TWO-DIMENSIONAL SUPERCONDUCTIVITY IN ULTRATHIN DISORDERED THIN FILMS

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Abstract

The status of the understanding of two-dimensional superconductivity in ultrathin, disordered thin films is reviewed. The different consequences of microscopic versus macroscopic disorder are stressed. It is shown that microscopic disorder leads to a rapid suppression of the mean-field transition temperature. The consequences of macroscopic disorder are not well understood, but a universal behavior of the zero-bias resistance as a function of field and temperature has been observed.

I. Introduction

Two-dimensional (2D) superconductivity has been of considerable interest for at least two decades, quite independent of the recent discovery of high-temperature superconductivity in the quasi-two-dimensional cuprate superconductors. Basically there are two reasons for this longstanding interest. Many novel effects are expected in two dimensions, and 2D superconductors are quite easy to make. Because of the relatively long coherence lengths typical of superconductivity (50 Å $< \xi(0) <$ 1000 Å), it is only necessary to make a relatively thin film to access the two-dimensional regime.

There are several types of 2D superconductors. On one side there are homogeneous thin films, in which any disorder occurs only on microscopic length scales. On the other, there are regular 2D Josephson junction arrays and superconducting lattices. In between these in a structural continuum are granular and inhomogeneous thin films, in which irregularities are present on macroscopic length scales. The importance of the distinction between microscopically and macroscopically disordered systems follows from the fact that disorder on length scales short compared to the superconducting coherence length $\xi(0)$ (*i.e.* on length scales smaller than the size of a Cooper pair) affect the BCS or mean-field transition temperature T_{co}, whereas, disorder on length scales large compared to $\xi(0)$ affect only the pair wavefunction or order parameter ψ. Of course, it is phase ordering of ψ that leads to actual superconductivity ($R = 0$). The true transition temperature of a superconductor $T_c \equiv T_c(R = 0)$ can be much less than T_{co}, in which case T_{co} corresponds to a crossover temperature at which pairs begin to form but not phase order. In this paper we are concerned with relatively homogeneous ultra-thin films of amorphous superconductors. Some of the other types are the subject of other papers in these Proceedings.

The basic phase diagram of a 2D superconductor with no macroscopic disorder is well known [1]. At zero applied magnetic field there is a Kosterlitz-Thouless (KT) vortex/antivortex unbinding transition at $T_c^{KT} < T_{co}$ below which true superconductivity exists. In a finite magnetic field no true superconductivity exists, but there is a vortex lattice at low temperatures that melts via a KT dislocation/antidislocation melting transition at a temperature $T_m^{KT} < T_c^{KT}$. For low fields T_m^{KT} is predicted to be field independent. Of greater current interest is how this picture is modified in the presence of macroscopic disorder.

Once macroscopic disorder or inhomogeneities are introduced, profound modifications to the above picture must arise. As shown originally by Larkin and Ovchinnikov, even arbitrarily weak disorder will disrupt the vortex lattice [2]. The correlated (lattice-like) regions are large for small disorder but get progressively smaller as the strength of the disorder increases. Of course, macroscopic disorder also brings vortex pinning and the possibility of zero resistance. It is also known that in the presence of disorder (macroscopic or microscopic) electrons localize and any metallic thin film must become an insulator as $T \to 0$. Accompanying such localization are enhanced Coulomb interactions associated with the decrease in screening in a localizing electron system. Thus, disorder both aids and hinders superconductivity. How this competition plays out is the central issue of this paper.

While a complete understanding is not at hand, one comprehensive schematic proposal has been put forth by M.P.A. Fisher and his coworkers [3]. Their proposed phase diagram is shown in Fig. 1. The idea is that in the presence of macroscopic disorder, the vortex lattice is destroyed but remains correlated (in a spin glass sense) with a correlation length that diverges as $T \to 0$. Moreover, only at $T = 0$ is the resistance truly equal to zero, *i.e.* $T_c = 0$. This idea, or close variants of it, have been discussed by several authors [1,4].

