## Positional disorder in Josephson-junction arrays: Experiments and simulations

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We present the results of measurements on proximity-effect arrays with deliberate positional disorder, characterized by a parameter  $\Delta^*$ . This system provides an experimental realization of an XY magnet with random Dzyaloshinskii-Moriya interactions. In measurements of resistance versus perpendicular magnetic field,  $R(f_0)$ , we find strong evidence for a critical field  $f_c \propto 1/\Delta^*$  beyond which phase coherence is destroyed. Our Monte Carlo simulations show evidence for reentrant behavior in the helicity modulus Y, but with Y always finite at the lowest temperatures. Such a critical field and reentrance were predicted by Granato and Kosterlitz.

A single Josephson junction is isomorphic to a pair of classical XY spins, and an array of junctions is therefore a realization of an XY spin system. This equivalence has prompted the use of two-dimensional junction arrays, in zero magnetic field, as model statistical mechanical systems for studying problems such as the Kosterlitz-Thouless transition.<sup>1,2</sup> A uniform array in a perpendicular magnetic field provides a realization of the uniformly frustrated XY magnet, with tunable frustration parametrized by f, the magnetic flux per plaquette in units of the flux quantum. The particular case of full frustration, where  $f = n + \frac{1}{2}$  (n an integer), has received a great deal of attention as a realization of Villain's "odd model,"<sup>3</sup> although theory<sup>4</sup> and experiment<sup>1,2</sup> are still far from complete in this area.

The XY model with nonuniform frustration<sup>5</sup> is of particular interest as a more realistic model for random magnetic systems. Again, the Josephson junction array provides a convenient model system—a junction array with positional disorder in a perpendicular magnetic field.

Granato and Kosterlitz (GK) recently considered<sup>6</sup> the effect of positional disorder, that is, displacing the superconducting sites from their average position r by an amount  $\mathbf{u}_{\mathbf{r}}$ , with probability distribution  $P(\mathbf{u}_{\mathbf{r}}) \propto \exp(-|\mathbf{u}_{\mathbf{r}}|^2/2\Delta^2)$ . This introduces correlated disorder in the plaquette areas and hence in f. In the Coulomb gas analogy, where vortices become charges, this leads to a quenched random distribution of dipoles of strength  $\mathbf{p}_r \propto \mathbf{u}_r$ . GK restrict their attention to the case where the average flux per cell  $f_0$  is an integer, in which case the system is equivalent to an XY magnet with random Dzyaloshinskii-Moriya interactions.<sup>7</sup> Invoking а renormalization-group analysis used in earlier work by Rubinstein, Shraiman, and Nelson,<sup>5</sup> GK predicted the qualitative phase diagram shown in Fig. 1. This phase diagram shows two striking effects. First, for fields  $f_0$ greater than a critical value  $f_c$  long-range phase coherence<sup>8</sup> is destroyed. The value of  $f_c$  is given by

$$f_c = \frac{1}{\sqrt{32\pi}} \frac{1}{\Delta} \approx \frac{0.10}{\Delta} . \tag{1}$$

Second, for fields  $f_0$  less than  $f_c$ , one should find two vortex-unbinding transitions. For temperatures below the

lower transition temperature  $[T_c^-(f_0)$  in Fig. 1], the quenched dipoles weaken the interaction between the mobile vortices so that some of the vortices are unbound, and there is no long-range phase coherence. For  $T_c^-(f_0) < T < T_c^+(f_0)$  the increased density of mobile vortices is sufficient to screen the quenched dipoles, so that all of the vortices are bound. Finally, for  $T > T_c^+(f_0)$ , the vortices are thermally unbound, as in a uniform array.

In this Rapid Communication, we report the first measurements of the critical field  $f_c$  and the first evidence based on Monte Carlo simulations for the reentrance.

We have fabricated arrays (typically  $50 \times 50$ ) of proximity-effect junctions, with controlled amounts of positional disorder. Figure 2(a) shows a section of a lithographic mask for a uniform array. The black crosses become superconducting Pb<sub>0.95</sub>Bi<sub>0.05</sub> islands in the actual samples, and the interisland coupling is provided by a continuous underlayer of Cu. Figure 2(b) shows a mask for a sample with positional disorder. Our procedure is to displace the centers of the crosses by a random amount, while keeping the junctions (the tips of the crosses) fixed on a regular lattice. Thus we introduce no deliberate disorder in the interisland coupling energies. For practical reasons, the site displacements have a *uniform* distribution, with a half-width  $\Delta^*$  in x and y directions. We have



FIG. 1. Schematic phase diagram for a 2D array with positional disorder. Vortex-unbinding transitions occur at  $T_c^{-}(f_0)$  and  $T_c^{+}(f_0)$ . In the region marked S (for "superconducting") the system shows long-range phase coherence. In the region labeled N (for "normal") this phase coherence is destroyed (Ref. 6).

