

Critical currents and order-disorder phase transition in the vortex states of $\text{YBa}_2\text{Cu}_4\text{O}_8$ with chemically introduced disorder

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A series of $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$ single crystals was measured to study the influence of site disorder on the transition line $H_{ss}(T)$ between quasicrystalline vortex lattice and highly disordered vortex glass, as well as on the maximum critical current density within the glass phase, j_c^{max} . When 32% of Ba is replaced by Sr, j_c^{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 5 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} decreases monotonically 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 cuprate superconductors it was established that there are at least two phases: a fluid (liquid or gas)² 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 up to a critical point.³⁻⁵ Recently, it was shown⁶⁻¹³ that there are two distinct solid phases. In low fields the vortices form a quasicrystalline lattice in a so-called Bragg glass,^{14,15} which is stable against the formation of dislocations. In higher fields they form a highly disordered, entangled¹ 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.^{16,17} Henceforth we will simply term it “glass.”

The high-field glass phase is caused by weak, random, and uncorrelated disorder due to pointlike defects such as oxygen vacancies.¹ 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 a different influence on the vortex matter, and can lead to additional phases, e.g., a “Bose glass” phase¹⁸ located at low fields.¹⁹ Correlated disorder may not be present in all crystals. In the following, when we simply speak of “disorder,” we are referring to weak random pointlike disorder.

The pinning induced by the disorder has two effects on the vortex lattice.²⁰ First, it localizes vortices by trapping them in low-lying metastable states, hardening the solid. Second, it promotes a transverse wandering of vortices from their ideal lattice positions. The mechanism of the destruction of the lattice, triggering the transition to the glass phase,^{15,20-23} is very similar to the mechanism of melting at higher temperature. Similar to the way the lattice loses trans-

lational order and melts into a fluid, when thermal fluctuations become of the order of the lattice spacing, it loses 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,^{22,23} of which there is some evidence in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (BSCCO).²⁴ This is not expected in less anisotropic systems, like $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Y123).^{22,23} There is considerable evidence that the phase transition between the lattice and the glass, like the melting transition between the lattice and the fluid, is of first order^{13,24-26} and is even a continuation of the melting line.²⁷

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. Calculations²⁰⁻²³ of H_{ss} , based on a Lindemann criterion, confirmed 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 the lattice and the glass found a systematic decrease of H_{ss} with increasing irradiation dose.^{7,12}

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. $\text{YBa}_2\text{Cu}_4\text{O}_8$ (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 eight per unit cell. Another

advantage of Y124 is that twinning, which leads to strong correlated disorder, does not exist in this compound.²⁸ Finally, Y124 has an intermediate anisotropy $\gamma \approx 12$,²⁸ 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.²⁹ Studies of compounds with chemical disorder also found increases of j_c and even the development of a second peak.³⁰

In this work, we study the influence of a substitution of isovalent Sr for Ba in $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$ on the lattice-to-glass transition line $H_{ss}(T)$, the irreversibility line $H_{irr}(T)$, and the critical current densities j_c . We provide 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. EXPERIMENT

Single crystals of Y124, with and without Sr substitution, were grown using a high-pressure technique; the details of crystal growth are reported elsewhere.³¹ Single crystals of $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$, with a Sr content up to $x=0.64$ or 32%, were obtained. The crystals were checked with energy dispersive x-ray analysis (EDX) and their structure analyzed with single-crystal x-ray analysis.³¹ The transition temperatures T_c of the crystals were determined with a superconducting quantum interference device (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 (10–90%) 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 noncommercial 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. A wait time of 5 min was chosen to avoid effects of the initial relaxation of the

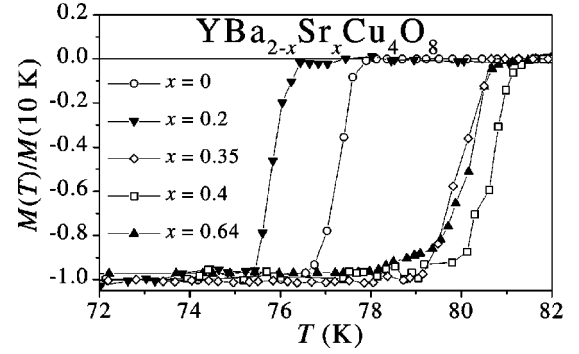


FIG. 1. Normalized $M(T)$ curves of the crystals used in this work, showing sharp superconducting transitions. The curves shown were measured in a magnetic field of $H=1$ Oe, under zero-field-cooled conditions.

