between the $\pm\frac{3}{2}$ levels (quadrupole states labeling) which occurs at very small effective γ. In the spin-$\frac{3}{2}$ case, the $|\Delta m|=3$ transition (the 0γ transition of Fig. 1) has the weak intensity.

We have been unable to detect any resonance at the 3γ position for a spin-$\frac{5}{2}$ nucleus experiencing a large quadrupole interaction (e.g., I^{127}) in a powder specimen.

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†Summer Faculty Participant; permanent address: Kalamazoo College, Kalamazoo, Michigan.

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QUANTUM TRANSITIONS AND LOSS IN MULTIPLY CONNECTED SUPERCONDUCTORS

A. H. Silver and J. E. Zimmerman

Scientific Laboratory, Ford Motor Company, Dearborn, Michigan

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This Letter reports on the observation of quantized flux flow across superconducting weak links.[1] This is accomplished by incorporating the weak link into a macroscopic superconducting ring and making use of the quantum behavior of this multiply connected superconductor. The quantized flux flow corresponds to direct transitions between the allowed states of the ring. Within this simple model we offer a criterion for the appearance of a fundamental resistive effect associated with the passage of flux across superconducting elements; this criterion was experimentally verified. Thus we have performed an elementary experiment on the dynamics of the quantized fluxoid which is pertinent to the recent discussions on voltages and resistance in superconductors.[2-5]

The experiment consists of measuring the current and voltage across a superconducting weak link or point contact in an otherwise macroscopic superconducting ring as shown in Fig. 1. The ring was made of niobium, the point contacts were adjustable 000-120 niobium screws,[6] and all experiments were performed at 4.2°K. With the superconducting ring open at point (b), the maximum supercurrent, i_c, of the contact at (a) is measured. Following this the ring is closed at (b) and the voltage across the link is measured as a function of the source current, I, and of maximum supercurrent, i_c.

The multiply connected superconducting states of the ring can be characterized by the quantum condition on the phase integral around the ring,[7] using standard notation,

$$\oint \vec{p}\cdot d\vec{l}=kh, \tag{1}$$

where $\vec{p}=2m\vec{v}+2e\vec{A}$ is the canonical momentum of the coherent electron pairs and k is an integer. Since the bulk portion of the ring is thick compared to the London penetration length, λ, we can always find a path such that $\int m\vec{v}\cdot d\vec{l}$ vanishes in the bulk superconductor. However, the thickness of the link is comparable to or

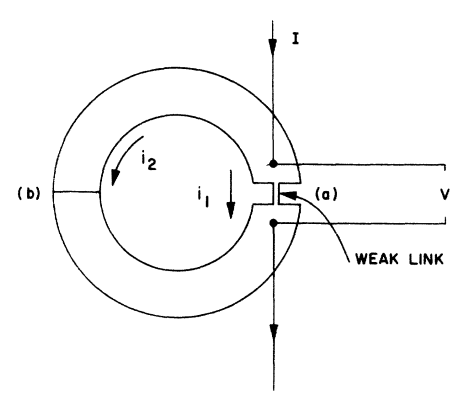

FIG. 1. Schematic of the macroscopic superconducting ring with the weak link. I is the total external current, i_1 the current in the weak link, and i_2 the current in the ring.

less than the penetration length, and therefore $\int m\vec{v}\cdot d\vec{l}$ will not vanish when evaluated through the weak link. We can then write,

$$\int_{\substack{\text{weak}\\ \text{link}}} \frac{m}{e}\vec{v}\cdot d\vec{l} + \Phi = k\Phi_0, \tag{2}$$

where Φ is the total enclosed magnetic flux and Φ_0 is the flux quantum $h/2e$. The precise relation of the current density j to the velocity, v, depends on the detailed nature of the weak link; e.g., for a Josephson tunneling junction[8] $j = j_0 \sin[(1/\hbar)\int(2m\vec{v})\cdot d\vec{l}]$. For simplicity let us assume $j = nev$, where n, the number of superconducting electrons, is a constant independent of v for all values of $|j| < i_c/\sigma$ and σ is the cross-sectional area of the link. If the current in the weak link is $i_1 = j\sigma$ and the link has length t, we can rewrite Eq. (2) as

$$(\mu_0\lambda^2 t/\sigma)i_1 + \Phi = k\Phi_0, \tag{3}$$

where $\mu_0\lambda^2 = m/ne^2$. The flux in the ring is just Li_2 if we neglect the inductance of the weak link. Therefore we can write the load-line equation

