Vortex-Lattice Melting in Nb

Matthew F. Schmidt, N. E. Israeloff, and A. M. Goldman

Center for the Science and Application of Superconductivity and School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455

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Detailed measurements and analysis of the temperature and magnetic field dependence of the magnetization and the magnetic relaxation of Nb films have been carried out. An irreversibility line has been identified, below which the remanent magnetization was found to decay logarithmically in time. The data were quantitatively compared to predictions of flux creep, vortex glass, and vortex-lattice-melting models. Only the vortex-lattice-melting model self-consistently explained the data.

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The magnetic-field–temperature (H-T) phase diagram for high-temperature superconductors has been the subject of intense interest. An irreversibility line (IL) and logarithmic decay of the remanent magnetization were first seen in ceramic LaSrCuO [1]. There has since been an ongoing debate over whether they are caused by glassy kinetics [2–4], flux creep [5–8], or vortex-lattice melting [9–13]. Although not necessarily expected [14], similar ILs and time-dependent magnetic properties have been observed in conventional type II superconductors [15,16]. While it is not clear that these features have the same origin as their counterparts in high-T_c materials—which are highly anisotropic and more heavily influenced by thermal fluctuations—conventional materials offer a simpler platform for the examination of the various models. In this Letter we present the results of detailed measurements and comprehensive analysis of the temperature and magnetic field dependence of the magnetization, and the decay of the remanent magnetization of Nb films. The analysis strongly suggests the irreversibility line in Nb films is a signature of vortex-lattice melting.

The sample studied in detail was rf sputtered onto an oxide layer on a Si substrate and had dimensions 5000 Å×2 mm×2 mm. The resistivity ratio was 5.9. Field-cooled (FC) and zero-field-cooled (ZFC) magnetization measurements were made in fields ranging from 10 to 3000 G. The procedure is well known and has been reported elsewhere [1,5,6,15,17]. The measurements were performed using a Quantum Design SQUID susceptometer with the sample aligned perpendicular to the magnetic field. The temperature control parameters of the susceptometer were adjusted to eliminate temperature overshoot at the sample during the ZFC measurement. To improve the precision of the data, temperature steps were limited to 0.05 K, and 16 scans were averaged at each temperature. A typical temperature sweep took 12 h. $T_c(H)$ was determined from the onset of diamagnetism, and $T^*(H)$, the irreversibility temperature, was defined as the lowest temperature where the difference between the FC and ZFC magnetizations was less than the standard deviation of the mean of the FC and ZFC measurements. Above T^* the magnetization was reversible; below T^* only the FC magnetization was thermal-history independent. (It has been reported that the FC magnetization of Nb *powder* can be thermal-history dependent below the irreversibility temperature [18], but no evidence of this was seen in our sample.)

In addition, the susceptometer's scan length was limited to 3.0 cm to minimize the effects of moving the sample through a nonuniform field—an experimental problem reported in earlier work using SQUID susceptometers on hysteretic samples [17,19]. (The fractional variation in field over this length is less than 0.0005.) All results reported here were found to be scan-length independent for scans less than 4.0 cm. A similar IL seen in vibrating reed experiments on Nb foils [16]—with the sample held stationary in the solenoid—provides additional evidence that this result is not an artifact of the scanning process.

Vanishing of the isothermal magnetic hysteresis has also been used to define an IL [15,17]. It has been reported that this method can give different results than the ZFC-FC method [15,20], but this was not seen in measurements on our sample. Since this method suffers from difficulty in producing the same field during the increasing and decreasing field sweeps due to flux trapping in the superconducting solenoid of the susceptometer [17], we concentrated on the ZFC-FC method.

Figure 1 shows the IL in the H-T plane. Houghton, Pelcovits, and Sudbo [9] used a nonlocal elasticity theory and a Lindemann criterion to derive an expression for the melting line in an anisotropic superconductor, which is given by

$$\frac{t_M}{(1-t_M)^{1/2}}\frac{\sqrt{b}}{1-b}\left(\frac{4(\sqrt{2}-1)}{\sqrt{1-b}}+1\right)=\alpha\,. \qquad (1)$$

Here α, the degree of susceptibility to thermal motion, is given by $\alpha=2\times10^5[H_{c2}(0)/T_c^2]^{1/2}(M/M_z)^{1/2}(c/\kappa)^2$, where $t_m=T_m/T_c(0)$, $b=H/H_{c2}(T)$, $H_{c2}(T)=H_{c2}(0)\times[1-T/T_c(0)]$, $\kappa\cong\lambda/\xi$ is the Ginzburg-Landau parameter, and M/M_z is the ratio of the in-plane and out-of-plane effective masses. The Lindemann criterion for melting is governed by the parameter c, which is the ratio of the mean-square thermal displacement of the flux lines from their equilibrium positions to the flux-line lattice