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 density³² j_c was calculated from the width of the hysteresis loop using an extended Bean model.³³ 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 $j_c = 50 \text{ A cm}^{-2}$ was used. For the crystals studied it corresponds to a criterion of in between 4.4 and 26 μemu , 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 linewidth.³¹ No evidence of an introduction of correlated disorder was found, and, since twin boundaries are absent in $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$, the assumption of the disorder in our crystals to be purely random and pointlike 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.³¹ 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.³⁴

An important structural modification due to Sr substitution is a decrease of the thickness of the blocking layer d_b ,

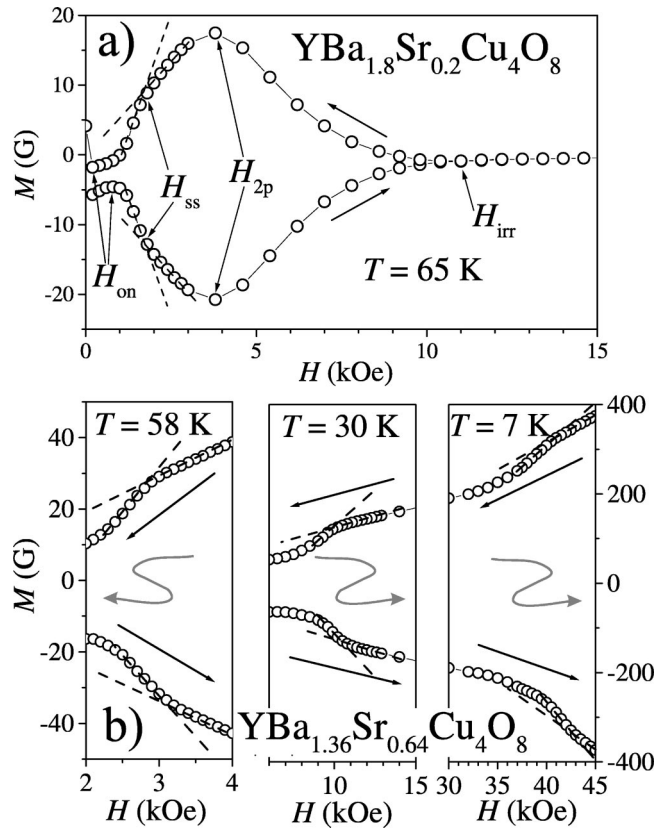


FIG. 2. (a) One of the hysteresis loops measured. The direction of the field change is shown by arrows. The onset of the second peak, the second peak maximum, and the kink in $M(H)$ between them are denoted as H_{on} , H_{2p} , and H_{ss} , respectively. Note that there are small differences between the respective fields between the two directions of field change (see the text and Fig. 10). 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.

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%,³¹ which leads to an improved coupling between the superconducting planes. Torque measurements indeed indicate a corresponding decrease of the anisotropy γ .³⁵

B. Critical current densities

Figure 2(a) 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 (also see 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 den-

sity 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 increases monotonically up to the highest substitution level measured, with the largest rate between $x = 0.4$ and $x = 0.64$.

When, upon raising the field, the lattice is destroyed in the glass state and the vortex system has an additional “dislocation degree of freedom,”²² 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.²³ Additionally, the entanglement itself was also suggested to increase critical current densities, due to an increased intervortex viscosity,³⁶ provided the barriers to flux cutting are enough high.^{21,22} In our case, a steep rise of the magnetization occurs just below the kink field, denoted H_{ss} in Fig. 2(a) (also see Sec. III D). Local magnetic measurements on a BSCCO crystal revealed a sharp increase in $|M(H)|$, followed by a monotonic decrease afterward.⁶ The so-called “second peak” (or “fishtail”) effect in BSCCO can thus be accounted for by an order-disorder transition. On untwinned Y123 crystals, a similar sharp increase in $|M(H)|$ was observed by local magnetic measurements,¹¹ but, contrary to the situation for BSCCO, $|M(H)|$ continues to increase upon increasing field, although with a slower rate, which leads to a sharp kink in $M(H)$. Only in even higher fields does the magnetization start to decrease with increasing field.

