

Percolation Transition in the Heterogeneous Vortex State of NbSe₂

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A percolation transition in the vortex state of a superconducting $2H - \text{NbSe}_2$ crystal is observed in the regime where vortices form a heterogeneous phase consisting of ordered and disordered domains. The transition is signaled by a sharp increase in critical current that occurs when the volume fraction of disordered domains reaches the value $P_c = 0.26 \pm 0.04$. Measurements on different vortex states show that, while the temperature of the transition depends on history and measurement speed, the value of P_c and the critical exponent characterizing the approach to it, $r = 1.97 \pm 0.66$, are universal.

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Recent imaging experiments [1] on vortex matter in superconductors have revealed a heterogeneous state consisting of domains of ordered and disordered phases [2,3]. The appearance of heterogeneity coincides with the observation of striking anomalies including the peak effect [4], nonlinear response [5], metastability, and history effects [6]. It is well known that multicomponent systems undergo a percolation transition when the volume fraction of one of the components reaches a critical value P_c , leading to the formation of a system-spanning cluster and to critical behavior [7]. In the case of the heterogeneous vortex system, because the critical current depends on the degree of order, the transport properties are sensitive to details of the spatial domain distribution and can lead to dramatic changes in the properties of the superconducting host when one of the domains percolates.

In this Letter, we report on experiments demonstrating the existence of a percolation transition in the vortex system in NbSe₂ and its effect on the transport properties. The experiments employed pulsed measurements of the voltage-current (V - I) characteristics together with a proposed two-phase model to obtain the fraction of disordered domains P . We find that the transition is signaled by a sharp rise in the critical current (I_c) at $P_c = 0.26 \pm 0.04$ and that the approach to P_c is characterized by a critical exponent $r = 1.97 \pm 0.66$. Our experiments show that the transition is uniquely determined by P_c and not by the thermodynamic parameters. Thus while the transition temperature varies with magnetic field and can be lowered with the application of a slow current ramp or increased by applying an ac current [5,8,9], the values of P_c and r are universal.

The data presented here were taken on a Fe-doped $2H - \text{NbSe}_2$ crystal with dimensions $3 \times 0.7 \times 0.03 \text{ mm}^3$, $T_c = 6.03 \text{ K}$, and transition width $\Delta T_c = 50 \text{ mK}$. The field was applied in the c direction, and the current was in the ab plane. The experiments employed a four-lead configuration with AgIn solder contacts to monitor the voltage response to current ramps and pulses. A $2 \mu\text{V}$ criterion

was used to define I_c . The voltage was measured with a low noise ($4 \text{ nV/Hz}^{1/2}$) fast amplifier and then digitized with a 100 MHz digital oscilloscope. The data were averaged over 10 runs (initiating with a zero field cooling cycle from above T_c for each run). The V - I curves were obtained point by point from the response to a current pulse as illustrated in Fig. 1(a). Each point on the V - I shown in Fig. 1(b) represents the voltage measured $3 \mu\text{s}$ after applying the current pulse. This procedure excludes the instrumental response which decays within $\sim 2 \mu\text{s}$, as well as heating effects which are negligible on time scales much shorter than the thermal diffusion time $\sim 10 \text{ ms}$ [10]. In addition to avoiding heating, the pulsed technique also eliminates effects due to current induced organization. Applied currents can induce reorganization of the vortex lattice by transforming one phase into another or introducing a disordered phase through the surface barrier at the sample edge [9,11,12]. Thus, unless the measurement is faster than the organization time, the result depends on the measurement speed. The relevant time scale is estimated from the time to move one lattice spacing. For $B = 0.5 \text{ T}$ and $V = 2 \mu\text{V}$ corresponding to the voltage resolution, the reorganization time $\sim 10 \mu\text{s}$ is longer than the pulsed measurement times. By comparison, in current ramped measurements I_c is reached within a time I_c/R , where R is the ramping rate, so that for $I_c = 50 \text{ mA}$ and $R = 1 \text{ mA/s}$ [referred to as the slow current ramp (SCR)], $I_c/R \sim 50 \text{ s}$ is much longer than the reorganization time. In the case of the fast current ramps (FCR), $R = 300 \text{ A/s}$, leading to $I_c/R \sim 150 \mu\text{s}$, which still allows for reorganization.

