

cients. For a plasma with a Maxwell distribution the radiation temperature should be a monotonically decreasing function with increasing magnetic field,⁷ and should be representative of black-body emission from the plasma. The fact that the plot in Fig. 2(d) exhibits pronounced maxima at cyclotron harmonics is evidence that the electron distribution is not Maxwellian.

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OBSERVATION OF QUANTUM PERIODICITY IN THE TRANSITION TEMPERATURE OF A SUPERCONDUCTING CYLINDER*

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Deaver and Fairbank¹ and Doll and Nabäuer² have shown experimentally that the flux which is trapped in a superconducting cylinder is an integral multiple of the unit $hc/2e$. It has been pointed out^{3,4} that this result follows because the free energy of the superconducting state is periodic in this unit of the flux if the electrons are paired in the manner described by the Bardeen-Cooper-Schrieffer (BCS) theory.⁵ The free energy of the normal state, on the other hand, is virtually independent of the flux. Consequently, the transition temperature T_c , which is the temperature at which the free energy of the normal and superconducting states are equal, must also be a periodic function of the enclosed flux ϕ . The magnitude of the change in T_c was calculated for a thin cylindrical sample using the BCS model in which the possible pairing of particles with net momentum was included. This calculation showed that the binding energy of each pair was reduced by the amount of energy required to provide the center-of-mass motion necessary to maintain the fluxoid,

$$\frac{1}{\hbar} \oint \left(m \vec{v}_s + \frac{2e}{c} \vec{A} \right) \cdot d\vec{s},$$

an integer. Each integer n corresponds to a different superconducting state characterized by a particular pairing arrangement and a different transition temperature. The transition temperature is found to vary as

$$\Delta T_c = \frac{\hbar^2}{16 m^* R_0^2} \left(\frac{2e}{\hbar c} \phi + n \right)^2.$$

The choice of n which gives the tightest binding and the highest transition temperature switches from 0 to -1, -1 to -2, etc., when ϕ is given by $\frac{1}{2}(hc/2e)$; $\frac{3}{2}(hc/2e)$, etc. We note also that the binding energy of the pair is a minimum at these points and varies periodically with the flux. At the transition temperature the penetration depth becomes infinite and consequently the flux ϕ , enclosed by the cylinder, is determined entirely by the external field. T_c is given by a periodic array of parabolas, each of which is centered on a flux unit (see Fig. 1). One can estimate the expected magnitude of ΔT_c by taking $m^* = m_e$ and a reasonable diameter of say 1 micron for the cylinder. ΔT_c is then approximately 5×10^{-5} K° which is of measurable magnitude in the liquid helium temperature range.

We have observed such an effect with a thin

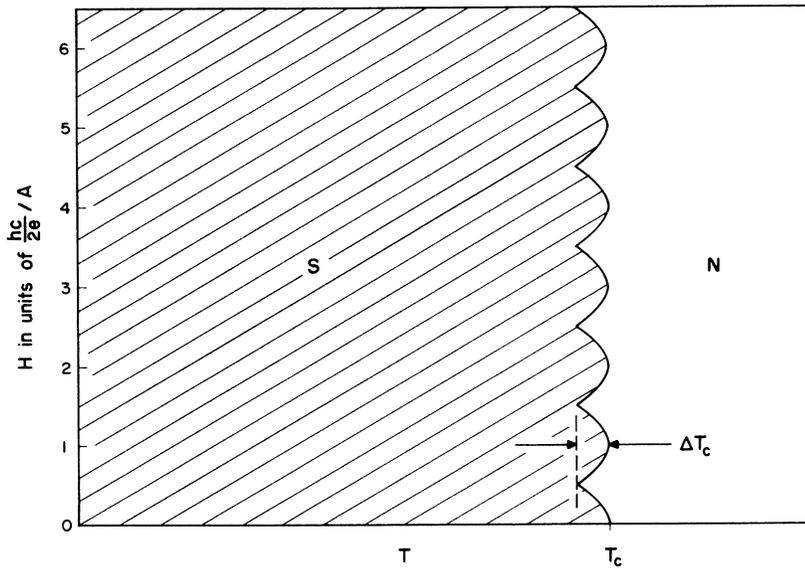


FIG. 1. Phase diagram for a thin cylindrical superconductor in an axial magnetic field. The scalloped edge of the superconducting phase results from the periodicity in the free energy of the bound pairs, in the magnetic flux through the cylinder.

