Structure of the flux lines lattice in NbSe₂: Equilibrium state and influence of the magnetic history

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We have performed small-angle neutron scattering of the flux line lattice (FLL) in a Fe-doped NbSe₂ sample which presents a large peak effect in the critical current. The scattered intensity and the width of the Bragg peaks of the equilibrium FLL indicate an ordered structure in the peak effect zone. The history dependence in the FLL structure has been studied using field-cooled and zero–field-cooled procedures, and each state shows the same intensity of Bragg scattering and good orientational order. These results strongly suggest that the peak effect is unrelated to a bulk-disordering transition and confirm the role of a heterogeneous distribution of screening current.

The peak effect in the critical current of a type II superconductor is a long-standing problem in the field of superconductivity. Since its observation,[1,2] no clear consensus has emerged as to its origin. Most models are based on the Pippard idea[3] who attributed the peak effect to the loss of rigidity of the flux lines lattice (FLL), which falls close to B_{c2} more rapidly than the pinning strength due to inhomogeneities. Larkin and Ovchinikov (LO) have treated the complex problem of the elastic FLL in the presence of random pinning centers.[4] The main issues are the statistical calculations of a volume V_c over which the lattice remains correlated and the corresponding pinning force per unit volume $F_p \propto V_c^{-1/2}$. In addition to the intense theoretical interest, there is the possible link between the FLL bulk disorder and the critical current. Other interpretations of the critical current deal with dominant pinning of flux lines by the surface,[5,6] without significant role of FLL bulk order. Recently, the explanation of the peak effect as a genuine phase transition has been addressed both theoretically and experimentally, and the peak effect is now considered as a classical example of an order-disorder transition in an elastic system.[7] A superconductor with a peak effect is remarkable by its anomalous transport properties. They have been tackled by quite thorough series of measurements in NbSe₂, with a large focus on the metastable properties.[8] Two coexisting macroscopic FLL states with different pinning strengths were observed[9] and the kinetics between these two states can explain most of the anomalous transport properties. Paltiel *et al.*[10] have also shown that the critical current in the peak effect zone is heterogeneous and more important close to the edge of the samples. However, the technique of Hall probe does not allow concluding on the structure of the FLL. The genuine nature of FLL states with the two different critical currents remains unknown even if one is usually assumed to be much more disordered following LO approach.[10] However, this latter assumption needs to be experimentally controlled using a structural probe: indeed, a correlation between the critical current and the FLL bulk order cannot be *a priori* assumed, as shown by different small-angle neutron-scattering (SANS) experiments.[11–13] When no change in FLL order is observed whereas the critical current is strongly modified,[12] an interpretation could be that the pertinent structural distortions are at a too small scale to be resolved. However, when important changes in the FLL order are seen without any modification of the critical current,[11,13] it shows more clearly that the related FLL order is not pertinent for the critical current and the dominant pinning mechanism was shown to be at the surface. SANS in the presence of a peak effect has been mostly used in Nb. Measurements show differences in the FLL structure after field cooling and zero-field cooling and it was proposed to arise from superheating and supercooling of metastable states as expected for a genuine first-order (melting) transition.[14,15] Other authors proposed that the results would be better understood involving a crossover into a disordered state.[16] The absence of intrisinc (thermally driven) melting transition in Nb is supported by the persistence of the Abrikosov lattice up to B_{c2} in a purest sample which does not present the peak effect.[17] Fe-doped NbSe₂ systematically displays a marked and extended peak effect, and then was studied as a school case using transport measurements.[8] In addition, no correlation was found between the topology of the FLL structure and the enhancement of the critical current in NbSe₂ using the decoration technique.[18] This makes this sample particularly interesting for a SANS study of equilibrium and metastable FLL in order to clarify their nature and the link with the pinning properties.

