Anomalous behavior of the second magnetization peak in $La_{1.81}Sr_{0.19}CuO₄$ single crystals: **Possible influence of two-band superconductivity**

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We report an anomalous temperature *T* variation of the field H_{on} for the onset of the second magnetization peak in La_{1.81}Sr_{0.19}CuO₄ single crystals with the external magnetic field *H* oriented parallel to the *c* axis. While the peak field H_p has a continuous decrease with increasing *T*, H_{op} exhibits a sudden decrease for $T \sim 11-15$ K. This behavior appears to be related to the particular *T* dependence of the superfluid density in the case of two-band superconductivity affecting the *T* variation of the elastic energy of the vortex system at low *H*.

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The widely investigated vortex phase diagram of hightemperature superconductors (HTSC) is directly related to the fundamental superconductor parameters, such as the magnetic penetration depth λ , the coherence length, the pinning strength, and the anisotropy factor γ .^{[1](#page-3-0)-4} Consequently, any particular change in these parameters should influence the shape of the transition lines. In clean HTSC the vortex system at low *T* organizes itself into a lattice, which melts at high T through a first-order transition,^{1,[2](#page-3-2)} whereas the vortex phase diagram of HTSC with pinning is dominated by an order-disorder transition[.3](#page-3-3)[,4](#page-3-1)

Significant progress for the understanding of the vortex phase diagram of HTSC in the presence of pinning has been made by considering the competition between the thermal energy, the pinning energy generated by the quenched disorder E_n , and the elastic energy of the vortex system E_{e^1} . If the thermal energy is small compared to E_{el} and E_p , when E_p overcomes *E*el a quenched-disorder-driven transition between the quasiordered vortex solid at low H (the Bragg glass, stable against dislocation formation) and a high-field disor-dered vortex phase is expected.^{3,[4](#page-3-1)} This is accompanied by the appearance of a second magnetization peak (SMP) , $5-7$ $5-7$ an increase in the absolute value of the irreversible magnetization with increasing *H* between H_{on} and H_p , well below the upper critical field. The proliferation of dislocations above H_{on} allows a better accommodation of vortices to the pinning centers. For static conditions, the order-disorder transition line at low *T* is roughly described by the equality $E_p(T,H)$ $=E_{\text{el}}(T,H).$ ^{[8](#page-3-6)}

There is no consensus yet about the nature of the SMP in La2−*x*Sr*x*CuO4. Since the superconductor parameters vary relatively slowly with *T* well below the critical temperature T_c , the SMP line derived from the above energy balance equation cannot have a strong *T* dependence in the low-*T* domain[.5](#page-3-4) However, the SMP line determined in standard dc magnetization measurements has a pronounced upward curvature even at low *T*. This behavior was explained by postulating that both thermally and quenched-disorder-induced fluctuations contribute to the destruction of the Bragg glass in a wide T range⁹ or by considering the square to rhombic vortex lattice transition as the source for the SMP.¹⁰ Alternatively, since in static conditions E_p is rather large (at least for *H* parallel to the *c* axis) and at low *T* it overcomes by far the thermal energy, it was argued in Ref. [11](#page-3-9) that the upward curvature in the *T* dependence of H_{on} and H_p in the low-*T* region is a dynamic effect caused by the finite current in the specimen during experiments, which reduces the effective pinning. In the dynamic condition characteristic to magnetization measurements, the appropriate energy balance relation should be

$$
U_p(J,T,H) \propto E_{\rm el}(T,H) \propto \lambda^{-2} H^{-1/2},\tag{1}
$$

where *J* is the density of the macroscopic currents induced in the sample, U_p (*J*,*T*,*H*) is the effective pinning energy, and λ is the in-plane magnetic penetration depth (with λ^{-2} proportional to the superfluid density appearing in E_{el} through the energy scale for the vortex line tension). In standard dc magnetization measurements *J* increases with decreasing *T* in the low-*T* range due to a lower overall relaxation in the time interval between the moment when *H* became stable and the moment at which the magnetization was measured. Following the behavior of the activation energy barrier in the vortex-creep process, U_p is a decreasing function of *J*. Equa-tion ([1](#page-0-0)) can thus explain the upward curvature of the orderdisorder transition line at low *T* in HTSC with pinning.

