

## Disorder, metastability, and history dependence in transformations of a vortex lattice

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Magnetic screening response of the superconductor 2H-NbSe<sub>2</sub> with varying pinning reveals a rich evolution of the peak effect from a history-independent sharp anomaly to a broad and strongly history dependent effect with internal structure. The results display a stepwise disordering of the vortex lattice through transformations affected by both thermal fluctuations and quenched disorder. [S0163-1829(99)03309-3]

The vortex matter<sup>1</sup> in the mixed state of type-II superconductors is an ideal system to study the interplay between interaction, thermal fluctuations, and quenched random disorder (in the form of pinning centers). The mean field theory yields an Abrikosov vortex lattice in the entire mixed state. Recent theoretical studies, including the effects of thermal fluctuations and quenched disorder, suggest the existence of phases such as vortex liquids<sup>2</sup> and various forms of “glassy” states.<sup>3</sup> Experimental evidence for pinning induced transformations in systems with varying quenched disorder is rare in comparison with that for thermally induced transitions, such as, melting.<sup>4</sup>

In this paper we show, first, the appearance of marked history dependence in the magnetic screening response in flux lattices that signifies effects of enhanced quenched disorder. Second, we report the occurrence of a sharp anomaly which marks a pinning-induced transformation between an ordered solid and a disordered solid in that part of parameter space where pinning is strong. The evolution of the resulting “phase diagram” illustrates the destabilization of the ordered phase by both quenched disorder and thermal fluctuations. Specifically, this destabilization is studied through the evolution of the peak effect (PE) phenomenon,<sup>5,6</sup> an anomalous enhancement of critical current  $J_c$  that occurs in close proximity of  $H_{c2}$  in low- $T_c$  systems and near the melting curve of the vortex lattice in high- $T_c$  systems.<sup>7</sup> Within the Larkin-Ovchinnikov<sup>8</sup> collective pinning scenario, the pinning force is given by

$$J_c B = \left( \frac{n_p \langle f_p^2 \rangle}{V_c} \right)^{1/2}, \quad (1)$$

where  $n_p$  and  $f_p$  are the density and strength of pins, respectively; the correlation volume  $V_c = R_c^2 L_c$ ,  $R_c = (c_{44}^{1/2} c_{66}^{3/2} r_f^2) / (n_p \langle f_p^2 \rangle)$  and  $L_c = (c_{44} / c_{66})^{1/2} R_c$ , where  $c_{66}$  and  $c_{44}$  are, respectively, the shear and tilt moduli and  $r_f$  is the pinning range. Although the exact origin of the PE is poorly understood,<sup>7,9</sup> it is assumed to signify a rapid collapse of  $V_c$ , i.e., an amorphization of the lattice. The PE is, therefore, ideally suited for the study of the effects of varying quenched disorder, in order to obtain insight into this amorphization process.

The system chosen is the low  $T_c$  system 2H-NbSe<sub>2</sub>,<sup>10</sup> studied widely in recent years due to its weak pinning nature.<sup>11–15</sup> Several recent experimental developments have added to the *puzzle of the peak effect*. First, a pronounced history dependence that usually marks a dominant role of disorder in systems, such as spin glasses, has been observed in transport and magnetization studies in some samples of 2H-NbSe<sub>2</sub>.<sup>12</sup> Second, these observations are similar to those in the mixed valent system CeRu<sub>2</sub> suggesting a generic phenomenon independent of differences in microscopic physics.<sup>13</sup> Third, a reentrance of the peak effect that resembles the reentrance of the theoretically proposed melting curve is observed in few samples of 2H-NbSe<sub>2</sub>.<sup>14</sup> Fourth, direct structural studies<sup>15</sup> show a disordering of the vortex lattice coincident with the onset of the PE. These results point to a complex interplay between quenched and thermal disorder in destabilizing the ordered vortex lattice, but the specific mechanisms remain obscure.