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FIG. 2. Sections of lithographic masks used to prepare arrays with (a)  $\Delta^* \approx 0$  and (b)  $\Delta^* = 0.10$ . Crosses become superconducting islands in actual samples.

fabricated samples with  $\Delta^* = 0.05$ , 0.10, 0.15, and 0.20 (in units of the lattice parameter *a*), as well as nominally uniform samples ( $\Delta^* \approx 0$ ). The masks were produced using electron-beam lithography on our scanning electron microscope (SEM), and samples were produced using photolithography and ion-beam etching.

In order to compare our results with theory we need a conversion factor relating  $\Delta$  and  $\Delta^*$ . We believe that the important quantity to consider is the strength of the dipoles  $\mathbf{p_r} \propto \mathbf{u_r}$ . Comparing the rms value of  $\mathbf{u_r}$  for the two cases, we obtain an approximate equivalence  $\Delta \leftrightarrow \Delta^*/\sqrt{3}$ .

The uniform arrays show rich structure in resistance versus magnetic field,  $R(f_0)$ , with strong oscillatory behavior of period  $\Delta f_0 = 1$  out to fields as high as  $f_0 = 44$ , and secondary minima at  $f_0 = \frac{1}{2}, \frac{1}{3}, \frac{2}{3}, \frac{1}{4}$ , and  $\frac{3}{4}$ , as observed in previous measurements.<sup>1,2</sup> Structure at such large fields is unusual for proximity-effect arrays, and indicates the high quality of our samples.

In contrast with the uniform case, the  $R(f_0)$  data for our disordered samples show oscillations only at low fields. The oscillation amplitude decreases with increasing field, with more disordered samples showing a more rapid decrease.

Figure 3 shows  $R(f_0)$  for two samples, with  $\Delta^* \approx 0$  and  $\Delta^* = 0.10$ . The complex background which modulates the resistance oscillations is due to the magnetic field modulating the coupling energy of the individual junctions. From the lower trace in Fig. 3, one can see that this actually suppresses resistance oscillations completely at the first maximum in the background,  $R_{\max}$ , at  $f_0 \approx 12$ , corresponding to the first minimum of the single-junction critical current. This suggests an empirical way to compensate for single-junction effects: We determine the oscillation amplitude  $\Delta R(f_0)$  as illustrated in the inset in Fig. 3, and introduce a rescaled oscillation amplitude  $\Delta R'(f_0) \equiv \Delta R(f_0)/[R_{\max} - R(f_0)]$ .



FIG. 3. Resistance vs magnetic field  $R(f_0)$  for  $\Delta^* = 0.10$  (upper trace) and  $\Delta^* \approx 0$  (lower trace), showing oscillations due to collective behavior, modulated by single-junction effects. Inset shows definition of oscillation amplitude  $\Delta R(f_0)$ .

Figure 4(a) shows  $\Delta R'(f_0)$  for samples with various amounts of disorder. The oscillation amplitudes decreases linearly with  $f_0$ , samples with greater disorder showing a steeper slope. Our experimental critical field  $f_c^{expt}$  is the field where a least-squares fit intercepts the line  $\Delta R' = 0$ . Figure 4(b) shows  $f_c^{expt}$  as a function of  $1/\Delta^*$ , and demon-



FIG. 4. (a) Amplitude of resistance oscillations vs  $f_0$  for various values of  $\Delta^*$ . Lines are least-squares fits, whose extrapolations to zero define experimental critical fields  $f_c$ . (b) Values of  $f_c$  from (a), plotted vs  $1/\Delta^*$ . Line is least-squares fit constrained to have zero intercept.

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strates the linear dependence predicted by (1), with  $f_c^{expt}\Delta^* \approx 0.95$ . This result is independent of temperature for data taken in the tail of the resistive transition, where structure is most pronounced.

