Fate of the Peak Effect in a Type-II Superconductor: Multicriticality in the Bragg-Glass Transition

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We have used small-angle neutron scattering (SANS) and ac magnetic susceptibility to investigate the global magnetic field H vs temperature T phase diagram of a Nb single crystal in which a first-order transition of Bragg-glass melting (disordering), a peak effect, and surface superconductivity are all observable. It was found that the disappearance of the peak effect is directly related to a multicritical behavior in the Bragg-glass transition. Four characteristic phase boundary lines have been identified on the H-T plane: a first-order line at high fields, a mean-field-like continuous transition line at low fields, and two continuous transition lines associated with the onset of surface and bulk superconductivity. All four lines are found to meet at a multicritical point.

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An outstanding question concerning the Abrikosov vortex state of type-II superconductors is whether a genuine order-disorder transition can still occur in vortex matter even though true crystalline order cannot be attained due to random pinning by impurities [1]. There are convincing theoretical arguments [2–5] and numerical evidence [6] suggesting that, instead of a true vortex crystal, a novel Bragg-glass phase with quasi-long-range order can exist in bulk samples with weak random pinning; hence, a true order-disorder transition can occur when the topological order of the Bragg glass is destroyed, by thermal fluctuations and/or random pinning.

However, it is still controversial as to whether a Braggglass melting (or disordering) transition is the underlying mechanism of the well-known anomaly of "peak effect" in weak-pinning type-II superconductors [7]. Recent neutron scattering experiments on Nb [8], and V₃Si [9], as well as STM studies of 2H-NbSe₂ [10] all suggested a disordering transition at the peak effect, and the phase transition appears to be first order [8]. However, it has been reported that some samples of similar quality, e.g., having only weak pointlike pinning centers, do not show a peak effect, nor a disordering phase transition [11]. This raises an obvious, but intriguing, question: Is the fate of the peak effect, i.e., appearing or disappearing, related to a multicritical behavior in the phase transition into the Bragg glass? In this Letter, we report the first direct evidence that the disappearance of the peak effect is related to a multicritical point on the Bragg-glass phase boundary.

Our experiment was carried out using the 30m SANS instruments NG7 and NG3 at the NIST Center for Neutron Research on a Nb single crystal (99.998% in purity) in which both the peak effect and the first-order Bragg-glass melting (disordering) transition were observed at the same temperatures [8]. The sample has a zero field $T_c = 9.16$ K, and an estimated Ginzburg-

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Landau parameter $\kappa_1(0) = 2.0$. The mean wavelength of the incident neutron beam was $\lambda = 6.0$ Å and the wavelength spread 11% (FWHM). The experimental configuration is shown in the inset of Fig. 1(a). A cadmium mask was used such that only the central portion of the sample was exposed to the incoming neutron beam. The scattered neutrons were captured by a 2D detector of 128×128 pixels (the pixel size is 0.5 cm by 0.5 cm) 15.3 m away from the sample. The dc magnetic field was applied in the direction of the incoming neutron beams using a horizontal superconducting magnet. A coil was wound on the sample to allow *in situ* ac magnetic susceptibility measurements.

Figure 1(a) shows the SANS data at H = 3.0 kOe. The Gaussian width data are obtained from fitting the measured (Bragg) intensity vs azimuthal angle to six Gaussian peaks evenly spaced 60° apart. It is clear that the azimuthal widths, a measure of orientational disorder in the vortex array, are strongly history dependent. Supercooling and superheating effects are observed for field-cooling (FC) and field-cooled-warming (FCW) paths, respectively. As reported previously [8], the disordered phase at $T > T_p$ and the ordered phase at $T < T_p$ are of their respective thermodynamic ground states. The abrupt change in the structure factor S(q) at the peak effect T_p depicts a symmetry-breaking phase transition from a vortex matter with short-range order to a Bragg glass with quasi-long-range order [8]. The phase transition is first order as evidenced by the strong thermal hysteresis in S(q). Compared to that at higher fields, the metastability region for H = 3.0 kOe is smaller but still pronounced.