$$\Phi = L(I - i_1), \tag{4}$$

which connects the internal parameters of the ring, i_1 and Φ, with the externally controlled current I. From Eqs. (3) and (4) we can construct Fig. 2 relating Φ and i_1 to the external parameter I. The parameter $\mu_0\lambda^2 t/\sigma L$ is assumed small compared to unity and the straight line describing each state k terminates abruptly at $|i_1| = i_c$. The maximum supercurrent i_c is an independent parameter which has been taken for illustration as $\frac{1}{4}$, $\frac{1}{2}$, and 2 units of Φ_0/L in Figs. 2(a), 2(b), and 2(c), respectively. The magnetic Gibbs free energy, G, is

$$-\tfrac{1}{2}Li_c^2 + L\int_0^I i_1 dI$$

and is plotted in units of $\Phi_0^2/2L$.

The data are concerned with transitions $\Delta k = \pm 1$ between adjacent states. These occur at $|i_1| = i_c$ and for a constant source current I. Consider Fig. 2(c) as an example of the general case $i_c > \Phi_0/2L$. The arrows show the variation of the flux, current, and free energy for a cycle of I between $\pm i_c$. Two irreversible transitions, each $\Delta\Phi = \pm\Phi_0$, occur during each half-cycle, and each transition produces a voltage pulse across the weak link such that

$$\int V dt = \pm\Phi_0; \quad i_c \geq \Phi_0/2L. \tag{5}$$

If the current is varied over the interval $(-i_c + \Phi_0/2L) \leq I \leq (i_c + \Phi_0/2L)$, only one transition will occur per half-cycle. An extension of this argument leads to a voltage whose amplitude is periodic in I with the period Φ_0/L. The hysteresis pattern of Fig. 2(c) further shows that these irreversible transitions represent a loss or resistive effect. Let us compare this behavior with the special case $i_c = \Phi_0/2L$ in Fig. 2(b). The transitions at $|i_1| = i_c$ produce the same voltage pulse as given by Eq. (5). However, because of the particular condition $i_c = \Phi_0/2L$, there is no hysteresis as I is cycled. The periodicity in the voltage is the same as in Fig. 2(c) but we should expect no loss effects. In the case $i_c < \Phi_0/2L$ for which Fig. 2(a) is an example, the transitions are between the quantized states k and the state $\Phi = LI$; hence, the voltage pulses are

$$\int V dt = \pm Li_c; \quad i_c < \Phi_0/2L, \tag{6}$$

and we expect smaller voltages, but still periodic in I and with no hysteresis and no loss.

The elementary predictions of this model can be summarized as follows. If the current

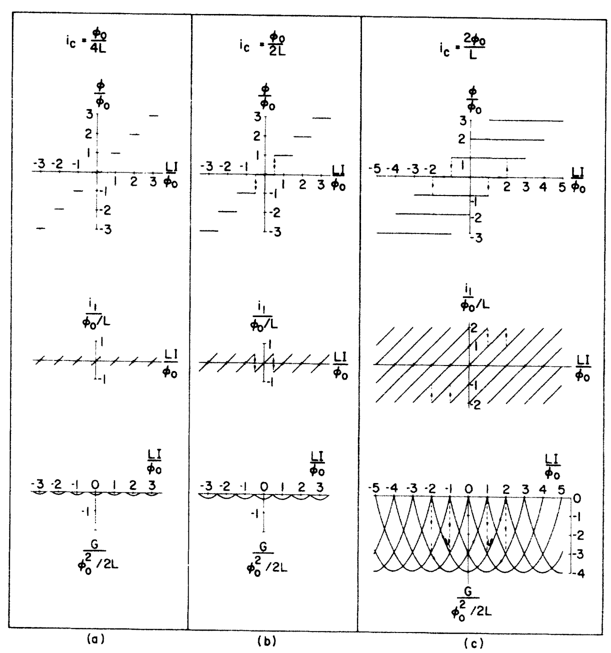

FIG. 2. Graphs of flux Φ, link current i_1, and magnetic Gibbs free energy G for the weak rings as a function of applied current I. A linear relation is assumed for the current density and the maximum supercurrent i_c is assumed to be $\frac{1}{4}$, $\frac{1}{2}$, and 2 times Φ_0/L in (a), (b), and (c), respectively.