In order to calculate P , we need to measure I_{co} and I_{cd} , the critical currents of the ordered and disordered lattices, respectively. The ordered lattice was prepared by zero field cooling (ZFC). Far below the peak effect the ZFC state is ordered and stable, and I_{co} is independent of measurement speed [8], as shown in Fig. 2 where we compare I_{co} obtained with various measurement speeds. The disordered

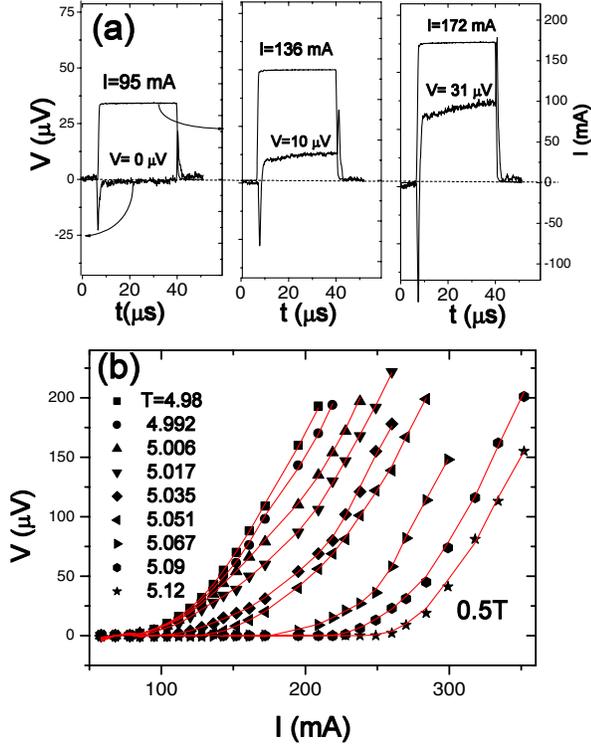


FIG. 1 (color online). (a) Time evolution of voltage in response to current pulses corresponding to 3 points on the V - I curve at $T = 5.035$ K and a field of 0.5 T. The narrow voltage spikes at the onset and removal of the pulse are instrumental. (b) V - I curves for vortex states prepared by ZFC at temperatures indicated in the legend and measured at $T_0 = 4.3$ K.

lattice is prepared by field cooling (FC). For the doped sample used here, the FC lattice forms a robust supercooled disordered state which does not evolve with time in the absence of external perturbations [5]. An applied current causes the lattice to reorganize, leading to a strong dependence of I_c on measurement speed as illustrated in Fig. 2. As can be seen in the inset, the voltage response does not evolve much immediately following the $2 \mu\text{s}$ duration of instrumental transients, but it grows significantly for longer times. We therefore assume that the voltage recorded $3 \mu\text{s}$ after the current onset represents the response of the pristine FC lattice and use this value to define I_{cd} .

The first step in the procedure to measure P was to prepare a ZFC lattice at a (variable) target temperature T . Subsequently, after waiting for ~ 1 min at T , the lattice was rapidly cooled to $T_0 = 4.3$ K where the pulsed V - I was measured point by point. The resulting pulsed V - I curves for several values of T are shown in Fig. 1(b). We note that the slope of each curve increases monotonically, saturating at a value of $R_{ff} \sim 2.4$ m Ω , the free flux flow resistance at T_0 . Moreover, although the measurement temperature T_0 was always the same, the shape of the V - I curves showed a strong dependence on the preparation temperature T . The shape of these curves and the values of