More remarkably, Fisher and his coworkers propose that at $T = 0$, superconductivity becomes destroyed if the strength of the disorder increases beyond some critical value. Specifically, they propose that there is a second-order superconductor/insulator transition at a sheet resistance $R_\square \simeq h/4e^2 \simeq 6000\Omega$. Even at smaller $R_\square$, there exists a critical magnetic field above which the insulating state is entered, as also shown in the figure. Implicit in this picture is the assumption that T_c is sufficiently smaller than T_{co} that one may speak of Cooper pairs that are localized into a Bose glass in the insulating phase. Moreover,the superconductor/insulator transition is associated with charge fluctuations (quantum fluctuations) in the superconductor at $T = 0$. This transition is

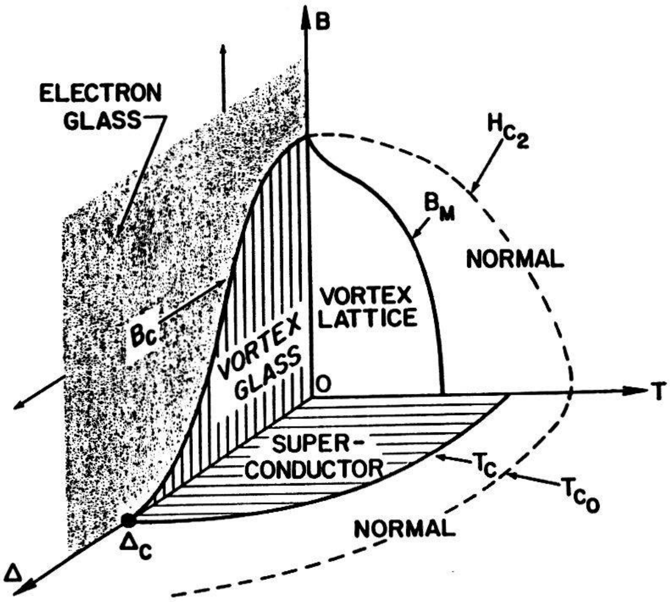

Fig. 1. Proposed schematic phase diagram of a 2D superconductor in the presence of disorder. (From Reference 3.)

presumably closely related to that discussed by Schon in these Proceedings for the case of highly undamped Josephson junction arrays.

With this background in mind, let us now turn to the properties of ultra-thin amorphous superconducting thin films to see what insight they provide on how superconductors are affected by disorder.

II. Superconducting Properties of Ultra-thin Amorphous Thin Films

A. Mean-field Transition Temperature T_{co}

Ultra-thin amorphous Mo-Ge superconducting films down to 10 Å thickness have been studied for some time now in the Stanford group beginning with the work of Graybeal [5]. They are co-sputtered onto amorphous substrates mounted on a rotating table so as to ensure maximal homogeneity. The films are quite homogeneous in the sense that most of the disorder is clearly on microscopic length scales. The electron mean-free path $\ell \simeq 3$ Å is much smaller than $\xi(0) \simeq 50$Å. On the other hand, as we shall see, the films have critical currents at $T = 0$ roughly on the order of 10^4 A/cm^2, and hence some macroscopic disorder must be present.

The uniformity of the films has been confirmed by x-ray diffraction (on thick films) and TEM cross-sectional micrographs of the ultra-thin films. Moreover, the resistivity of these films ($\rho = 250\mu\Omega$-cm) is found to be constant independent of thickness, except for small changes ($< 10\%$) consistent with of the theory of localization. Thus there is no