cylinder of tin. A clearly defined series of parabolic variations of the transition temperature was observed as the magnetic field was changed. The parabolas were regularly spaced at intervals of $hc/2e$ in the magnetic flux, in agreement with the results of the experiments on quantized flux by Deaver and Fairbank and Doll and Nabäuer.

The cylinder of tin was prepared in the following way. A drop of G.E. 7031 cement was held on the ends of two wires and the wires were then rapidly drawn apart to arm's length. A thin filament of cement was formed which extended from one wire to the other. After some hours of practice we succeeded in drawing a filament of approximately 1 micron in diameter. A portion of this filament was laid carefully onto a glass slide over a slot 3 mm wide by 10 mm long which had been cut in the slide. The filament was cemented in place with a dilute solution of the same cement. The slide was then mounted in a high vacuum evaporator on a rotating "spit" so that the axis of rotation of the "spit" was along that of the filament. In this way it was possible to evaporate metal over the complete perimeter of the filament, the slot in the slide being used to expose the underside of the filament to the evaporating metal. Earlier experience had shown that tin did not adhere well to the filament; indeed, tin films less than 900 Å thick were found to be noncontinuous. For this reason, a thin layer of gold about 25 Å thick was first evaporated onto the filament. This was not electrically continuous but provided a surface to which the tin could adhere. A layer of tin 375 Å thick was then evaporated at a pres-

sure of 3×10^{-6} mm onto the gold substrate. This formed a continuous amorphous film which was electrically conducting.

Electrical contact was made to each end of the cylinder by cementing thin copper wires onto the glass slide and then covering them and the surrounding tin film with a coat of silver paint. The slide was mounted on a bakelite base inside a copper tube which was split lengthwise. The whole assembly was fitted inside a copper solenoid in a glass Dewar system. The system was precooled and then filled with liquid helium. The temperature of the helium bath could be controlled to about 10^{-4} K° with a diaphragm manostat in the pumping line. The resistance of the tin cylinder was measured by passing a constant current of about $10 \mu\text{A}$ through it and measuring the potential drop across it with a microvoltmeter. The transition from the normal to the superconducting state occurred at about 3.45°K and extended over a temperature range of about 0.05K°. Temperatures in this region were measured with a carbon resistor thermometer placed within one centimeter of the filament and calibrated against the vapor pressure of helium.

If the transition region of a superconductor is spread over a finite temperature interval, there is no unique transition temperature T_c ; however, one may be defined by some criterion such as the temperature at which the resistance falls to one-half of its normal value. Alternatively, if the variation in the resistance is measured, one can calculate the change in the transition temperature ΔT_c from the slope of the resistance versus tem-

perature curve. This is the technique we used. Small changes in the resistance of the tin film were observed as the axial magnetic field was varied and these were interpreted later as changes in the transition temperature.

The results we obtained are shown in the accompanying figures. To obtain these photographs, the magnetic field at the sample was varied sinusoidally at 25 cps and the potential across the filament observed on an oscilloscope while a constant current of $25 \mu\text{A}$ was flowing through it. Similar results were obtained from dc measurements with the microvoltmeter. Figure 2 is the variation of the resistance with magnetic field and clearly shows a series of parabolas superimposed upon a quadratic background. The upper trace is a measure of the magnetic field, with zero field at the center of the picture. In Fig. 3 an enlarged

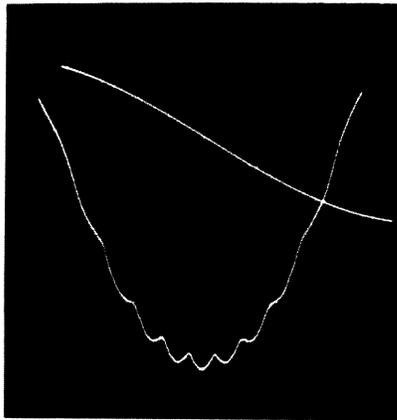


FIG. 2. Lower trace: variation of resistance of tin cylinder at its superconducting transition temperature as a function of magnetic field. Upper trace: magnetic field sweep.