Our sample is a large single crystal of 2*H*-NbSe₂ doped with 100 ppm of Fe (T_c=5.80 K), which was used in Ref. 13. SANS experiments were performed at ILL (Grenoble-France) using D22 and D11 spectrometers. The neutron beam was parallel to the magnetic field and no external current was applied. In this geometry, the higher angular reso-
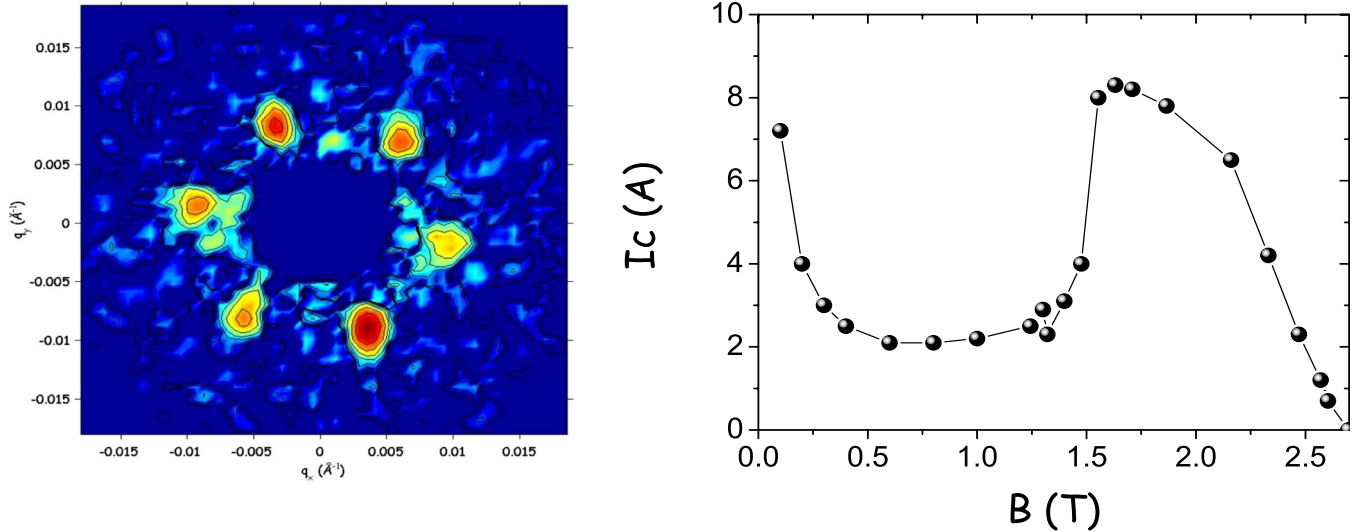
FIG. 1. (Color online) Left: 2D pattern of the ZFC FLL ($B=0.4$ T, $T=2$ K) obtained by rocking the sample through the Bragg conditions and after a subtraction of the normal-state background. Right: critical current $I_c(B)$ measured after ZFC at $T=2$ K in superfluid helium and showing the sharp increase in critical current: the peak effect at $B_{peak} \approx 1.5$ T.

lution is obtained on the rocking curves which measure correlation along the flux lines. The neutron wavelength was $\lambda=8$ and 10 Å with a spread $\Delta\lambda/\lambda \approx 10\%$. Depending on the diffraction vector Q of the FLL Bragg planes and in order to optimize the intensity different collimation and sample-detector distances were used. The beam was collimated in the middle of the sample. Background scattering obtained from the sample in the normal state ($B>B_{c2}$) can be subtracted to reveal the two-dimensional (2D) FLL image (Fig. 1). All the measurements were made at a temperature $T=2$ K using field cooling (FC: magnetic field applied at $T>T_c$) or zero-field cooling (ZFC: magnetic field applied at $T=2$ K) procedures.

A crucial point for a quantitative interpretation of the results is the value of the upper critical field B_{c2}. It was measured using different techniques [magnetization, resistivity, $V(I)$ curves] on a small crystal cleaved from the large one. $V(I)$ curves were also performed on the whole crystal used for SANS measurements. All measurements show a sharp increase in the critical current after ZFC at $B=1.4$ T and a second critical field $B_{c2} \approx 2.55$ T at $T=2$ K, as shown in Fig. 1 (ZFC procedure avoids much of metastability when a peak effect is present). We observe also strong evidences of surface superconductivity for $B>B_{c2}$. One can estimate the field where the peak effect occurs as the midpoint of the abrupt increase in $I_c(B)$. This gives $B_{peak} \approx 1.5$ T, i.e., $B_{peak}/B_{c2} \approx 0.6$ in agreement with previous reports.[19] First series of SANS measurements relate to the structure of FLL in the area of the peak effect to see if there exists a transition toward a disordered phase. Such measurements were made for applied magnetic fields from $B=0.4$ T to 2 T using the ZFC procedure to directly compare with the critical current measurements shown in Fig. 1. For fields larger than $B>2$ T, the scattered intensity from the FLL became too small to measure with counting times of 4 h on D22 but this value is however much higher than B_{peak}. Such measurements always show a hexagonal lattice with sharp Bragg