We found that the thermal hysteresis of S(q) observed in SANS is strongly field dependent, and the metastability region disappears completely at low fields. Figure 1(b) shows the azimuthal width data for H = 2.0 kOe. For comparison, the real part $\chi'(T)$ of the ac magnetic



FIG. 1. (a) Temperature and history dependence of azimuthal widths of the (1, -1) diffraction peak at $H_{dc} = 3.0$ kOe. The widths are obtained by Gaussian fits. The dashed line is the peak-effect temperature T_p at this magnetic field based on ac magnetic susceptibility measurements. Inset: Experimental configuration. (b) Temperature and history dependence of azimuthal widths of (1, -1) diffraction peak at $H_{dc} = 2.0$ kOe. The ac susceptibility data are also shown for reference. Definitions of $T_p(H)$, $T_{c2}(H)$, and $T_{c3}(H)$ (see below) are shown.

susceptibility is also shown in Fig. 1(b). The dip in $\chi'(T)$ is a well-established signature of the peak effect [12–14]. The history dependence of the Bragg-peak width is detectable only within 0.1 K of the peak-effect temperature T_p .

A similar trend is observable in the history dependence of the radial widths of the Bragg peaks, as shown in Fig. 2, obtained by fitting a single Gaussian function to the q dependence of the SANS intensity. At 3.0 kOe, there is a pronounced thermal hysteresis in the radial widths. At 2.0 kOe, however, the hysteresis is barely discernable. At an even lower field of 1.0 kOe (data not shown), the thermal hysteresis in S(q) is undetectable.



FIG. 2. Temperature and history dependence of the radial widths of the diffraction peaks at (a) $H_{dc} = 3.0$ kOe and (b) $H_{dc} = 2.0$ kOe.

At H = 1.0 kOe, a very sharp peak effect (the onset-toend width = 40 mK) is still present. Thus, we believe the phase transition at 1.0 kOe is still first order but the metastability region is too narrow to be resolved in SANS (the temperature resolution in SANS \approx 50 mK). Nevertheless, the diminishing hysteresis in the Braggglass transition in the low-field regime suggests that the phase transition is becoming continuous and meanfield-like.

The fact that the transition into the Bragg glass is first order at high fields, but mean-field-like at low fields, strongly suggests the existence of a multicritical point on the phase boundary bordering the Bragg-glass on the H-T phase diagram. We show below that this multicritical behavior is directly related to the appearance and the disappearance of the peak effect. Figure 3 shows a three-dimensional plot of the $\chi'(T)$ as a function of temperature and magnetic field in the field range of 0–5.12 kOe. At high fields, there is a pronounced peak effect, a characteristic dip in $\chi'(T)$. With decreasing field, the peak effect becomes narrower and smaller. For H < 0.8 kOe, there is only a single kink in $\chi'(T)$ corresponding to the mean-field transition $H_{c2}(T)$. According to the $\chi'(T)$ data in Fig. 3, there is no reentrant peak effect at low fields, in contrast to that in 2H-NbSe₂ [15] and YBCO [16]. The peak effect simply vanishes here.

At higher temperatures above the peak-effect temperature $T_p(H)$ [or $H_p(T)$, used interchangeably], there is a smooth step in $\chi'(T)$. This step, $T_{c3}(H)$ [or $H_{c3}(T)$], defined in Fig. 1(b), is the onset of surface superconductivity. The separation between T_p and T_{c3} grows larger with increasing magnetic field. Upon cooling, below $T_{c3}(H)$ and towards $T_p(H)$, the screening effect in $4\pi\chi'(T)$ increases gradually. Nevertheless, a less welldefined characteristic temperature $T_{c2}(H)$ can be identified to mark the onset of bulk superconductivity [see Fig. 1(b) for definition]. Note that the notation $T_{c2}(H)$ is used for H > 0.8 kOe, while $H_{c2}(T)$ for H < 0.8 kOe.

The results of Fig. 3 are summarized in a new phase diagram of Bragg-glass superconductivity in Nb as shown in Fig. 4. The measured ratio of H_{c3}/H_{c2} at low temperatures is about 1.60, slightly smaller than the expected value of 1.695 by Saint-James and de Gennes [17]. This is likely due to the nonideal cylinder surface being not exactly parallel to the field. The crossing of $H_{c3}(T)$ and $H_{c2}(T)$ lines below T_c was observed previously [18,19],



FIG. 3. Three-dimensional (3D) magnetic field and temperature dependence of the real part of the ac susceptibility $4\pi\chi'(T)$. $H_{\rm dc}||H_{\rm ac}$. Note that two values of ac fields were used in the measurements. For $H_{\rm dc} < 3.0$ kOe, $H_{\rm ac} = 1.7$ Oe and f = 1.0 kHz and, for $H_{\rm dc} > 3.0$ kOe, $H_{\rm ac} = 7.0$ Oe and f = 1.0 kHz. The two straight lines are hand drawn as guides for the eye. For the ac fields used, T_p is independent of the ac field amplitude [14].

and has been interpreted as due to a depressed BCS gap function near the surface [20].

