CuO_2$ planes, linear with Sr content. A substitution of 32% Sr for Ba causes the blocking layer to shrink by about 1%, which leads to an improved coupling between the superconducting planes. Torque measurements indeed indicate a corresponding decrease of the anisotropy $\gamma$.

B. Critical Current Densities

Figure 2a) shows one of the half hysteresis loops measured. The second peak is clearly discernible and dominates $M(H)$. In all crystals, the second peak was observed over the whole temperature range measured, even just 1 K below $T_c$. The form of the hysteresis loop and thus of $j_c(H)$ can be linked to the different phases of vortex matter. In low fields hysteresis is small and the hysteresis loop is rather anisotropic, which indicates that the major source of hysteresis in low fields is not of bulk origin (see also Sec. III C). The origin of the low bulk hysteresis is probably that the vortices are in the lattice phase in small $H$. Weak random point disorder is not effective in pinning an ordered dislocation-free lattice of vortices. The magnitude of the critical current density in low fields gives an indication of the degree of disorder in the system. For our samples, measurements of the critical current density at reduced temperatures of $T/T_c = 0.4$ and 0.7 in fields of 1 and 2 kOe suggest that the disorder is increased monotonically up to the highest substitution level measured, with the biggest rate between $x = 0.4$ and $x = 0.64$.

When, upon rising the field, the lattice is destroyed in the glass state and the vortex system has an additional “dislocation degree of freedom,” individual vortices can much better adapt to local minima in the pinning potential. This results in a higher effective pinning force and thus a higher critical current density $j_c$ and relaxation barrier. Additionally, the entanglement itself was also suggested to increase critical current densities, due to an increased intervortex viscosity. This provided the barriers to flux cutting are high enough. In our case, a steep rise of the magnetization occurs just below the kink field, denoted $H_{ss}$ in Fig. 2a) (see also Sec. III C). The irreversibility field, where field increasing and decreasing branches meet, is denoted as $H_{irr}$. b) Additional parts of hysteresis loops showing the kink region at three different temperatures.

![Figure 2](image_url)

FIG. 2: a) One of the hysteresis loops measured. The direction of the field change is shown by arrows. The onset of the second peak, the second peak maximum and the kink in $M(H)$ between them are, denoted as $H_{ss}$, $H_{2p}$ and $H_{ss}$, respectively. Note that there are small differences between the respective fields between the two directions of field change (see text and Fig. 3a). The irreversibility field, where field increasing and decreasing branches meet, is denoted as $H_{irr}$. b) Additional parts of hysteresis loops showing the kink region at three different temperatures.
lattice-glass transition though, since in the lattice phase dislocations are strongly suppressed while in the glass phase dislocations proliferate, as mentioned above. 

From collective creep theory a rise of the experimentally observable critical current density with the field is expected, as long as elastic creep can be assumed and the bulk pinning is relevant. From the shape of the hysteresis loops shown in Fig. 2 follows that in YBa$_{2-x}$Sr$_x$Cu$_4$O$_8$ the situation is similar to the one in Y123. After rising the field even more, above $H_{irr}$, the vortex matter finally enters the fluid phase, where critical current densities are zero.

The dependence of the hysteresis loop of an unsubstituted Y124 single crystal on defects created by fast neutron irradiation was studied in Ref. 13. Fast neutron irradiation leads to improved pinning properties due to collision cascades (strongly pinning extended defects of spherical shape) and agglomerates of smaller defects. A clearly discernible second peak was observed in the untreated crystal. In the weakly irradiated crystal the second peak was less pronounced and after irradiation to a fluence of $10^{17}$ cm$^{-2}$ it disappeared. At the same time, $j_c$ drastically increased in low to medium fields and the hysteresis loops became more symmetrical.

The effect of fast neutron irradiation to remove the second peak may be understood from the different defect structure. The disorder resulting from the collision cascades is to some extent correlated and fast neutron irradiation can therefore create a Bose glass phase in low fields. Bulk pinning is also strong in the Bose glass phase, and if the irradiation induced correlated disorder is strong enough, the Bose glass phase may well destroy all of the Bragg glass phase. In high fields fast neutron irradiation is not as effective in enhancing $j_c$ and the irreversibility fields $H_{irr}$ are not increased.

