

Nucleation of superconductivity in thin type-I foils

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The expulsion of flux in five type-I materials in a slow, continuously decreasing, perpendicular magnetic field provides evidence for the possible existence of a barrier in the superconductive transition. The variation of the observed critical fields with temperature yields Ginzburg-Landau parameter determinations for the materials which suggests their behavior to be more strongly type I than generally considered.

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The magnetic cycling of a type-I superconductor is fundamentally hysteretic:^{1,2} the first-order transition permits superheating and supercooling states. For thin flat samples in a perpendicular field, the hysteresis is even more pronounced because of a demagnetization-generated, geometrical edge barrier,³ which inhibits the penetration of flux in increasing field. A topological hysteresis in the intermediate-state flux structures is also observed between crossing the phase line in increasing or decreasing field.⁴

It is commonly assumed that no similar barrier exists in decreasing field,^{5,6} and that the expulsion of flux is governed by the basic tenets of phase transitions. In the nucleation regime ($H_a > H_{c2}$), only seeds of the superconductive phase with size larger than a critical radius evolve; smaller seeds collapse.^{7,8} In the spinodal regime ($H_a \leq H_{c2}$), there is no free-energy barrier to nucleation of the superconducting phase, and arbitrarily small seeds may evolve. This description, however, fails to treat the general nucleation of the superconductive state during a continuous decrease of the applied field. Neither does it include the effects of short- or long-range interactions, nor effects associated with demagnetization or surface nucleation.

Recent experiments on a tin foil in a continuously decreasing applied field using a fast-pulse induction technique observed the first expulsion of magnetic flux to occur at H_{c3} ,⁹ which the authors then discounted as coincidental. We here report an examination of the superconductive transition of several type-I materials, listed in Table I, at several temperatures in a gradually decreasing magnetic field using fast-pulse techniques. The results generally confirm H_{c3} as the first flux expulsion field, and suggest the existence of a barrier to the expulsion of flux. The measured critical fields themselves, moreover, yield determinations of the Ginzburg-Landau parameter $\kappa(T_c)$ for the materials in agreement with those obtained from measurements on superconducting spheres, and a factor ~ 2 below those derived from the more accepted magnetization measurements on thin films or foils (which agree with BCS estimates).

TABLE I. Superconductive parameters of the study materials, from Ref. 10, except for rhenium (Ref. 11).

Parameter	Lead	Tantalum	Rhenium	Tin	Indium
ξ_0 (μm)	0.10	0.097	0.15	0.23	0.38
λ_L (μm)	0.035	0.032	0.068	0.034	0.025
T_c (K)	7.2	4.5	1.7	3.7	3.4
H_c (G)	803	829	205	305	282

The fast-pulse measurement technique has been described in detail elsewhere.^{9,12,13} The samples were cut from 98.8–99.999 % pure, annealed, pinhole-free metallic foils of 10–125 μm thicknesses (d). Each foil was placed within a rectangular copper pickup loop of 800 μm width, in contrast to Ref. 9 where the tin strip was electroplated on only one loop branch. The loop is transformer bridged to a charge-sensitive fast amplifier: a low-frequency cutoff (10 kHz) on the bandwidth prevents the recording of flux variations at the sweep rate of H_a . Generally, only fast flux changes within the loop are recorded: the nucleation of a flux bundle creates a discontinuity in the flux intersecting the sense loop; the variation is a δ function in time, and a response to the step variation is obtained as long as the change is shorter than the nanosecond rise time of the preamplifier. Imposition of a discriminator threshold above the noise level defines the minimum recordable amount of flux change, which we estimate at a few hundred ϕ_0 . Extrapolation of the measurements to zero threshold yields a noise-free determination of the characteristic transition fields.