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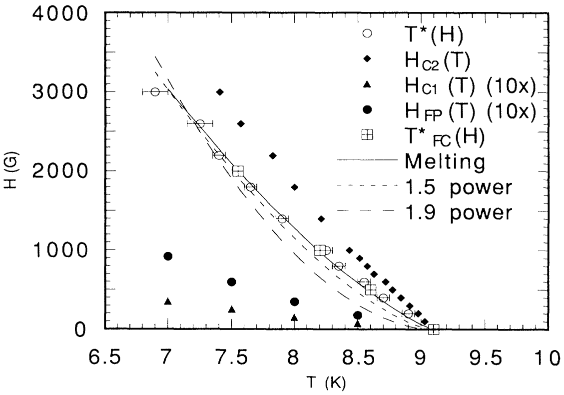

FIG. 1. The H-T phase diagram. The open circles with error bars are the ZFC-FC irreversibility temperatures T^*. The solid line is the melting prediction of Ref. [9], and the dashed lines are depinning lines. The squares are $T^*_{FC}(H)$, the temperatures where the FC remanent moment has decreased to the equilibrium value. Note: The lower critical field (H_{c1}) and full-penetration field (H_{FP}) have been scaled by a factor of 10 for improved visibility.

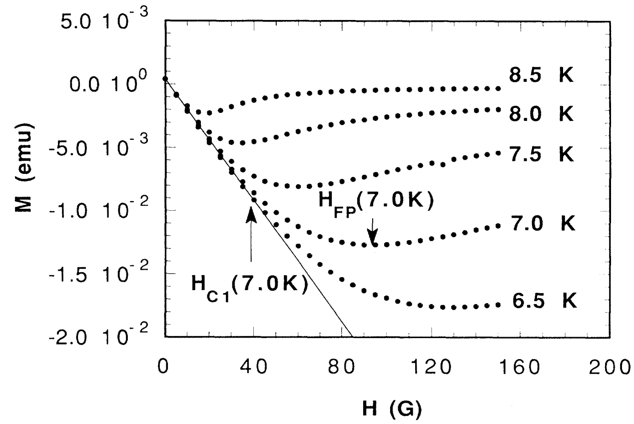

FIG. 2. Magnetization vs field for five temperatures. The lower critical field (H_{c1}) and full-penetration field (H_{FP}) for $T = 7.0$ K are indicated.

spacing. Suenaga *et al.* [15] have found good agreement between Eq. (1) and measurements on Nb_3Sn and Nb-Ti magnet wire, but were unable to make a meaningful comparison in the case of Nb because the difference between T_c and T^* had become too small. The parameters $T_c(0) = 9.11$ K and $H_{c2}(0) = 15\,200$ G were determined by fitting the equation $H_{c2}(T) = H_{c2}(0)[1 - T/T_c(0)]$ to the $H_{c2}(T)$ data shown in Fig. 1. The solid line is a single-parameter fit of Eq. (1) to the IL; the short-dashed line is a fit of the $\frac{3}{2}$-power-law prediction from the depinning theory of Yeshuran and Malozemoff [5]. (The de Almeida-Thouless glass model [21] suggested by Ref. [1] predicts the same $\frac{3}{2}$ power dependence.) While both curves look reasonable at first glance, it is clear upon closer examination that the melting line provides the better fit.

Since $M/M_z = 1$ for an isotropic superconductor, we can determine the Lindemann number c if κ is known. The lower critical field $H_{c1}(T)$ can be determined from a plot of magnetization M vs field H, but the procedure is complicated by the high demagnetization factor of the sample [22,23]. Figure 2 shows a typical plot of M vs H. M increases linearly with H at low fields, but the peak is rounded and there is no sharp maximum. M deviates from the linear dependence when the flux begins to penetrate at the outer edge of the sample [23], so this field is defined as H_{c1}. M reaches a maximum at a field $H_{FP}(T)$ when the flux penetration has reached the center of the sample. $H_{c1}(T)$ has been included in the H-T plane of Fig. 1. Defined in this manner it represents a *lower* bound for the bulk material, and combined with $H_{c2}(T)$, gives an *upper* bound for κ of 10.8.

This value of κ, together with the fitted parameter α, gives an *upper* bound of $c = 0.04$ for our Nb sample. This is somewhat smaller than the values determined by Suenaga: 0.065 and 0.1 for Nb_3Sn and Nb-Ti magnet wire, respectively [15]. It is substantially smaller than the value $c = 0.4$ determined by Houghton for a BSCCO sample. This is not unexpected however. In Ref. [10], Brandt offers several reasons why the Lindemann number might be lower than the oft-quoted value $c = 0.1$. He uses a nonlocal elasticity theory and a Lindemann criterion to obtain an expression similar to Eq. (1) for isotropic superconductors. He compares this to a different melting criterion based on thermal fluctuations of the shear strain and concludes: "smaller values $c \approx \frac{1}{20}$ appear thus more realistic." In addition, he offers five (rather technical) reasons why c might be even smaller still.