FIG. 1. Main panel: dc magnetization curves $M(H)$ of $La_{1.81}Sr_{0.19}CuO₄ single crystals upon increasing H (oriented paral$ lel to the *c* axis) around the onset field H_{on} (indicated by an arrow) for *T* between 6 and 25 K (with the step in *T* of 1 K). A careful data inspection shows the intersection of the $M(H)$ curves for *T* \sim 11–15 K and $H \sim$ 10–17 kOe leading to a nonmonotonous $M(T)$, as illustrated in the inset.

Here we report an anomalous decrease in H_{on} with increasing *T* between \sim 11 and \sim 15 K for La_{1.81}Sr_{0.19}CuO₄ single crystals with *H* oriented parallel to the *c* axis. Our analysis of the nature of the SMP in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ indicates that the observed behavior can be associated with the existence of an inflection point in the *T* dependence of $\lambda^{-2}(T)$ in the above *T* range, as reported in Ref. [12,](#page-3-10) affecting the *T* variation of the elastic energy of the vortex system at low *H*.

The investigated overdoped single crystals $(x \approx 0.19)$ were grown by the traveling solvent floating zone technique.^{[13](#page-3-11)} The magnetization *M* was measured with *H* applied in zero-field cooling conditions and oriented along the *c* axis using a commercial Quantum Design magnetic property measurement system. The onset of the diamagnetic signal at low *H* occurs at $T_c \approx 29$ K and the transition width is around 1 K. In the (H, T) domain considered below, M was identified with the irreversible magnetization. The magnetization relaxation $M(t)$ was registered with the magnet in the persistent mode, the relaxation time *t* was considered to be zero when magnet charging was finished, and the first data point was taken at $t = t_1 = 60$ s.

Figure [1](#page-1-0) (main panel) illustrates the dc magnetization curves $M(H)$ around H_{on} for *T* between 6 and 25 K obtained upon increasing *H* with the magnet in the hysteresis mode. For every $M(H)$ curve set, the step in H was the same $(1 \text{ or } 1)$ 0.2 kOe in our experiments). As can be seen, for *T* \sim 11–15 K and $H \sim$ 10–17 kOe, *M* is almost independent of *T*. This should appear as a "plateau" in the *T* variation of the real part of magnetic susceptibility, which has been observed for different doping levels in the overdoped region of $La_{2-x}Sr_xCuO_4$.^{[14](#page-3-12)} A careful data inspection reveals the inter-

FIG. 2. Main panel: The evolution of the SMP at *T*= 20 K around the peak field H_p (indicated by an arrow) with increasing relaxation level (decreasing the current density J in the sample). We included here the data from the magnetization relaxation curves. At high relaxation levels H_p is shifted to lower values. When SMP is registered with a magnet in the hysteresis mode, $H_p \sim 11$ kOe and decreases to \sim 9 kOe after a relaxation time $t \sim 1600$ s. The inset shows the normalized vortex-creep activation energy U^* vs $1/J$ (log–log plot), with $U^* = -Td \ln(t)/d \ln(|M|)$ determined from the magnetization relaxation $M(t)$ measured in a *t* interval of \sim 5000 s $(H=15)$ kOe and $T=20$ K). The plastic creep behavior clearly appears for $H > H_p$, and the exponent $p = -0.5$. The dashed line represents the fit of the data with the plastic creep $U^*(J)$ relation (see text).

section of the $M(H)$ curves, leading to a nonmonotonous $M(T)$ in the above (H, T) domain [significantly below the $H_p(T)$ line] as illustrated in the inset.

