

Fluctuations of the Resistance and Resistance Levels in the Intermediate State of Superconductors*

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It is demonstrated that fluctuations of the resistance in the current-induced intermediate region of both hard and soft superconductors are caused by normal or superconducting domain motion. The amplitude and frequency of the resistance fluctuations have been studied as a function of supercooling, and the velocity of the domain boundary propagation estimated. Definite evidence for both stable and unstable resistance (i.e., energy) levels in the intermediate state is reported. The observations made on vanadium and tantalum wires have a bearing on certain aspects of the properties of superconducting solenoids, in particular, the relation of the critical current to the external magnetic field.

I. INTRODUCTION

RANDOM variations of the resistance in the intermediate state of hard superconductors have been previously reported^{1,2} near the superconducting critical current value. Instability of the intermediate state of some soft superconductors has been observed³ under certain circumstances and such noise was thought to be the major obstacle in attaining the predicted sensitivity of superconducting bolometers.^{2,4}

Using the niobium powder method, Schawlow⁵ has demonstrated that the size of the domains in the intermediate state was considerably larger in hard superconductors than in soft superconductors for the same thickness of material studied. It was, therefore, concluded that the interphase domain boundary energy in hard superconductors is much greater than in soft superconductors and is in excess of that expected on the basis of Landau's theory.⁶ Since such a configuration of domains in hard superconductors would therefore possess a very large free energy, instability is more likely to occur. Schawlow⁵ and deSorbo⁷ have found evidence of this instability, through their observation of abrupt spontaneous changes of the domain boundaries. Their experiments were performed with or without varying external magnetic fields.

The interphase boundary energy becomes a major part of the total free energy for soft superconductors of small dimensions. Consequently, they would be expected to exhibit instability of domain configuration. In fact, calculations of the critical sizes of soft superconductors for such an instability have been made by the author.⁴

It seems reasonable to suggest, therefore, that fluctuations of the resistance are a consequence of the

instability of the domain configuration when the interphase boundary energy is a predominant part of total free energy in the intermediate state of superconductors. It is the purpose of the present paper to present experimental evidence to support this suggestion. The description of the fluctuation pattern of different superconductors is presented and related to certain characteristic parameters of the intermediate state. The existence of resistance levels and instability regions is reported for the intermediate state and correlated to the fluctuation patterns observed.

II. EXPERIMENTAL METHOD

The four-probe method was used in our investigation of resistance fluctuations. Current was supplied from a battery through a series of fixed and variable resistors. All current joints were spot-welded to the sample and the potential leads were attached to the sample either with the current leads or at some distance removed from them. The fluctuation patterns observed were independent of the method of lead attachment. A six-dial Rubicon thermofree potentiometer with a galvanometer of 0.5- μ V sensitivity was used in the measurement of sample resistance. A Tektronix oscilloscope with a photographic camera was used to measure the initial rise time of the fluctuation pulses, and a Varian recorder of 10-mV span to record the amplitude of resistance fluctuations.

III. GENERAL FEATURES OF RESISTANCE FLUCTUATIONS

The shape of the pulse, its initial rise time, width, and amplitude differ greatly from one superconductor to another. Even for the same superconductor, these parameters vary with the dimension of the superconducting specimen and the degree of supercooling. Large hysteresis effects were observed in the investigated specimen; the insert in Fig. 2 shows a typical loop. It should be noted that the resistance appears at the critical current, i_c , increases with increasing current and decreases with decreasing current, disappearing at some value i_d which is $\ll i_c$.

* A part of the present work was reported at the Baltimore Meeting of the American Physical Society, March 1962 [Bull. Am. Phys. Soc. **7**, 175 (1962)].

¹ R. Weber, Phys. Rev. **72**, 1241 (1947); B. Kaplan and J. G. Daunt, *ibid.* **89**, 907 (1953); D. Baird, Can. J. Phys. **37**, 120 (1959).

² D. H. Andrews, R. M. Milton, and W. deSorbo, J. Opt. Soc. Am. **36**, 520 (1946).

³ E. R. Andrew, Proc. Roy. Soc. (London) **A82**, 194 (1948).

⁴ B. Lalevic, J. Appl. Phys. **31**, 1234 (1960).

⁵ A. Schawlow, Phys. Rev. **101**, 573 (1956).