In what follows, we report on the PE in single crystal samples of 2H-NbSe<sub>2</sub> of varying purity. The samples studied here represent three classes of NbSe<sub>2</sub> crystals with resistance

ratio  $R_{300\text{K}}/R_{8\text{K}}$  varying between 30 and 10. Crystals grown from the purest starting materials, notably nearly tantalum free ( $<10$  ppm of Ta) Nb, hereafter referred to as sample X, has the weakest pinning and the sharpest PE, reported earlier.<sup>10</sup> Sample Y represents typically clean crystals with intermediate pinning (grown using Nb with  $\sim 50$ – $100$  ppm of Ta impurity), in which the reentrant PE is readily observed.<sup>14</sup> Sample Z was grown from commercial grade starting materials (with about 200 ppm of Fe impurities in Nb) and taken from the same batch of samples in which the pronounced history dependence of the pinning properties were observed.<sup>12,13</sup>  $J_c$  at 4.2 K and in  $H=10$  kOe varied from a few amps/cm<sup>2</sup> for the sample X to  $\sim 10^3$  amps/cm<sup>2</sup> for sample Z, leading to the variation of the pinning parameter  $J_c/J_0$  between  $10^{-6}$  to  $10^{-3}$ , where  $J_0$  is the depairing current density. Comparison with the Ginzburg parameter<sup>10</sup>  $G_i(=(1/2)(k_B T_c/H_c^2 \xi^3 \epsilon)^2) \sim 10^{-4}$  suggests that this is an interesting range in parameter space: quenched disorder is small compared to thermal fluctuations for the cleanest sample X, whereas the two sources of disorder are clearly comparable for the most strongly pinned sample Z.<sup>12</sup> Moreover, the zero field transition temperature  $T_c(0)$  is reduced to 6 K in sample Z due to the presence of magnetic impurities, but all physical characteristics related to PE scale with the respective  $T_c(0)$  and  $H_{c2}$  values in different samples.<sup>10,12</sup>

The PE is readily seen in the magnetic screening response through the in-phase component of the ac susceptibility  $\chi'$ , given by a generalized critical state model<sup>17</sup> relation:<sup>18</sup>

$$\chi' \approx -1 + \left( \frac{\alpha \cdot h_{ac}}{J_c} \right), \quad (2)$$

where  $\alpha$  is a geometry and size dependent factor. The rapid anomalous increase in  $J_c$  at PE leads to the sudden enhancement of diamagnetic response in  $\chi'$ . An example of a very sharp PE is shown in Fig. 1 for the sample X in an applied field of 4 kOe. We characterize the (anomalous) PE region by the onset temperature  $T_{pl}$  and the peak temperature  $T_p$ . In the example shown, the width of the anomalous region,  $\Delta T = T_p - T_{pl} \approx 20$  mK. Another 20 mK above  $T_p$ , the sample shows a differential paramagnetic effect<sup>19</sup> (DPE) at  $T \geq T_{irr}$  in the inset of Fig. 1, which implies the reversible magnetic response (above  $T_{irr}$ ). Note that the entire PE region, between  $T_{pl}$  and  $T_{irr}$ , is narrower than even the zero-field superconducting transition, as is evident in Fig. 1.

How this sharp anomaly undergoes changes with varying pinning is studied through the evolution of the three features ( $T_{pl}$ ,  $T_p$ , and  $T_{irr}$ ) spanning the anomalous PE regime. We focus on the three representative samples, X, Y, and Z, in the field range above 1 kOe. The low field ( $<1$  kOe) behavior, especially the evolution of the reentrance of the peak effect with pinning is discussed elsewhere.<sup>20</sup> Data were taken for two different thermomagnetic histories of the system, field-cooled (FC) and zero-field cooled (ZFC). All data were recorded during the warm-up cycle.

Figure 2 shows the details of the evolution of PE in high field range. First, the cleanest sample X, shows a sharp PE in a field of 15 kOe [see Fig. 2(a)], but more significantly, over the entire field ( $20$  kOe  $< H < 1$  kOe) and temperature range we did not observe any difference between the FC and ZFC

