Small Angle Neutron Scattering and Magnetization Measurements in the Cubic $(\mathrm{K,Ba})\mathrm{BiO}_3$ Superconductor

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We report on both small angle neutron scattering and magnetization measurements in the *cubic* $(\mathrm{K,Ba})\mathrm{BiO}_3$ superconductor. We show that the flux lines are ordered into a hexagonal lattice at low field ($B < B_e \sim 0.6$ T) and that the diffracted intensity continuously drops to zero as the magnetic field is increased towards B_e. This intensity fall is attributed to a static Debye-Waller factor associated with disorder induced longitudinal fluctuations of the vortex lines. We show that B_e lies close to the onset of the anomalous "peak effect" in magnetization measurements, suggesting that this peak is related to the change in the structure of the vortex solid. [S0031-9007(99)09428-4]

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One of the most intriguing features that has been observed in high T_c superconductors is the presence of peak effect in the critical current located at a field much smaller than the upper critical field H_{c2}. The origin of this peak remains very controversial as it is still unclear if it should be associated to some intrinsic change in the vortex structure or rather to an inhomogeneous relaxation of the *apparent* critical current. The situation is even more complicated in highly anisotropic cuprates as one has to take into account a possible decomposition of the vortices into 2D pancakes at high fields. However, this peak effect has also been observed in the perfectly isotropic (cubic) $(\mathrm{K,Ba})\mathrm{BiO}_3$ system [1] ($T_c \sim 32$ K for optimally doped samples) and is thus not directly related to the quasibidimensionality of the cuprates. In order to check whether this peak is indeed associated to some change in the vortex structure we have thus performed both small angle neutron scattering (SANS) and magnetization measurements on the same single crystal. SANS measurements are a very powerful tool in the investigation of the nature of the vortex structure [2–5] and have, for instance, been used to get a detailed description of the evolution of the vortex morphology as a function of the applied magnetic field in the region of the peak effect in Nb single crystals [6].

The SANS experiment was carried out on the D11 line at the ILL-Grenoble on a large (~300 mg) single phased nonoptimally doped crystal grown by electrochemical crystallization ($T_c \sim 23.3$ K). The magnetic field was parallel to the incident neutron beam and the [111] axis of the crystal. The penetration length (λ) and superconducting coherence length (ξ) have been deduced from the reversible part of the magnetization loops: $\lambda(0) \sim 3600 \pm 300$ Å and $\xi(0) = 45 \pm 5$ Å [i.e., $H_{c2}(0) \sim 16$ T] in this sample. This λ value is very similar to the one which has been reported recently in this system [7].

As shown on Fig. 1 (at $T = 3$ K), the SANS experiment confirmed the existence of a hexagonal flux line lattice (FLL) in the $(\mathrm{K,Ba})\mathrm{BiO}_3$ system at low field. However, the intensity of the diffraction peaks drops down very rapidly with increasing field. For a perfect FLL, the intensity of the (1,0) peak obtained by integrating over the sample and scattering angles (i.e., by rocking the sample through the Bragg condition) is given by [2]

$$I_{10} = 2\pi\phi(\mu/4)^2 \frac{V\lambda_n^2}{\Phi_0^2 q_{10}} F_{10}^2 , \qquad (1)$$

where ϕ is the incident neutron flux, μ is the magnetic moment of the neutron in nuclear magnetons (= 1.91), V is the volume of the sample, λ_n is the neutron wavelength (= 10 Å), Φ_0 is the flux quantum, q_{10} is the (1,0) reciprocal lattice vector, and F_{10} is the form factor of the (1,0)