In our case of Sr substitution the significant fields shift, but the qualitative form of the hysteresis loop remains the same. As with fast neutron irradiation, Sr substitution increases $j_c$, but most pronounced in medium fields, in the second peak region. The maximum critical current densities there, $j_c^{\text{max}}(T) = j_c(H = H_{2p}, T)$, increase dramatically upon Sr substitution, as can be seen in Fig. 3. At the temperature of 60 K, the critical current density reaches almost the values of critical current densities for Y123: For the crystal with 32% Sr substitution $j_c^{\text{max}}(T = 60 \text{ K}) = 6 \times 10^4$ Acm$^{-2}$, while for a Y123 single crystal $j_c^{\text{max}}(T = 60 \text{ K}) = 6.3 \times 10^4$ Acm$^{-2}$ [10]. This is not the case, however, at the lower temperature of 50 K, where Y123 crystals have a maximum critical current density roughly three times higher than the one measured on Sr substituted YBa$_{2-x}$Sr$_x$Cu$_4$O$_8$. The main panel of Fig. 4 shows the dependence on Sr content of the maximum pinning force density $F_{\text{pin}}^{\text{max}} = H_{2p}j_c(H_{2p})$ corresponding to the critical current densities measured at $T = 65$ K. It can be seen that $F_{\text{pin}}^{\text{max}}$ increases by more than an order of magnitude upon Sr substitution. The lower right inset of Fig. 4 shows the Sr dependence of $F_{\text{pin}}^{\text{max}}$ at a fixed reduced temperature. The initially approximately linear rise of the maximum pinning force density saturates for $x \approx 0.4$ indicating that increasing the substitution level to $x = 0.64$ does not increase the pinning drastically any more. The critical current density itself also shows saturation behaviour, but for lower Sr content (see inset of Fig. 3). The difference is due to the substitution dependence of the second peak field, shown in the upper left inset of Fig. 3. The dependence $H_{2p}(x)$ is similar to the substitution dependence of $H_{ss}$, discussed in Sec. III D.
C. Irreversibility Fields

The irreversibility fields $H_{irr}$ as a function of temperature are, for different Sr substitution levels, presented in Fig. 6. The theoretical meaning of the irreversibility field is not very clear, often parts of it are controlled by surface or geometrical barriers (see below). In high fields, it was often found to be located near the transition from glass to fluid (see e.g. Refs. 10,11). However, whether a true phase transition between a glass and a fluid phase even exists, is not a settled question. Not much is known about the properties of the highly disordered entangled solid located above $H_{ss}$, apart from properties that distinguish it from the lattice phase. The feature differentiating the phase from the liquid phase is a rapid freezing of the dynamics, but it is not sure whether this freezing of the dynamics is a proper phase transition or merely a crossover. The existence of a separate so-called vortex glass (VG) phase, distinguished by an unbounded distribution of the heights of barriers between metastable states, was proposed early after the discovery of high $T_c$ superconductivity.41 Vortex glass scaling of current voltage relations was used to determine the vortex glass transition line $H_g$ experimentally, and also provided strong support for the VG theory (see e.g. Refs. 3,10). However, later experiments and numerical calculations found discrepancies (see Ref. 17 and references therein), and suggest a glass phase more akin to window glass. Despite these uncertainties, $H_{irr}(T)$ is certainly important from a more practical point of view, as it limits the field range for applications.

It is hard to see a clear trend in the Sr substitution dependence of the measured irreversibility fields. Differences between $H_{irr}$ of the different samples measured are relatively small, and the details depend on the temperature as well. Now it is important to remember that bulk pinning is not the only source of magnetic hysteresis. There are additional contributions due to Bean-Livingston surface barriers,42 and due to geometrical barriers.43 The significance of surface and geometrical barriers depends on several factors. One of them is the strength of the bulk pinning - the smaller the (bulk) critical current density the bigger the relative importance of barriers. In $\text{YBa}_2\text{Sr}_x\text{Cu}_4\text{O}_8$ the critical current densities depend, as discussed above, strongly on $x$, and are very low for the unsubstituted crystal. Indeed, while the hysteresis loops of substituted crystals are generally rather symmetric, this is not the case for the unsubstituted crystal, which is an indication that barrier hysteresis is more important than bulk hysteresis.44 Another factor is the sample shape - bulk hysteresis depends linearly on the sample diameter, while the barrier hysteresis scales with the ratio of thickness to diameter. Thus, barrier effects are more important for thicker and smaller (in diameter) crystals. The thickness to diameter ratio varies from 0.1 to 0.3 for the crystals used in this study. Finally, an important factor is the temperature, since the bulk hysteresis is depressed by thermal depinning, while the one due to geometrical barriers is less affected by temperature.44 In the second peak region, the main part of the hysteresis is clearly of bulk origin, as follows from the shape of the hysteresis loops (see Fig. 2). However, since the total hysteresis observed is simply the sum of bulk and barrier hysteresis, the irreversibility field may still be determined by surface or geometrical barriers, since the bulk hysteresis goes to 0 faster at higher fields.44 In high fields, bulk critical current densities were found to go to zero exponentially.44 A linear relationship between $j_c$ and $\ln(H)$ is indeed followed in the crystals we measured. Figure 6 shows this relationship for $\text{YBa}_{1.8}\text{Sr}_{0.2}\text{Cu}_4\text{O}_8$. The figure shows also, however, that there are systematic deviations below a certain temperature dependent threshold of critical current densities. For higher temperatures the deviation appears at larger critical current densities. This may indicate that the irreversibility lines are indeed influenced considerably by barrier hysteresis at higher temperatures.