is given by $I = I_0 + i_c \cos\omega t$, there will be a voltage at the frequency ω which is amplitude modulated as a function of I_0 with the period Φ_0/L. For $i_c \le \Phi_0/2L$ the amplitude of this voltage varies linearly with i_c, and the voltage and current are 90° out of phase so that no loss will occur. For $i_c > \Phi_0/2L$ there will be an in-phase component of the voltage and consequently resistance. Since the width of a hysteresis loop is $2[i_c - \Phi_0/2L]$ and the height is Φ_0, the energy lost in traversing a single hysteresis loop between adjacent states is $2i_c\Phi_0 - \Phi_0^2/L$, and this is identical with the total irreversible change in the free energy. Hence the loss is linear in i_c for $i_c > \Phi_0/2L$.

Figure 3 is typical of the direct experimental data obtained with a current source $I = I_0 + i_0 \cos\omega t$, where $\omega/2\pi = 30$ Mc/sec, and a critical current of approximately Φ_0/L. The figure is a set of oscilloscope traces of the rectified 30-Mc/sec voltage across the weak link taken in succession as the current amplitude i_0 is increased in steps of approximately $\Phi_0/4L$. The oscilloscope is swept in time and I_0 is slowly varied over a range slightly greater than $4\Phi_0/L$, hence there are four periods in each sweep of I_0. The ordinate measures the full rectified voltage with the lowest line representing zero voltage for $i_0 = 0$. As i_0 is increased, the average voltage level increases because there are more transitions $\Delta\Phi = \pm\Phi_0$ in each

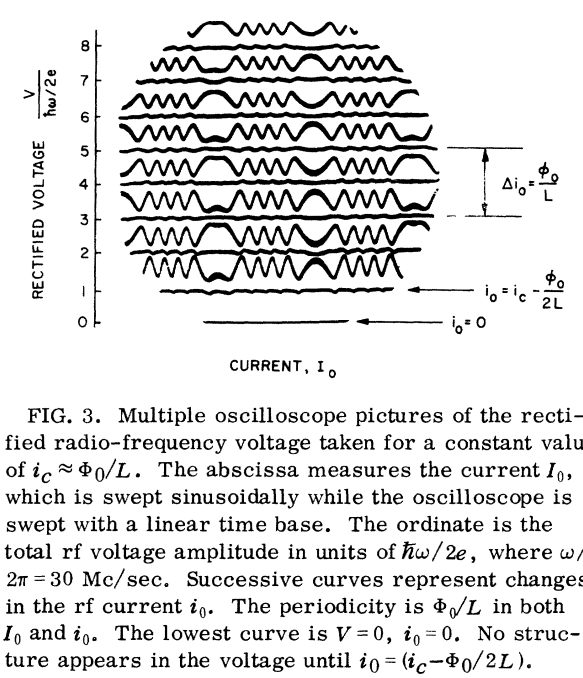

FIG. 3. Multiple oscilloscope pictures of the rectified radio-frequency voltage taken for a constant value of $i_c \approx \Phi_0/L$. The abscissa measures the current I_0, which is swept sinusoidally while the oscilloscope is swept with a linear time base. The ordinate is the total rf voltage amplitude in units of $\hbar\omega/2e$, where $\omega/2\pi = 30$ Mc/sec. Successive curves represent changes in the rf current i_0. The periodicity is Φ_0/L in both I_0 and i_0. The lowest curve is $V = 0$, $i_0 = 0$. No structure appears in the voltage until $i_0 = (i_c - \Phi_0/2L)$.

cycle of the 30-Mc/sec current. However, the amplitude modulation, which is the periodic signal plotted as a function of I_0, decreases slowly with an alternation of sign. This is a result of the generation of harmonics of 30 Mc/sec and can be shown to be consistent with the simple model we have presented.

We have determined that for $i_c < \Phi_0/2L$, the voltage signals as shown in Fig. 3 are 90° out of phase with the source current $i_0 \cos\omega t$ and that the amplitude of the voltage is approximately proportional to i_c. For $i_c > \Phi_0/2L$ we observe an in-phase component demonstrating resistive effects in agreement with the model presented here. Loss occurs when there is a discontinuous and irreversible change in the free energy. Since the magnetic free-energy difference between the singly and multiply connected rings is zero when these transitions occur, we apparently have a mechanism for interaction between the normal and superconducting states and there can be energy flow. The energy loss at each transition is given by the irreversible change in G at the transition.