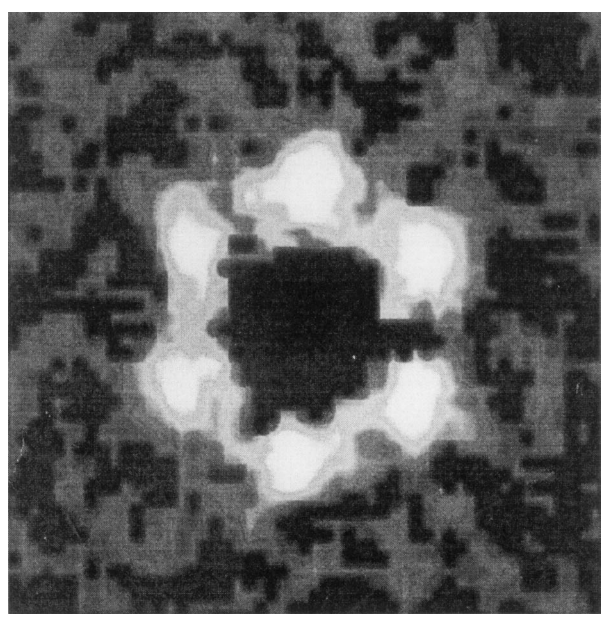

FIG. 1. Neutron diffraction pattern from the flux line lattice in $(\mathrm{K,Ba})\mathrm{BiO}_3$ at $T = 3$ K and $H = 0.1$ T.

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reflection which is related to λ through

$$F_{10} = \frac{B}{1 + (q_{10}\lambda)^2} \,. \qquad (2)$$

As already pointed out by Cubitt *et al.* in BiSrCaCuO [4], we did not observe any change in either the width of the rocking curve ($\sigma \sim 0.28° \pm 0.02°$) or the transverse or radial size of the diffraction spots (within the accuracy of our data) but only a fall in the amplitude of the peak and, as shown in Fig. 2, even after correcting for the $1/q_{10}$ field dependence of I_{10}, the intensity still drops rapidly with field. This fall has been first attributed to a decomposition of the vortices into 2D pancakes in BiSrCaCuO [4]. However, such an explanation can obviously not hold in our cubic system, and we have thus assumed that this field dependence is due to a static Debye-Waller factor associated with the fluctuations of the vortex lines in the presence of point disorder. The diffracted intensity will thus be given by [8]

$$I_{\mathrm{mes}} = I_{10} \exp(-2\pi^2 \langle u^2 \rangle / a_0^2) \,, \qquad (3)$$

where a_0 is the lattice spacing and $\langle u^2 \rangle$ the mean square displacement of the vortices which may include both fluctuations in the position of the vortex lines from their ideal positions and uncorrelated fluctuations (wiggling) along the vortex lines [9]. The magnetic field dependence of $\langle u^2 \rangle^{1/2}/a_0$ is shown on Fig. 3: The solid triangles have been obtained from Eqs. (1)–(3) taking $\lambda = 3600$ Å, whereas the open triangles (dotted line) would correspond to $\lambda = 4300$ Å, i.e., assuming that the FLL is perfectly well ordered at 0.1 T. We have also reported on Fig. 2 the $\langle u^2 \rangle^{1/2}/a_0$ values deduced from Eq. (3) for BiSrCaCuO [4] and YNi_2B_2C [5] samples assuming that $I_{\mathrm{mes}} = I_{10}$ at low field (the field axis has been rescaled to the field B_e corresponding to a vanishingly small diffracted intensity). As shown, for all samples, $\langle u^2 \rangle^{1/2}/a_0$ increases with field, and the diffracted intensity finally drops beyond the experimental resolution for $\langle u^2 \rangle^{1/2}/a_0 \geq 0.3$. As mentioned above, the difference at low fields may be related to an ill-defined λ value but is most probably due to the presence of fluctuations in our sample even at low field. Note that $I_{\mathrm{mes}}(T, B)/I_{\mathrm{mes}}(T = 0, B)$ is field independent on a large temperature range (small deviations can be observed at high temperature due to the decrease of the entanglement field with T—see below) indicating that the Debye-Waller factor is mainly static in our system. However, as shown in the inset of Fig. 2, $I_{\mathrm{mes}}(T) \sim 1/\lambda(T)^4$ decreases faster than the predictions of the two fluid model (dotted line) and, as already observed by Yethiraj *et al.* in YBaCuO samples [3], $I_{\mathrm{mes}}(T)$ can actually be well described by a $[1 - (T/T_c)^n]^2$ law with $n \sim 2.3$ (solid line) instead of $n = 4$ in the two fluid model.