peaks and a diffraction vector Q corresponding to the applied magnetic field. For each field value, we have measured the full rocking curve to get the scattered intensity of the (1,0) Bragg peak of the FLL. Each rocking curve was fitted by a Lorentzian (Fig. 2) in order to extract the integrated scattered intensity and rocking curve width.
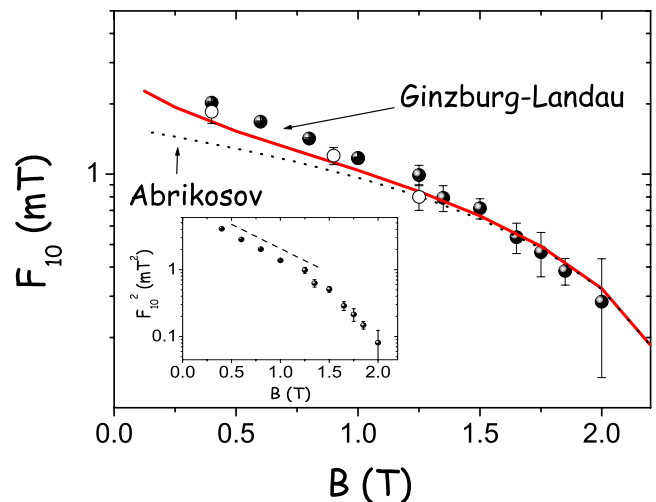
The intensity for a (h,k) reflection is

FIG. 2. (Color online) The full points are the experimental Fourier component F_{10} deduced from the scattered intensity I_{10} of the ZFC FLL and compared to the Abrikosov and to the Ginzburg-Landau models ($B_{c2}=2.5$ T, $\lambda=120$ nm). The empty points are experimental points corresponding to the FC FLL. In the inset is shown the square of the Fourier component to illustrate the expected crossover at $B \approx 0.4.B_{c2}=1$ T from first-order Ginzburg-Landau correction to the London model at low field (shown as a straight line in this scale) to the Abrikosov line.

$$I_{hk} = 2\pi V \phi (\gamma/4)^2 \lambda_n^2/\phi_0 |F_{hk}|^2/Q_{hk}, \qquad (1)$$

where V is the illuminated sample volume, ϕ the incident flux, γ the magnetic moment of the neutron, and ϕ_0 the flux quantum. For a quantitative analysis of the scattered intensity at high fields, the Abrikosov limit can be used in the range $0.4.B_{c2} \leq B \leq B_{c2}$ (Ref. [20]), where the Fourier factor of the hexagonal vortex lattice can be expressed as

$$F_{hk} = (-1)^{\nu} e^{-3^{-1/2}\pi\nu} \mu_0 M, \qquad (2)$$

where $\nu = (h^2 + k^2 + hk)$ and $\mu_0.M = (B - B_{c2})/\beta_a (2\kappa^2 - 1)$ according to the Abrikosov limit ($\beta_a = 1.16$ for the hexagonal lattice, κ is the Ginzburg-Landau parameter).