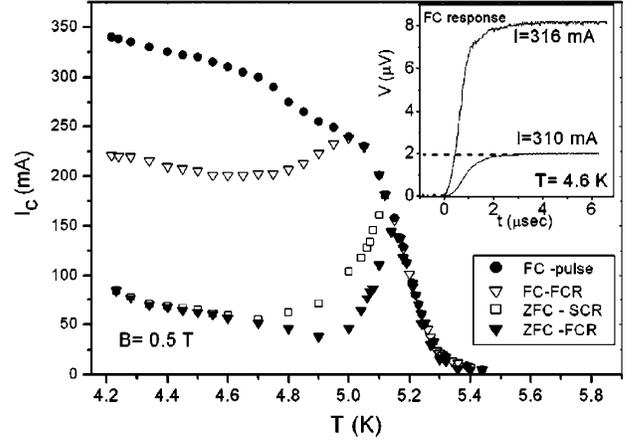


FIG. 2. Dependence of critical currents on measurement speed and method of preparation. Solid circles: FC state obtained with short current pulses; open triangles: FC states measured with FCR; solid triangles: ZFC measured with FCR. Open squares: critical currents measured with SCR are the same for FC, ZFC and other methods of preparation. The inset shows the response of an FC state to short pulses at $T = 4.6$ K. The critical current $I_c(4.6 \text{ K}) = 310$ mA is defined by a response of $\sim 2 \mu\text{V}$.

I_{co} and I_{cd} are used together with the two-phase model to obtain $P(T)$.

In the two-phase model the domains of ordered phase (OP) and disordered phase (DP) are characterized by critical current densities J_o and J_d , respectively, with $J_d > J_o$ [13]. Before vortices start moving, the current density in the sample has to be either zero or equal to the local critical current density, which can take one of the two values [14], J_d or J_o . Vortex motion first occurs at a current for which there exists a cross section (transverse to the current) within which all vortices are subject to their respective critical current density. Therefore the critical current corresponding to a cross section $A(x)$ at position x along the current flow and containing a fraction $\alpha(x)$ of DP is

$$I_c(x) = \{\alpha(x)J_d + [1 - \alpha(x)]J_o\}A + I_s. \quad (1)$$

Here I_s is a surface current resulting from the Bean-Livingston surface barrier [11]. We have assumed that both the cross section and I_s are uniform along the sample so that $A(x) = A$ and $I_s(x) = I_s$, but the model can be generalized to a situation where this is not so as long as vortices do not intersect. The global critical current is determined by the cross section containing the minimal fraction of DP, α_m . As long as a cross section exists that does not intersect a domain of DP, i.e., $\alpha_m = 0$, the corresponding value of I_c will be the same as that of a sample with all vortices in the ordered phase $I_o = J_o A + I_s$. Just above the percolation transition, after the first system-spanning cluster of DP has formed, $\alpha_m > 0$ and $I_c > I_o$. Thus we can obtain the value of α_m for an arbitrary distribution of domains from the values of I_c , J_o , and J_d . Although I_o and I_d can be measured directly, this is not the

case for the critical current density $J_c = (I_c - I_s)/A$, which contains a surface component not directly measured. Since direct measurements of I_s are quite difficult, we use quantities that do not explicitly depend on it. We note that I_s is a single-vortex property and should not depend on the degree of order in the vortex state, so it would have the same value for the OP, DP, and the heterogeneous state. This leads to a cancellation of I_s from Eq. (1), resulting in a simplified expression for α_m which involves only the measured quantities, provided all are measured at the same temperature: $I_c(T) = I_o + \alpha_m[I_d(T) - I_o(T)]$. In order to take advantage of this simplification and minimize the reorganization effects, all measurements were carried out at the base temperature $T_0 = 4.3$ K.