It has been suggested [10] that the formation of dislocations may become energetically favorable for $\langle u^2 \rangle^{1/2}/a_0 > c$, where c is a numerical factor of the order of $\sim$0.2–0.3 (in analogy with the Lindemann number) in good agreement with our experimental value. In the so-called single vortex regime (i.e., at low fields), this condition is satisfied when the magnetic field reaches the entanglement field B_e which is of the order of [10]

$$B_e \sim c^5 \, \frac{\Phi_0}{\xi^2} \left(\frac{L_c}{\xi} \right)^3 , \qquad (4)$$

for isotropic samples (L_c is the collective pinning length i.e., $u \sim \xi$ for a vortex length $L \sim L_c$). Taking $c \sim$ 0.25–0.3 and $B_e \sim 6000$ G, one gets an estimation of the

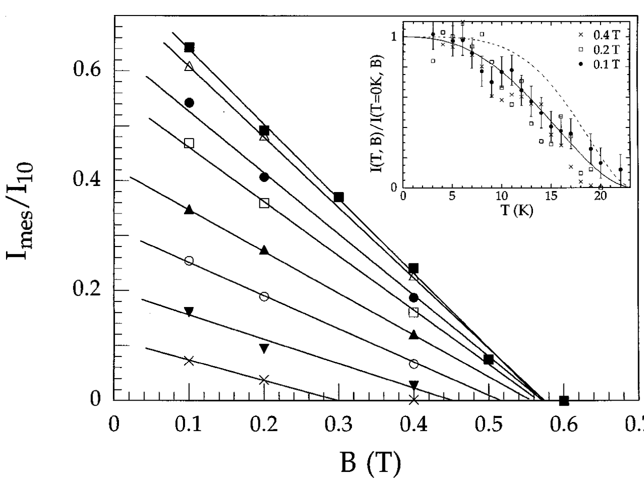

FIG. 2. Field dependence of the diffracted intensity after correcting for the $1/q_{10}$ field dependence in Eq. (1) (from the top to the bottom $T = 3, 5, 8, 10, 13, 15, 18, 20$ K—the lines are a guide to the eyes). In the inset: Temperature dependence of the diffracted intensity renormalized at $T = 0$ K at $H = 0.1$, 0.2, and 0.4 T; the dotted line is a two fluid model fit to the data and the solid line a $[1 - (T/T_c)^{2.3}]^4$ fit.

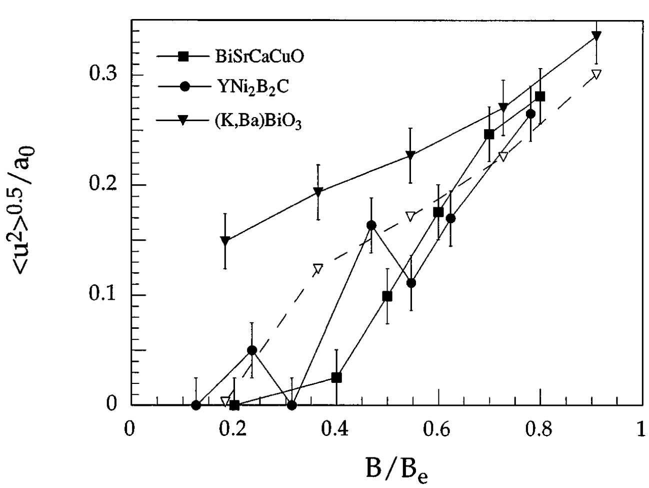

FIG. 3. Magnetic field dependence of the vortex displacement $\langle u^2 \rangle^{1/2}/a_0$ deduced from Fig. 2 using Eq. (3) (the lines are a guide to the eyes) for (K, Ba)BiO_3 (solid triangles, $\lambda = 3600$ Å; and open triangles, $\lambda = 4300$ Å—see text for details), YNi_2B_2C from [5] (circles) and BiSrCaCuO from [4] (squares).

collective pinning length $L_c \sim 70-90$ Å at low temperature. This small L_c value shows that there is a rather large amount of weak pinning centers in our sample which are probably related to an inhomogeneous potassium distribution. Note that this entanglement transition can be observed either for large disorder, i.e., small L_c values [(K, Ba)BiO$_3$] or a large anisotropy (BiSrCaCuO [11] or NdCeCuO [12]), and may actually be shifted towards very high fields in high quality YBaCuO single crystals.

