Irreversibility Temperatures of Nb_3Sn and Nb-Ti

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For fine multifilamentary Nb_3Sn and Nb-Ti wires, it was found that there exists a surprisingly large temperature region (e.g., ~1 and 0.5 K at 2 T for a Nb_3Sn and Nb-Ti wire, respectively) below the mean-field critical-magnetic-field line $H_{c2}(T)$ where the magnetization is reversible during a cycle of warming and cooling. The magnetic-field dependence of the lower boundary of this temperature region, i.e., the irreversibility temperature $T_r(H)$, was shown to follow theoretical predictions for flux-lattice melting based on an analysis of thermally induced flux-line fluctuations calculated with nonlocal lattice elasticity and the Lindemann melting criterion.

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Soon after the discovery of superconductivity in a La-Ba-Cu oxide, Müller, Takashige, and Bednorz[1] showed that in this material there exists a large temperature region below the mean-field critical-magnetic-field line $H_{c2}(T)$ where the motion of magnetic flux lines is reversible. The lower temperature boundary of the region is called the irreversibility temperature $T_r(H)$. A similar observation was also made for single-crystal $YBa_2Cu_3O_7$ by Yeshurun and Malozemoff.[2] These measurements prompted a number of experimental as well as theoretical studies to understand the nature of reversible flux-line motion in the new oxide superconductors.[3] Currently, the debate focuses on whether the irreversibility line $T_r(H)$ in the T-H plane represents a phase transition of the flux-line system (e.g., melting of the crystalline flux lattice[4-6] or of a disordered, glasslike flux lattice[7-9]) or a temperature region where the classical Kim-Anderson flux creep[10] is enhanced due to high operating temperatures.[11] Although some of the more recent work tends to support the latter possibility in the high-T_c oxides,[12] the question is far from completely answered.[3]

Regardless of the mechanisms for the observed easy flux-line movement in these oxides, it seems to be generally agreed that the main cause for the phenomenon is the very weak electronic coupling between the superconducting (CuO_2) layers (a large anisotropy in electronic properties), in conjunction with other related properties such as the short superconducting coherence length ξ and the large magnetic-field penetration depth λ (e.g., $\xi \cong 1.5$ nm and $\lambda \cong 150$ nm in the a-b plane of $YBa_2Cu_3O_7$).[3,13] Also, it is generally believed that the temperature region of reversible flux motion in metallic superconductors would be too small to be easily observed and of any significance,[14] except for some specially prepared materials such as InO,[14] amorphous Mo-Ge,[15] and Nb-Ge.[16] On the contrary, we report here the observation of surprisingly large temperature regions of reversible magnetization in state-of-the-art Nb_3Sn and Nb-Ti multifilamentary wires. The measurements were made with a dc magnetometry technique as the temperature was cycled under zero-field and field-cooled conditions. In contrast to the results for the high-T_c oxides,[12] these results agree surprisingly well with the predictions of an expression for the melting temperature $T_M(H)$ of the flux-line lattice derived by Houghton, Pelcovits, and Sudbø[5] using a nonlocal elasticity theory of the flux lattice. The details of the experimental observations are discussed below.

For the present measurement of the irreversibility temperatures, commercial multifilamentary Nb_3Sn and Nb-Ti wires were selected. The Nb_3Sn wire consists of ~7400 Ti-alloyed (~2 at.% of the Nb replaced) Nb_3Sn filaments (3-4 μm in diameter) embedded in a Cu-Sn alloy matrix.[17] For Nb-Ti, several commercial wires were used. The sizes of the filaments in a Cu matrix ranged from ~2.8 to ~24 μm, and the values of J_c at 50 kG varied among these wires from ~700 to ~3000 A/cm^2. In addition, in order to study the effect of variations in the value of the Ginzburg-Landau constant κ on the irreversibility temperatures, a fine multifilamentary (~3 μm) cold-drawn Nb wire was also studied.