Vortex lattices with different fractions of DP at 4.3 K were obtained by preparing the ZFC lattice at a (variable) temperature T , and then cooling it to T_0 . The difference between these lattices is reflected in the V - I curves in Fig. 1(b) which are qualitatively different from each other in spite of having the same measurement temperature. In other words, cooling a ZFC lattice from the preparation temperature to T_0 creates a frozen replica of the state at T [5]. This is consistent with the fact that the OP and DP must be separated by an energy barrier that far exceeds the thermal energy in order to exhibit the experimentally observed stability and coexistence [1]. If the vortex lattice does not change its composition upon further cooling, then $\alpha_m(T_0) = \alpha_m(T)$, and therefore $\alpha_m(T)$ can be obtained directly from the measured critical currents at T_0 :

$$\alpha_m(T) = [I_d(T_0) - I_o(T_0)]/[I_c(T, T_0) - I_o(T_0)]. \quad (2)$$

Here $I_c(T, T_0)$ is the critical current of the ZFC state prepared at T and measured at T_0 , while $I_o(T_0)$ and $I_d(T_0)$ are the critical currents in the OP and DP, respectively, measured and prepared at T_0 . To test the validity of the model, we used this value of α_m to calculate the critical current of the ZFC state at T : $I_c(T) = I_o(T) + \alpha_m[I_d(T) - I_o(T)]$, and compared it to the directly measured value. The results of this calculation [open circles in Fig. 3(b)] are in good agreement with the measured values of $I_c(T)$ [solid triangles in Fig. 3(b)].

To obtain the value of P we need to analyze the shape of the V - I curves. The response to a current I can be expressed as $V(I) = R_{ff} \int_0^I [I - I_c(x)] dx$, where x is in units of sample length and the integrand is zero for $I < I_c(x)$. By using Eq. (2) and the fact that $P = \int_0^1 \alpha(x) dx$, we obtain the response in the free flux flow regime where all vortices are in motion $V(I) = R_{ff}[I - I_o - (I_d - I_o)P]$, leading to $P = (I_{\text{eff}} - I_o)/(I_d - I_o)$, where I_{eff} is the $V = 0$ intercept of the linear portion (free flux flow regime) of the V - I curve. This gives a straightforward geometrical procedure to extract P from the V - I curve. Alternatively, P is obtained from the curvature of the V - I curves $P = \frac{1}{R_{ff}(I_d - I_o)} \int_{I_c}^{I_{cf}} (I - I_o) \frac{\partial^2 V}{\partial I^2} dI$. Here, I_{cf} is the current at which the slope recovers to the value of the free flux flow

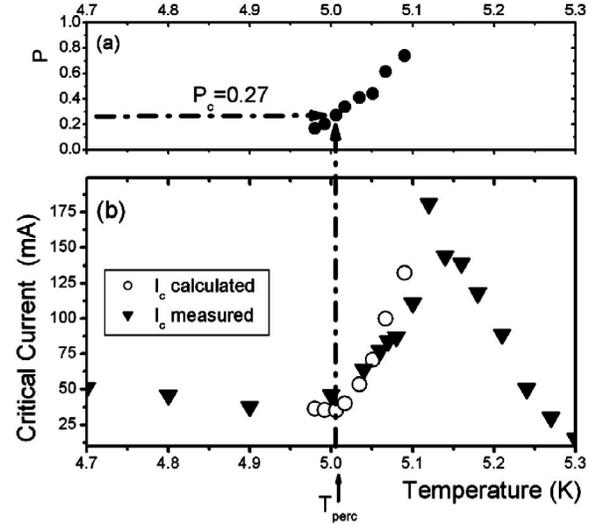


FIG. 3. (a) Temperature dependence of fraction of disordered phase, P . (b) Temperature dependence of measured and calculated I_c . The onset of increase in critical current (onset of the peak), marked by the arrow, is identified with the percolation transition at $P_c = 0.27$.

resistance. Both methods give the same values for P within experimental accuracy.