The measurements were performed in a single-shot ³He refrigerator at temperatures between 0.33 and 4.2 K, with an overall measurement uncertainty of better than 0.5%. The magnetic field was applied perpendicularly to the sample by a coil external to the refrigerator, with a homogeneity of 1% over the sample area and relative precision of better than 2×10^{-4} . The activation of a gate is synchronized with the magnetic field step command, so that pulses originating on the pickup loop are recorded in the appropriate field bin. Due to the large inductance of the magnet coil, the signal is integrated in a linearly varying field; the sweep rate was varied from a minimum of 0.5 G/s to 250 G/s, although the results reported herein were systematically obtained with a rate of ≤ 3 G/s.

After zero-field cooling of the samples, measurements were performed by recording all pulses above the discriminator threshold during increase of H_a at a constant rate from zero field to well above the thermodynamic critical field $H_c(T)$ and subsequent return to zero. Data were recorded separately for each direction of the field sweep as a function of H_a . The data acquired during the field increase were used to assess the foil quality and measurement threshold level by monitoring the flux penetration profile, which in the absence of noise yields a zero signal until the first penetration field is reached.

A typical differential curve of the normal (N) \rightarrow superconducting (S) transition results is shown in Fig. 1. The abscissa is given in reduced applied field $h_a(T) = H_a/H_c(T)$. The

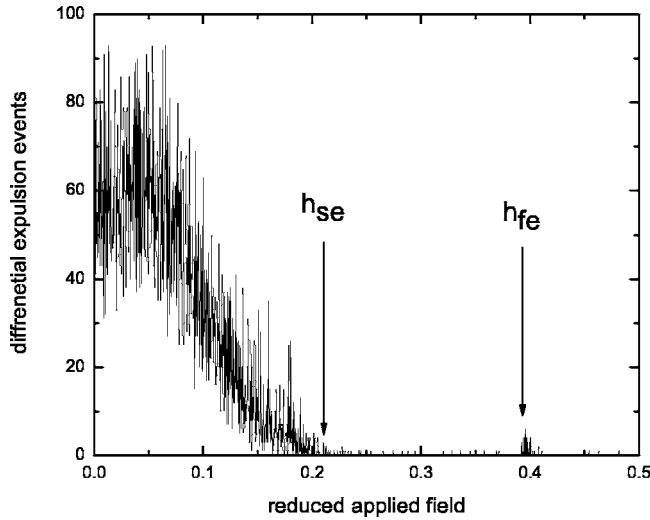


FIG. 1. Typical differential of the $N \rightarrow S$ transition signal obtained with a $50 \mu\text{m}$ tin foil following zero-field cooling to 0.350 K and ramping of the applied magnetic field to well above H_c .

event count at each h_a corresponds to a single-field-step decrease. The transition is characterized by three regimes demarcated by two characteristic fields. For the lowest thresholds, there is a characteristic first expulsion field H_{fe} indicated by a narrow signal, followed by an absence of events for further decrease of H_a . This field disappears with increasing threshold, suggesting it to consist of small flux expulsions. There is, in general, no signal above h_{fe} , except in cases where a direct correlation can be made with perimeter metallurgical defects.

The characteristic second expulsion field H_{se} is indicated by a rapid signal onset; this field persists with higher threshold measurements, although the number of events below h_{se} is severely reduced. The largest-amplitude pulses appear at the lowest applied fields.

Similar transition curves were obtained with all materials studied, for various aspect ratios and at different temperatures. Variation of the strip positioning relative to the pickup loop, including mounting the foil on a single branch of the loop, yielded no qualitative differences at the level of experimental uncertainty.

Within uncertainties, there is typically no significant variation of the characteristic fields with sample thickness, as shown in Fig. 2 for tin and rhenium. This identifies the two fields as intrinsic to the materials.

For type-I materials, there are in general only two intrinsic fields associated with the phase transition, H_{c2} and H_{c3} . Near T_c , H_{c2} can be written as¹

$$H_{c2}(t) = \frac{\phi_0}{2\pi\xi^2(t)}, \quad (1)$$

where ϕ_0 is the flux quantum, $\xi(t) = \xi_0(1-t)^{-1/2}$, and $t = T/T_c$ is the reduced temperature. In Fig. 3 we show the variation of h_{se} with $1/(\xi^2 H_c)$ for the different materials and temperatures, assuming $H_c(t) = H_c(0)(1-t^2)$. The lower line indicates the behavior anticipated from Eq. (1) with ξ_0 and T_c taken from Table I, and identifies H_{se} with H_{c2} .