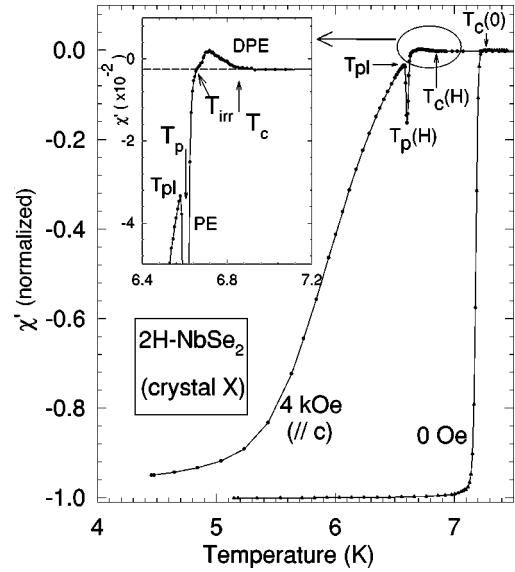


FIG. 1. In-phase ac susceptibility  $\chi'$  [ $f=211$  Hz,  $h_{ac}=1$  Oe (rms)] in nominal zero field and applied field ( $H_{dc} \parallel c$ ) of 4 kOe in the purest crystal X [ $T_c(0)=7.22$  K] of 2H-NbSe<sub>2</sub>. The PE peak has been identified and various temperatures marked. The inset reveals the presence of a paramagnetic ( $\Delta M/\Delta H > 0$ ) peak due to DPE (Ref. 19) sandwiched between  $T_{irr}(H)$  and  $T_c(H)$ .

states of the system. In other words, the vortex system approaches a unique state regardless of its preparation history.

Second, the sample Y, with intermediate pinning (an enhancement in  $J_c$  by an order of magnitude over the crystal X at the same value of H of 15 kOe), shows a qualitatively

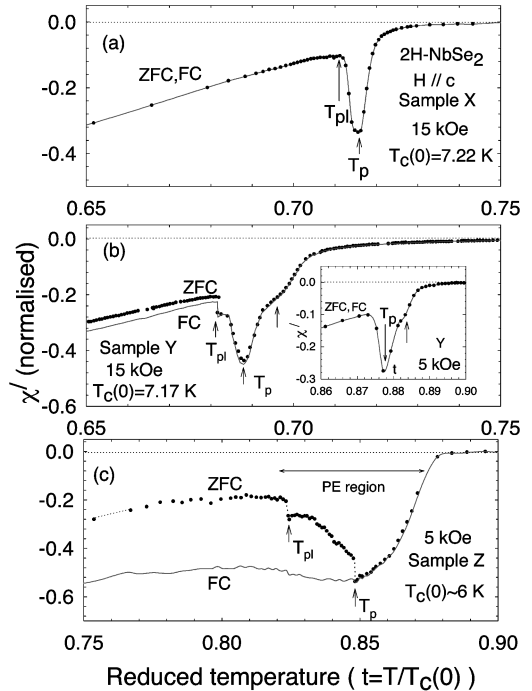


FIG. 2. ac susceptibility values for vortex states prepared in ZFC (see data points) and FC (see continuous curves passing through closely spaced data points) modes in three crystals of 2H-NbSe<sub>2</sub>. Figure (c) clarifies the identification of the PE region and the temperatures  $T_{pl}$  and  $T_p$  in the crystal Z, which also shows pronounced history dependence in  $\chi'$  up to  $T_p$ .

different behavior [see Fig. 2(b)]. A small but measurable difference in  $\chi'$  is observable between the ZFC and the FC states. In both cases, the onset of PE happens discontinuously, through a small step at the onset, marked  $T_{pl}$ . Moreover, a small shoulder appears on the high temperature side of the main dip (in  $\chi'$ ), marking yet another small peak effect superimposed on the rapidly increasing  $\chi'$  above  $T_p$ . Interestingly, however, the same crystal Y studied at a lower field of 5 kOe, shown in the inset of Fig. 2(b), shows no difference between ZFC and FC states and the results are qualitatively the same as in the cleaner sample X.

Third, the sample Z, with  $J_c$  another factor of 50 larger, shows a pronounced difference between the two histories of the system in the lower field of 5 kOe [see Fig. 2(c)]. The ZFC [ $\chi'(T)$ ] curve shows a sharp onset of the PE at  $T_{pl}$ ;  $\chi'$  continues to decrease with increasing  $T$  and shows a second sharp drop at  $T_p$ , above which the history dependence in  $\chi'$  disappears. The FC curve on the other hand shows little signature of disordering in the same region, it has a very broad and insignificant peak effect and it merges with the ZFC curve above  $T_p$ .