It is not yet decided what kind of quenched disorder plays the decisive role for the occurrence of the SMP in La2−*x*Sr*x*CuO4. There is a widespread belief that twin boundaries are responsible for the pronounced SMP appearance when *H* is oriented along the *c* axis. Alternatively, the possible (charge) phase separation should be considered¹⁴ since this can supply a relatively strong δT_c pinning, accounting for the occurrence of the SMP with a decreasing $H_p(T)$ even for *T* very close to T_c .

First we show that the SMP in $La_{1.81}Sr_{0.19}CuO₄$ enters the quite general class based on the destruction of the Bragg glass, where the dynamic effects are included in Eq. (1) (1) (1) through $U_p(J)$. Figure [2](#page-1-1) (main panel) illustrates the evolution of H_p with increasing relaxation level (decreasing J in the sample) at $T=20$ K. The shift of H_p to lower values at high relaxation levels is obvious and this was proved for *T* = 15 K and even close to T_c (at $T = 26$ and 27 K). The same behavior was reported for $H_{on}^{9,11}$ $H_{on}^{9,11}$ $H_{on}^{9,11}$ $H_{on}^{9,11}$. Since the SMP occurs when the decrease in the pinning energy with increasing *H* is slower than $E_{el}(H)$, ^{[15](#page-3-13)} the shift of the SMP to lower *H* values at low J is in agreement with Eq. (1) (1) (1) .

The scenario for the appearance of the SMP through

FIG. 3. Main panel: *T* variation of the peak field H_p and of the absolute value of the peak magnetization $|M_p|$ for increasing *H*, and the onset field $H_{on}(T)$ for increasing $H(H_{on\uparrow})$ and decreasing $H(H_{on}$). The inset illustrates the *T* dependence of the absolute value of the magnetization at the onset field $|M_{on}|$ for increasing *H* and that of the remnant magnetization M_r . Although H_p , $|M_p|$, and M_r decrease continuously with increasing T , an anomalous decrease in $H_{on}(T)$ for *T* between \sim 11 and \sim 15 K occurs. The latter generates a double slope change in $M_{on}(T)$. For a better comparison, $4\pi|M_p|$ (in the main panel) and $4\pi M_r$ (in the inset) were multiplied by convenient factors.

the continuous destruction of the Bragg glass between H_{on} and H_p implies that for $H > H_p$ the vortex-creep process should be plastic.^{15[–18](#page-3-14)} Here $M(t)$ is very helpful if one determines the normalized vortex-creep activation energy $U^* = -Td \ln(t)/d \ln(|M|)$. The meaning of U^* appears using the parametrization of the actual vortex-creep activation energy *U* from Ref. [19:](#page-3-15) $U(T, H, J) = (U_c/p)[(J_c/J)^p - 1]$, where U_c is the characteristic pinning energy and the exponent p is identified with the (positive) collective pinning exponent μ in the case of elastic (collective) vortex creep, 20 and $p < 0$ for plastic creep. With the above equation for *U* and keeping $J(\propto|M|)$ as explicit variable, one can derive $U^*(J)$ using the general creep relation²¹ $U = T \ln(t/t_0)$, where t_0 is the time scale for creep.²² For the elastic-creep domain one obtains $U^*(J) = U_c(J_c/J)^\mu$, whereas in the plastic creep regime $U^*(J) \propto (J_c/J)^p$ (with an opposite variation with *J*).

The inset of Fig. [2](#page-1-1) shows U^* vs $1/J$ for $H=15$ kOe and $T=20$ K, with U^* determined from $M(t)$ registered in a *t* interval of \sim 5000 s. *J* was extracted from |M| with the Bean model.[23](#page-3-19) The plastic creep behavior clearly appears for *H* H_p and the exponent *p* practically coincides with the plastic creep exponent proposed in Ref. 17 ($p = -0.5$). The above results strongly support Eq. (1) (1) (1) as the starting point for the interpretation of the SMP in $La_{2-x}Sr_xCuO_4$.