⁶ L. Landau, J. Phys. (U.S.S.R.) **11**, 129 (1937).

⁷ W. deSorbo, Bull. Am. Phys. Soc. **5**, 290 (1960).

Despite the variety of the forms of resistance fluctuations, there seems to be a systematic trend in their behavior. For example, a shorter superconducting wire of a given diameter has more uniform amplitudes of resistance fluctuations, and the slope of the pulse is smoother. When a critical length is reached, characteristic for a given superconductor, the amplitudes become uniform and the pulse has a single slope. This occurred for 0.01-in.-diam vanadium wire less than 3 cm in length and for 0.002-in. tantalum wire less than 1.5 cm. However, for tin wires of 0.003-in. diam less than 1.5 cm in length, the slope and amplitude were nonuniform for all lengths of wire used. The variety of the resistance fluctuation patterns obtained for the same length of vanadium, tantalum, and tin is exhibited in Fig. 1.

We will now describe the properties and the pattern of resistance fluctuations for vanadium, tantalum, and tin.

A. Vanadium

Vanadium wire of 0.01-in. diam and 99.8% purity, supplied by Mackay Corporation, and 0.011-in. diam of 99.9% purity obtained from Bram Corporation, were investigated at 4.2°K.

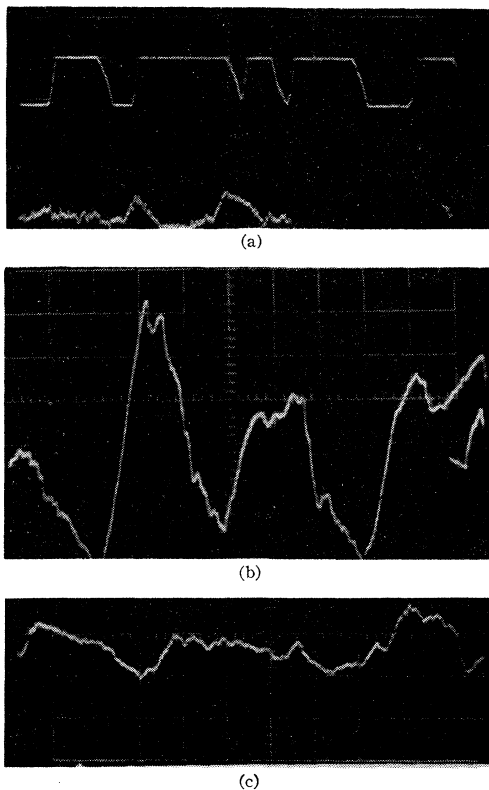


FIG. 1. The resistance fluctuation patterns in the intermediate state of (a) vanadium, (b) tantalum, and (c) tin. The vertical deflection is the voltage across the specimen and one big square of the horizontal deflection is equal to 0.05 sec for vanadium, 0.1 sec for tantalum, and 0.5 sec for tin.

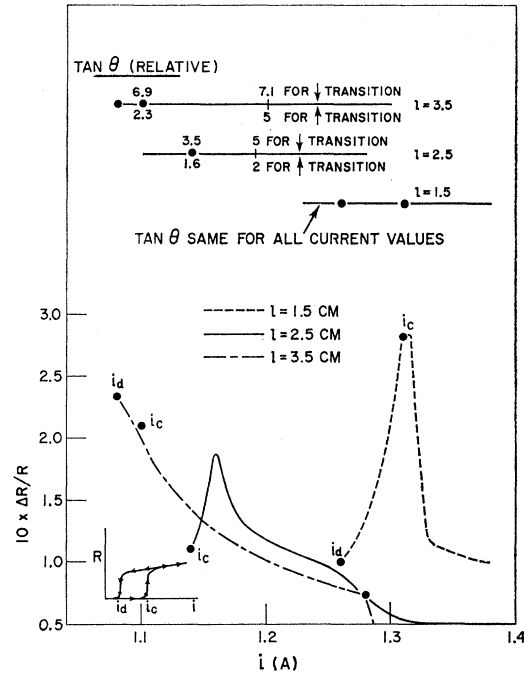


FIG. 2. Plot of the relative changes $\Delta R/R$ of the resistance fluctuations and the tangent for various lengths of 0.01-in.-diam vanadium wire.