Independently of vortex matter phase transitions, the second peak or fishtail effect in Y123 was attributed to a cross-over from elastic to dislocation-mediated plastic creep.^{8,37} This mechanism is closely connected to the lattice-glass transition, however, since in the lattice phase dislocations are strongly suppressed while in the glass phase dislocations proliferate, as mentioned above.³⁸ From collective creep theory a rise of the experimentally observable critical current density with the field is expected,^{1,37} 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, it follows that in $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$ the situation is similar to the one in Y123. After raising 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. 39. 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

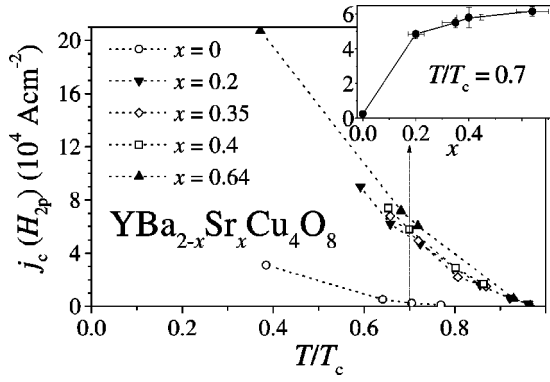


FIG. 3. Critical current density at H_{2p} vs reduced temperature for different Sr contents. It can be seen that unsubstituted Y124 has much lower maximum critical current densities than substituted Y124, at all temperatures. Inset: $j_c(H_{2p})$ vs Sr content x at $T/T_c = 0.7$.

strong in the Bose glass phase, and, if the irradiation-induced correlated disorder is strong enough, the Bose glass phase may well destroy the entire 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 the effect is most pronounced in medium fields, in the second peak region. The maximum critical current densities there, $j_c^{\max}(T) \equiv j_c(H = H_{2p}, T)$, dramatically increase upon Sr substitution, as can be seen in Fig. 3. At a temperature of 60 K, the critical current density reaches almost the values of critical current densities for Y123: for a crystal with 32% Sr substitution $j_c^{\max}(T = 60 \text{ K}) = 6 \times 10^4 \text{ A cm}^{-2}$, while for a Y123 single crystal $j_c^{\max}(T = 60 \text{ K}) = 6.3 \times 10^4 \text{ A cm}^{-2}$.⁴⁰ 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 higher³⁹ than the one measured on Sr-substituted $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$. The main panel of Fig. 4 shows the dependence on Sr content of the maximum pinning force density $F_{pin}^{\max} = H_{2p} j_c(H_{2p})$, corresponding to critical current densities measured at $T = 65 \text{ K}$. It can be seen that F_{pin}^{\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_{pin}^{\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 $x = 0.64$ no longer drastically increases the pinning. The critical current density itself also shows a saturation behavior, but for lower Sr content (see the 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. 4. 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.

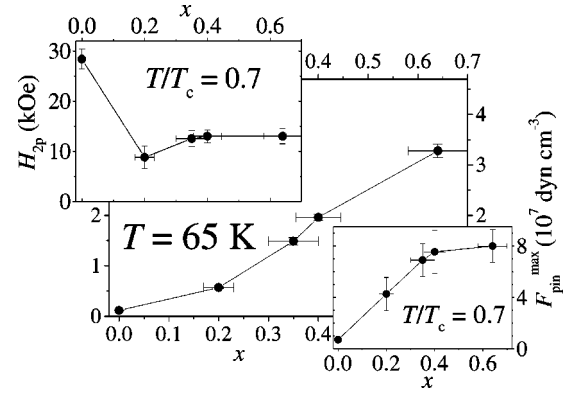


FIG. 4. Dependence of the maximum pinning force density $F_{pin}^{\max} = H_{2p} j_c(H_{2p})$ on Sr content, at a temperature of 65 K. The maximum pinning force density increases by more than an order of magnitude upon substituting 32% of Sr for Ba. Upper left inset: second peak field vs Sr content at $T/T_c = 0.7$. Lower right inset: maximum pinning force density vs Sr content, at $T/T_c = 0.7$.