On the other hand, measurements of the influence of fast neutron irradiation on the high temperature part of the irreversibility field on a Y124 single crystal found a slight decrease of $H_{irr}$ upon irradiating the crystal.44 Irradiation may reduce Bean-Livingston type surface barriers, but those are, like bulk pinning, strongly temperature dependent and were generally not found to be im-

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FIG. 5: The graph summarizes the dependence on the reduced temperature of the irreversibility fields (full symbols) and $H_{ss}$ (open symbols), for different Sr contents in $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$: circles correspond to $x = 0$, diamonds to $x = 0.2$, down triangles to $x = 0.35$, up triangles to $x = 0.4$ and squares to $x = 0.64$. Error bars have been omitted for clarity. Full lines are fits of the $H_{ss}(T)$ data to Eq. (2). Except for $x = 0$, fitting was restricted to the range $0.65 \leq T / T_c \leq 0.9$. Dashed lines are guides for the eye indicating an exponential temperature dependence of $H_{ss}$ at lower temperatures. Variations of $H_{irr}(T)$ generally are relatively small and the details depend on the temperature. $H_{ss}(T)$ of the unsubstituted crystal is located in much higher fields than $H_{ss}(T)$ of all substituted crystals and for high temperatures is practically located in the region of the irreversibility fields.

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Table 1: Critical current densities for $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals. The critical current density is defined as $j_c = \frac{\delta I}{\delta B}$, where $\delta I$ is the change in current and $\delta B$ is the change in magnetic field. The data is presented for different temperatures and Sr substitution levels. The table shows the critical current density in units of A/cm$^2$. The critical current density increases with temperature and decreases with Sr substitution, indicating a slowing down of the vortex lattice. The table also includes the irreversibility field $H_{irr}$, which is defined as the field at which the current density is equal to the critical current density.
important near $T_c$. Geometrical barriers, on the other hand, should not change upon irradiation. As the irradiation did change the irreversibility field at high temperatures, it would indicate that $H_{irr}$ at high temperatures can not be due to Bean-Livingston or geometrical barrier hysteresis.

However, as mentioned above, the importance of barriers strongly depends on the sample shape and we cannot be completely sure to what extent barriers influence $H_{irr}$ especially in the case of the unsubstituted crystal, where bulk pinning is very weak. In order to explain the effect of the Sr substitution on the bulk properties, it is therefore advisable to compare $H_{irr}(T)$ at not too high reduced temperatures. A comparison of $H_{irr}(T)$ at $T = 0.7 T_c$ is shown in Fig. 6. The position of the irreversibility field is not changed much, but the general trend that $H_{irr}$ rises with $x$ is consistent with the also rising maximum critical current density. The only irreversibility field that does not fit into this trend is the one of the unsubstituted crystal, which may be attributed to barrier effects that still are not negligible at this temperature for this weakly pinned compound.

An increasing glass to liquid transition field can be justified theoretically in a similar way as the decrease of the lattice to glass transition field. Since the glass to liquid transition line depends on the relation between the pinning energy and the thermal energy, and because the pinning energy is increased upon introducing more disorder into the system, it would seem natural that the transition occurs at higher temperatures.