The following conclusions emerge directly from these data. (i) *The entire peak regime broadens significantly with increased pinning (the onset at  $T_{pl}$  for a given  $H$  moves to lower reduced temperature) and considerable structure appears;* (ii) *history dependence is unambiguously a consequence of increased pinning;* (iii) *for a given sample, increasing  $H$  is equivalent to increasing the effects of quenched disorder as has been suggested before.*<sup>21</sup>

That the FC state shows a larger screening response implies, through the relation of Eq. (2), that this state has a larger  $J_c$ , consistent with the transport and magnetization results.<sup>12</sup> We note that Eq. (1) then implies that the FC state has a smaller correlation volume  $V_c$  (since neither  $n_p$  nor  $f_p$ , both microscopic quantities for the same realization of quenched disorder, can depend on the history of the system). Thus, the two states with different correlations represent two metastable states of the vortex system. Direct structural results of neutron scattering studies<sup>22</sup> also show that the FC state has a smaller vortex correlation volume than the ZFC state. Thus the FC state shows less conspicuous PE, i.e., further disordering with increasing  $T$  could be absent in this already disordered metastable state. By contrast, the more ordered ZFC branch reveals the details of the disordering process of the FLL in the sharp anomalies at  $T_{pl}$  and  $T_p$ , which marks two abrupt drops in  $V_c$ . In order to understand the nature of these anomalies, we focus on their location in the  $(H, T)$  space.

Figure 3 shows the resulting location of the anomalous peak regime for the three samples under study. We plot the loci of the three features mentioned above, the onset of the PE at  $(H_{pl}, T_{pl})$ , the location of the peak of the PE at  $(H_p, T_p)$  and the apparent irreversibility line  $(H_{irr}, T_{irr})$ . Clearly, the anomalous PE regime is extremely narrow for sample X such that for this sample the thickness of  $(H_p, T_p)$  curve drawn in Fig. 3 nearly encompasses the  $(H_{pl}, T_{pl})$  and  $(H_{irr}, T_{irr})$  data points as well. The PE region becomes measurably broader in sample Y and it significantly broadens more for sample Z. Further, the onset of the PE moves farther away from the normal state not only for the more strongly pinned sample Z, but also for a given sample at

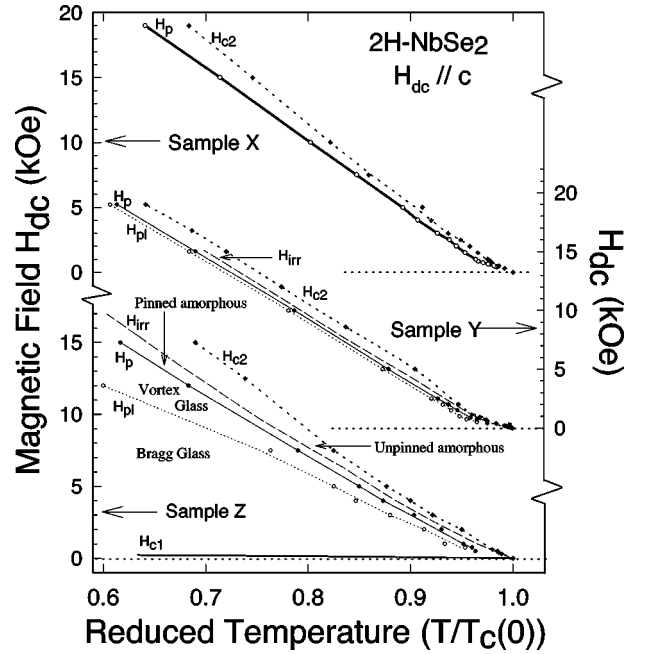


FIG. 3. A comparison of magnetic “phase diagrams” in three crystals X, Y, and Z of 2H-NbSe<sub>2</sub>. The  $H_{pl}$ ,  $H_p$ ,  $H_{irr}$ , and  $H_{c2}$  lines correspond to  $T_{pl}(H)$ ,  $T_p(H)$ ,  $T_{irr}(H)$ , and  $T_c(H)$  values, as indicated in Fig. 1 and Fig. 2. The PE region in crystal X is very narrow, thus only the  $H_p(T)$  line is drawn in this case (see text).

higher  $H$  and lower  $T$ . This provides a key evidence that the onset of the PE is strongly dominated by quenched disorder. For a comparison, the peak of the PE remains much closer to the normal state phase boundary, suggesting a greater role of thermal fluctuations.