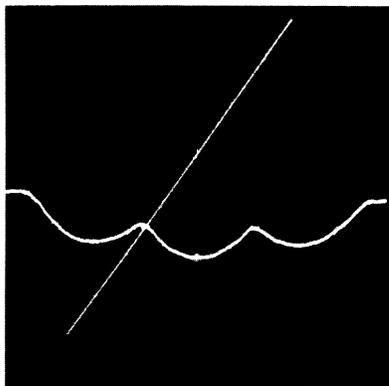


FIG. 3. Enlarged view of parabolic variation of the resistance of tin cylinder for pairs in quantum states -1 , 0 , and $+1$. Straight line is magnetic field variation with zero field at the center of the picture.

view of the parabolas, corresponding to pairing of the particles in the states $n = -1$, 0 , and $+1$, is shown. The minima occur at values of the magnetic field of -14 , 0 , and $+14$ gauss. The diameter of the filament had been measured previously with an optical microscope and found to be 1.4 ± 0.1 microns which gives a value of 13 ± 2 gauss for the field which would correspond to one flux unit $hc/2e$. The uncertainty in the diameter results from the difficulty in interpreting the diffraction pattern in the microscope. We could measure no change in this pattern of the filament over its entire length indicating that the filament was of uniform cross section. This suggests that one could usefully determine the diameter to greater precision with an electron microscope and hence determine the flux quantum to a few percent. In Fig. 4 the quadratic background has been reduced to a linear term by electrically differentiating the signal to the oscilloscope. In this way one can see more clearly the parabolas in the region where the quadratic term is large. From this picture one can see the strictly periodic nature of the minima over a region of seven parabolas. From our dc measurements we found that the maximum excursion of the transition temperature ΔT_c was 5×10^{-4} K° (after subtracting the quadratic background). This was somewhat greater than we expected and is probably attributed to the effective mass of the electrons being smaller than that of the true mass, and to some corrections from a more exact theory.

In summary we state the following results and conclusions from this experiment:

1. The quantum periodicity in the free energy of a superconducting pair predicted by Byers and

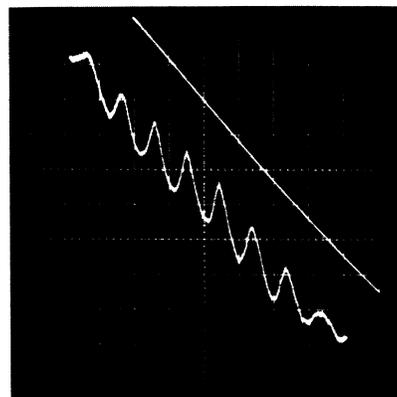


FIG. 4. Lower trace: variation of the resistance of tin cylinder with magnetic field after differentiation which reduces the quadratic background to a linear background. Upper trace: magnetic field sweep.

Yang⁴ has been directly observed and this was found to be the same order of magnitude predicted above.

2. Further evidence of electron pairs has been obtained in support of the BCS theory.

3. There was no indication (at least for the tin film measured) of uneven steps in the flux quanta of the type shown by Doll and Näbauer's results on lead.³

4. The quadratic background in the variation of the transition temperature gives evidence of the weakening of the binding of the pair in a magnetic field.

5. This type of experiment provides a means of determining to great precision the magnitude of the flux quanta by measuring the minima of the observed parabolas.

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couragement of B. S. Deaver and W. M. Fairbank in this experiment. We are grateful also to J. R. Schrieffer and W. T. Sommer for stimulating discussions.