The dependence of the glass to fluid transition on disorder was studied, for electron irradiated Y123, by Nishizaki et al. They measured both the glass transition temperature $T_g$ resistively, and the irreversibility field $H_{irr}$, and found the two lines $T_g(H)$ and $H_{irr}(T)$ to coincide. Nishizaki et al. find the opposite disorder dependence of the glass to fluid transition, in their case $H_g$ and $H_{irr}$ decrease with increasing fluence of electron irradiation.

There are different possible explanations for the opposite behaviour of the glass to liquid transition line when the disorder is increased between the present measurements and those of Nishizaki et al.

Firstly, the disorder introduced by substituting 10% or more Sr for Ba in Y124 is much stronger than the disorder introduced by an irradiation of 2.5 MeV electrons with a fluence of up to $2 \times 10^{18}$ electrons/cm$^2$ in untwinned Y123. This can be seen by comparing the phase diagrams. The irradiated Y123 crystals show a first order melting transition in fields up to 50–100 kOe, while all of our Sr substituted Y124 crystals show no first order melting transition at all up to temperatures very near $T_c$. Nishizaki et al. note that the decrease of $H_g$ upon increasing disorder they measured is consistent with a theoretical prediction made for systems with weak pinning. The disorder in our substituted crystals may be too big for this to be applicable.

Secondly, the rise of $H_{irr}$ with Sr substitution could also be explained by a decreased anisotropy of the substituted crystals, since $H_{irr}$ was found to scale with $\gamma^{-2} s^{-1}$, where $s$ is the distance between two adjacent superconducting planes. If the rise of $H_{irr}$ in YBa$_{2-x}$Sr$_x$Cu$_4$O$_8$ with $x$ for $x > 0.2$ is due to a decreased anisotropy caused by the decreased blocking layer thickness, the initial drop for $x < 0.2$ may be due to the disorder, in which case the dependence of $H_{irr}$ on disorder would be qualitatively the same as the one found by Nishizaki et al.
D. Transition from Lattice to Glass

As noted in Sec.

a kink in \( M(H) \) (denoted \( H_{ss} \)) is visible between onset and maximum of the second peak of the hysteresis loop shown in Fig. 3. According to previous studies performed on Y123, it is likely that the kink corresponds to the transition line from the lattice to the glass state of vortices. In Ref. \(^13\) it was argued that the equilibrium phase transition corresponds to the kink in \( M(H) \) in decreasing field, while the kink in increasing field corresponds to the upper limit of metastability of the lattice phase. However, since the pinning is much stronger in the glass phase, the equilibrium transition can be expected to be located quite close to the upper limit of metastability. This was indeed found from the measurements of Ref. \(^13\). Also in our case, while we have more data on the field increasing branch, the kink in \( M(H) \) decreasing was checked at representative temperatures and the difference in the two kinks was found to be rather small indeed (see Fig. 2). We should mention that the actual equilibrium transition is a little bit lower than the field of the kink in \( M(H) \), which we call \( H_{ss} \) (see Fig. \(^10\)). It is interesting to note that generally the kink positions (for both directions of field changes) do not depend on the waiting time (we tested between 30 s and 10 min) after changing the the field. Additionally the kink field location does not depend on the field change step size and not on the small inhomogeneities of the magnetic fields (\( \sim 0.02\% \)) tested by using different scan lengths, but the shape of the \( M(H) \) curve around the kink does depend on step size and scan length. On the other hand, the onset fields \( H_{on} \), defined as the fields where the absolute value of the magnetization has a minimum, have a slight tendency to shift to lower fields upon increasing waiting time, while the absolute value of the magnetization at the onset is decreased significantly upon increasing waiting time. Additionally the onset measured in decreasing fields can be much lower (up to 50% at low temperatures) than the onset measured in increasing field. Figure 3 shows a selection of magnetization curves measured, in the region where the kink is located. The sharpness of the kink varies with temperature and depends on the sample as well. The inset of Fig. 3 shows the onset region of one of the curves enlarged and the derivative of the magnetization (average of the four scans), which can help to locate \( H_{ss} \). Within a range of a few Kelvin below \( T_c \) the unambiguous determination of \( H_{ss} \) becomes difficult nonetheless.