The most striking aspect of Fig. 4 is that all four lines, $H_p(T)$, $T_{c2}(H)$, $H_{c2}(T)$, and $H_{c3}(T)$, meet at a multicritical point (MCP). To determine the nature of a MCP, one needs to know how many of these lines are second-order phase transitions. In the theory of Saint-James and de Gennes [17], $T_{c3}(H)$ is a continuous phase transition. For H < 0.8 kOe, the linear temperature dependence of $H_{c2}(T)$ follows the expected behavior of a Ginzburg-Landau mean-field transition line [21]. This is a line of continuous phase transitions. For H > 0.8 kOe, the peak effect $T_p(H)$ traces out a line of first-order transitions. Across this line, the thermal hysteresis in the structural factor S(q) of the vortex matter can be observed, especially striking at high fields.

The nature of the $T_{c2}(H)$ line is less clear. The vortex matter is liquidlike structurally in the shaded part of the phase diagram. Whether this disordered vortex matter is a distinct thermodynamic phase from the normal state is still being debated (see [7] and references therein). If $T_{c2}(H)$ is a true second-order phase transition, e.g., as in a vortex glass transition [22], the MCP in Fig. 4 would appear to be a tricritical point. On the other hand, we



FIG. 4. The phase diagram of a weak-pinning Nb crystal for the $H_{dc}||\langle 111 \rangle$ crystallographic direction. The upper solid circles (the crosses correspond to measurements using $H_{ac} =$ 1.7 Oe, see Fig. 3 captions) are the peak of the peak effect and the first-order transition line; the lower ones are the mean-field transition. The open diamonds (two sets are for two values of ac fields, see Fig. 3) are H_{c3} . The multicritical point is indicated by the large filled circle. The H_{c1} data (triangles) are estimated from the first penetration in the ac susceptibility data. All lines are hand drawn as guides for the eye.

found that the measured slopes of the four phase boundaries near the MCP cannot satisfy the requirements [23] imposed on a tricritical point by thermodynamics, but are consistent with those for a bicritical point. This leads to an important conclusion that only one of the two lines, $T_{c2}(H)$ or $H_{c3}(T)$, can be related to the MCP.

For $T_{c2}(H)$ to be a second-order line for the bicritical point, its slope has to be larger (in magnitude) than that of $H_{c2}(T)$, such that the thermodynamics rule [24] that no phase can occupy more than 180° of the phase space around a MCP is satisfied. However, this is not observed in our data as shown in Fig. 4. For the $H_{c3}(T)$ line to be the relevant one, the ratio of specific-heat jump at $H_{c2}(T)$ over that at $H_{c3}(T)$ should be 43.6. While this large ratio is consistent with the existing specific-heat data on Nb [25], presently there are no reliable specific-heat data near a crossing point of $H_{c2}(T)$ and $H_{c3}(T)$ to allow us to make a quantitative comparison.

We should point out that a similar critical point has also been observed in platelet geometries such as MgB₂ [26,27] crystals for which $H_{c3}(T)$ is not expected to play a role in the critical point. In high- T_c YBCO, a disappearance of the first-order transition was also observed in the low-field regime, and was interpreted as a critical end point [28]. If our critical point in Fig. 4 is also interpreted as a critical end point, $T_{c2}(H)$ would not be a true phase transition, and the meeting of $H_{c3}(T)$ at the MCP would be purely coincidental.

In summary, we found that, in a Nb crystal in which a peak effect in ac magnetic susceptibility and a first-order melting (disordering) transition in SANS were found to coincide previously, both effects disappear at a low field. It is suggested that the appearance or absence of a peak effect in a type-II superconductor may be directly correlated with a multicritical point (MCP) on the Bragg-glass phase boundary. The existence of a MCP, at which the peak effect vanishes, suggests that the origin of the peak effect is related to a second-order phase transition at a higher temperature. In the sample studied, it appears that the MCP may be related to surface superconductivity.

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