5. 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 and 12). However, whether a true phase transition between the glass phase and the fluid phase even exists, is not a settled question. Not much is known about the properties of the highly disordered entangled solid located

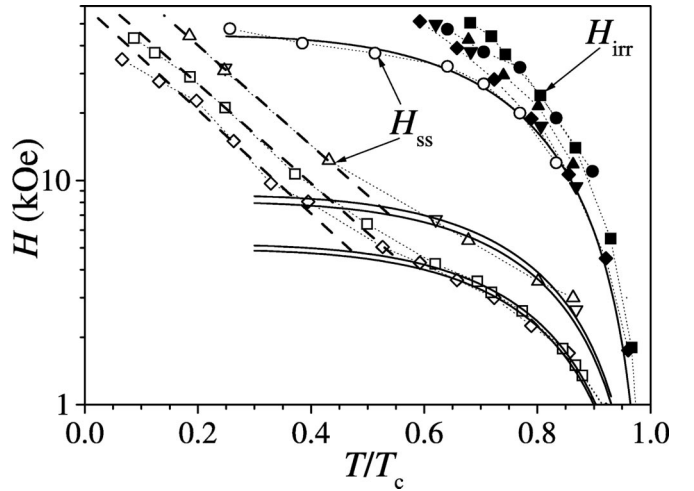


FIG. 5. The graph summarizes the dependence on the reduced temperature of the irreversibility fields (full symbols) and H_{ss} (open symbols), for different Sr contents in $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$: circles correspond to $x = 0$, diamonds to $x = 0.2$, down triangles to $x = 0.35$, 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.

above H_{ss} , apart from properties that distinguish it from the lattice phase. The feature differentiating the glass 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.¹⁶ Vortex glass scaling of the 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 and 10). However, later experiments and numerical calculations found discrepancies (see Ref. 17 and references therein), and suggested 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^{41,42} and 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 larger the relative importance of barriers. In $YBa_{2-x}Sr_xCu_4O_8$ the critical current densities depend strongly on x , as discussed above, 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.⁴² 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.^{42,45} In high fields, bulk critical current densities were found to go exponentially to zero.⁴⁶ A linear relationship between j_c and $\ln(H)$ is indeed followed in the crystals we measured. Figure 6 shows this relationship for $YBa_{1.8}Sr_{0.2}Cu_4O_8$. The figure also shows, 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

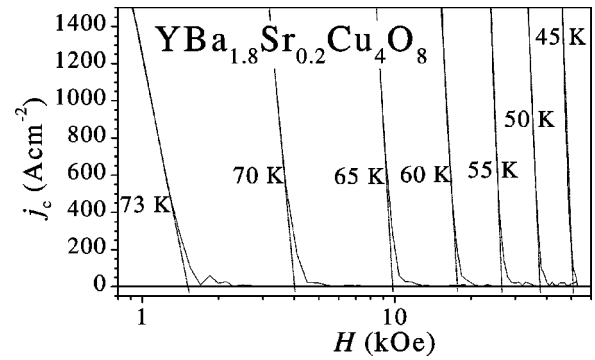


FIG. 6. Critical current density vs logarithm of the magnetic field, for a sample with 10% Sr substitution. The relationship is linear for high critical currents. Below a temperature-dependent threshold, deviations appear. Dashed lines extrapolate the linear relationship to $j_c=0$.

densities. This may indicate that the irreversibility lines are indeed considerably influenced 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.⁴⁷ Irradiation may reduce Bean-Livingston-type surface barriers, but those are, like bulk pinning, strongly temperature dependent, and were generally not found to be 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, this would indicate that H_{irr} at high temperatures cannot 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