Also, in recent Monte Carlo simulations [24] Ryu *et al.* computed a melting line for Houghton, Pelcovits, and Sudbo's BSCCO sample. Their criterion for melting was based on the disappearance of in-plane translational order monitored by the Fourier transform of the density-density correlation function at the first Bragg point. They obtained the value of c corresponding to this criterion by computing the rms deviation of the vortices from their equilibrium positions, thus obtaining a field-dependent Lindemann number. They found that c decreases with decreasing flux line density, and hence, field. It ranged from 0.45 to 0.1 as the field went from 10^6 to 10^2 G. More important than the specific value, however, is *the trend* of their results: As the field shifts downward and the flux line density decreases, the interlayer coupling becomes relatively stronger than the in-plane correlations, yielding a very fragile three-dimensional lattice of straighter flux lines—*and a lower Lindemann criterion.* This suggests that the Lindemann criterion for Nb might indeed be much lower than the value found for the BSCCO sample, which would be consistent with our results. The $\frac{3}{2}$-power-law depinning line suggested by Yeshuran and Malozemoff (YM) in Ref. [5] also has strong implications for remanent magnetization decay studies. To derive the $\frac{3}{2}$ law, they assumed that the activation energy U_0 scales as $H_c^2 a_0^2 \xi$ where $a_0 = 1.075 \times (\Phi_0/B)^{1/2}$ is the flux lattice spacing. Using $H_c \propto 1-t$ and $\xi \propto (1-t)^{-1/2}$, they found $U_0(T) = U_0(0)(1-t)^{3/2}/B$. [In the Anderson-Kim theory [25], U_0 is assumed to scale as $H_c^2 \xi^3$, which leads to $U_0 \propto (1-t)^{1/2}$.] By combining this with the thermal-activation dependent critical current, $J_c = J_{c0}[1-(kT/U_0)\ln(Bd\Omega/E_c)]$ [26], and requiring that the critical current be zero, they found the depinning line was given by $H \propto (1-t)^{3/2}$.

The temperature dependence of the activation energy U_0 can be independently determined from magnetic relaxation studies. We performed FC magnetization decay measurements for temperatures from 4.5 to 9.2 K and in fields of 0 to 2000 G. Typical results are shown in the inset to Fig. 3. A magnetic field was applied while the sample was above T_c. The sample was then cooled below T_c and T^*, and the field was decreased by $|\Delta H| = 1000$ G. The resulting remanent magnetization decayed logarithmically on time scales of up to a few hours. The normalized logarithmic decay rates S versus temperature for final fields of 0, 500, 1000, and 2000 G are shown in Fig. 3. The decay rates were normalized to the magnetization measured 10 min after setting the final field to avoid introducing transient effects due to the magnet itself. Comparison with the full-penetration field $H_{\rm FP}(T)$ shown in Fig. 1 shows that $|\Delta H| = 1000$ G ensures the entire sample is in the critical state. (A partial critical state would complicate interpretation of the results [27].) The decay rates all increase monotonically up to a temperature $T^*_{\rm FC}(H)$, somewhat less than $T_c(H)$, at which point the initial remanent moment has decreased to the FC equilibrium value and no decay can be discerned. This is in contrast to the results for high-T_c materials, where a peak in the decay rate versus temperature was observed [8]. In Ref. [8], Hagen and Griessen explain this peak within the flux-creep picture by introducing a distribution of activation energies. The absence of a peak in our data suggests a single-activation-energy model is adequate. The four $T^*_{\rm FC}(H)$ values have also been plotted in the H-T plane of Fig. 1. They are in excellent agreement with the ZFC-FC IL and with the melting line of Ref. [9]. This is to be expected if the ZFC-FC and isothermal-hysteresis ILs are to be coincident.

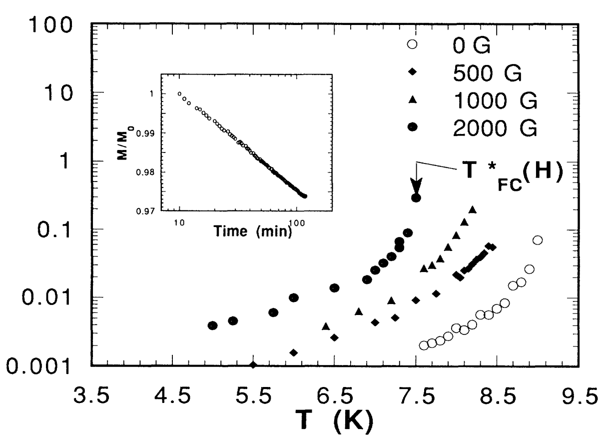

FIG. 3. Normalized logarithmic decay rate $S = -(1/M_0)dM/d\ln t$ vs temperature. The decay rates were normalized to the remanent moment M_0 measured 10 min after changing the field. Above $T^*_{\rm FC}(H)$ the initial remanent moment has decreased to the FC equilibrium value and no decay is observed. The inset shows a typical logarithmic decay of the normalized remanent moment M/M_0 for a final field $H=0$ and $T=6.0$ K.

