Evidence for a quantum-vortex-liquid regime in ultrathin superconducting films

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We report the observation of activated dissipation in ultrathin superconducting films near the superconductor-to-insulator transition (SIT) in magnetic fields. The activation energy T_0 scales with the mean field transition temperature, film thickness, and logarithm of the applied field, in qualitative agreement with the activation of defects in a vortex solid. We find T_0 extrapolates to zero at H_0, a field well below the upper critical field implying the existence of a quantum-vortex-liquid regime. We discuss the implications of this result for the magnetic field tuned SIT in this system. [S0163-1829(96)51446-3]

Recent extensive studies of type II superconductors have revealed the vortex phase diagram to be extremely rich. It contains a variety of phases depending on such variables as the dimensionality, intrinsic disorder, and electronic properties of the superconductor.[1] In particular, it has been observed that thermal fluctuations can lead to the melting of the vortex lattice into a liquid at a field below the upper critical field H_{c2}.[2] It has been proposed that quantum fluctuation effects can be strong enough to melt the vortex solid in disordered superconductors.[3] By inducing fluctuations in the amplitude of the superconducting order parameter, the disorder increases the fluctuations of the vortices about their equilibrium positions. Experimental evidence for the melting of the vortex solid into a quantum vortex liquid (QVL) has been obtained on a 3D disordered superconductor.[4] In 2D the QVL phase has been predicted to occur at high fields in superconducting films with very high normal state sheet resistances $R_N \sim R_Q = 6.45$ kΩ.

Interestingly, the presently accepted phase diagram for disordered superconductors in 2D does not include a QVL phase. This $T=0$ phase diagram, based on a model due to Fisher, contains a superconducting ($R=0$) vortex glass phase at low fields that condenses into an insulating ($R=\infty$) vortex superfluid phase, which is distinct from a QVL phase, at high fields.[5] A single critical field marks the boundary of this superconductor-to-insulator transition (SIT). Quantum fluctuations in the phase of the superconducting order parameter are responsible for this transition and fluctuations in the amplitude are considered irrelevant.[5,6] This theory accounts well for the scaling behavior of data obtained in InO_x (Ref. 6) and Josephson Junction (JJ) arrays,[7] systems in which the order parameter amplitude is understood not to be susceptible to fluctuations. The same scaling analysis has been applied to a number of other systems in which the order parameter is small and a field tuned SIT is observed.[8,9]

It has been suggested recently, however, that the above picture may not apply to certain systems such as ultrathin homogeneous films in which the order parameter amplitude is suppressed by disorder and likely to fluctuate.[10,11] In this work we discuss transport data on Bi/Sb and PbBi/Ge films as evidence that a QVL regime exists in the 2D vortex phase diagram in such systems. The data also suggest an alternative physical interpretation of the SIT in which the vortex solid phase melts at a field below the critical field for the SIT. In the data, at low fields, the resistance is thermally activated at low temperatures and the field dependence of the activation energy is consistent with the motion of defects in a vortex solid. However, the characteristic field at which the activation energy scales to zero falls well below H_{c2}, the field at which all evidence of superconducting fluctuations has disappeared. Recent theories suggest that this result can be attributed to quantum fluctuation effects[3,12,13] and the appearance of a QVL regime.[3,12]

We have performed transport measurements on two materials, $Pb_{0.9}Bi_{0.1}$/Ge and Bi/Sb and tunneling measurements on PbBi/Ge. All materials were thermally evaporated at low temperature, $T\sim 8$ K. 0.6 nm Ge or 1.2 nm Sb were predeposited on fire-polished glass in each experimental run to serve as underlayers for producing ultrathin films with homogeneous properties. Films were in thermal contact with a dilution refrigerator and located at the center of an 8T superconducting solenoid which generates a magnetic field oriented perpendicular to the film plane. Film thicknesses were measured *in situ* with a calibrated quartz crystal. Four terminal transport measurements were performed using standard low frequency techniques. Additional voltage contacts measured the adjacent square of film to check film quality after each deposition. The resistances shown here are excitation voltage independent and were checked with dc *IV* curves. Tunneling measurements have been described previously.[10,11]

Many details of the mixed state transport properties of high R_N films in systems like PbBi/Ge and Bi/Sb have been established previously.[10,11] Increasing the magnetic field broadens the resistive transition until a field H^c, at which the resistance of the film does not increase or decrease by more than 5% over the measured range of temperature (down to $T/T_{c0} \sim 0.1$, where T_{c0} is the zero field, superconducting transition temperature; the temperature at which the film resistance has dropped to half the normal state resistance). H^c has been observed to scale roughly with the size of the energy gap measured by tunneling in zero field.[14] Tunneling measurements at H^c reveal the presence of superconducting fluctuations by the appearance of a small gap in the quasiparticle spectrum around the Fermi energy in both PbBi/Ge,[11] and similar behavior is expected in Bi/Sb.[10] The strongly suppressed order parameter in the vicinity of H^c contributes to the broadening of the resistive transitions in field by fluctua-