The fluctuations are large in the vicinity of the critical current, and they persist for currents well above the critical value. The amplitude of fluctuations for longer vanadium wires increases in the hysteresis region with decreasing current. Above the critical current, the amplitudes decrease linearly with increasing current. For shorter wires, the amplitudes of fluctuations are larger at the critical current and decrease rapidly for smaller or larger values of current. Such a behavior is exhibited in Fig. 2, where there are plotted the relative changes of the resistance fluctuations, $\Delta R/R$, against current for various lengths of 0.01-in.-diam vanadium wires.

On the same figure the value of the tangent of the initial rise time is given. The tangent is determined from the slope of the oscilloscope trace during the initial rise of a fluctuation. It can be seen that, for longer wires, the relative value of the tangent of initial rise time decreases with decreasing current. For the same value of the current, the tangent of initial rise time is greater for the upper resistance transition. For the shorter wires, the difference in the relative value of the tangent of initial rise time for different currents becomes small, and at the length of 1.5 cm the tangent has the same value for all currents and for either type of transition. These observations will be discussed in Sec. V and will be used to derive the values for the velocity of propagation of a phase boundary.

The critical current value seems to increase with decreasing length of wire. The reproducibility of the

TABLE I. Characteristic parameters of the intermediate state of 0.01-in.-diam vanadium wire.

| L (cm) | i_c (A) | i_d (A) | φ | i_{dn} (A) | R/R_n |
|-------------|--------------|--------------|--------------|-----------------|---------|
| 3.5 | 1.10 | 1.08 | 0.33 | 1.28 | 0.934 |
| 2.5 | 1.14 | 1.13-1.10 | 0.0199-0.069 | 1.40 | 0.603 |
| 1.5 | 1.305 | 1.26-1.20 | 0.137-0.20 | 1.48 | 0.58 |

critical current, i_c , is better for longer samples and seems to be related to φ , the degree of supercooling. φ is usually defined as $1 - (H_d/H_c)^2$, where H_c is the critical magnetic field and H_d the value of magnetic field for which the specimen undergoes a superconducting transition as the magnetic field is decreased below the critical value, ($H_d \ll H_c$). In the case of a current induced transition, since $H_c = 2i_c/cr$ (Silsbee rule), the degree of supercooling can be therefore written as $\varphi = 1 - (i_d/i_c)^2$, where i_d is the value of the current at the end of hysteresis region (see insert in Fig. 2). In Table I, the values of the characteristic parameters for the intermediate state, i_c , i_d , and φ are listed. Also included in the table are current values above the critical value at which the resistance fluctuations disappear, i_{dn} , and also R/R_n for different lengths of 0.01-in. vanadium wire.

The two values for i_d , and consequently for φ , for the samples 2.5 and 1.5 cm in length indicate that the 0.01-in. vanadium wire may exhibit superconductivity for a value of current between these limits which are

a function of the rate of current change. The lower current limit is obtained by choosing the slowest rate of current change. A somewhat similar instability is encountered on increasing the current before reaching the critical current value. For certain values of current lower than the critical value, the resistance may suddenly increase, and fluctuations of large amplitude occur. After a few seconds, the resistance drops to a value approximately 10 times smaller and remains at this value until the critical current is reached. This state of affairs can be easily induced when a rapid increase of current is imposed.

As can be seen from Table I, the ratio of the resistance at the critical current to that in the normal region (R/R_n) decreases with decreasing length of wire. This decrease eventually saturates at the value 0.58.

B. Tantalum

Measurements on tantalum wire of 0.002- and 0.005-in. diameter of 99.98% purity, obtained from Bram and Fansteel Corporation, were made at 4.2°K.

Fluctuations of large amplitude occur deeper in the hysteresis, and the amplitude increases with decreasing current. This is to be compared with vanadium, where the amplitude of fluctuations was large at the critical current value. For longer wires, the amplitude of fluctuations is relatively small at the critical current; but, for shorter wires, the amplitudes are larger and the resistance fluctuations persist above the critical current, decreasing linearly with increasing current. The relative changes of the resistance fluctuations $\Delta R/R$ for different lengths of 0.002-in. tantalum wire are plotted against current in Fig. 3. The relative values for the tangent of initial rise time of a pulse for different current values are also given in Fig. 3. For the longer specimen, the value of the tangent decreases with decreasing current, and for shorter wires remains approximately at the same value for all current values.