Figure [3](#page-2-0) (main panel) illustrates $H_p(T)$, the *T* variation of the absolute value of the peak magnetization $|M_p|$ for increasing *H*, and $H_{on}(T)$ for increasing and decreasing *H* in the case of $La_{1.81}Sr_{0.19}CuO₄$ single crystals. The inset shows

the *T* dependence of the absolute value of the magnetization at the onset field $|M_{on}|$ for increasing *H* and that of the remnant magnetization M_r . As can be seen, H_p , $|M_p|$, and M_r decrease continuously with increasing *T*, whereas H_{on} exhibits an anomalous decrease with increasing *T* between \sim 11 and \sim 15 K. The latter generates a double slope change in $M_{on}(T)$, which is easily understood in terms of the energy balance equation. The possible origin of the anomalous shift of H_{on} to lower *H* values (which can explain the peculiar evolution of the SMP form Fig. 1) is discussed below.

It is tempting to connect the anomalous $H_{on}(T)$ decrease from the main panel of Fig. [3](#page-2-0) to a pinning increase at the two-dimensional (2D)–three-dimensional (3D) crossover recently considered for $La_{1.81}Sr_{0.19}CuO₄$ single crystals in Ref. [24.](#page-3-21) As known, this crossover appears when the coherence length along the *c* axis overcomes the interlayer spacing *s*. This should manifest for *H* up to the 3D–2D crossover field $B_{2D} \sim \Phi_0 / \gamma^2 s^2$.^{[22](#page-3-18)} With the reported γ value for $La_{1.81}Sr_{0.19}CuO₄$ [~10 (Ref. [25](#page-3-22))] B_{2D} is roughly one order of magnitude higher than H_p for $T \sim 11-15$ K. However, H_p , M_p , and M_r in Fig. [3](#page-2-0) show no sudden change. In the dynamic conditions characteristic to dc magnetization measurements the effect of a change in the pinning strength (for static conditions) on $U_p(T)$ will be smeared out due to the opposite contribution of $J(t_1, T)$ to $U_p(T)$. This assertion results directly from the general creep relation, 21 with $U_p[J(t_1), H, T] = U[J(t_1), H, T] = T \ln(t_1/t_0)$, and is confirmed by the fact that $U^*(T)$ determined from the relaxation of M_r (not shown) increases continuously with T in the T range of interest (as expected for elastic vortex creep well below T_c).

Besides the pronounced shift of H_{on} to lower values with increasing *T* between \sim 11 and \sim 15 K, the decrease in the magnetization relaxation rate with increasing *H* above H_{on} may also contribute to the peculiar evolution of the SMP from Fig. [1.](#page-1-0) However, it is impossible to explain the anomalous $H_{on}(T)$ decrease from the main panel of Fig. [3](#page-2-0) in terms of magnetization relaxation effects. The continuous $U_p(T)$ variation (discussed above) and Eq. (1) (1) (1) suggest that the $H_{\text{on}}(T)$ anomaly could be related to a rapid decrease of E_{el} for $T \sim 11-15$ K due to a particular $\lambda^{-2}(T)$ variation. This was recently reported for $La_{1.81}Sr_{0.19}CuO₄$, where muon spin rotation experiments revealed an inflection point in $\lambda^{-2}(T)$ for T around $10-15$ K, associated with the presence of two superconducting gaps (with d -wave and s -wave symmetry).^{[12](#page-3-10)} Such an interpretation is supported by the fact that a well developed inflection point in $\lambda^{-2}(T)$ appears at low *H* (up to a few kilo-oersted),^{[12](#page-3-10)} mainly affecting $H_{on}(T)$.

In summary, we have observed an anomalous $H_{on}(T)$ decrease in the *T* interval between \sim 11 and \sim 15 K for $La_{1.81}Sr_{0.19}CuO₄$ single crystals with *H* oriented along the *c* axis. The analysis of vortex dynamics across the SMP and the use of the energy balance relation for dynamic conditions suggest that this is caused by a sudden decrease in the elastic energy of the vortex system in the above *T* range confirming the existence of an inflection point in the *T* variation of the superfluid density.¹²

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