The fluctuations can be divided into two types of displacements [9]: fluctuations in the position of the vortex lines and *uncorrelated* wiggling along the vortex line. However, modifications of the lattice parameter are expected to show up in the width of the rocking curve as a $[2\theta(\delta a_0/a_0)]^2$ contribution [13] to σ^2 (where θ is the diffraction angle) in clear disagreement with our experimental data (σ = const $\sim 0.28° \pm 0.02°$). This suggests that, in our case, u is related to uncorrelated wigglings of the vortex lines which show up as an "apparent" field dependent λ. The characteristic longitudinal fluctuation length L_0 can be deduced from u in the single vortex regime assuming that [14] $\langle u^2\rangle^{1/2} \sim \xi(L_0/L_c)^{0.6}$ and one gets $L_0 \sim a_0$ for our experimental u, ξ, and L_c values in good agreement with [10] ($L_0 \sim 2\epsilon a_0$). Note that L_0 is much shorter than the longitudinal correlation length ξ_L which can be deduced from the width of the rocking curve: $\xi_L \sim 1/q_{10}\sigma \sim 30a_0$.

The field B_e can be estimated by extrapolating the diffracted intensity linearly to zero. As shown on Fig. 2, B_e is almost temperature independent on a large temperature range and finally decreases in the vicinity of T_c. A temperature independent entanglement field has actually been obtained by Monte Carlo simulation in the so-called 3D-*XY* regime (by analogy with random ferromagnets) [15]. However, neglecting thermal fluctuations which would smooth out the static disorder, B_e is expected to vary as L_c^3/ξ^5 [see Eq. (4)]. As pointed out by Giller *et al.* in NdCeCuO crystals [12], a decrease of B_e is consistent with the δ-T_c pinning model (i.e., pinning induced by fluctuations in the critical temperature) for which $L_c \sim \xi^{2/3}$ and $B_e(T) \sim 1/\xi(T)^3$ (in contrast with the δ-l pinning regime for which B_e is expected to increase with field: $B_e \sim \xi$ [14]). As an example, we have reported on Fig. 4 a fit to our data using Eq. (4) with $\xi(T) = \xi_0/[1 - (T/T_c)^4]^{1/2}$ [12]. Note that Eq. (4) has been derived in the single vortex regime and, even if it will probably provide a good estimation of B_e, its application in our system may not be completely correct since $L_0 \sim a_0$ and interactions between vortices should thus not be neglected anymore.

Finally, we are comparing the magnetic field B_e obtained from the SANS experiment to the field corresponding to the onset of the increase in the current density J deduced from the width ΔM of the magnetization loops. Indeed, we have shown [16] that the nonlinear response of the ac susceptibility can be very well described by the bulk pinning model and ΔM is thus directly proportional to the bulk critical currents [17]. As shown on Fig. 5, the central peak can be well described by a power law behavior, and the field B_e deduced from the SANS experiment coincides very well with the onset of the deviation from this power law dependence. Even if it is much more difficult to define a criterium for the increase of J in the (K, Ba)BiO$_3$ system than it is in BiSrCaCuO [11] and NdCeCuO [12] (for which the magnetization loops reveal a very sharp onset of the current density), the field B_e clearly lies in the vicinity of the peak anomaly. Note also that magnetic loops are nonequilibrium measurements which may be affected by creep phenomena leading to a time dependent position of the peak. However, we have shown that the shape of

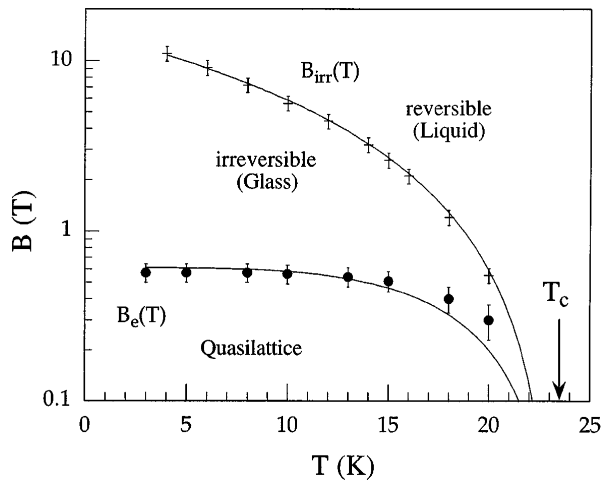

FIG. 4. Low field part of the H-T phase diagram of the (K, Ba)BiO$_3$ system. The field B_e (circles) is obtained by extrapolating the diffracted intensity to zero [the solid line is a fit to the data following Eq. (4)] and the irreversibility field B_{irr} (crosses) is obtained from the magnetization loops [B_{irr} corresponds to $\Delta M = 1/1000$ of its maximum value—the solid line is a $(1 - T/T_c)^{1.6}$ fit to the data].