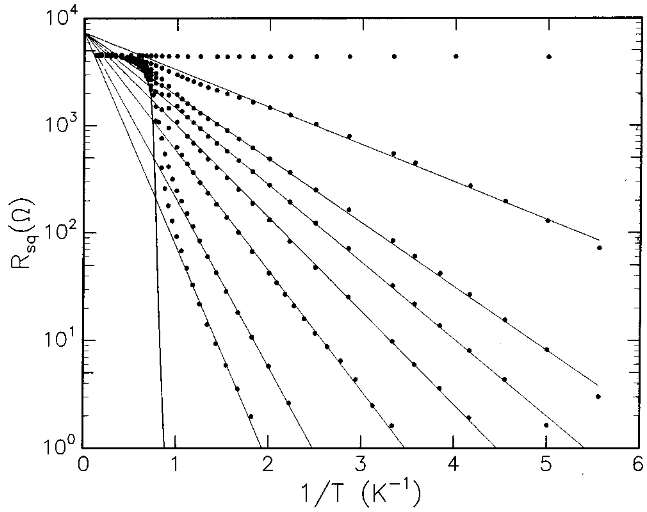

FIG. 1. Arrhenius plot of thermally activated dissipation in a Bi/Sb film with T_c=1.43 K and H^c=1.64 T in magnetic fields of 0.05 T, 0.1 T, 0.2 T, 0.3 T, 0.4 T, 0.5 T, and 0.8 T. Solid lines are least-squares linear fits to the activated region using a fixed intercept R_0.

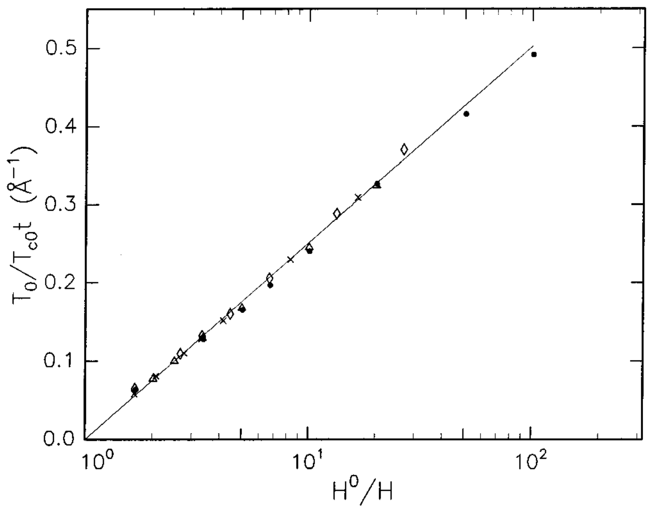

FIG. 3. Data from Fig. 2 collapsed onto a single line by dividing T_0 by $T_{c0}t$ and H by H_0, the field at which T_0 extrapolates to zero.

tions in a small quantity: the superfluid density. This claim is supported by empirical estimates which indicate that the number of pairing states in a coherence volume is on the order of 1.[11] At fields well above H^c, the film exhibits the logarithmic decrease of conductance with decreasing temperature characteristic of an insulating 2D disordered electronic system.

Here, we report that at fields well below H^c all of our Bi/Sb and PbBi/Ge films with $T_{c0}<3$ K exhibit activated dissipation. That is, when $R(T)\ll R_N$ it follows $R_0\exp(-T_0/T)$ over more than two decades of resistance as shown by the Arrhenius plot in Fig. 1. For ease of discussion, we will concentrate on the properties of four Bi/Sb films with $0.7<T_{c0}<1.7$ K and $7.1>R_N>4.1$ kΩ. We can make reasonable fits to these data, to extract T_0, by assuming a field independent prefactor, R_0. These least-squares fits to the activated portions are shown in Fig. 1. The value of R_0 was obtained by fitting the lowest temperature data for a field in the middle of the range. As shown in Fig. 2, T_0 exhibits a field dependence consistent with $\ln(1/H)$ over more than a decade of H for the four Bi/Sb films. The slope of the ln dependence, E_0, is comparable to T_{c0} (see Table I). The data in Fig. 2 can be collapsed onto a single straight line by dividing all T_0's of a film by its T_{c0} and film thickness t and scaling the applied field by its H_0, the field at which $T_0(H)$ extrapolates to zero (see Fig. 3). In all cases the characteristic field H_0 is less than H^c (see Table I). In PbBi/Ge films, H_0 is less than or nearly equal to H^c. We summarize our results for the activated region with the following expression for $R(T,H)$:

$$R(T,H)=R_1\exp[-(E_0/T)\ln(H_0/H)(1-T/T_{co})], \quad (1)$$

where the factor $(1-T/T_{c0})$ incorporates a temperature dependence to the activation barrier that is consistent with the activated form of the data. R_1 depends on field. The measured prefactor is given by

$$R_0=R_1(H_0/H)^{\alpha}, \quad (2)$$

with $\alpha=E_0/T_{c0}$. Empirically, R_0 depends weakly on magnetic field implying that,

$$R_1\propto H^{\alpha}. \quad (3)$$

For all of the Bi/Sb and PbBi/Ge films that we have investigated, $0.67<\alpha<1$ (see Table I for Bi/Sb film results), so that R_1 is nearly proportional to H. We have formulated the description in this manner to compare to recent theories (see below) which calculate the dissipation in this regime using the Bardeen-Stephen prefactor.

There have been a number of observations of activated resistance in other 2D and quasi-2D superconductors.[7,15–19] Systems with activation energies that depend logarithmically on H when $H\ll H_{c2}$ and scale with $T_{c0}t$ include high-T_c

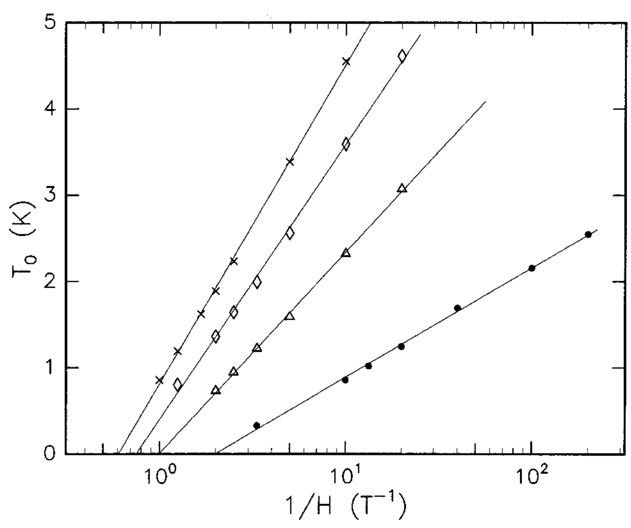

FIG. 2. Activation energy, T_0, plotted vs $\log(1/H)$ for four Bi/Sb films. (Circles) T_{c0}=0.69 K, (triangles) T_{c0}=1.15 K, (diamonds) T_{c0}=1.43 K, and (crosses) T_{c0}=1.63 K. The solid lines are least-squares fits. The slopes and x-axis intercepts are listed in Table I.

TABLE I. Parameters for four Bi/Sb films all from the same experimental run. $R_Q=4.12$ kΩ.

R_N/R_Q	t (nm)	T_{c0} (K)	H^c (T)	H_0 (T)	E_0/T_{c0}
1.72	0.76	0.69	0.78	0.51	0.8
1.29	0.82	1.15	1.33	1.01	0.88
1.10	0.87	1.43	1.64	1.33	0.97
0.995	0.9	1.63	1.9	1.66	0.99

compounds, layered structures, and amorphous thin films such as MoGe.[16–19] These data have been compared to models of thermally activated flux flow due to the motion of defects in a flux line lattice. The defects considered in the two most relevant models are edge dislocation pairs[1] and vortex/antivortex pairs.[20] Both lead to activation energies which depend on T_{c0} and t and logarithmically on H. Physically, these dependences have their roots in vortex-vortex interaction energies. Explicitly, the energy required to separate two overlapping vortices by a distance a_0 is[21]

$$U_{vv}=\frac{\Phi_0^2 t}{8\pi^2\lambda^2}\ln\left(\frac{a_0}{\xi}\right)\propto\ln\left(\frac{H}{2\pi H_{c2}}\right), \tag{4}$$

where λ is the penetration depth, ξ is the superconducting coherence length, Φ_0 is the flux quantum, and a_0 is the vortex lattice spacing.[22] In a dirty superconductor, $\lambda^{-2}\propto N(E_F)v_F l T_{c0}$ so that the coefficient of the logarithm scales like T_{c0}. $N(E_F)$ is the density of states at the Fermi energy, v_F is the Fermi velocity, and l is the elastic mean free path.[22] The characteristic field scale, $2\pi H_{c2}$, appears in both models. The dislocation model, due to Feigelman and co-workers, predicts a prefactor of the logarithm, E_0, that agrees quantitatively with experiment[16–19] while the vortex-antivortex unbinding model of Jensen *et al.* predicts activation energies that are more than an order of magnitude too high.[20]