In the single-activation-energy picture, when the decay rate is small compared to the magnetization, $-1/S = U_0(T)/kT - \ln(t_b/\tau_0)$ can be used to ascertain the temperature dependence of the activation energy U_0 [28]. Here t_b is the normalization time, and $1/\tau_0$ is an attempt frequency for hopping, typically on the order of 10^{10} Hz [8]. We assumed $U_0(T) = U_0(0)(1-t)^m$—which is consistent with both the Anderson-Kim and the YM assumptions—and fitted the equation to the data for the four fields. Results are shown in Fig. 4. We found $m = 1.1 \pm 0.1$ for all four fields, which is inconsistent with the YM result $m = 1.5$. When we fitted only the higher-temperature data, nearer the irreversibility line, *m decreased*, deviating even farther from the YM prediction. We also repeated the depinning-line analysis of Ref. [5], but using the measured temperature dependence of U_0 from the FC decay data. Where YM assumed the flux bundle volume contained one power of the coherence length ξ, and two powers of the flux lattice spacing a_0, we relaxed the criteria and allowed an unknown power (d) of the coherence length, and assumed $U_0(T)$ scales as $H_c^2 a_0^{3-d} \xi^d$ (this is equivalent to YM when $d=1$). This led directly to $U_0 \propto (1-t)^{2-d/2}/B^{(3-d)/2}$ and $H \propto (1-t)^{(4-d)/(3-d)}$. Thus, if our IL was to be the YM depinning line, our FC decay result $U_0(T) \propto (1-t)^{1.1}$ (im-

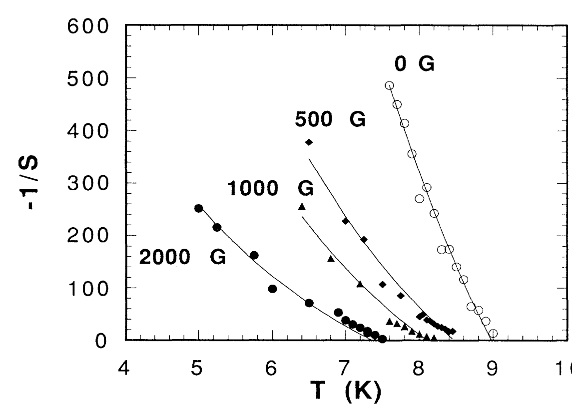

FIG. 4. Inverse logarithmic decay rate vs temperature. The solid lines are results from fitting by $-1/S = U_0(T)/kT - \ln(t_b/\tau_0)$ where $U_0 \propto (1-t)^m$. The logarithm had little effect on the quality of the fit and was fixed at 30 ($\tau_0 \approx 10^{-10}$). The best fit was obtained for $m = 1.1 \pm 0.1$.

plying $d \approx 1.8$) suggested we should see $H \propto (1-t)^{1.9}$. This curve is also shown in Fig. 1, with the linear coefficient adjusted to obtain the best fit. It fits worse than the $\frac{3}{2}$ power law. When we reversed the analysis and found the best-fit power law for the IL, the resulting exponent (1.2) led to the nonsensical result $d=-2$. This is not to say that flux creep cannot describe the magnetization decay *below* the IL, but it does suggest that the IL itself is not due to depinning. (Coexistence of flux creep and melting in some two-dimensional systems has also been suggested by resistance measurements on ultrathin Nb_3Ge films [29].)

A recent study suggestive of a vortex glass state in high-T_c materials involves a scaling analysis of the I-V characteristics [4]. Van der Beek [30] used the I-V scaling forms to interpret magnetization decay data for BSCCO. His scaling exponents were consistent with the resistive measurements and theoretical predictions. We undertook a similar analysis for our Nb sample, the details of which will be reported elsewhere, but found no evidence for a vortex glass transition.

In conclusion, we have identified the irreversibility line in Nb with the vortex-lattice melting line. The data are inconsistent with the depinning theory of Yeshuran and Malozemoff [5], but agree well with the melting model of Houghton, Pelcovits, and Sudbø [9]. The resulting value of the Lindemann criterion is lower than has been reported in high-T_c materials, but this is consistent with both the predictions of Brandt [10] and recent Monte Carlo simulations of Ryu *et al.* [24].

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