We propose the following scenario to explain these results. The FLL is a relatively well-ordered solid below  $T_{pl}$ , an elastically deformed solid,<sup>23</sup> akin to a *Bragg glass*, free of topological defects; in this regime the elastic energy dominates over both pinning and thermal fluctuations. With increasing  $T$ , elastic energy decreases faster than pinning, as in the original Pippard<sup>5</sup> scenario of PE. At  $T_{pl}$ , pinning energy overcomes elastic energy as has been contemplated in several recent theoretical scenarios,<sup>24</sup> and the system undergoes a transition into a topologically defective glassy state, such as a *vortex glass*. This is consistent with the structural evidence<sup>15</sup> of a disordering of the ordered lattice at this line. Increased disorder stabilizes the defective glassy phase between  $T_{pl}$  and  $T_p$ , as can be seen from the rapid increase of  $\Delta T$  both among samples X, Y, and Z and for sample Z between low and high fields. As pinning increases, pinning energy overcomes the elastic energy at progressively lower (reduced) temperature and thus the regime of stability of the disordered vortex glass phase above  $T_{pl}$  expands at the expense of the ordered phase below  $T_{pl}$ .

The understanding of the anomaly at  $T_p$  is more complex. One explanation is that at  $T_p$ , the thermal energy overcomes the elastic energy and the FLL system further disorders into an amorphous state which, in the absence of pinning, would be equivalent to a vortex liquid. A fit using the Lindemann melting equation,<sup>16</sup>  $B_m(T) = \beta_m (c_L^4/G_i) H_{c2}(0) (T_c/T)^2 [1 - (T/T_c) - (B_m/H_{c2}(0))]^2$ , where  $\beta_m = 5.6$ , yields a very reasonable Lindemann parameter  $c_L = 0.24$  for sample X

over the entire  $(H, T)$  space reported in this study. For sample Y, the same fit works satisfactorily only up to 15 kOe with a value of  $c_L = 0.19$ . Forcing the fit to higher  $H$  and lower  $T$  yields even lower values of  $c_L$ . For sample Z, the fit is satisfactory for an even smaller range, i.e., up to  $H = 7$  kOe with a  $c_L = 0.17$ , above which the fit is rather poor. This suggests that the disordering of the FLL is also influenced by quenched disorder, as would be expected from progressively larger  $J_c/J_o$  values in samples Y and Z. In the absence of a clear theoretical understanding of how to combine the effects of quenched disorder and thermal fluctuations, we tentatively attribute the drop in correlation at  $T_p$  to an amorphization of the vortex array influenced by both thermal and quenched disorder. This differs from the original Larkin-Ovchinnikov scenario<sup>8</sup> where the amorphization is caused by quenched disorder alone.

Although macroscopic history dependence disappears above  $T_p$ , the vortex system still remains “pinned” and may be in a *pinned amorphous/pinned* liquid phase. Note the onset of reversibility at  $T_{irr}$  is nearly coincident with  $T_p$  in sample X, but is significantly above  $T_p$  for sample Z. This shows that  $T_{irr}$  marks the final crossover when thermal energy overcomes pinning and one obtains a pinning-free liquid which occurs at higher reduced temperatures (relative to corresponding  $T_p$ ) for systems with stronger pinning. One cannot eliminate the possibility, especially for sample Z, that the system reverts to individually pinned vortices [given by  $J_c/J_o \approx (G_i)^{1/3}$ ] above  $T_p$ .<sup>16</sup>

Importantly, the pinned amorphous “phase” above  $T_p$  provides a clue to the strong history dependence below  $T_p$ . As the system is prepared in the FC state, the system is pinned below  $T_{irr}$  in the amorphous state and, because of pinning, fails to explore the phase space when it is further cooled. Thus the FC state below  $T_p$  retains the frozen-in short-range correlation and is thus analogous to a super-

cooled amorphous phase that has fallen out of equilibrium. In the cleanest system X, for example, the PE occurs within a very narrow temperature window. Most notably,  $T_{irr}$  is nearly coincident with  $T_p$  and thus the pinned amorphous phase is only marginally stable. In this case, due to the weak nature of pinning, the system is able to explore the available phase space and find the “equilibrium” state regardless of the history.

The ZFC state, by contrast, is prepared at low  $T$  where a large field is applied. Thus the vortices enter the sample from the edges at high velocity, where the pinning is unimportant,<sup>10</sup> but intervortex interaction remains operative. If an ordered state is one of the metastable ground states in this part of the phase space, the system is able to find it.