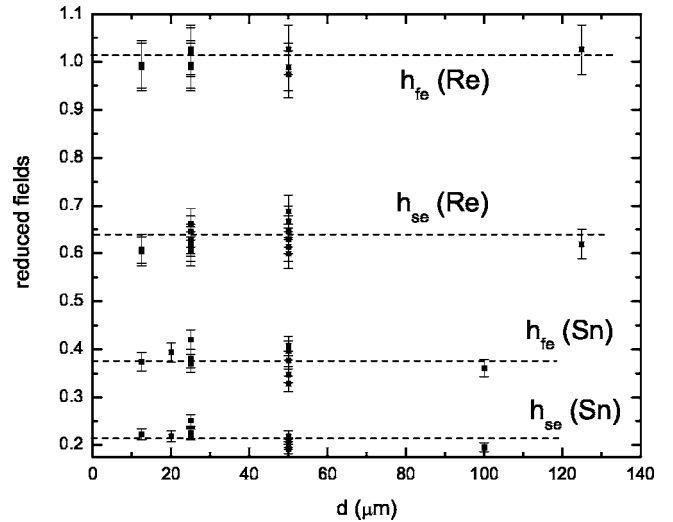


FIG. 2. Characteristic reduced $N \rightarrow S$ transition fields for different tin and rhenium sample thicknesses, at 350 mK .

If H_{se} corresponds to H_{c2} , then Fig. 2 suggests that $H_{fe} \sim 1.7H_{se}$ corresponds to H_{c3} . Figure 3 also displays h_{fe} for the various materials, with the associated line corresponding to a slope of $1.7(\phi_0/2\pi)$, providing strong support for this identification.

The first expulsion of flux at H_{c3} without a complete collapse of the normal state implies the spontaneous nucleation of superconductivity in a surface sheath of width $\sim \xi(T)$ over at least a part of the foil perimeter distant from the corners, corresponding to the creation of a narrow flux-free band near the foil edge. Although not associated with a barrier, such a band has been observed in magneto-optic studies of Pb, Sn, and In films^{14,15} in decreasing field. A similar band in increasing field is commonly associated with the geometric barrier,^{3,15} which separates the foil perimeter from the intermediate-state structure created by penetrated flux that is

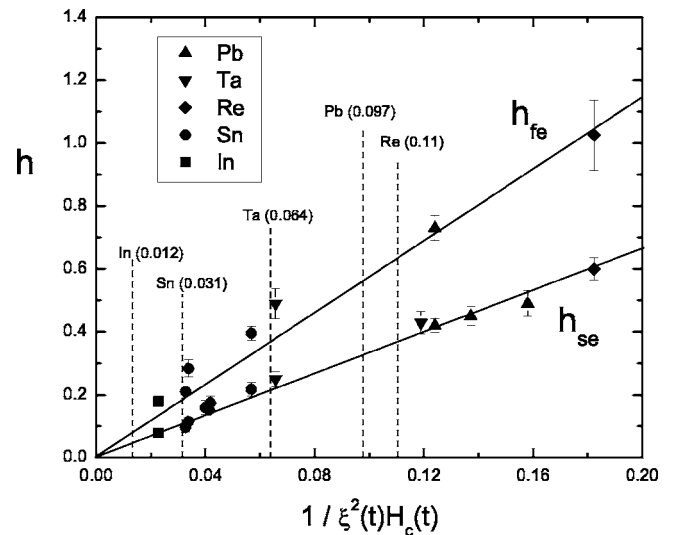


FIG. 3. $N \rightarrow S$ transition fields h_{se} and h_{fe} for different materials and temperatures; the dotted lines correspond to the abscissa at T_c , providing a lower limit to each material's response.