Figure 3 indicates that there is a large hysteresis region for all lengths of wire, and the value of critical current increases with decreasing length of wire. Typical data for 0.002-in.-diam tantalum wire in the intermediate state are given in Table II.

It is observed that the absolute values of the parameters given in Table II may change from sample to sample, but the ratio of these parameters for corresponding lengths seems to be constant. Again, the range of values for i_d or φ indicates that tantalum wire may become superconducting for any value of the current between these limits and depends on the rate of current change. As before, the ratio R/R_n decreases with decreasing length of the wire.

C. Tin

Tin wires of 0.003- and 0.001-in. diam and 99.99% purity were obtained from Mackay Corporation.

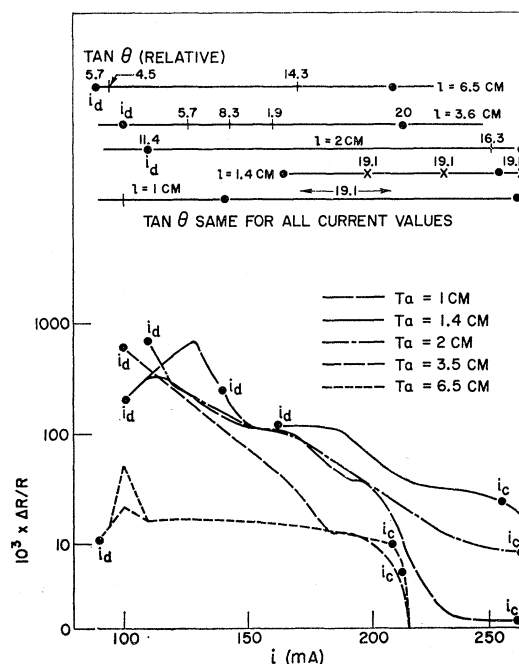


FIG. 3. Plot of the relative changes $\Delta R/R$ of the resistance fluctuations and the tangent of the initial rise time of a pulse for various lengths of 0.002-in.-tantalum wire.

No fluctuations were observed in tin at the transition temperature. However, they did occur at lower temperatures, at the lower end of the hysteresis region. This behavior is completely different to that previously described for vanadium and tantalum. The probability of occurrence of fluctuations increases with increasing degree of supercooling, the amplitudes and width of the pulses depending on the value of the specimen current. For large values, the width of the pulse is narrow and of smaller amplitude. For lower current values (i.e., at the end of the hysteresis region), the pulses are wider and of larger amplitudes. In contradistinction to the vanadium and tantalum wires, the pulses have irregular shapes even for very short wires. The superconducting transition often occurs through a series of fluctuations near the end of the hysteresis region.

The current intervals h for 0.003-in.-diam tin wires in the hysteresis region, where the fluctuations occur, are given in Table III, as a function of temperature.

The results indicate that the value of the current interval h , in the fluctuation region, seems to increase with increasing supercooling coefficient.

TABLE II. Characteristic parameters of the intermediate state of 0.002-in.-diam tantalum wire.

| L (cm) | i_c (mA) | i_d (mA) | φ | i_{dn} (mA) | R/R_n |
|-------------|---------------|---------------|-------------|------------------|---------|
| 6.5 | 208 | 90 | 0.845 | ... | 0.95 |
| 3.5 | 212 | 122-100 | 0.67-0.77 | ... | 0.82 |
| 2 | 262 | 120-100 | 0.79-0.824 | 288.0 | 0.8 |
| 1.4 | 252 | 162-160 | 0.587-0.556 | 265.0 | 0.8 |
| 1 | 258 | 137-102 | 0.576-0.85 | 275.0 | 0.75 |

Fluctuations were observed with 0.001-in. tin wires. However, it was difficult to obtain reproducible results with these specimens.

IV. NATURE OF RESISTANCE FLUCTUATIONS

In order to observe the behavior of a domain system during resistance fluctuations in the intermediate state, an insulated bismuth wire of 0.006-in. diam and 3-cm length was placed alongside the 0.01-in.-diam vanadium wire of 3.5-cm length. The change in the resistance in bismuth wire is approximately proportional to the square of the magnetic field, and the resistance of a length of bismuth wire should be proportional to the average of the square of the field over the length of the wire.⁸ Any change in the current distribution along the wire brought about by changes of domain configuration during the resistance fluctuations should be reflected in the change of a bismuth-wire resistance. The four-probe method was used as before on both the bismuth and vanadium wires. The potential leads were fed into

⁸ D. Shoenberg, *Superconductivity* (Cambridge University Press, New York, 1952).