After normalization to the incident neutrons flux, F_{10} can be extracted from Eq. (1). We choose to fix $B_{c2} = 2.55$ T which was robustly determined, and only one parameter κ is adjustable. F_{10} compares favorably at high fields with the Abrikosov form of the intensity with $\kappa \approx 12$ (Fig. 2), giving $\lambda = \kappa\xi = 136$ nm with $\xi^2 = \phi_0/(2\pi B_{c2}) \approx 128$ nm². To describe the data at the lowest fields, we used numerical corrections from Ginzburg-Landau solution as described in Refs. [20] and [21]. Since it converges to the Abrikosov limit for $B/B_{c2} \geq 0.4$, there is no supplementary fitting and the agreement with the experimental data is satisfactory (Fig. 2). The values of λ and ξ are consistent with the literature considering that previous estimates of λ are quite scattered over the broad range 70–250 nm (Ref. [22] and references herein). The full width at half maximum (FWHM) of the rocking curves was analyzed with the correction of instrumental resolution mainly due to wavelength spread and the beam divergence. It can be analytically estimated as a first approach using a Gaussian approximation.[23] For a peculiar setup, we find, for example, FWHM (resolution)=0.16° for $B = 0.4$ T, to compare with FWHM (rocking-curve)=0.22°, and an intrinsic with $\Delta\omega$ can be extracted.[24] Finally, a longitudinal correlation length $d \approx a_0/\pi\Delta\omega$ is deduced. As shown in Fig. 3, we have found that $d \approx 11 \pm 4$ μm is roughly constant for all field values (the crystal is 200 μm thick). Note that since FWHM is close to the instrumental resolution, the value of d has rather the meaning of a lower bound.[25] This value is however already far larger than the one deduced from a criteria used to indicate the appearance of dislocations in the FFL [the order state is expected to be unstable for $d \leq 20a_0$ (Ref. [26]) and we measure $d > 200a_0$]. Due to the poor resolution in the positional correlation function in our SANS geometry we cannot distinguish between a perfect lattice of flux lines, a Bragg glass state with algebraically decaying translational order,[27] or a multidomains structure which fractures at large scale.[28] Detailed investigations of correlation functions of the FLL using a high-resolution geometry indicate some large scale fracturing of FLL even in best quality Nb samples.[29] We summarize the most important result of this part: our analysis of FLL Bragg peaks stabilized after ZFC shows that the FLL order is unchanged and robust far inside the peak effect where the critical current has strongly increased.

In NbSe₂, FC states show different properties than ZFC states.[8] In particular, they present a large and metastable critical current and are macroscopically heterogeneous even
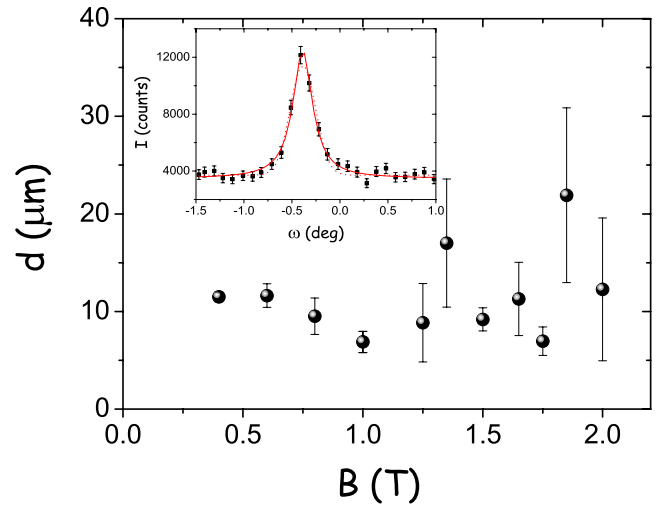

FIG. 3. (Color online) Correlation length d deduced from the rocking curve width of a first-order Bragg peak as function of the magnetic field ($T = 2$ K) after ZFC. In the inset is shown a typical rocking curve better fitted with a Lorentzian ($B = 1.25$ T). No background is subtracted.