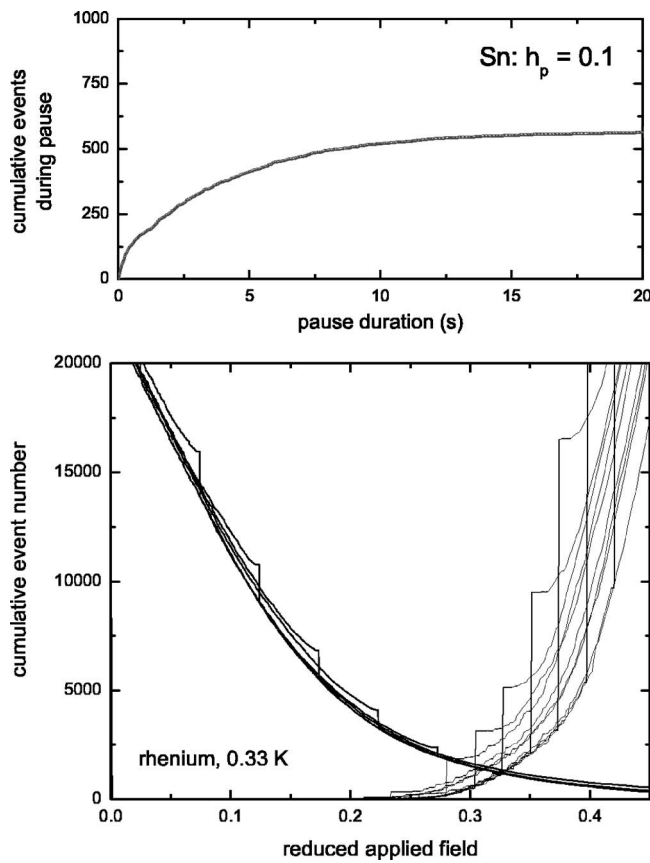


FIG. 4. Effect of a pause insertion at various fields in field increases and decreases, for 25 μm rhenium at 330 mK. The step results from flux changes occurring during the pause, which are recorded separately as shown in the top figure.

driven to the minimum of the barrier potential near the foil center. As indicated by Fig. 2, the band in decreasing field is not of geometric origin.

With the existence of a perimeter band, further nucleation of superconductive zones is technically impeded until the spinodal regime is reached and nucleation in the bulk of the foil becomes feasible. The fact that a signal below H_{c2} is observed at all is indication of a continuing barrier: flux expulsion occurring at the rate of the field decrease alone is not observable with this technique.

Additional indications of a barrier existence come from pauses inserted at various $h_a < h_{c2} = 0.62$ in the field ramp, shown in Fig. 4; for $h_a > h_{c2}$, no signal is recorded. The vertical lines are discontinuities between pause initiations

and ends, during which signal was recorded in separate files (the event count of the ramp resumption begins at the event number of the last pause event; the small plateau at the outset results from the response time of the electronics); as seen in Fig. 4, this is exponentially saturating in time, and results from the decay of eddy currents in the magnet and refrigerator.

A larger-amplitude, identical response is observed (Fig. 4) with pauses inserted in the penetration branch, which is generally interpreted as the relaxation of the system to an equilibrium state resulting from the lowering of the perimeter field by the penetrating flux, raising the geometrical barrier again.^{3,15} Once equilibrium is established, no further signal is recorded; further field increase is required to reinitiate the penetration of flux. In the $N \rightarrow S$ transition, the superconductive zones similarly continue to nucleate following cessation of the down ramp as a result of eddy currents, with expulsion of the displaced flux, until an equilibrium is established across the foil.