Using the approximate conversion  $\Delta \leftrightarrow \Delta^*/\sqrt{3}$  discussed earlier, our data thus give a result for the critical field,  $f_c^{\text{expt}} \approx 0.95/\Delta^* \approx 0.55/\Delta$ . The experimental critical field as defined here is much larger than predicted by (1). As noted above, however, the theoretical  $f_c$  corresponds to the destruction of long-range phase coherence. By contrast,  $f_c^{expt}$  measures the destruction of phase coherence on a length scale  $L \sim a$ . This short-range coherence is much less easily destroyed and therefore gives a larger critical field, although with the same dependence on disorder,  $\Delta^*$ . This can be quantified further by looking at the destruction of structure in  $R(f_0)$  at rational values  $f_0 = p/q$ , which is due to coherence on a length scale  $L \sim qa$ . Although the present theory is only valid for  $f_0$  an integer, data at  $f_0 = n \pm \frac{1}{2}$  and  $n \pm \frac{1}{3}$  suggest that one can define experimental critical fields  $f_c^{expt}(q)$  for these higher-order effects. Empirically we find  $f_c^{expt}(q)\Delta = c_1 + (c_2/q^2)$ , with  $c_1$  and  $c_2$  constants, and that extrapolating to  $q = \infty$  gives a result  $f_c^{\text{expt}}(\infty) \Delta = c_1 \approx 0.06$ , in quite good agreement with (1).

To further our understanding of this system we have also performed Monte Carlo simulations of XY spin systems with positional disorder. Our starting point is the Hamiltonian for the frustrated XY spin system,  $H = -J\sum_{(i,j)}\cos(\theta_i - \theta_j - \psi_{ij})$ . Here  $\psi_{ij} = (2\pi/\phi_0)\int \mathbf{A} \cdot dl$ , is the line integral of the magnetic vector potential,  $\mathbf{A}$ , between the centers of nearest-neighbor superconducting sites *i* and *j*, and  $\phi_0 = hc/2e$  the superconducting flux quantum. Positional disorder is introduced by imagining each site of the lattice to be displaced by a random amount, given by a Gaussian distribution of width  $\Delta$ . This leads to randomness in the  $\psi_{ij}$ 's.

Using the Metropolis rule<sup>9</sup> on small (typically  $16 \times 16$ ) lattices, with periodic boundary conditions, we calculated the helicity modulus Y using Eq. (3.6) of Ref. 10 as a function of temperature, magnetic field, and disorder  $\Delta$ .<sup>11</sup> The helicity modulus gives the rise in the system free energy in response to a long-wavelength twist imposed on the phases  $\theta$ , and is proportional to the effective superfluid density for an array. Whereas in the pure system Y increases monotonically as temperature decreases, we expect that in the disordered case Y will decrease and actually go to zero again at low temperatures.

Our most extensive simulations are for a value of  $\Delta = 9.9736 \times 10^{-4}$ , so that the theoretical critical field (1) is 100. This enables us to consider integer values of  $f_0$  very close to  $f_c$ . Figure 5 shows some of our results for the helicity modulus of a  $16 \times 16$  array. The upper curve is for  $f_0=0$ , where the positional disorder has no effect. As temperature decreases there is a monotonic increase in Y, with the usual finite-size-broadened rise at  $T_c \approx J$ . The lower curve shows results for  $f_0=98$ , and represents an average over 37 disorder realizations. At such a field we should be at the tip of the theoretical phase boundary, where the reentrant transition temperature  $T_c^{-1}(f_0)$  should be a maximum. The results show complex behavior, with Y increasing, then decreasing over a narrow tem-



FIG. 5. Simulation results for the helicity modulus, Y, of a  $16 \times 16$  disordered array. The value of  $\Delta$  is such that  $f_c = 100$ . The upper curve is for  $f_0 = 0$  and the lower for  $f_0 = 98$ .

perature range around T/J = 0.5, and then finally increasing again as temperature decreases. The largest fluctuations in Y also occur near T/J = 0.5, where Y is reentrant, as is typical of thermodynamic quantities near a phase transition. This reentrant behavior is confined to a narrow range of fields close to  $f_c$ . For example, our results for  $f_0=80$  and  $f_0=120$  show no evidence for reentrance but exhibit the same overall background shape as for  $f_0=98$ , with Y increasing monotonically as temperature decreases. Results at  $f_0=96$  are inconclusive, showing only a slight dip in Y at  $T/J \approx 0.3$ , with a maximum in the fluctuations at the same temperature.