In order to measure the magnetization, the wires were cut into segments of length ~6 mm and several lengths were placed horizontally (perpendicular to applied fields) in a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, Inc.). The measurement sequence for $T_r(H)$ consisted of cooling to 4.5 K in zero field, applying a magnetic field, and then warming well above $T_c(H)$ and cooling to 4.5 K with the same applied field. The incremental temperature steps for warming and cooling were 0.2 and 0.1 K (for Nb_3Sn and Nb-Ti, respectively) well below $T_r(H)$ and 0.1 and 0.05 K near and above $T_r(H)$. Also, the measurement of temperature at the specimen location by suitable placement of a thermometer indicated that there was no overshoot in temperature during the cycle. The scan length of the specimen for the measurement of the magnetic moments was 10 mm to minimize the spatial variation of the magnetic field in which the specimen had to travel during the measurement. At this scan length the variation is estimated to be 10^{-3}%. Examples of the results of such a measurement near $T_r(H)$ and $T_c(H)$ are shown in Fig. 1, indicating the criteria which were used for determining the irreversibility temperature $T_r(H)$

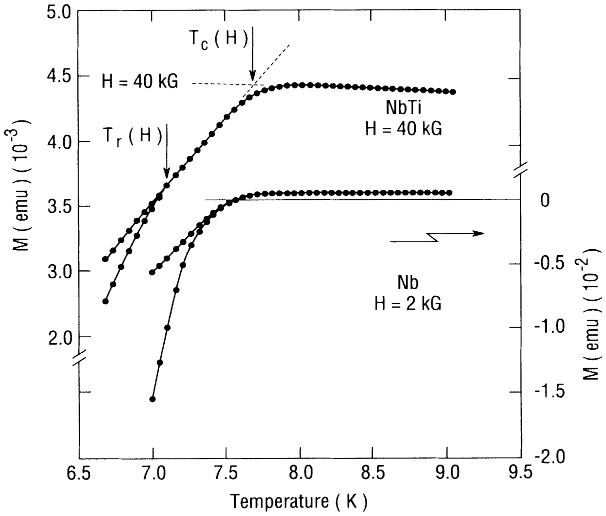

FIG. 1. An example of the variation in the magnetic moments of Nb-Ti ($\sim$2.8 μm) and cold-drawn Nb ($\sim$3 μm) wires as temperature is cycled, cooling in zero magnetic field and heating in an applied field as noted. Also, the criteria for the irreversibility and critical temperatures $T_r(H)$ and $T_c(H)$ [$H_{c2}(T)$] are shown.

and the critical temperature $T_c(H)$ [i.e., the mean-field critical magnetic field $H_{c2}(T)$]. We define $T_r(H)$ as the temperature at which an observable deviation is noted in the magnetic moments after the temperature cycle. It was found that by this criterion the difference in the moments for the warming and the cooling cycle was consistently $\sim$0.1% of the values at $T_r(H)$. For $H_{c2}(T)$, as also shown in Fig. 1, a standard linear extrapolation method was used.

In order to ensure that the observed reversibility is not due to a lack of the sensitivity in determining $T_r(H)$, we plotted the difference in the moments $\Delta M(T)$ between the warming and the cooling cycle as a function of the temperature. The value of $\Delta M(T)$ decreases very rapidly, rather than asymptotically, as $T_r(H)$ is approached and becomes zero well below the critical temperature $T_c(H)$. This indicates that there is no question about the existence of the temperature region where $\Delta M(T) = 0$.

The results of the measurements for $T_r(H)$ and $T_c(H)$ as a function of applied magnetic field up to 50 kG are summarized in Figs. 2 and 3 for a Nb-Ti and a Nb_3Sn wire, respectively. These figures illustrate the surprisingly large temperature ranges of reversible flux motion in these wires. In fact, they are as large as $\sim\frac{1}{3}$ of that for $YBa_2Cu_3O_7$ (Ref. 12) in the same range of reduced temperature t [$=T_r(H)/T_c(0)$].