Observation of the critical fields permits an examination of the Ginzburg-Landau classifications of the materials via $\kappa(t) = (\sqrt{2})^{-1} h_{c2}(t)$ with $H_{c2} \rightarrow H_{se}$.² The results of this analysis for all materials are shown in Table II, in comparison with those previously extracted (where available) from previous measurements on thin films or foils¹⁶⁻¹⁸ and microspheres,^{19,20} and BCS estimates based on Table I parameters. In those cases with insufficient temperature measurements, the κ has been estimated from Fig. 3 based on the overall agreement of the results with Eq. (1). Since $\xi^2 H_c = \xi_0^2 H_c(0)(1+t)$, there exists a lowest abscissa for each material (shown as dotted vertical lines in Fig. 3) corresponding to T_c , which constitutes a lower limit on κ . In all cases, the derived κ are consistent with those from the microsphere measurements,^{19,20} but significantly below the tabulations and thin film and foil magnetization measurements.

The discrepancy is not related to field calibrations, as verified using a triaxial Hall magnetometer; moreover, the first penetration fields measured during field increase are in good agreement with geometric barrier predictions.²¹ The measured residual resistivity ratios of the samples varied between 60 and 450, consistent with impurities and lattice imperfections not playing a dominating role in the results. These in any case would tend to decrease the electron mean free path ($\langle l \rangle$), increasing all κ by an additional $\kappa_+ \sim \lambda_L(0)/\langle l \rangle$. The results might also be explained by an insufficient experimental sensitivity to small-amplitude pulses associated with smaller flux jumps at or below noise

TABLE II. Survey of the Ginzburg-Landau parameters for the various materials. The tabulated κ are obtained from the BCS $\kappa = 0.96\lambda_L(0)/\xi_0$ in the clean limit; the spheres from Ref. 20 and, the films and foils from Refs. 16 and 17. “-” denotes no existing measurement.

κ	Lead	Tantalum	Rhenium	Tin	Indium
Tabulated	0.43	0.32	0.44	0.16	0.17
Thin film or foil	0.34	0.36	-	0.15	0.13
Microsphere	0.25(8)	-	-	0.086(2)	0.066(3)
This work	0.23(3)	0.15(2)	0.26(3)	0.09(1)	0.036(4)

level, except for the fact that the fields represent zero-noise extrapolations.

The discrepancy in κ between spheres and thin film or foil determinations has been known for some decades, but to the best of our knowledge remains unexplained. Curiously, the thin film and foil results are in fact in better agreement with κ derived from h_{c3} via $\kappa(t)=(1.695\sqrt{2})^{-1}h_{c3}(t)$, and also agree in general with the lower-temperature results of both the spheres and this paper. The low-temperature measurements in rhenium indicate $H_{c2} < H_c < H_{c3}$, characteristic of type-1 $\frac{1}{2}$ materials ($\kappa \geq 0.42$), despite the determination of $\kappa_{Re} = 0.26 \pm 0.03$ for which $H_{c2} < H_{c3} < H_c$.^{1,2} This suggests a variation of the transition order with temperature, which has possibly important ramifications since κ is then less a fundamental property of the superconductor than a simple ratio between the two characteristic lengths in the description, both of which vary with temperature and yield results consistent with the observed κ determinations. Variation of the order of the transition with the temperature is predicted in recent renormalization-based reformulations of basic superconductive theory,²² which include fluctuations in the involved gauge and scalar fields, and result in a dividing line between type-I and type-II behavior at $\kappa = 0.8/\sqrt{2}$, with a magnetic response that can be varied between type I and type II simply by temperature change. This variation has been seen in nitrogen-doped Ta ($\kappa = 0.665$).¹⁸

The possible change in transition order with temperature

in turn has impact on current studies of quenched phase transitions in superconductive systems as a means of obtaining information on the formation of topological defects as seeds of large-scale structure in higher-energy cosmological transitions.^{23,24} The defect creation and distribution depend heavily on whether they arise from gauge or scalar field fluctuations,^{25,26} and on the order of the transition.^{27–29}

In summary, the nucleation of superconductivity in planar foils in decreasing field, while characterized by the customary critical fields of the phase transition, appears to be accompanied by a barrier to the expulsion of magnetic flux. The observed critical field variations with temperature further suggest a possible change in the transition order with temperature. Given the implications of these observations, further experiments to confirm or deny them are encouraged.

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