There appears, then, to be some mechanism which is counteracting the expected reentrance, leading to a finite value of Y as T goes to zero, even for  $f_0 > f_c$ . The rise at low temperature may result from the finite size of our simulated spin system. According to GK, order should be destroyed at low temperatures due to the presence of free vortices. Since the vortices are thermally activated, however, there will be fewer present at low temperatures. In fact, in a small sample there is a strong probability that there are actually no vortices present, so that the helicity modulus does not go to zero. Further simulations of larger arrays might clarify the role of finite sample size but are currently impractical for our computer facilities given the need for extensive disorder averaging. Alternatively, the finite value of Y at low temperatures might be due to the pinning of vortices by the disorder, since pinned vortices will not destroy phase coherence. In this regard, Nelson has pointed out to us that this system is formally identical to a two-dimensional (2D) random binary mixture of hard spheres, where the disorder is due to the presence of a random admixture of large spheres, which disrupt translational order. Experiments on such a system showed a finite shear modulus (analogous to the helicity modulus) despite the absence of translational order, because the dislocations (analogous to vortices) were pinned by the large spheres over laboratory time scales.<sup>12</sup>

We are currently looking for evidence of reentrant behavior in our experiments, as well as extending our simulations to clarify the nature of the reentrant behavior in finite samples. We are pleased to acknowledge useful discussions with J. M. Kosterlitz, E. Granato, D. Stroud, Jean S. Chung, D. R. Nelson, and S. P. Benz during the course of this work. We are also indebted to H. Rogalla for writing the *e*-beam lithography software, and to P. Sokol for providing computer time. This work was supported in part by the National Science Foundation, through Grants No. DMR-86-14003 and No. DMR-84-04489, and used facilities supported in part by the Office of Naval Research, Contract No. N00014-83-K-0383.

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- <sup>1</sup>C. J. Lobb, Physica B 126, 319 (1984), and references cited therein.
- <sup>2</sup>Ch. Leemann, Ph. Lerch, B.-A. Racine, and P. Martinoli, Phys. Rev. Lett. **56**, 1291 (1986); R. K. Brown and J. C. Garland, Phys. Rev. B **33**, 7827 (1986); B. J. van Wees, H. S. J. van der Zant, and J. E. Mooij, *ibid.* **35**, 7291 (1987); D. J. Van Harlingen and K. N. Springer, Bull. Am. Phys. Soc. **32**, 690 (1987).
- <sup>3</sup>J. Villain, J. Phys. C 10, 1717 (1977).
- <sup>4</sup>S. Teitel and C. Jayaprakash, Phys. Rev. B **27**, 598 (1983); T. C. Halsey, J. Phys. C **18**, 2437 (1985); M. Yosefin and E. Domany, Phys. Rev. B **32**, 1778 (1985); M. Y. Choi and D. Stroud, *ibid.* **32**, 5773 (1985); E. Granato and J. M. Kosterlitz, *ibid.* **33**, 4767 (1986).
- <sup>5</sup>M. Rubinstein, B. Shraiman, and D. R. Nelson, Phys. Rev. B 27, 1800 (1983).

- <sup>6</sup>E. Granato and J. M. Kosterlitz, Phys. Rev. B 33, 6533 (1986).
- <sup>7</sup>T. Moriya, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1963), Vol. I, Chap. 3.
- <sup>8</sup>Throughout this paper we use the term "long-range phase coherence" to refer to the algebraic order which occurs in two-dimensional XY systems.
- <sup>9</sup>See, for example, *Monte Carlo Methods in Statistical Physics,* edited by K. Binder (Springer, Heidelberg, 1974).
- <sup>10</sup>W. Y. Shih, C. Ebner, and D. Stroud, Phys. Rev. B **30**, 134 (1984).
- <sup>11</sup>M. Y. Choi, Jean S. Chung, and D. Stroud [Phys. Rev. B 35, 1669 (1987)] have performed both simulations and a renormalization-group analysis for the case of full frustration,  $f_0 = \frac{1}{2}$ , with positional disorder. They found no evidence for reentrant behavior but some evidence for a critical field.
- <sup>12</sup>D. R. Nelson, M. Rubinstein, and F. Spaepen, Philos. Mag. A 46, 105 (1982).



FIG. 1. Schematic phase diagram for a 2D array with positional disorder. Vortex-unbinding transitions occur at  $T_c^-(f_0)$  and  $T_c^+(f_0)$ . In the region marked S (for "superconducting") the system shows long-range phase coherence. In the region labeled N (for "normal") this phase coherence is destroyed (Ref. 6).