Qualitatively, these theories can account for the activated behavior in Bi/Sb films as indicated by the collapse of the data in Fig. 3. Moreover, our observation that $R_1\propto H^\alpha$ with $\alpha\cong 1$ is strong evidence that the dissipation due to the motion of these defects follows the Bardeen-Stephen flux flow dependence in agreement with the models. Our measurement of E_0, however, falls between the two predictions suggesting that the defects involved in our films differ from those considered in Refs. 1 and 20.

An interesting quantitative deviation from these models is the result that $H_0<2\pi H_{c2}$. It implies that the length scale relevant to vortex-vortex interactions has become greater than $(2\pi)^{1/2}\xi_0$, [cf. Eq. (4)]. Recent theoretical work suggests disorder induced quantum fluctuation effects as the source of this deviation. Blatter and Ivlev have argued that strong disorder creates quantum fluctuations which smear the vortex cores making their effective radius greater than ξ_0.[3] A calculation by Spivak and Zhou indicates that there are large mesoscopic fluctuations in H_{c2} in 2D disordered superconductors. These spatial variations would result in a distribution of vortex sizes.[13] These pictures provide a connection between the evidence for order parameter amplitude fluctuations[11] and the enhancement of the characteristic length scale for interactions in the vortex solid.

The observation that H_0 is even less than H_{c2} leads to the interpretation that these quantum fluctuations are sufficient to melt the vortex solid. Explicitly, H_0 is the field at which the defect activation energy extrapolates to zero and as such provides an upper field limit for a vortex solid phase at any temperature. Thus, in the midst of a field regime where superconducting fluctuations are evident, the vortex solid melts.[23] At zero temperature it melts solely due to disorder induced quantum fluctuations. Presumably, the resistance of the melted phase is finite and proportional to R_N since the activated dissipation at lower fields follows a Bardeen-Stephen dependence.[24]

Recent theories discuss a quantum vortex liquid regime in 2D films with $R_N\sim R_Q=h/4e^2$. Blatter and co-workers have predicted that a QVL phase exists at $T=0$ from the argument that quantum motion of vortices can melt the lattice.[3] By including quantum fluctuations they were able to account quantitatively for the shape of the melting curve, $H^m(T)$, for 3D amorphous Nb_3Ge and explain why the melting field fell below the upper critical field at the lowest temperatures and highest magnetic fields. In Ikeda's theory, field driven quantum melting occurs when quantum fluctuations are strong enough to dominate the classical (thermal) fluctuation mode. A QVL regime arises and the mean field description becomes invalid.[12] Neither theory makes a prediction about the zero temperature transport in this regime.

The existence of a QVL regime for $H<H^c$ affects the interpretation of experiments on homogeneous superconducting films at the magnetic field tuned SIT. The success of the model of Fisher and co-workers[5] in describing the SIT in JJ arrays and InO_x films is expected because the order parameter amplitude appears to be well defined ($H^c\ll H_{c2}$).[6,7] However, a number of groups have scaled transport data in which H^c is much closer to H_{c2} and found similar exponents raising the possibility that the Fisher model applies.[8,9,14] In contrast, our results suggest that if a phase transition occurs at H^c in a homogeneous superconducting system, then it is a transition from a vortex liquid state with finite resistance to some other phase.[25]

We do not know what the phase is above H^c. Our earlier tunneling measurements suggest that its transport is dominated by paraconductance or amplitude fluctuation effects. Given the vortex liquid state below H^c, we speculate that H^c separates regimes in which phase fluctuations due to vortex motion and amplitude fluctuations dominate the transport properties. Spivak and Zhou describe a phase that may apply to the paraconductance dominated regime.[13] Their theory shows that disorder induced mesoscopic fluctuations give rise to spatial fluctuations in the upper critical field and they predict that, at fields approaching the average H_{c2}, a film consists of superconducting regions separated by normal regions. We speculate that, at and above H^c, the vortex cores overlap so strongly that the normal regions percolate across the film.