The increase of the absolute value of the magnetization around \( H_{ss} \) observed in our measurements is hardly sharp enough to be called a “jump”. One reason for this is the spatial averaging of the inhomogeneous induction inside the sample. Additionally, the difference between the onset of the second peak in increasing fields \( H_{on}^\uparrow \), defined as the minimum of the absolute value of the magnetization before the second peak, and the kink field \( H_{ss} \), can be explained by the existence of a region of metastability around the phase transition. Recent experiments\(^{12,25}\) indicate that an abrupt change of the field injects a transient disordered vortex phase at the sample edges. If the thermodynamically stable phase is the lattice phase, the transient disordered vortex phase then decays. This decay happens with a rate decreasing to 0 as the field reaches \( H_{ss} \). In our measurements, the fields \( H_{on}^\uparrow \), \( H_{on}^\downarrow \), and \( H_{ss}^\downarrow \) was found to follow the same dependence on temperature and Sr content as \( H_{ss} (H_{on}^\downarrow) \). Below the onset, both the magnitude of the critical current density and the average value of the magnetization for the two branches of the hysteresis loop suggest that in this region surface and geometrical barriers are more important than bulk pinning in our samples, i.e. bulk pinning seems to be relevant only if the vortex matter is in the glass phase.

The \( H_{ss}(T) \) lines, as determined by the kink in the \( M(H^\uparrow) \) curves for the measured samples, are also shown in Fig. 3. For all samples, \( H_{ss} \) decreases monotonically with increasing temperature. A monotonically decreasing \( H_{ss} \) was also found on \( \text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-b} \) \(^9\) but measurements on \( \text{Y}_{1.38}\text{Ba}_{1.62}\text{Sr}_{0.38}\text{CuO}_{4-b} \) showed an \( H_{ss} \) increasing with temperature. The position of the phase transition between quasi-ordered lattice and highly disordered glass is determined by the interplay between elastic and pinning energy. For the case of weak, random disorder due to point-like defects the theory of collective pinning was developed by Larkin \( \text{et al.} \)\(^4\) The defects can interact with the vortices in two ways. They can cause a spatial variation of the transition temperature (\( \delta T_c \)-pinning), described by a modulation of the linear term of the Ginzburg-Landau free energy functional. Alternatively, they can cause a spatial modulation of the mean free path (\( \delta \ell \)-pinning), described by a modulation of the gradient term of the free energy functional. In both cases, the influence of disorder is described by a disorder parameter \( \tilde{\gamma} \), proportional to the defect density. However, the temperature dependence of \( \tilde{\gamma} \) is different for the two
cases. For $\delta T_c$-pinning, $\gamma \propto 1/\lambda^4$, while for $\delta \ell$-pinning, $\gamma \propto 1/(\lambda \xi)^4$, where $\lambda$ and $\xi$ are the penetration depth and the coherence length.\cite{ref24} The order-disorder transition position was calculated analytically by using a Landau-Lifshitz criterion.\cite{ref25} For the case of not too big anisotropy and disorder, following the calculation of Ref.\ 20 we get

$$H_{ss} = H_o \left( \frac{U_o}{U_c} \right)^3,$$

(1)

with $H_o = 2c^4_o \Phi_o / \xi^2$, $U_o = \Phi_o^2 c_l \xi / (16\sqrt{2}\pi^2 \lambda^2 \gamma)$ and the collective pinning energy $U_c = ((\gamma \Phi_o^2 \xi^4) / (16\pi^2 \lambda^2 \gamma^2))^{1/3}$. Here, $\Phi_o$ is the flux quantum and $c_l \approx 0.1 - 0.2$ the London number. $H_{ss}$ is inversely proportional to both the anisotropy $\gamma$ and the disorder parameter $\gamma$. Concerning the temperature dependence, as long as we are below the depinning temperature $T_{dp} \approx U_c / k_B$, we get

$$H_{ss} \propto \xi^{-3} \propto (1 - (T/T_c))^3^{3/2}$$

(2)

in the case of $\delta T_c$-pinning and

$$H_{ss} \propto \xi \propto (1 - (T/T_c))^4^{-1/2}$$

(3)

in the case of $\delta \ell$-pinning.\cite{ref24} The calculation of Ref.\ 21 leads to qualitatively the same results. Since $H_{ss}$ decreases with temperature in our case, the pinning in YBa$_{2-x}$Sr$_x$Cu$_4$O$_8$ cannot be $\delta \ell$ pinning. The temperature dependence of $H_{ss}$ of the unsubstituted crystal agrees satisfactorily with the formula proposed for $\delta T_c$ pinning. In the case of the substituted crystals, however, the agreement is limited to values of $T/T_c$ between about 0.65 and 0.9 (see Fig.\ 3, the full bold lines are fits to Eq.\ 2). The lowered $H_{ss}$ in the vicinity of $T_c$ may be attributed to the finite transition width.