for magnetic fields smaller than B_{peak} (Refs. [8] and [13]). This is a quite unusual case since FC leads usually to a more perfect lattice in samples without the peak effect. Comparison between FC and ZFC scattering intensity was here tricky. Rocking the sample through the expected Bragg angle after FC gives a scattered intensity significantly reduced. One might conclude that much of the intensity was incoherently scattered from a strongly disordered-supercooled state. However, as noted in Ref. [30] when measuring CeRu₂, if the Bragg peaks are strongly broadened, all the intensity is not recovered for restricted angular scans. This is also the case if the Bragg angle has changed. Measurements performed with large enough angular scans show that most of the intensity in the FC rocking curve was surprisingly outside the regular Bragg conditions (Fig. 4). The shape of the rocking curve is difficult to extract because it emerges only slightly from the background. However, an analysis with two similar Bragg peaks rotated from the theoretical Bragg angle is most satisfactory. Even if two peaks in the rocking curve indicate a very peculiar FLL structure, the sum of integrated intensity of the two peaks corresponds to the intensity expected for the regular Abrikosov lattice measured after ZFC (Fig. 2).[31] The Q value is also unchanged and its radial width is resolution limited. Since the intensity in the RC is preserved, all the "disorder" should be along the longitudinal direction, without affecting the orientational quality of the FLL. This is consistent with measurements of the orientational width of the Bragg peaks in the plane of the detector. They remain resolution limited ($\Delta\psi = 13 \pm 1°$ in the azimuthal direction) for the FC case (Fig. 5). Due to this relatively poor azimuthal resolution we do not know if the FC FLL presents an orientational order as good as the ZFC FLL. But clearly, the FC FLL is not a supercooled glasslike state which should lead to a degenerated orientational order (a ring of scattering).[14] The double peak structure was first observed in Ref. [13]. It implies that the FLL is tilted from the magnetic-field direc-
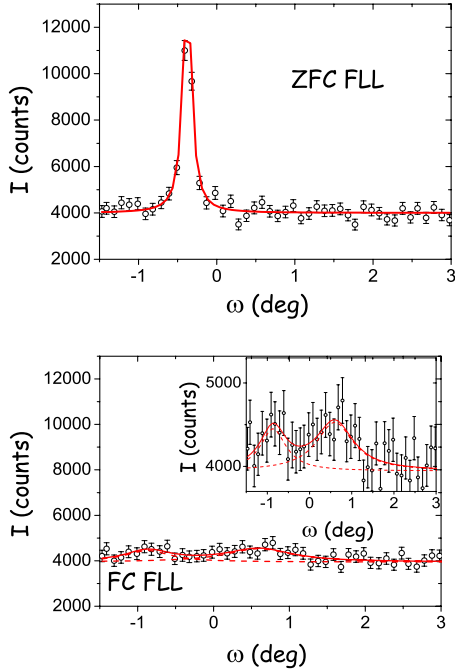
FIG. 4. (Color online) Top: rocking curve measured after a zero-field cooling for $B=0.4$ T. The fit is a Lorentzian with FWHM $=0.13\pm0.01°$. Bottom: rocking curve measured after a field cooling for $B=0.4$ T with the same scale as the top figure. The inset is a zoom showing two Bragg peaks. The fit is with two Lorentzians of FWHM$_1=0.7\pm0.2°$ and FWHM$_2=1.0\pm0.2°$ which are roughly symmetric with respect to the theoretical Bragg angle. No background is subtracted. The Q value is the same for FC and ZFC FLL.

FIG. 5. (Color online) Azimuthal scan for FC and ZFC states ($B=0.4$ T). Despite ω rocking curves are very different, the orientational order remains resolution limited as demonstrated by the mean value $\Delta\psi$ of the peaks FWHM (ZFC: $\Delta\psi=12.98\pm0.50°$, FC: $14.09\pm1.38°$). For the FC case, all the six peaks are observed without performing full scans for all the (h,k) reflections because the lattice is in Bragg condition for an extended angular range.