TABLE III. Characteristic parameters of the intermediate state of 0.003-in.-diam tin wire.

| T (°K) | i_c (mA) | φ | h (mA) |
|-------------|---------------|-----------|-------------|
| 3.645 | 145 | 0.77 | 20 |
| 3.61 | 210 | 0.801 | 68 |
| 3.59 | 240 | 0.82 | 70 |

a double-beam oscilloscope, and for the bismuth wire a preamplifier was found necessary. One can see from Fig. 4 that the potential changes across the bismuth wire closely follow the resistance fluctuations and correspond to a variation in magnetic field of approximately $\pm 6\%$. Since the total current through the vanadium and bismuth wires is constant, the changes of the magnetic field over the length of bismuth wire can be assumed to be caused by the changes in the current distribution in the vanadium wires. If the density of current across the cross section of vanadium wire were axially symmetric, there would not have been any change in magnetic field along the bismuth wire. However, if the domain configuration of vanadium wire changes during the resistance fluctuations, the distribution of the current across the cross section of vanadium wire would also change. If the flow of the current in the superconducting regions of vanadium wire is assumed to be confined to the surface, then the

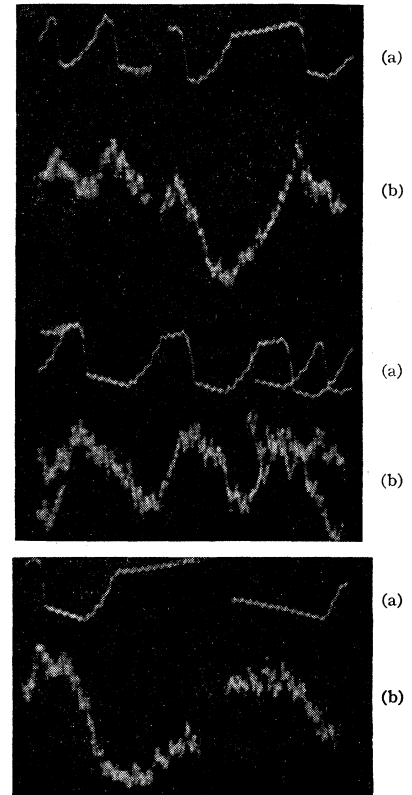


FIG. 4. (a) The resistance fluctuations in the intermediate state of a vanadium wire and (b) the corresponding potential changes across the bismuth wire.

The instability of the intermediate state of tin wire carrying current in a horizontal or transverse magnetic field has been discussed by Andrew,³ who also remarks on the tendency of the intermediate state in a hysteresis region to preserve its domain configuration during the change of current; i.e., the resistance remains the same for a considerable change of current. This corresponds,

in principle, to our statement of resistance levels. The resistance levels have been also observed in thin tin films⁹ and in thin indium wires.¹⁰

The velocity of propagation of a superconducting domain boundary in bulk tin in a horizontal magnetic field was measured by Faber.¹¹ He obtained 10 cm/sec for the velocity of the longitudinal domain propagation, as compared to the present value of 4 cm/sec. The difference may be accounted for by the different configuration of the magnetic field and of the domain distribution in the two experimental arrangements. Ittner¹² has reported measurements on the velocity of normal phase propagation in tantalum. The reported values for velocity are much larger than ours. However, the magnetic field geometry and the experimental conditions were quite different than in the present experiments.