In Fig. 3(a) we plot the temperature dependence of P . In Fig. 3(b) we show that the percolation threshold as signaled by the onset of increasing I_c occurs at $P_c = 0.27 \pm 0.04$. Beyond this point I_c continues to grow, as the disordered regions expand with increasing P , until the entire sample becomes disordered. The values of α_m calculated from Eq. (1) are shown in Fig. 4(a) together with a power law fit to $\alpha_m \propto (P - P_c)^r$. The exponent r characterizes the behavior of the minimal cross section close to P_c and is not one of the standard critical exponents. However, as we show below, the behavior of α_m near P_c can be mapped onto the conductivity near the metal-insulator transition, and therefore r should be the same as the conductivity exponent t [7,15]. The analogy is drawn by considering a binary insulator-conductor sample that is topologically identical to the heterogeneous vortex sample. This is obtained by mapping all the DP domains onto a conducting phase with conductivity σ_c and all the OP ones onto an insulating phase. Just above the percolation transition the conductivity is dominated by the bottlenecks forming at the cross section containing a minimal amount of conducting phase, α_m , so that the effective conductivity, σ , is proportional to $\sigma_c \alpha_m$. This is valid in the limit where the bottlenecks are much narrower than any other conducting cross section. Near the percolation transition the conductivity will thus be $\sigma \propto \alpha_m \propto (P - P_c)^t$, where t is the conductivity exponent. In 3D systems the value $t = 2$ is expected to be universal [6]. Fitting our data over a range of P close to P_c we find $r = 1.78 \pm 0.52$, which is consistent with this model.

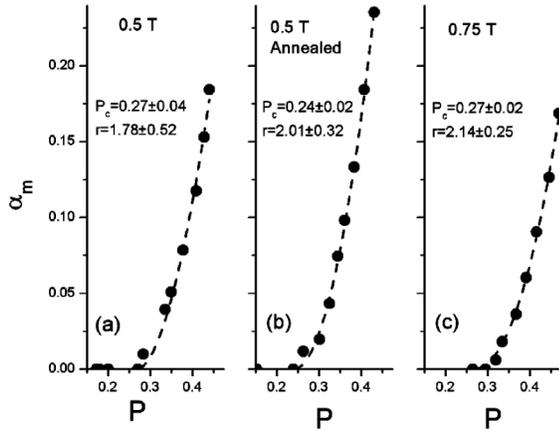


FIG. 4. Dependence of the minimal cross section, α_m , on the fraction of disordered phase, P , together with fits to $\alpha_m \propto (P - P_c)^r$ for three vortex lattices. (a) Lattice prepared by ZFC at 0.5 T. (b) Current annealed lattice at 0.5 T. (c) ZFC lattice at 0.75 T.

A salient feature of percolation is that it is governed by a single parameter P_c and thus provides a stringent test for identifying the transition. To apply this test here we repeated the experiments with vortex states prepared by different methods. Current annealed vortex states were prepared by applying a slow current ramp to the ZFC state at T before cooling to T_0 . The onset of increase in I_c for the annealed states (open squares in Fig. 2) is shifted to a lower temperature compared to the unannealed ZFC states [16], yet if this is the signature of percolation we should find the same value of P_c . Indeed, as shown in Fig. 4(b), although the temperature of the onset, 4.7 K, is lower than that in the unannealed lattice, $P_c = 0.24 \pm 0.02$ and the exponent $r = 2.01 \pm 0.32$ are the same within experimental error. Another set of measurements carried out on the unannealed ZFC vortex lattice but at a different field, $B = 0.75$ T [Fig. 4(c)], resulted in $P_c = 0.27 \pm 0.02$ and $r = 2.14 \pm 0.25$. Again these results are in good agreement with the data at $B = 0.5$ T, confirming the universality of the transition. Comparing to other systems we find that the value (averaged over the three experiments) of the percolation threshold $P_c \sim 0.26 \pm 0.04$ and the exponent $r = 1.97 \pm 0.66$ places the vortex percolation transition in the universality class of overlapping random spheres (the “inverse Swiss cheese” model [7]).

The experiments described here demonstrate that the onset of increase in critical current associated with the peak effect is the signature of a static percolation transition [17] when domains of disordered phase form a system-spanning cluster. They further show that while the temperature and field defining the onset of the peak effect depend on history and measurement speed, the percolation

transition is uniquely defined by the volume fraction of disordered phase, P_c , and by the exponent r .

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