tion due to magnetic-field components in the plane. A nonsymmetric distribution of superficial screening currents was proposed as a qualitative interpretation.[13] The information obtained here (the preserved intensity of the whole FLL after FC and the absence of bulk transition in the ZFC FLL when crossing the peak) are consistent with this scenario. We have tried the usual "shaking" procedure consisting in a variation in the magnetic field in the form of a sinusoidally damped oscillation in order to improve the FLL spatial order.[32] The effect was only marginal, likely due to the nontrivial nature of the FC FLL structure. Contrarily, a thin Bragg peak centered at the theoretical Bragg angle is recovered when passing a large dc overcritical current in the sample.[13] A possible reason of the negligible effect of the shaking is that the screening currents are too large to be affected by the currents induced by the field oscillation. Another one is that the screening currents have to be polarized in one direction to recover a homogeneous distribution and a regular FLL. Note that our results appear consistent with the edge contamination model[10] and with the observation of two coexisting macroscopic vortex states,[9] to the extent that the supercooled state with the high critical current stabilizes surface currents only and is unrelated to a bulk transition. Such nonhomoge-
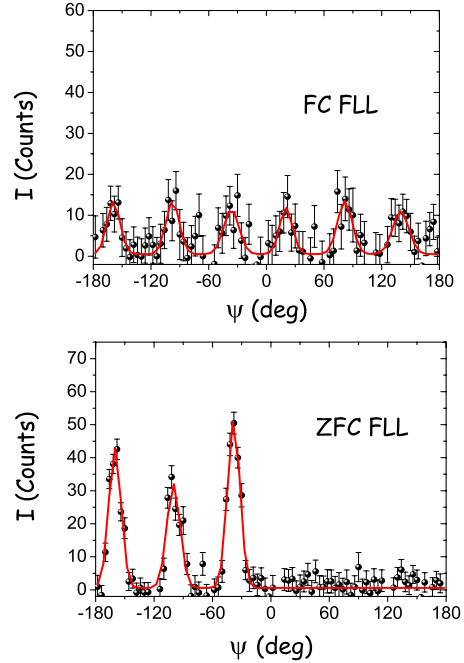
neous distribution of surface currents is also consistent with the complicated current flow necessary to induce the interface dynamics between the coexisting FLL states.[9] The origin of two competing surface currents is unclear but may arise from a coexistence of two separate mechanisms contributing to the surface transport current in the mixed state. Such a scenario was proposed some years ago from angular magnetotransport measurements coupled with surface treatments in conventional type II superconductors[33] and deserves a peculiar attention.

In conclusion, we have studied the FLL structure in a Fe-doped NbSe$_2$ crystal which shows a large peak effect in the critical current. The ZFC FLL is always ordered. The FC FLL is notably different but does not present the characteristics of a bulk, glassylike, and disordered state. These results are extremely difficult to conciliate with a fracturing or melting transition of the FLL usually proposed as the cause of the peak effect and calls for alternative interpretations. A direct consequence of our SANS study of FLL structures is that the peak effect may correspond to an anomalously large surface or boundary current developing in the peak. It can be frozen over macroscopic zones of the sample and leads to the two-states mixture responsible for the metastable transport measurements.

[1] M. A. R. Le Blanc and W. A. Little, Proceedings of Seventh International Conference Low Temperature Physics, edited by G. M. Graham and A. C. Hollis Hallett (Toronto University Press, Toronto, 1961), Vol. 362.

[2] T. G. Berlincourt, R. R. Hake, and D. H. Leslie, Phys. Rev. Lett. **6**, 671 (1961).

[3] A. B. Pippard, Philos. Mag. **19**, 217 (1969).

[4] A. I. Larkin and Y. N. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979).

[5] T. Hocquet, P. Mathieu, and Y. Simon, Phys. Rev. B **46**, 1061 (1992).

[6] A. Pautrat, J. Scola, C. Goupil, Ch. Simon, C. Villard, B. Domengès, Y. Simon, C. Guilpin, and L. Méchin, Phys. Rev. B **69**, 224504 (2004).

[7] T. Giamarchi and S. Bhattacharya, in *Vortex Phases in "High Magnetic Fields: Applications in Condensed Matter Physics and Spectroscopy"*, edited by C. Berthier *et al.* (Springer-Verlag, New York, 2002), Vol. 9, p. 314.

[8] M. J. Higgins and S. Bhattacharya, Physica C **257**, 232 (1996) and older references therein; M. Marchevsky, M. J. Higgins, and S. Bhattacharya, Nature (London) **409**, 591 (2001); Y. Paltiel, E. Zeldov, Y. Myasoedov, M. L. Rappaport, G. Jung, S. Bhattacharya, M. J. Higgins, Z. L. Xiao, E. Y. Andrei, P. L. Gammel, and D. J. Bishop, Phys. Rev. Lett. **85**, 3712 (2000).

[9] M. Marchevsky, M. J. Higgins, and S. Bhattacharya, Phys. Rev. Lett. **88**, 087002 (2002).