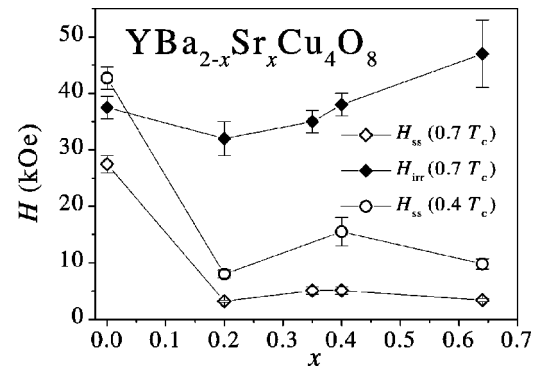


FIG. 7. Sr dependence of H_{ss} at temperatures of $0.7 T_c$ (open diamonds), $0.4 T_c$ (open circles), and $H_{irr}(0.7 T_c)$ (full diamonds). For both temperatures H_{ss} drops significantly by a small substitution of Sr for Ba, but remains essentially constant upon further Sr substitution. H_{irr} also drops by a small Sr substitution, but less significantly. It rises again upon further substitution, and is higher for $x=0.64$ than for $x=0$. The x error bars have been omitted for clarity.

pinning is very weak. In order to explain the effect of the Sr substitution on the bulk properties, it is therefore advisable to compare $H_{\text{irr}}(T)$ at not too high reduced temperatures. A comparison of $H_{\text{irr}}(T)$ at $T=0.7 T_c$ is shown in Fig. 7. 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 way similar to that for 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.*¹² They measured both the glass transition temperature T_g resistively, and the irreversibility field H_{irr} , and found that the two lines $T_g(H)$ and $H_{\text{irr}}(T)$ coincide. Nishizaki *et al.* found an opposite disorder dependence of the glass-to-fluid transition; in their case H_g and H_{irr} decreased with an increasing fluence of electron irradiation.

There are different possible explanations for the opposite behavior of the glass-to-liquid transition line when the disorder is increased between the present measurements and those of Nishizaki *et al.* First, 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 2×10^{18} electrons/cm² 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.* noted that the decrease of H_g upon increasing disorder they measured is consistent with a theoretical prediction made for systems with weak pinning.⁴⁸ The disorder in our substituted crystals may be too large for this to be applicable.

Second, 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.² If the rise of H_{irr} in $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$, 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. III B, 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. 2. According to previous

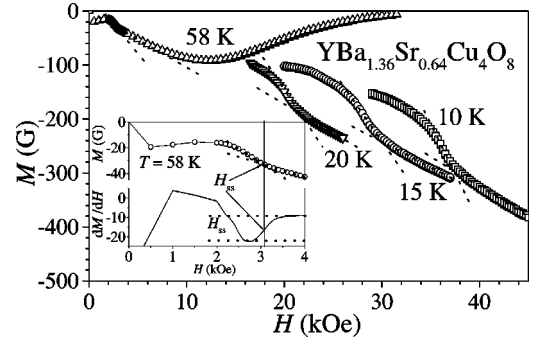


FIG. 8. $M(H)$ curves for increasing field near the onset of the second peak. Dashed lines are guides for the eye pinpointing the kink in the magnetization H_{ss} . The inset shows a magnification of the onset region of a $M(H)$ curve and its derivative at $T=58$ K. H_{ss} is located at the position of the steepest increase of dM/dH .

studies performed on Y123,^{10–12} 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 increasing field branch, the kink in decreasing H 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 $M(H^\uparrow)$, 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 or 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 is at 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 8 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. 8 shows an enlarged image of the onset region of one of the curves and the derivative of the magnetization (average of the four scans), which can help to locate H_{ss} . Within a range of a few K below T_c , the unambiguous determination of H_{ss} nonetheless becomes difficult.