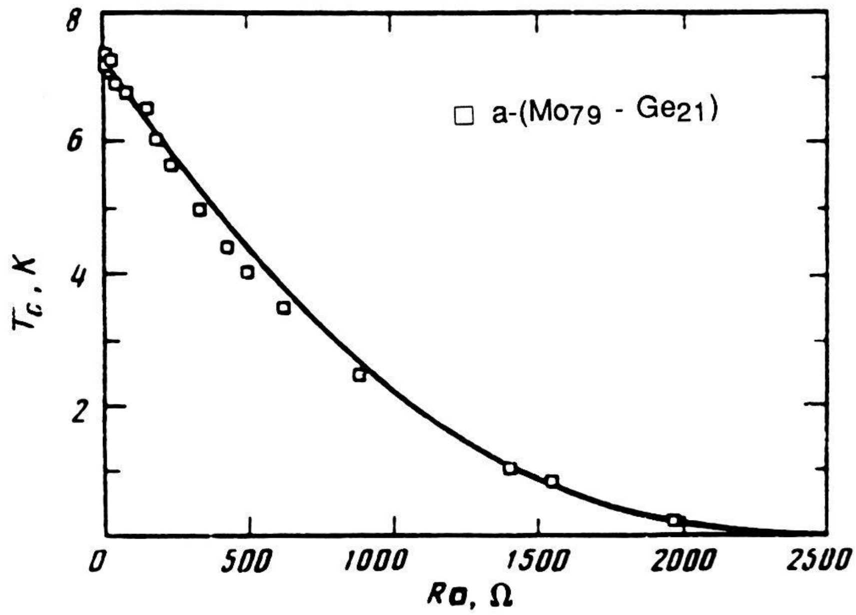

Fig. 2. Observed reduction of the mean field T_c of amorphous Mo-Ge thin films as function of sheet resistance (Ref. 5) compared with theory of Ref. 8. (From Reference 8).

reason to believe that the bulk electronic properties of the films have changed as they are made thinner. To this extent, any changes in the superconducting properties of the films can be associated with the progressive two-dimensionality of the films as they are made thinner.

Figure 2 shows how the mean-field T_{co} decreases as film thickness is reduced. The data are plotted as a function of sheet resistance, $R_\Box = \rho/d$, but since ρ is constant this is equivalent to a plot *vs.* d^{-1} where d is the film thickness. The solid line is a theoretical prediction to be discussed below. In these data, T_{co} was defined as the midpoint of the resistive transition, which was shown to be effectively equal to the temperature at which the fluctuation conductivity would diverge in all cases where a check was made.

Clearly T_{co} is rapidly depressed as a function of $R_\Box$. This depression of T_{co} is much faster as a function of $R_\Box$ than the decrease in T_c^{KT} with $R_\Box$ expected theoretically [6]. Hence, different physics is involved. We have interpreted this decrease in T_{co} with $R_\Box$ as a result of the enhanced Coulomb interactions expected due to the poorer screening associated with diffusing (as opposed to balistic) electrons, particularly in reduced dimensions.

The reduction of T_{co} with $R_\Box$ expected due to these enhanced Coulomb interactions has been studied extensively by Fukuhama and coworkers as a perturbation on BCS theory [7]. Physically the reduction is due to a combination of a reduced density of states, corrections to μ^* and depairing due to an increase in the inelastic electron-electron scattering rate. The relative contributions of each of these factors is not so clear, however. Finkelshtein has argued that as $R_\square$ increases ($R_\square > 0.012h/e^2 \simeq 500\Omega$) a scaling theory of the Coulomb interactions is required [8]. A fit of Graybeal's data to this more complete theory is shown by the solid line in Fig. 2.

Quite independent of any theoretical fit, it is important to emphasize that T_{co} decreases so rapidly with $R_\square$ in these amorphous systems that it is very small before the sheet resistance at which the superconductor/insulator transition shown in Fig. 1 is expected. While not in themselves proving one way or the other whether such a transition exists, these results dramatically illustrate the need to distinguish between microscopic and macroscopic disorder. Concomitantly, they suggest it is important to establish on what length scale the disorder reflected in $R_\square$ actually arises. For a striking contrast to the behavior observed with amorphous Mo-Ge alloys, see the paper by Hebard in these Proceedings on In-oxide films.

While the fit to theory shown in Fig. 2 is obviously quite good, it does not rule out the possibility that the reduction of T_c is due to a proximity effect. To show that the observed reductions in T_{co} are not due solely to a proximity effect, and to establish an upper limit on the length scale over which screening is weakened in these ultra-thin films, Missert *et al.* carried out a careful comparison of the T_{co} of amorphous Mo-Ge single layers and multilayers [9]. In the multilayers, extra screening of any given layer is provided by adjacent layers. Also, motion into the third dimension may be permitted, depending on the nature (insulating or normally conducting) and thickness of the nonsuperconducting layers in the mulilayer. In making the comparison, over and under layers of the nonsuperconductor were included in the single layer in a fashion such that any proximity effect would be identical to that in the multilayer. The results unambiguously show that T_{co} increases when electrons are permitted to move in three dimensions. They also set an upper limit of $\sim$ 20 Å for the screening length in these materials.

Thus the case for enhanced Coulomb interactions reducing T_{co} seems on firm ground, provided the theory is consistent with such a short screening length. The lack of long-range Coulomb effects demonstrated by the results of Missert is also consistent with the reduction of the transition temperature being a mean-field effect, rather than related to the quantum fluctuation driven superconductor/insulator transition proposed by Fisher and coworkers. Since a sheet resistance of $\sim$6000Ω would require depositing a uniform 4 Å thick film, a direct test of whether this transition occurs in a homogeneous, microscopically-disordered amorphous Mo-Ge film seems impossible. Measurements in very high magnetic fields are of obvious relevance, however.