The reversibility temperatures can also be measured by cycling the magnetic field at constant temperature and by determining the value of H at which the hysteresis width $\Delta M(H)$ vanishes. We have done this for a

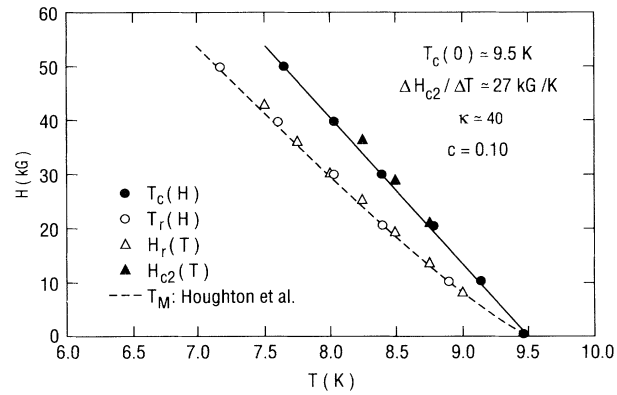

FIG. 2. The irreversibility temperature $T_r(H)$ and the critical temperature $T_c(H)$ [or the mean-field critical-field line $H_{c2}(T)$] as a function of temperature for Nb-Ti ($\sim$24 μm). The solid line is the melting temperature $T_M(H)$ calculated with Eqs. (1) and (2) using the parameters indicated. The open triangles are the irreversibility fields $H_r(T)$ as determined from hysteresis measurements at constant temperature.

Nb-Ti wire and the results are also included in Fig. 2 as the irreversibility field $H_r(T)$. Ideally, the $H_r(T)$ and $T_r(H)$ data should coincide, and this is very nearly the case for the Nb-Ti wire. However, similar measurements on Nb_3Sn showed a difference between $T_r(H)$ and $H_r(T)$, as shown in Fig. 3; this is very puzzling and requires further study to resolve the discrepancy.

In order to study further the nature of flux motion in the specimen, the flux-creep rate at 1 and 2 T was measured for times up to $\sim 3.5\times10^4$ s for Nb_3Sn, following the procedure described in Ref. 18, for $T \gtrsim 2.0$ K. The decay of the hysteretic moment $\Delta M(H)$ follows the well-known $\ln t$ dependence, except for the initial $\sim 3.5\times10^2$ s. The results of these measurements show that the flux-line "mobility," $[d(\Delta M)/d(\ln t)]\Delta M$, as determined

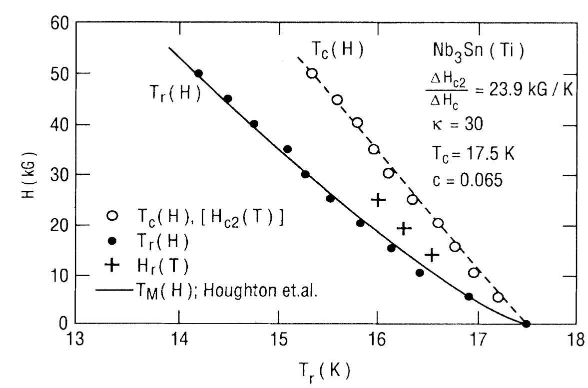

FIG. 3. $T_r(H)$ and $T_c(H)$ [$H_{c2}(T)$] for Nb_3Sn ($\sim$3.5 μm) and the melting temperature $T_M(H)$ from Eqs. (1) and (2). The crosses are the irreversibility fields $H_r(T)$ as determined from hysteresis measurements at constant temperature.

by the creep rate increases very rapidly near the irreversibility temperature, confirming the easy motion of the flux lines in this temperature range.