[10] Y. Paltiel, E. Zeldov, Y. N. Myasoedov, H. Shtrikman, S. Bhattacharya, M. J. Higgins, Z. L. Xiao, E. Y. Andrei, P. L. Gammel, and D. J. Bishop, Nature (London) **403**, 398 (2000).

[11] P. Thorel, R. Kahn, Y. Simon, and D. Cribier, J. Phys. (France) **34**, 447 (1973).

[12] A. Pautrat, C. Simon, C. Goupil, P. Mathieu, A. Brulet, C. D. Dewhurst, and A. I. Rykov, Phys. Rev. B **75**, 224512 (2007).

[13] A. Pautrat, J. Scola, C. Simon, P. Mathieu, A. Brulet, C. Goupil, M. J. Higgins, and S. Bhattacharya, Phys. Rev. B **71**, 064517 (2005).

[14] X. S. Ling, S. R. Park, B. A. McClain, S. M. Choi, D. C. Dender, and J. W. Lynn, Phys. Rev. Lett. **86**, 712 (2001).

[15] N. D. Daniilidis, S. R. Park, I. K. Dimitrov, J. W. Lynn, and X. S. Ling, Phys. Rev. Lett. **99**, 147007 (2007).

[16] G. P. Mikitik and E. H. Brandt, Phys. Rev. Lett. **89**, 259701 (2002); Phys. Rev. B **64**, 184514 (2001).

[17] E. M. Forgan, S. J. Levett, P. G. Kealey, R. Cubitt, C. D. Dewhurst, and D. Fort, Phys. Rev. Lett. **88**, 167003 (2002).

[18] Y. Fasano, M. Menghini, F. de la Cruz, Y. Paltiel, Y. Myasoedov, E. Zeldov, M. J. Higgins, and S. Bhattacharya, Phys. Rev. B **66**, 020512(R) (2002).

[19] W. Henderson, E. Y. Andrei, M. J. Higgins, and S. Bhattacharya, Phys. Rev. Lett. **77**, 2077 (1996).

[20] E. H. Brandt, Phys. Rev. Lett. **78**, 2208 (1997).

[21] A. Yaouanc, P. Dalmas de Réotier, and E. H. Brandt, Phys. Rev. B **55**, 11107 (1997).

[22] W. Henderson, E. Y. Andrei, M. J. Higgins, and S. Bhattacharya, Phys. Rev. Lett. **80**, 381 (1998).

[23] R. Cubitt, E. M. Forgan, D. McK. Paul, S. L. Lee, J. S. Abell, H. Mook, and P. A. Timmins, Physica B **180-181**, 377 (1992); P. Harris, B. Lebech, and J. S. Pedersen, J. Appl. Crystallogr. **28**, 209 (1995).

[24] We use a Gaussian approximation to add the resolution contributions (FWHM) in quadrature whereas the RC are better fitted by Lorentzian so the correction is an approximation.

[25] M. R. Eskildsen, C. D. Dewhurst, B. W. Hoogenboom, C. Petrovic, and P. C. Canfield, Phys. Rev. Lett. **90**, 187001 (2003).

[26] T. Klein, I. Joumard, S. Blanchard, J. Marcus, R. Cubitt, T. Giamarchi, and P. Le Doussal, Nature (London) **413**, 404 (2001).

[27] T. Giamarchi and P. Le Doussal, Phys. Rev. B **52**, 1242 (1995).

[28] G. I. Menon, Phys. Rev. B **65**, 104527 (2002).

[29] M. Laver, E. M. Forgan, A. B. Abrahamsen, C. Bowell, Th. Geue, and R. Cubitt, Phys. Rev. Lett. **100**, 107001 (2008).

[30] A. Huxley, R. Cubitt, D. McPaul, E. Forgan, M. Nutley, H. Mook, M. Yethiraj, P. Lejay, D. Caplan, and J. M. Penisson, Physica B **223-224**, 169 (1996).

[31] Note that the error bars are large due to the small ratio counts/background (Fig. 4) (we estimate that 10% is the typical relative uncertainty).

[32] S. J. Levett, C. D. Dewhurst, and D. McK. Paul, Phys. Rev. B **66**, 014515 (2002).

[33] P. S. Swartz and H. R. Hart, Jr., Phys. Rev. **137**, A818 (1965).