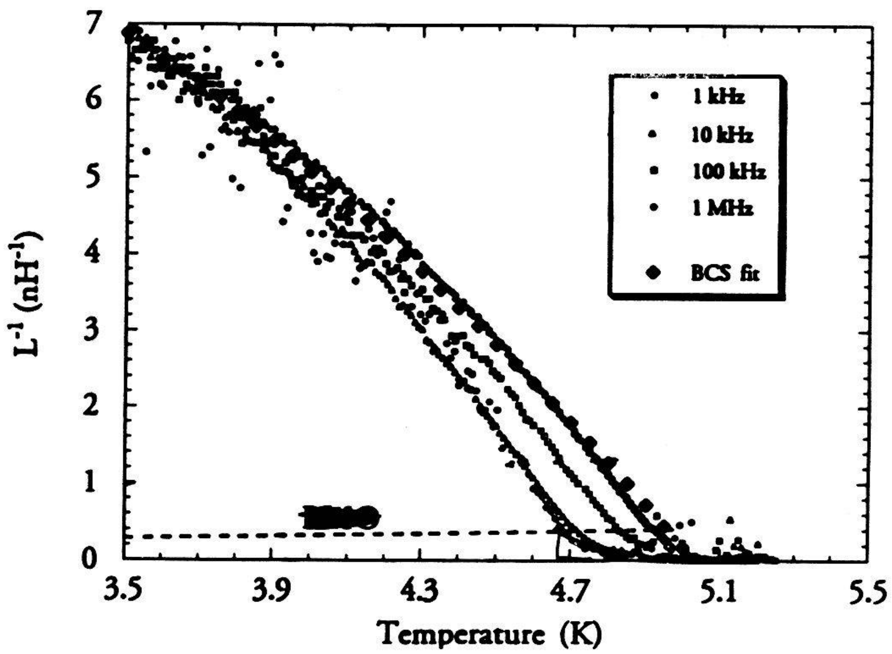

Fig. 3. Measured inverse sheet kinetic inductance $L_{K\square}^{-1}$ of a 60 Å thick amorphous Mo-Ge film versus temperature.

B. Effects on the Pair Wavefunction

More recently we have been examining the macroscopic quantum behavior of these ultra-thin disordered films. Some of these results are only preliminary while others are well established. One obvious question is whether the zero-field KT vortex/anti-vortex unbinding transition occurs in these films. As a first step in answering this question, Hahn and White have measured the sheet inductance $L_{K\square}$ of some of our films [10]. Earlier measurements were also made by Missert collaboratively with Hebard and Fiory [11]. The technique used was the familiar mutual inductance measurement.

The data for a 60 Å thick sample are shown for various measurement frequencies in Fig. 3, plotted in such a way to test the Kosterlitz-Nelson universal relation

$$kT_c^{KT} = \frac{\pi}{2}\frac{\hbar^2}{2m}n_s^{2D}(T_c^{KT}) = \frac{\pi}{2}\left(\frac{\hbar}{2e}\right)^2 L_{K\square}^{-1}$$

between T_c^{KT} and the superfluid sheet density n_s^{2D} (written in terms of the experimetally accessible quantity $LL_{K\square}$). The KT transition should occur where the dashed straight line (proportional to T) intersects the data for $L_{K\square}^{-1}$ at the lowest measurement frequencies. No transition is evident, although the data are well below the BCS mean-field behavior observed for the highest frequencies. A BCS fit to the high frequency data is also shown in the figure. It should be noted that the frequency dependence of the intersection points for the various curves is consistent with that expected from the dynamical theory of the KT transition, even if the predicted jump in n_s^{2D} is not seen.

We do not currently understand why no KT transition is observed, although macroscopic inhomogenities in the films are an obvious possibility. Unfortunately, there are no well-developed theoretical treatments of the effects of disorder on the KT transition.

In a preliminary way Hahn and White have also looked for the KT melting transition in an applied magnetic field. Again, mutual inductance measurements were used. The peak in the dissipative part of the signal was used as an indication of a change in the dynamics of the vortices and studied as a function of field and temperature. The results are described in the paper by White *et al.* in these Proceedings. The conclusion is that while the data show a frequency dependence consistent with a KT melting transition at a finite T_m^{KT}, no final conclusion can yet be drawn.