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.¹³ Recent experiments^{25,26} indicated 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 occurs with a rate decreasing to 0 as the field reaches H_{ss} . In our measurements, the fields H_{on}^{\uparrow} , $H_{\text{on}}^{\downarrow}$, and $H_{\text{ss}}^{\downarrow}$ were found to follow the same dependence on temperature and Sr content as H_{ss} (H_{ss}^{\uparrow}). 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 $H_{\text{ss}}(T)$ lines, as determined by the kink in the $M(H^{\uparrow})$ curves for the measured samples, are also shown in Fig. 5. For all samples, H_{ss} decreases monotonically with increasing temperature. A monotonically decreasing H_{ss} was also found on $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$,⁹ but measurements on Y123 (Refs. 10–13) showed an H_{ss} increasing with temperature. The position of the phase transition between quasiordered lattice and highly disordered glass is determined by the interplay between elastic and pinning energy. For the case of weak, random disorder due to pointlike defects, the theory of collective pinning was developed in Ref. 49. The defects can interact with the vortices in two ways.¹ They can cause a spatial variation of the transition temperature (δT_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 (δl 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 δT_c pinning, $\tilde{\gamma} \propto 1/\lambda^4$, while for δl pinning, $\tilde{\gamma} \propto 1/(\lambda\xi)^4$, where λ and ξ are the penetration depth and the coherence length.¹ The order-disorder transition position was calculated analytically by using a Lindemann criterion.^{20,21} For the case of not too large anisotropy and disorder, following the calculation of Ref. 20, we obtain

$$H_{\text{ss}} = H_0 \left(\frac{U_0}{U_c} \right)^3, \quad (1)$$

with $H_0 = 2c_L^2\Phi_0/\xi^2$, $U_0 = \Phi_0^2 c_L \xi / (16\sqrt{2}\pi^2\lambda^2\gamma)$, and the collective pinning energy $U_c = [(\tilde{\gamma}\Phi_0^2\xi^4)/(16\pi^2\lambda^2\gamma^2)]^{1/3}$. Here Φ_0 is the flux quantum, and $c_L \approx 0.1-0.2$ the Lindemann number. H_{ss} is inversely proportional to both the anisotropy γ and the disorder parameter $\tilde{\gamma}$. Concerning the

temperature dependence, as long as we are below the depinning temperature $T_{\text{dp}} \approx U_c/k_B$, we obtain

$$H_{\text{ss}} \propto \xi^{-3} \propto [1 - (T/T_c)^4]^{3/2} \quad (2)$$

in the case of δT_c pinning, and

$$H_{\text{ss}} \propto \xi \propto [1 - (T/T_c)^4]^{-1/2} \quad (3)$$

in the case of δl pinning.⁵⁰ The calculation of Ref. 21 leads to qualitatively the same results. Since H_{ss} decreases with temperature in our case, the pinning in $\text{YBa}_{2-x}\text{Sr}_x\text{Cu}_4\text{O}_8$ cannot be δl pinning. The temperature dependence of H_{ss} of the unsubstituted crystal agrees satisfactorily with the formula proposed for δT_c pinning. In the case of substituted crystals, however, the agreement is limited to values of T/T_c between about 0.65 and 0.9 [see Fig. 5; 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. 5, with some indications of a flattening at the lowest temperatures measured. This behavior 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}^{\uparrow} at low temperatures was also found on a $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-x}$ single crystal by Andrade *et al.*,⁵¹ 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_{ss} 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 $[1 - (T/T_c)^4]^{3/2}$ 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 as being composed of stacks of “pancake” vortices.¹ Only below the two-dimensional/three-dimensional (2D/3D) crossover field $H_{2D} \approx \Phi_0/(\gamma^2 s^2)$, 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, $H_{2D} \approx 77$ kOe is estimated well above the upper limit of the fields attainable in our magnetometers. Also, $H_{\text{ss}} \propto \xi^{-5/2}(\xi^{5/2})$ for 2D and δT_c (δl) pinning,²⁰ 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, pointlike disorder should rather (additionally) suppress H_{ss} , while the influence of correlated disorder should be more visible at high temperatures or low fields.¹⁹ Figure 9 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

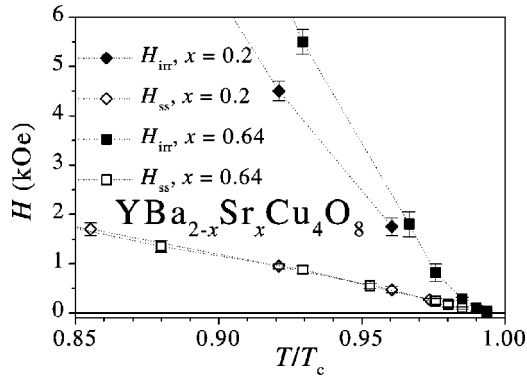


FIG. 9. H_{ss} and H_{irr} in the vicinity of T_c , for substitutions of 10% and 32% Sr.