Some of the present observations are pertinent to the work on hard superconductors, such as Mo-Re or Nb-Zr, used for the production of high magnetic fields.^{13,14} Here again the rate of current increase fixes the critical value of field necessary to destroy superconductivity, the value of critical field being much lower if the current is changed abruptly. This corresponds to our excitations of an unstable resistance level by a sudden change of current at a value lower than critical. The graphical study of current versus magnetic field¹³ for the Mo-Re alloys reveals a behavior which can be related to our observation of energy-resistance levels. That is, for the same value of the magnetic field, there is a range of current value necessary for destruction of superconductivity, and it is a function of rate of current change. These levels have been referred to as "unstable adaptation"¹³ and have qualitative similarity with the instability regions mentioned above. The very low resistance values^{13,14} observed in Mo-Re and Nb-Zr have an analogy in our very low and stable resistance level in vanadium and tantalum, owing probably to the splitting of superconducting domains into fine threads. The presence of fine superconducting threads is believed to be the consequence of the negative characteristic length^{8,15} Δ of the interphase boundary. The characteristic length Δ is defined either as¹⁶ $\Delta = (8\pi/H_c^2)\alpha_{ns}$ (α_{ns} is the energy per unit volume of boundary between superconducting and normal region) or as¹⁷ $\Delta \approx (\xi - \lambda)$, where ξ is the coherence length and λ penetration depth of a magnetic field.

VIII. DISCUSSION

A number of qualitative generalizations can be made from the present experiment with regard to noise in superconductors. However, further discussion of the phenomena requires an adequate dynamic theory of the intermediate state of a superconductor. The static London¹⁸ theory of current-induced intermediate state predicts a domain-structure configuration which has been experimentally proven. In this theory it is assumed that the geometrical boundaries of the materials are larger than the domain size. However, for a specimen with a dimension smaller than the domain size, as is presently the case, the predictions of London's theory have not been satisfied.¹⁸ We assume that our specimens fall into this category on the basis of the domain size observations^{5,7} on bulk material. Therefore, it is doubtful if the domain-structure configuration, as proposed by London, can be used as a basis for discussion of the presently observed resistance fluctuations.

Before briefly considering the dynamic models, we will discuss the analogy of the superconducting transition with the vapor-liquid transition, i.e., the supercooling of vapor below its transition temperature. One can imagine the equivalent supercooling effects in superconductors to occur because of the difficulty of growth of a nucleus of stable phase. The conditions for growth, equilibrium, or shrinkage of a superconducting nucleus⁸ are

$$\begin{aligned} (1 - H^2/H_c^2) &> \Delta/d + \partial\Delta/\partial n && \text{(growth)} \\ &= \Delta/d + \partial\Delta/\partial n && \text{(equilibrium)} \\ &< \Delta/d + \partial\Delta/\partial n && \text{(shrinkage),} \end{aligned} \quad (1)$$

where Δ is the characteristic length, n the demagnetization coefficient, and d the smallest dimension of the superconducting nucleus. We believe that these conditions are sufficiently general to be applicable to our case. The supercooling will become unstable when the right-hand side of (1) is smaller than the left, and when this inequality is satisfied the metal will undergo a transition to the superconducting state.

The probability of the growth of a superconducting nucleus through thermal fluctuations⁸ is given by

$$\omega \propto A \exp(-\Delta G/kT), \quad (2)$$

where ΔG is the Gibbs free energy excess of the nucleus. Expression (2) can then be rewritten in the form:

$$\omega \propto A \exp \left\{ - \left[\frac{4}{3} \frac{(H_c^2 - H^2) \Delta^3}{kT \varphi^3} \right] \right\}. \quad (3)$$

Our observations are in qualitative agreement with this expression as far as the dependence on the degree of supercooling is concerned. However, in order to obtain a sufficient probability for occurrence of fluctuations, the characteristic length Δ would have to be

¹⁸ F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1954), Vol. 1.

⁹ A. M. Kolchin, Soviet Phys.—JETP **13**, 1083 (1961).

¹⁰ W. Love, Phys. Rev. **85**, 715 (1952).

¹¹ T. E. Faber, Proc. Roy. Soc. (London) **214**, 392 (1952).

¹² W. B. Ittner, Phys. Rev. **111**, 1483 (1958).

¹³ M. A. R. LeBlanc, Phys. Rev. **124**, 1423 (1961).

¹⁴ T. G. Berlincourt, R. Hake, and D. Leslie, Phys. Rev. Letters **6**, 671 (1961).

¹⁵ B. B. Goodman, Phys. Rev. Letters **6**, 597 (1961).

¹⁶ L. Landau, J. Phys. U.S.S.R. **7**, 99 (1943).