Much more definitely known is how the dc zero-bias resistance of these films behaves as a function of field and temperature in the region above the inferred T_m^{KT}. Figure 4 shows some typical I-V curves measured by Graybeal on a 50Åthick Mo-Ge film as a function of reduced temperature [12]. Clearly the I-V's become increasingly nonlinear as temperature is increased. At the lowest temperature a critical current of $\sim 230\mu$A corresponding to $J_c \simeq 10^4$ A/cm^2 can be defined above which classical Bardeen-Stephen flux flow is seen. Also clearly evident at the high temperatures is a finite zero-bias resistance.

The field and temperature dependence of this zero-bias resistance is shown in an Ahrenius plot in Fig. 5. The resistance is clearly thermally activated over the entire range of the measurement. Qualitatively this is consistent with the fact that $T > T_m^{KT} > T_G = 0$ where T_G is the vortex glass transition temperature. Much more interesting is that Graybeal was able to show that the data exhibit a universal behavior over the entire range of measurement. As described in Ref. 12, the data follow the empirical law

$$R_{I\rightarrow O} = 0.5 R_N \exp\left\{-\frac{A(T_{c2}(H)-T)}{H^{2/3}R_{\Box}}\cdot\frac{1}{T}\right\}$$

As also described in Ref. 12, all aspects of this law can be understood on simple physical grounds except the $H^{-2/3}$ field dependence of the activation energy. Since the density of votices is proportional to H, this $H^{-2/3}$ dependence contains information about the nature of the correlated vortex motion of the system. Note that a similar experiment on high-T_c YBCO/PBCO multilayer yields a $\log H$ dependence, as discussed by Fisher in these Proceedings.

An even more intriguing aspect of the correlated nature of the vortex motion in these 2D superconductors is shown in Fig. 6, based on very recent data of White [13]. The figure shows the bias current dependence of the differential resistance of films identical to those studied by Graybeal. At low I, Graybeal's result is recovered. However, as the

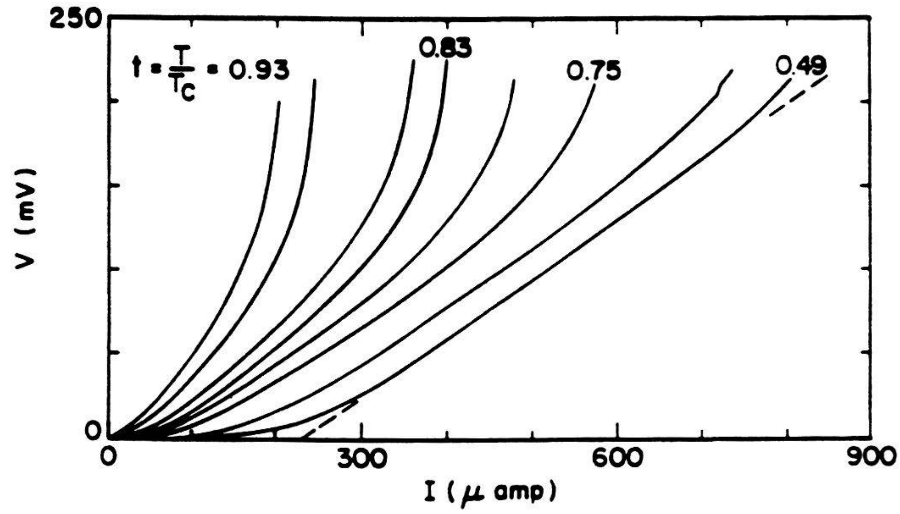

Fig. 4. I-V curves of a thin film of amorphous Mo-Ge for various reduced temperatures. (From Reference 12.)