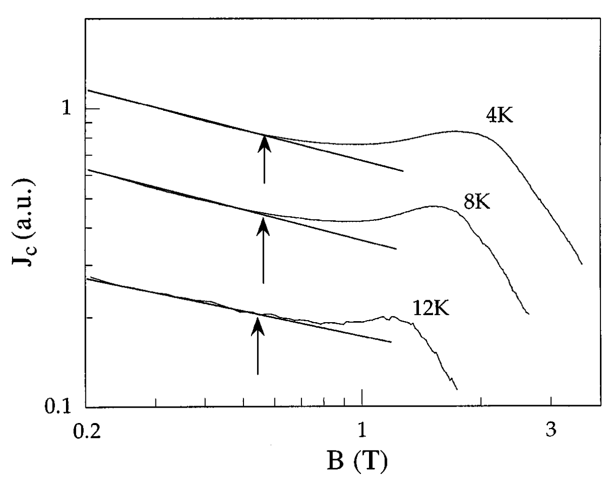

FIG. 5. Magnetic field dependence of the current density at $T = 4$, 8, and 12 K. The arrows are marking the position of the field B_e corresponding to the complete loss of diffracted intensity in the SANS experiments.

the magnetization curves becomes independent of the experimental time scale below $\sim T_c/2$ in (K, Ba)BiO_3 [1]. Similarly, it has been shown recently [18] that the peak may split into two different peaks between $\sim$70 and 80 K in YBaCuO: P_h (high field peak) and P_l (low field peak), where P_l is visible up to T_c in twinned samples and is related to a change in the vortex dynamics, whereas P_h is independent of the time scale (i.e., probably related to B_e) and tends towards the so-called critical point of the H-T phase diagram as usually observed in BiSrCaCuO samples [11]. The presence of this critical point is an important feature of the H-T phase diagram in cuprates as it marks the crossover from a first order lattice-liquid transition at low fields to a second order glass-liquid transition at high fields. However, as shown in Fig. 4, this point is absent (i.e., shifted down to T_c) of the H-T phase diagram in the (K, Ba)BiO_3 system. There is thus no direct transition from the solid to the liquid in agreement with our magnetotransport data which could be well described by the vortex glass scaling theory on the *entire* field range [19]. The static disorder thus probably plays a crucial role in the melting process as expected from the rather small thermal fluctuations in (K, Ba)BiO_3 (the Ginzburg number $G_i \sim 10^{-4}$–10^{-5} is intermediate between high T_c oxides and conventional low T_c materials). The H-T phase diagram of Fig. 4 is in very good agreement with recent London Langevin simulations [20] and may be characteristic of disordered systems. B_{irr} has been deduced from the magnetization curves, and we have shown in other samples that this field coincides very well with the vortex glass transition line in (K, Ba)BiO_3.

The evolution of the morphology of the vortex structure with field in our system is actually quite different from the one that has been observed in Nb single crystals [6]. Indeed, in this latter case, it has been shown that the in-plane positional order is first completely lost close to the minimum of J_c (the vortex phase becomes hexatic) and that the orientational order is finally lost at the location of the peak but the corresponding amorphous phase is still constituted of rigid lines. In (K, Ba)BiO_3 we did not observe any significant change in the shape of the diffraction spots but only a fall in the intensity which is completely lost close to the onset of the peak due to the *entanglement* of vortices.

In summary, we have shown that there exists an ordered hexagonal lattice for $B < B_e(T)$ in the cubic (K, Ba)BiO_3 superconductor. The loss of diffracted intensity with increasing field coincides well with the onset of the increase in the current density deduced from magnetic measurements suggesting that the peak effect is indeed related to a change in the vortex structure.

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