The “phase diagram(s)” in Fig. 3 can be compared with theoretical expectations. It is essentially identical to the common theoretical phase diagram<sup>16</sup> with an added solid-to-solid transition at the  $(H_{pl}, T_{pl})$  line. This transition, likely similar to the Bragg glass-vortex glass or elastic glass-plastic glass transition postulated recently in several theoretical scenarios,<sup>18,21,23,24</sup> provides a close correlation between theory and experiment. These results, provide the crucial link between the history independent weak pinning regime (sample X) to a history dependent, stronger pinning regime (sample Z) via an intermediate pinning regime (sample Y). The sharp solid-to-solid transition is clearly seen in our experiment in that part of the  $(H, T)$  space where history dependence, i.e., pinning effects are strong. Furthermore, the disappearance of the disordered solid phase in the cleanest sample and its pronounced expansion in  $(H, T)$  space with increasing effective pinning clearly establishes the pinning-driven nature of the transition at  $T_{pl}$ . The second sharp anomaly at  $T_p$ , marking the final step in the amorphization of the vortex array, appears to have contributions from both thermal and quenched disorder and needs further study.

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<sup>1</sup>A. A. Abrikosov, Sov. Phys. JETP **5**, 1174 (1957).

<sup>2</sup>D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988).

<sup>3</sup>D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).

<sup>4</sup>A. Schilling *et al.*, Nature (London) **382**, 791 (1996).

<sup>5</sup>A. B. Pippard, Philos. Mag. **19**, 217 (1969); P. W. Anderson, *Basic Notions in Condensed Matter Physics* (Addison-Wesley, New York, 1983), pp. 162–163.

<sup>6</sup>R. Wordenweber, P. H. Kes, and C. C. Tsuei, Phys. Rev. B **33**, 3172 (1986) and references therein.

<sup>7</sup>C. Tang *et al.*, Europhys. Lett. **35**, 597 (1996) and references therein.

<sup>8</sup>A. I. Larkin and Yu. N. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979).

<sup>9</sup>A. I. Larkin *et al.*, Phys. Rev. Lett. **75**, 2992 (1995).

<sup>10</sup>S. Bhattacharya and M. Higgins, Phys. Rev. Lett. **70**, 2617 (1993); Physica C **257**, 232 (1996) and references therein.

<sup>11</sup>L. A. Angurel *et al.*, Phys. Rev. B **56**, 3425 (1997); D. Anna *et al.*, Physica C **218**, 238 (1993); Europhys. Lett. **25**, 225 (1994); **25**, 539 (1994).

<sup>12</sup>W. Henderson *et al.*, Phys. Rev. Lett. **77**, 2077 (1996); G. Ravi-

kumar *et al.*, Phys. Rev. B **57**, R11 069 (1998).

<sup>13</sup>S. S. Banerjee *et al.*, Phys. Rev. B **58**, 995 (1998).

<sup>14</sup>K. Ghosh *et al.*, Phys. Rev. Lett. **76**, 4600 (1996); S. Ramakrishnan *et al.*, Physica C **256**, 119 (1996).

<sup>15</sup>T. V. Chandrasekhar Rao *et al.*, Physica C **299**, 267 (1998); for similar conclusions in Nb, see P. L. Gammel *et al.*, Phys. Rev. Lett. **80**, 833 (1998).

<sup>16</sup>G. Blatter *et al.*, Rev. Mod. Phys. **66**, 1125 (1994).

<sup>17</sup>C. P. Bean, Rev. Mod. Phys. **36**, 31 (1964).

<sup>18</sup>X. S. Ling and J. Budnick, in *Magnetic Susceptibility of Superconductors and other Spin Systems*, edited by R. A. Hein, T. L. Francavilla, and D. H. Leibenberg (Plenum, New York, 1991), pp. 377–388.

<sup>19</sup>R. A. Hein and R. A. Falge, Jr., Phys. Rev. **123**, 407 (1961).

<sup>20</sup>S. S. Banerjee *et al.*, Europhys. Lett. **44**, 91 (1998).

<sup>21</sup>G. I. Menon and C. Dasgupta, Phys. Rev. Lett. **73**, 1023 (1994); see also V. M. Vinokur *et al.*, Physica C **295**, 209 (1998).

<sup>22</sup>A. D. Huxley *et al.*, J. Phys.: Condens. Matter **5**, 7709 (1993); Physica B **223&224**, 169 (1996).

<sup>23</sup>T. Nattermann *et al.*, Phys. Rev. Lett. **64**, 2454 (1990); T. Giamparchi and P. Le Doussal, *ibid.* **72**, 1530 (1994); Phys. Rev. B **52**, 1242 (1995).

<sup>24</sup>M. Gingras and D. A. Huse, Phys. Rev. B **53**, 15 193 (1996); D. Ertas and D. R. Nelson, Physica C **272**, 79 (1996).