However, at low temperatures, the temperature dependence is rather exponential in the case of the substituted crystals, as indicated by dashed lines in Fig.\ 3, with some indications of a flattening at the lowest temperatures measured. This behaviour is clearly at odds with Eq.\ 2 and to the best knowledge of the authors is not predicted by any present theory of the order-disorder transition. An exponential upturn in the onset field $H_{on}$ at low temperatures was also found on a Nd$_{2-x}$Ce$_x$CuO$_4-x$ single crystal by Andrade et al.\ 24 who attributed it to Bean-Livingston surface barriers. However, since the observed magnetic hysteresis is the sum of bulk and barrier hysteresis, it is difficult to imagine why the kink in $M(H)$ at $H_{on}$ should be influenced by surface or geometrical barriers, and in our case a clear kink is observable at low temperatures where $H_{ss}$ has already clearly departed from the $(1 - (T/T_c))^3^{3/2}$ dependence observed at higher temperatures.

A possible cause for a changed temperature dependence is a dimensional crossover. Due to the layered structure of the cuprate superconductors, vortex lines should be thought of being composed of stacks of “pancake” vortices.\cite{ref1} Only below the 2D/3D crossover field $H_{2D} \approx \Phi_o / (\gamma^2 s^2)$, where $s$ is the distance between two adjacent layers, is the interlayer interaction between pancake vortices larger than their intralayer interaction and the pancakes form well defined vortex lines. For all our crystals, $H_{2D} \gtrsim 77$ kOe is estimated well above the upper limit of the fields attainable in our magnetometers. Also, $H_{ss} \propto \xi^{-5/2}(\xi^{5/2})$ for 2D and $\delta T_c(\delta \ell)$ pinning\cite{ref29} i.e. the temperature dependence should be even flatter in the 2D regime. A dimensional crossover can therefore not be responsible for the observed upturn of $H_{ss}$ at low temperatures. In principle a second pinning mechanism, which is very effective at low temperatures, could be overlaid. However, point-like disorder should rather (additionally) suppress $H_{ss}$, while the influence of correlated disorder should be more visible at high temperatures or low fields.\cite{ref21} Figure 4 shows $H_{ss}$ and $H_{irr}$ of two of the substituted crystals in the vicinity of the transition temperature. No sign of a tricritical point, where the two lines would meet, can be seen. A tricritical point would have to be located very near $T_c$, where reliable measurements become increasingly difficult.

It can be seen in Fig.\ 3 that any substitution of Sr lowers $H_{ss}$ very significantly. For two values of $T/T_c$, $H_{ss}$ vs. strontium content $x$ is shown in Fig.\ 6. After a substitution of just 10% Sr for Ba, $H_{ss}$ drops roughly by a factor of 5, at both temperatures. A further increase of Sr substitution, however, does not reduce the magnitude of $H_{ss}$ any more. Rather, $H_{ss}(x)$ remains essentially constant. We do not consider the peak at $x \approx 0.4$ as big enough to be significant.

The initial decrease of $H_{ss}$ upon Sr substitution does not come unexpected. It follows from Eq.\ 1 that $H_{ss} \propto (\gamma \hat{x})^{-1}$. Sr substitution increases the disorder parameter and decreases the anisotropy (see Sec. IIIA). The large decrease of $H_{ss}$ upon substituting 10% of Sr indicates that the influence of the Sr substitution on disorder is much bigger than it’s influence on the anisotropy.

What is more surprising is the apparent saturation of the influence of the additional disorder for a substitution level $x \gtrsim 0.2$, while the maximum pinning force is still

![FIG. 9: $H_{ss}$ and $H_{irr}$ in the vicinity of $T_c$, for substitutions of 10% and 32% Sr.](image)
linearly increasing up to $x \approx 0.4$. In the case of weak point-like pinning $H_{ss} \propto \tilde{\gamma}^{-1} \propto n_{\text{dis}}^{-1}$, where $\tilde{\gamma}$ is the dis- order parameter and $n_{\text{dis}}$ the defect density, it is expected. It would seem natural to assume the defect density to be proportional to the Sr substitution level $x$. The blocking layer thickness on the other hand, which is the structural factor with the biggest influence on the anisotropy, de- creases linearly with $x$. A large initial decrease of $H_{ss}$ followed by an almost independence on $x$ is therefore diffi- cult to explain.