In summary, we have reported measurements of field dependent thermal activation energies, T_0, for dissipation in ultrathin superconducting films with $R_N\sim R_Q$. For both Bi/Sb and PbBi/Ge films, T_0 scales with film thickness, T_{c0}, and logarithmically with field. H_0 is the field at which the activation energy extrapolates to zero. These dependencies are explained by models of defects in a vortex solid phase but comparison to the models implies that disorder induced quantum fluctuations cause the field scale H_0 to fall well below H_{c2}. At fields near and above H_0, we have suggested that the vortex solid melts, and enters a vortex liquid regime that persists to zero temperature. We argue that the resistance in this regime is finite. We speculate that the existence of this QVL may be important to the $T=0$ phase diagram of the 2D field tuned SIT.

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[1] M. V. Feigel'man, V. B. Geshkenbein, and A. I. Larkin, Physica C **167**, 177 (1990); G. Blatter *et al.*, Rev. Mod. Phys. **66**, 1125 (1994).

[2] W. K. Kwok *et al.*, Phys. Rev. Lett. **69**, 3370 (1992); P. Gammel, A. F. Hebard, and D. J. Bishop, *ibid.*, **60**, 144 (1988).

[3] G. Blatter and B. Ivlev, Phys. Rev. Lett. **70**, 2621 (1993); G. Blatter *et al.*, Phys. Rev. B **50**, 13 013 (1994).

[4] P. Berghuis and P. H. Kes, Phys. Rev. B **47**, 262 (1993).

[5] M. P. A. Fisher, Phys. Rev. Lett. **65**, 923 (1990).

[6] A. F. Hebard and M. A. Paalanen, Phys. Rev. Lett. **65**, 927 (1990); A. F. Hebard, *Strongly Correlated Electronic Materials: The Los Alamos Symposium* (Addison-Wesley, Reading, MA, 1993).

[7] H. S. J. van der Zant *et al.*, Phys. Rev. Lett. **66**, 2531 (1991); **69**, 2970 (1992). The field dependence of the barrier was not established in this work.

[8] G. T. Seidler *et al.*, Phys. Rev. B **45**, 10 162 (1992); S. Tanda *et al.*, Phys. Rev. Lett. **69**, 530 (1992); Y. Liu *et al.*, Phys. Rev. B **47**, 5931 (1993).

[9] A. Yazdani and A. Kapitulnik, Phys. Rev. Lett. **74**, 3037 (1995).

[10] R. C. Dynes *et al.*, Phys. Rev. Lett. **57**, 2195 (1986); J. M. Valles, Jr., R. C. Dynes, and J. P. Garno, *ibid.* **69**, 3567 (1992).

[11] S.-Y. Hsu, J. A. Chervenak, and J. M. Valles, Jr., Phys. Rev. Lett. **75**, 132 (1995); J. M. Valles, Jr., S.-Y. Hsu, and J. A. Chervenak, J. Phys. Chem. Solids **56**, 1809 (1995).

[12] R. Ikeda, Int. J. Mod. Phys. B **10**, 601 (1996).

[13] B. Spivak and F. Zhou, Phys. Rev. Lett. **74**, 2800 (1995).

[14] Shih-Ying Hsu, Ph.D. dissertation, Brown University, 1995.

[15] A previous report, Y. Liu *et al.*, Phys. Rev. Lett. **68**, 2224 (1992), showed resistive transitions in similar systems exhibiting $R \propto \exp(T)$ which they attributed to quantum tunneling of vortices. We do not observe this signature in our data and neglect such effects in our analysis.

[16] T. T. M. Palstra *et al.*, Phys. Rev. Lett. **61**, 1662 (1988).

[17] O. Brunner *et al.*, Phys. Rev. Lett. **67**, 1354 (1991); Y. Suzuki *et al.*, Phys. Rev. B **52**, 6858 (1995).

[18] W. R. White, A. Kapitulnik, and M. R. Beasley, Phys. Rev. Lett. **70**, 670 (1993).

[19] X. G. Qiu *et al.*, Phys. Rev. B **52**, 559 (1995).

[20] H. J. Jensen *et al.*, Europhys. Lett. **20**, 463 (1992).

[21] $U_{vv}=(\varepsilon_0 c^2 \Phi_0^2 t/2\pi\lambda^2)\ln(a_0/\xi)$ (SI units).

[22] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, Florida, 1980).

[23] H_{c2}, though experimentally difficult to measure exactly, is taken to be the field at which all superconducting fluctuations in the resistance and the tunneling density of states are quenched.

[24] J. Bardeen and M. J. Stephen, Phys. Rev. **140**, A1197 (1965).

[25] Other workers, D. Ephron *et al.*, Phys. Rev. Lett. **76**, 1529 (1996), observed large deviations from activated dissipation at low fields and temperatures and came to a similar conclusion about the SIT in homogeneous MoGe films. Our systems, however, do not exhibit this deviation in the same limit of R/R_N, R_N/R_Q, and T/T_c.