¹⁷ A. Pippard, Proc. Cambridge Phil. Soc. **47**, 617 (1951).

less than 10^{-5} cm. This value is much smaller than the accepted value of 4×10^{-5} cm for tin at these temperatures, and smaller than the value of 1.7×10^{-5} cm which Andrew³ has deduced from measurement on tin wires of similar diameter. The assumption of a negative Δ (i.e., negative surface energy) would suffice to explain the creation of superconducting nucleus,¹¹ but not the sustained resistance fluctuations observed in the present measurements.

Although dynamic theories¹⁹⁻²¹ of the intermediate state are not, in general, in agreement with the experiment, they do provide, at present, the only direct physical model for a discussion of resistance fluctuations.

Beck¹⁹ was able to demonstrate the instability of domain boundaries, when the latter were subjected to a periodic perturbation. He has obtained the conditions for the instability assuming negative surface energy, an assumption certainly not acceptable in the case of tin. Dzialoshinskii²⁰ criticized Beck's assumption of negative surface energy and has obtained by a variation of the free energy under a periodic perturbation the following expression for the displacement of domain boundaries:

$$\langle (\delta Z)_{av} \rangle^2 = (4kT/\Delta H_c^2) \ln(\Delta/\lambda). \quad (4)$$

For tin, this expression gives the value $\langle \delta Z \rangle_{av} \sim 10^{-7}$ cm, which corresponds to a very small change in current and is much too small to explain the amplitude of observed fluctuations. The value of Δ is not known for vanadium and tantalum, and, consequently, one cannot calculate the numerical value of $\langle \delta Z \rangle_{av}$.

In Gorter's²¹ dynamic theory of intermediate state, the following assumptions have been made:

¹⁹ F. Beck, Phys. Rev. **98**, 852 (1955).

²⁰ I. E. Dzialoshinskii, Soviet Phys.—JETP **3**, 980 (1956).

²¹ C. J. Gorter, Physica **23**, 45 (1957); C. J. Gorter and M. L. Potters, *ibid.* **24**, 169 (1958).

(a) Domain boundaries are parallel to the electric field.

(b) Domain boundaries move in a direction perpendicular to the current and local magnetic field.

(c) The voltage drop observed in the direction of current is produced by nonhomogeneous distribution of currents. This leads to a Lorentz force. Due to the boundary motion, it would be expected that an induced voltage would be produced. This induced voltage would be responsible for a voltage drop along the wire.

With these assumptions, Gorter has been able to deduce the London expression for the change of resistance in the intermediate state. When applied to the case of supercooling, a harmonic perturbation will be generated and Gorter has calculated the voltage variation in the intermediate state of tin. The maximum amplitude of voltage variation calculated is about $5 \mu\text{V}$, and the velocity of sideways boundary propagation $V = 200$ cm/sec. Although Gorter's theory is in disagreement with observations on domain configuration in bulk material,⁵ a value of $5 \mu\text{V}$ for the voltage fluctuations is obtained. Although this predicted value is close to ours (0.2 mV), it is still out by factor of 40. There is also a very large discrepancy between his predicted value of 200 cm/sec for the velocity of domain boundary propagation and the present observed value of 4 cm/sec.

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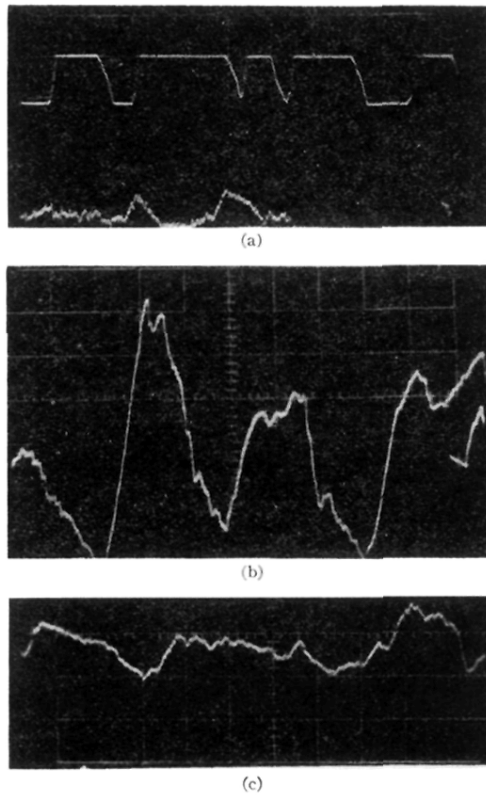


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