As shown above, there exists surprisingly large temperature ranges in which flux-line motion is reversible in Nb_3Sn and Nb-Ti. The question is whether the onset of reversible motion is due to melting of a crystalline flux lattice or of a glass-to-liquid phase transition, or simply is due to the enhanced rate of thermally activated flux motion. It has been believed that the flux-line lattice in the low-T_c superconductors should melt near $H_{c2}(T)$, so close to $H_{c2}(T)$ that it cannot easily be measured. We will, however, examine this possibility more carefully. In particular, we compare our results with a theoretical expression for the melting temperature $T_M(H)$ which was derived on the basis of the Lindemann "law" of melting of Houghton, Pelcovits, and Sudbø[5] in the nonlocal elasticity regime. Brandt[4] also derived similar results for melting, but the former authors provide an explicit expression for $T_M(H)$. Thus, we compare our data with their melting criterion:

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

Here, α indicates the degree of susceptibility to thermal agitation of the motion of flux lines and is given by

$$\alpha=2\times 10^5(H_{c2}^0/T_c^2)^{1/2}(M/M_z)^{1/2}\kappa^{-2}c^2 , \qquad (2)$$

where $t_M=T_M/T_c(0)$, $b=H/H_{c2}(T)$, $H_{c2}(t)=H_{c2}^0(1-t)$, $\kappa\cong\lambda/\xi$, and M/M_z is the electronic mass anisotropy (which equals 1 in the case of Nb_3Sn and Nb-Ti). Also, c is Lindemann's criterion for melting, defined by $\langle u^2(T)\rangle^{1/2}\cong ca_0$, where $\langle u^2(T)\rangle^{1/2}$ is the mean-square thermal displacement of a flux line and a_0 is the flux-line lattice spacing. c is ~ 0.1 for the melting of metals. In order to compare the calculated $T_M(H)$ with our results on $T_r(H)$ for Nb_3Sn and Nb-Ti, we used the following values: For Nb_3Sn, $T_c=17.5$ K, $\Delta H_{c2}/\Delta T=23.9$ kG/K, and $\kappa=30$,[19] and for Nb-Ti, $T_c=9.5$ K, $\Delta H_{c2}/\Delta T=27$ kG/K, and $\kappa=40$.[20] The values of $\Delta H_{c2}/\Delta T$ were determined from the present measurements and agreed well with the previously measured values.[19,20] Since the only unknown parameter in Eqs. (1) and (2) is Lindemann's constant c, we have varied the value of c to find the best fit with the data. As shown in Figs. 2 and 3, very good fits between the data and Eq. (1) were found with $c=0.1$ and 0.065 for Nb-Ti and Nb_3Sn, respectively. The fits are quite sensitive with respect to variations in c. For example, variations of ± 0.005 cause significant deviations in the fitting. Also, these values of c are reasonable, since values for the melting of atomic lattices are approximately 0.1. However, the implication of the difference in the values of c between Nb_3Sn and Nb-Ti is not clear.

These data are in surprisingly good agreement with the predicted melting temperature $T_M(H)$ even though the temperature ranges which were studied extend beyond the limit of applicability for Eq. (1). Also, because of strong pinning in these high-J_c materials the correlation length for positional order used in the flux-line lattice is expected to be quite small, and the applicability of Eq. (1) in such cases is questionable. Thus, we further examine the plausibility of Eq. (1) by considering $T_r(H)$ for a fine-filamentary pure Nb wire. Since the reversibility is strongly dependent on α, and hence on the value of κ, which is an order of magnitude smaller for Nb ($\kappa\sim 3$–5) than Nb-Ti or Nb_3Sn, it is expected that the temperature region between $T_r(H)$ and $T_c(H)$ will be significantly smaller in the case of Nb. As expected, the measurements of $T_r(H)$ for these wires, shown in Fig. 1, indicated that the reversible region, i.e., $T_c(H)-T_r(H)$, was quite small (≤ 0.2 K) in the same reduced-temperature and field range in which the Nb-Ti wires were measured. Although the difference in temperature between $T_c(H)$ and $T_r(H)$ has become sufficiently small that a meaningful comparison of $T_r(H)$ with Eq. (1) was not possible, the result also appears to support $T_r(H)$ being the melting temperature $T_M(H)$ since the κ dependence of $T_r(H)$ qualitatively follows the prediction of Eq. (1).