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 from Fig. 5 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. 7. 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 further. Rather, $H_{ss}(x)$ remains essentially constant. We do not consider the peak at $x \approx 0.4$ to be large enough to be significant.

The initial decrease of H_{ss} upon Sr substitution is not unexpected. It follows from Eq. (1) that $H_{ss} \propto (\gamma\tilde{\gamma})^{-1}$. Sr substitution increases the disorder parameter and decreases the anisotropy (see Sec. III A). The large decrease of H_{ss} upon substituting 10% of Sr indicates that the influence of the Sr substitution on disorder is much larger than its influence on the anisotropy.

What is more surprising is the apparent saturation of the influence of the additional disorder for a substitution level $x \geq 0.2$, while the maximum pinning force is still linearly increasing up to $x \approx 0.4$. In the case of weak pointlike pinning, $H_{ss} \propto \tilde{\gamma}^{-1} \propto n_{dis}^{-1}$, where $\tilde{\gamma}$ is the disorder parameter and n_{dis} the defect density, is expected.¹ It would seem natural to assume the defect density to be proportional to the Sr substitution level x . On the other hand, the blocking layer thickness, which is the structural factor with the greatest influence on the anisotropy, decreases linearly with x .³¹ A large initial decrease of H_{ss} , followed by the almost independence on x , is therefore difficult to explain.

A downward shift of H_{ss} upon introducing more disorder was also found in electron-irradiated crystals of BSCCO (Ref. 7) and Y123.¹² The notable difference between both 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. 7 and 12. It may be that in our substituted crystals the disorder is already too large for the applicability of formulas derived for weak disorder.