If the relation between j and v is not linear,[9,10] or if $\mu_0\lambda^2 t/\sigma L$ is not small compared to unity, then the critical current for which loss occurs will be different from $\Phi_0/2L$. For a Josephson junction irreversibility[6] occurs for $i_c = \Phi_0/2\pi L$. The linear model adopted here and the Josephson relation probably represent the limiting forms for j, and hence the critical value of i_c will be between these two extremes.

These results show that the voltages associated with the motion and trapping of fluxoids are neither always lossy nor always conservative. If one considers the loss to be associated with the superconducting weak link, then the appearance of this loss depends on the external superconducting load line, namely the ratio Li_c/Φ_0. In relation to the general problem of voltages and resistive effects in type-II superconductors one could consider a weakly connected ring as a grossly inhomogeneous superconductor. On the other hand, consideration of loss in single superconducting elements would be equivalent to the limit $L \to \infty$ in our experiment.

The experiments reported here indicate the existence of discrete states for the multiply connected superconductor characterized by Eq. (1). The selection rule $\Delta k = \pm 1$ is rigorously obeyed at least for i_c of the order of Φ_0/L. The critical condition for the appearance of resistance in the superconducting ring is $i_c \geq \Phi_0/2L$. This resistance is of a fundamental nature related to the trapping of flux and a change of state of the superconductor.

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COEXISTENCE OF ANTIFERROMAGNETISM AND SUPERCONDUCTIVITY*

D. K. Finnemore, D. C. Hopkins,† and P. E. Palmer

Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa
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Matthias and co-workers[1-4] have reported a close relationship between the occurrence of superconductivity and dilute ferromagnetism. For the particular case of La doped with the magnetic impurity Gd, the curve for the superconducting transition temperature, T_c, appears to intersect the curve for magnetic ordering temperature, T_O, at approximately 0.5°K and 1% Gd. As shown in Fig. 1, their results for T_O differ slightly depending on the method of measurement. The solid line was determined from the peak of the differential magnetization curves,[3] and the dashed line was determined from the occurrence of a remanent magnetization.[1] On the basis of the existence of a remanence, the ordering was identified as ferromagnetic. The purpose of this note is to present magnetization data for LaGd alloys which indicate that the ordering in low applied fields is not ferromagnetic but is a helical or other antiferromagnetic structure.

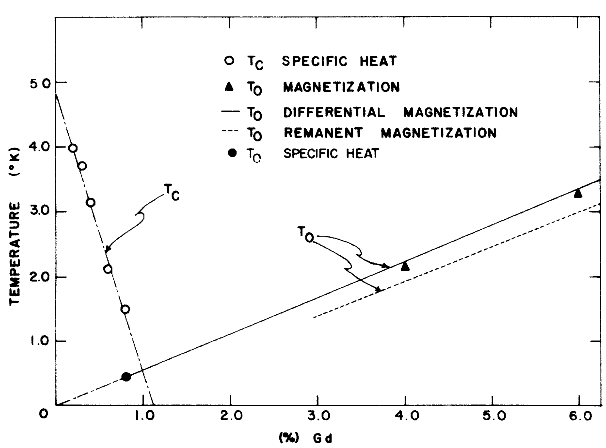

FIG. 1. Superconducting and antiferromagnetic transition temperatures for LaGd alloys.

The present measurements are made by pulling a spherical sample throught a uniform magnetic field between two counter-wound 16 400-turn coils. The induced signal is detected with a ballistic galvonometer. The system is calibrated by measuring a superconducting sphere in the same geometry and by assuming its susceptibility to be $-1/4\pi$. LaGd samples with the dhcp structure are cut from alloys prepared in the same manner as the specific-heat samples described earlier.[5] Any strain introduced in the cutting process trends to maintain or enhance the desired phase. As a final step in preparation, a layer approximately 0.005 in. thick is electropolished from the surface.

Figure 2 shows the constant-field magnetization curves for La-4.0 at.% Gd in fields up to 50 Oe. The curves clearly break from the Curie-Weiss behavior and go through a maximum near 2°K. This peak is rather broad, as might be expected for a solid-solution alloy, but its general character is similar to the paramagnetic-to-helical transitions in erbium[6] and terbium.[7] The tendency to antiferromagnetism persists up to fields of the order of 1000 Oe, but the maximum occurs below our measuring range. A La-6.0 at.% Gd sample (insert in