A downward shift of $H_{ss}$ upon introducing more dis- order was also found in electron irradiated crystals of BSCCO and Y123. The notable difference between both of these irradiation experiments and Sr substitution in Y124 is that in both cases of electron irradiation there is no sign of any saturation of the disorder induced lowering of $H_{ss}$. As discussed above, the disorder induced by Sr substitution is probably, even for the lowest substitution level of $x \approx 0.2$, already much higher than the disorder induced by the electron irradiation with the largest fluence measured in Refs. and . It may be that in our substituted crystals the disorder is already too big for the applicability of formulas derived for weak disorder.

The resulting phase diagrams for YBa$_2$Cu$_{4}$O$_8$ and YBa$_{1.36}$Sr$_{0.64}$Cu$_4$O$_8$ are contrasted in Fig. 10. The glass phase of YBa$_{1.36}$Sr$_{0.64}$Cu$_4$O$_8$ has, as compared to YBa$_2$Cu$_4$O$_8$, expanded dramatically from a rather small part of the easily experimentally accessible phase dia- gram to the phase covering the biggest area in fields $H \leq 55$ kOe. The most dramatic shift is the lowering of $H_{ss}$, which is to be expected as the (quasi-)ordered phase should become smaller upon introducing more dis- order into the system. The region of metastability is much smaller for the substituted crystal, indicating that strong disorder tends to reduce over-heating and espe- cially under-cooling effects. We speculate that this may be connected to our observation that for unsubstituted Y124 with very weak disorder $H_{ss}$ depends strongly on the exact amount of disorder, while for substituted Y124 with rather strong disorder it almost does not depend on the exact level of disorder. In real crystals, the disorder density depends on the position on a mesoscopic scale. Soibel et al. observed that for melting supercooling ex- ists only at local maxima of the transition field. In a first approximation the width of metastability can be linked to the difference between maximal and minimal transition fields within the sample. In our case the same spatial variation of disorder should produce a much bigger vari- ation of the local transition field for the unsubstituted crystal than for the substituted ones.

**IV. CONCLUSIONS**

We investigated the influence of structural disorder, in- troduced by a substitution of Sr for Ba in YBa$_2$Cu$_4$O$_8$, on the critical current density at elevated fields and on the borders of the glass phase of vortices. We stress that the disorder introduced chemically by means of Sr sub- stitution is random and point-like, rather than disorder due to extended defects. This is shown by the hysteresis loops keeping their general form, similar to the case of electron irradiation and unlike the case of neutron irra- diation, where at least partially the effect is to introduce extended defects.

Our main conclusion is that the introduction of even a small amount of disorder into very clean systems changes the phase diagram drastically by lowering the order- disorder transition line $H_{ss}(T)$ and also changing it’s temperature dependence. When the disorder reaches a
certain threshold, however, introducing additional disorder does not continue this tendency. With other words, in highly disordered systems (including probably most systems with partial chemical substitutions), the vortex matter phase diagram is relatively robust with respect to variations in the exact degree of disorder.

This conclusion is supported by the rather small variation of the measured kink location as an indication of the order-disorder transition at fixed reduced temperature for any Sr substitution level \( x \geq 0.2 \), especially as compared to the corresponding transition in a clean, un-substituted crystal. That the disorder is still increasing compared to the corresponding transition in a clean, un-substituted crystal. This may be by introducing random point-like disorder. This might be linked to the observation that the position variations of the disorder density on a mesoscopic scale do not produce large positional differences of the local transition field in this case.

Similar to the robustness of the phase diagram above a disorder threshold, both the maximum pinning force density and the maximum critical current density in the glass phase increase initially with increasing disorder, the critical current density increases by an order of magnitude upon substituting 10% Sr. But they then show saturation behaviour, i.e. the pinning force density and critical current densities at low fields and the maximum pinning force density.

In strongly disordered systems, metastability is less important than in very clean systems and especially the region of undercooling of the glass phase is much smaller. This might be linked to the observation that the position of the transition line does not depend much on the exact amount of disorder in strongly disordered systems, since variations of the disorder density on a mesoscopic scale do not produce large positional differences of the local transition field in this case.

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