In addition, we also examined the effect of variations in the values of J_c on $T_r(H)$. Houghton, Pelcovits, and Sudbø do not consider the effect of flux pinning on $T_M(H)$, treating it simply as a phase transition from a crystal to a liquid, and in this view it is expected that variations in J_c (i.e., the pinning strength) would have very little effect on $T_r(H)$. Thus, a comparison was made between the sizes of the temperature range of reversible flux motion for two Nb-Ti wires with values of J_c differing by a factor of 4. The range of reversibility was essentially the same in both cases. We also measured $T_r(H)$ for similar Nb-Ti wires with different filament sizes (~ 2.8 to ~ 24 μm) and, within 0.05 K, no difference was found in the width of the reversible temperature region. This result ensures that the observed reversible region is not due to field penetration within the penetration depth $\lambda(T)$ near $T_c(H)$. In addition, this result can also be used to argue against $T_r(H)$ being the depinning line, since, in the creep model, complete flux penetration in the larger filaments takes more time than in the smaller filaments; $T_r(H)$ is thus expected to be higher in the larger one than in the smaller one, in contrast to the above observation.

In addition to the depinning-line and the lattice-melting interpretations discussed above, there are other possibilities for the observed irreversibility lines, e.g., the glass-to-liquid phase transition[8] and the untangling of flux lines.[9] However, the temperature of the onset of reversible flux motion, which was predicted by the latter theories, strongly depends on the existence of large values of the electronic mass anisotropy, and thus it is likely that this concept is not applicable to Nb_3Sn and

Nb-Ti since $M/M_z = 1$ for these materials.

As discussed above, the observed irreversibility temperatures $T_r(H)$ in Nb-Ti and Nb_3Sn show very strong correlation with the lattice-melting temperatures predicted by Houghton, Pelcovits, and Sudbø.[5] However, this raises a number of questions. The first is why such a large reversible temperature region was not observed earlier. In the past, flux-lattice melting was possibly observed only in In/InO_x by mechanical spectroscopy,[14] and in amorphous Mo-Ge (Ref. 15) and Nb-Ge (Ref. 16) by resistivity measurements. However, it is also true that the resistive transitions of type-II superconductors broaden significantly under high magnetic fields, particularly for high-κ materials.[21] Although the observed transition widths for Nb_3Sn (Ref. 21) were less than what is observed here, and, as assumed in the past, it is likely that the inhomogeneity of the specimens contributed to the broadening, it is possible that a portion of the broadening of the resistive transitions can also be associated with melting. Particularly if one considers the difference in the level of sensitivity of the measurements, this may not be unreasonable.

Finally, if the irreversibility temperatures are indications of the melting of the flux-line lattice, perhaps the most puzzling result is the fact that in the case of the Nb_3Sn wire the irreversibility-magnetic-field line $H_r(T)$, which was determined by the hysteresis measurements at a constant temperature, is well above the $T_r(H)$ line (see Fig. 3). If the $T_r(H)$ line is truly a melting temperature, it is expected that hysteresis should not exist above that temperature, i.e., $T_r(H) = H_r(T)$ (as was found for the Nb-Ti wire). Although these two measurements, $H_r(T)$ and $T_r(H)$, are not the same with respect to the establishment of the magnetic-field profiles during measurements, it is difficult to explain the difference solely based on the lattice-melting concept, and this requires further study. However, it should be noted that even though $H_r(T)$ is closer to $H_{c2}(T)$ than is $T_r(H)$, the $H_r(T)$ line is still substantially below the H_{c2} line and this difference is much greater than what was previously expected.

In summary, results for the magnetically measured irreversibility lines $T_r(H)$ in conventional superconductors, particularly Nb-Ti, strongly suggest melting of the flux-line lattice, because of their general agreement with the predictions of the theory of melting by Houghton, Pelcovits, and Sudbø,[5] in contrast to the results for the oxides for which the $T_r(H)$ lines do not agree with melting theories and are interpreted as being depinning lines.[12] These studies suggest that the observation of the lattice melting is only possible by a dc magnetometry technique if the flux pinning is sufficiently strong such that the depinning lines are pushed above the melting line and become unobservable.

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