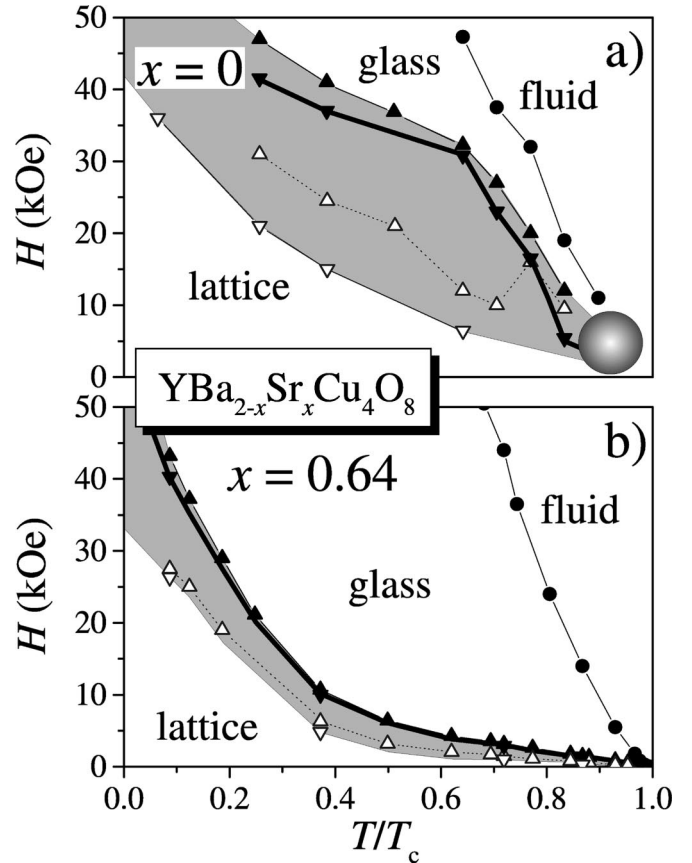


FIG. 10. Phase diagrams of the crystals with the lowest [$x=0$, (a)] and highest [$x=0.64$, (b)] Sr substitution. The quasiordered lattice phase at low temperatures and fields is separated from the highly disordered glass phase by the transition line H_{ss}^l (full down triangles, thick line). Around the transition is a region of metastability (Ref. 13) where both phases can coexist (shaded). The lower limit of metastability is marked by H_{on}^l (open down triangles) and the upper limit, very close to the equilibrium transition, by H_{ss}^u (full up triangles). Also shown is H_{on}^u (open up triangles). The irreversibility field H_{irr} (full circles) is assumed to correspond roughly to the transition line between glass and fluid phases. Both H_{ss} and H_{irr} decrease monotonically with increasing temperature. For the crystal with $x=0$ the region of a possible tricritical point was not measured, and is blurred in the figure. For the crystal with $x=0.64$ a tricritical point could not be detected and would have to be located very near T_c .

The resulting phase diagrams for $\text{YBa}_2\text{Cu}_4\text{O}_8$ and $\text{YBa}_{1.36}\text{Sr}_{0.64}\text{Cu}_4\text{O}_8$ are contrasted in Fig. 10. The glass phase of $\text{YBa}_{1.36}\text{Sr}_{0.64}\text{Cu}_4\text{O}_8$ has, as compared to $\text{YBa}_2\text{Cu}_4\text{O}_8$, expanded dramatically from a rather small part of the easily experimentally accessible phase diagram to the phase covering the largest 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 disorder into the system. The region of metastability is much smaller for the substituted crystal, indicating that strong disorder tends to reduce overheating and especially undercooling 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.*⁵² observed that, for melting, supercooling exists 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 larger variation of the local transition field for the unsubstituted crystal than for the substituted ones.

IV. CONCLUSIONS

We investigated the influence of structural disorder, introduced by a substitution of Sr for Ba in $\text{YBa}_2\text{Cu}_4\text{O}_8$, 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 substitution, is random and pointlike, 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 irradiation, 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 its 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 (probably including 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, unsubstituted crystal. That the disorder is still increasing with increasing substitution level for $x > 0.2$ is indicated both by an increasing NQR linewidth,³¹ by increasing critical current densities at low fields, and by the increasing 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.

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 initially increase with increasing disorder, and the critical current density increases by an order of magnitude upon substituting 10% Sr. But they then show a saturation behavior, i.e., the pinning force density and critical current densities can be raised only up to a certain point by introducing random pointlike disorder. This may be relevant for possible applications where high critical current densities are required.

It should be noted that current detailed theories of the order-disorder transition generally assume the disorder to be very weak. Our measurements indicate that in systems with a high amount of disorder, the phase diagram, and in particular the order-disorder transition, differ qualitatively from the weak disorder case. Theoretical investigations beyond the weak disorder limit would be very helpful.

Both the pinning of unsubstituted Y124, and the pinning of Sr substituted Y124 is likely to be of δT_c , rather than δl , type, indicated by the temperature dependence of the order-disorder transition at intermediate temperatures. For the substituted crystals this can be expected, since there are variations of T_c upon Sr substitution. However, the exponential-like temperature dependence of the order-disorder transition of substituted crystals at low temperatures is also emphasized. The dependence cannot be linked to the influence of surface barriers, at least not in any straightforward way, the same dependence of all significant fields (onsets and clearly discernible kinks for both field directions) suggests it is a true bulk property of the transition. We stress that the observed temperature dependence is at odds with currently published theoretical formulas.

Note added in proof. Recent measurements⁵³ on $\text{La}_{2-x}\text{Sr}_x\text{CO}_4$ found a temperature dependence of H_{ss} similar to the one reported here on Sr substituted Y124. The authors propose a crossover between disorder-induced and thermal melting as an explanation for the unusual T dependence. We should point out, however, that in our case the unusual upturn of $H_{ss}(T)$ at low T is stronger in crystals with higher disorder, contrary to what is expected by the model proposed in Ref. 53.

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