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PARAMAGNETISM OF Fe IMPURITIES IN TRANSITION METALS*

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For some time it has been known that Fe or Co impurities in Pd have associated with them large magnetic moments.¹ Clogston *et al.*² deduce from the Curie-Weiss behavior of the magnetic susceptibility of a 1% Fe in Pd alloy a moment of $10 \mu_B$ (Bohr magnetons), while from magnetization measurements at low temperatures Bozorth *et al.*³ obtain a moment of 9-10 μ_B in $\text{Co}_{0.001}\text{Pd}_{0.999}$. The susceptibility measurements have been interpreted by the above authors and by Anderson,⁴ Wolff,⁵ and Clogston⁶ as implying the existence of localized magnetic moments associated with each Fe or Co impurity. We report here experiments utilizing the Mössbauer effect in very dilute solid solutions of Fe^{57} in Pd and in other transition series metals which demonstrate that the magnetic moment associated with the Fe impurities is local, in the sense that each Fe impurity acts in an applied external field at low temperatures like an isolated magnetic moment. The moment is found to be (in Pd) about $12.6 \mu_B$ and the spin about 13/2, so the gyromagnetic ratio is about two.

Our technique involves studying the hyperfine splitting of the Fe^{57} nuclei as functions of an external field H and of temperature. Since the internal field is known to be proportional to the

(time) average value of the z component of the electronic spin of Fe,⁷ measurements of the splitting permit determination of the electronic polarization of the Fe. We find in certain metals that the splitting vanishes in zero external field at all temperatures studied (1.5°K to 4°K), and increases as the external field is increased up to a saturation value, corresponding to complete polarization, at large fields and low temperatures. The splitting is found to be a function of H/T , and is representable by Brillouin functions⁸ appropriate to pure paramagnetism. The local magnetic moment, the associated angular momentum, and the saturation internal field are parameters of the Brillouin function, and our values given for these quantities represent the best fit of the experimental data.

The Pd sources were prepared by depositing ion exchange column purified, carrier-free Co^{57}Cl (HCl) onto high-purity Pd foil. The sources were reduced in hydrogen at 900°C and then diffusion annealed in vacuum for about 50 hours at 1200°C. Other sources mentioned below were prepared similarly. (Co^{57} decays by electron capture followed by gamma transitions to the 14-keV level of Fe^{57} .) The nuclear physics aspects of the 14-keV Fe^{57} gamma-ray

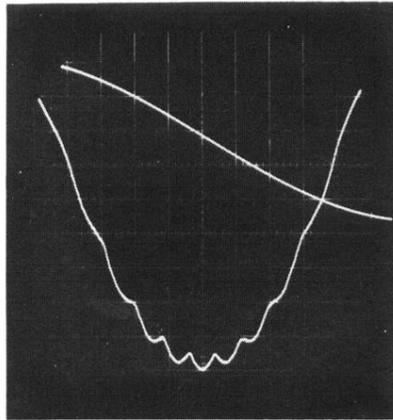


FIG. 2. Lower trace: variation of resistance of tin cylinder at its superconducting transition temperature as a function of magnetic field. Upper trace: magnetic field sweep.

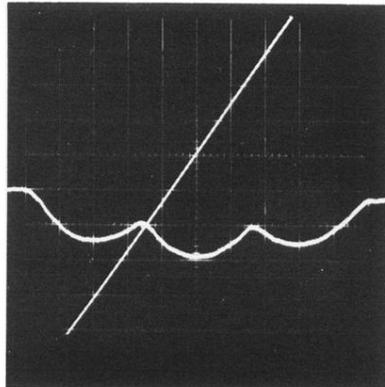


FIG. 3. Enlarged view of parabolic variation of the resistance of tin cylinder for pairs in quantum states -1 , 0 , and $+1$. Straight line is magnetic field variation with zero field at the center of the picture.

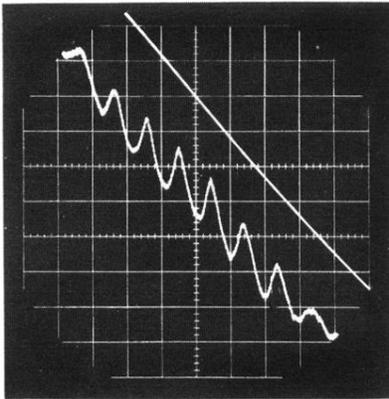


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