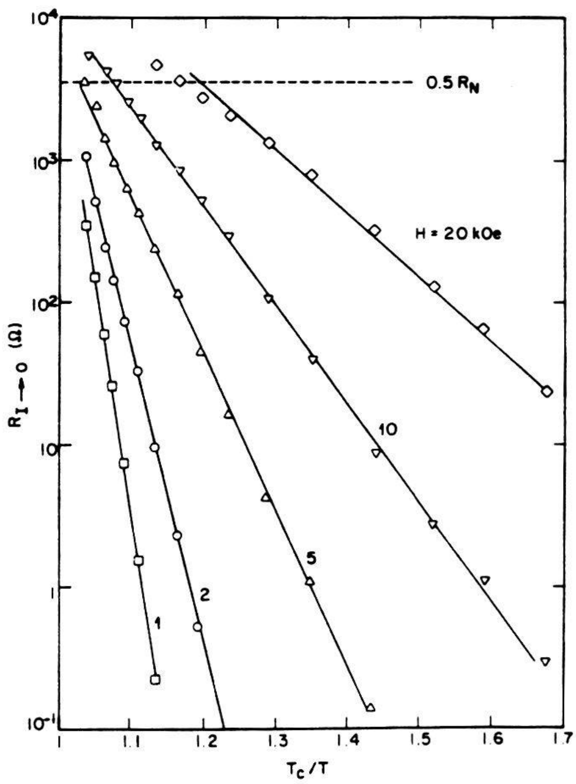

Fig. 5. Zero-bias resistance of the same film as in Fig. 4 as a function of temperature for various magnetic fields. (From Reference 12.)

bias current increases, there is a rise in dV/dI to an intermediate plateau before the final rise to the Bardeen-Stephen flux flow regime. While the data are still preliminary, they suggest that the application of a bias current to the vortices in a 2D superconductor, destroys their correlations in two stages and not continuously. Clearly, the temperature and field dependence of this decorrelation process will be of great interest. Hopefully, it

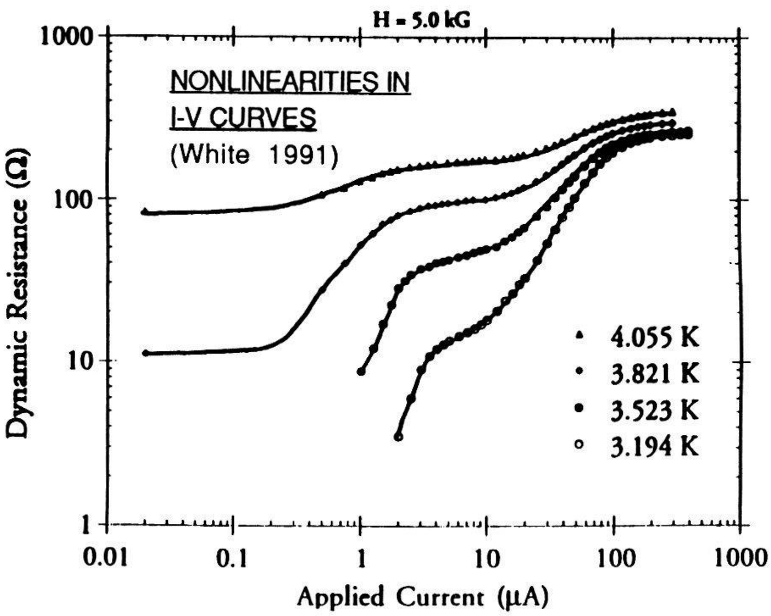

Fig. 6. Bias current dependence of the differential resistance of a thin amorphous Mo-Ge film such as shown in Figs. 4 and 5. Note two-stage crossover to classical flux flow.

will provide a means of distingushing between the various theories of these correlations.

In summary, the detailed behavior of relatively homogeneous micoscopically disordered 2D superconductors is becoming progressively clearer from the experimental point of view. There can be no doubt that the mean-field T_{co} is dramatically reduced by microscopic disorder, and that almost certainly this reduction is due to enhanced short-range Coulomb interactions. When some macroscopic disorder is present, as evidenced by a modest (10^4 A/cm^2) zero-temperature critical current density, the resistance of these films in a finite applied field remains finite for temperatures down to $\sim 1/2\ T_{co}$. Also at this level of macroscopic disorder there is no well-defined signature of the zero-field KT vortex/antivortex unbinding transition. There are, however, very definite correlations in the motion of the vortices at finite field that exhibit a very characteristic dependence on the vortex density. Moreover, the evidence to date suggests that those correlations may be destroyed in a two-step process under the presence of a bias current.

Acknowledgements

The author would like to thank his various Stanford collaborators over the years for many seminal discussions. He also acknowledges recent clarifying discussions with A. Hebard, C. Lobb, G. Schon, J. Mooij, and Ø. Fischer. This work originally supported by the NSF and currently supported by the AFOSR.

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