Fermi-Surface Topology Changes in In Alloys Deduced from the Behavior of T_c under Strain*

D. R. Overcash, [†] M. J. Skove, and E. P. Stillwell Department of Physics, Clemson University, Clemson, South Carolina 29631 (Received 13 January 1971)

We have measured the effect of dilute alloying on the strain (ϵ) dependence of the superconducting transition temperature T_c of In whiskers. Alloy contents x ranged up to 0.1-at. % Ag, 1.5-at. % Sn, and 3-at. % Cd. The addition of Ag and Sn did not affect $\partial T_c/\partial \epsilon$, but the addition of Cd had a large effect. The initial slope of the T_c -vs- ϵ curve, $(\partial T_c/\partial \epsilon)_{\epsilon=0}$, decreased with increasing Cd content until at 0.9 at. % it was a minimum of 15 mK/%, onefourth of its value for pure In. As x increased from 0.9- to 2-at. % Cd, $(\partial T_c/\partial \epsilon)_{\epsilon=0}$ increased to almost its value for pure In. The curves T_c vs ϵ were linear in ϵ for $0 \le x \le 0.3$ and for x > 2 at. %. For $0.3 < x \le 0.9$ -at. % Cd, $\partial^2 T_c/\partial \epsilon^2$ was negative, while for 0.9 < x < 2-at. % Cd it was positive. For alloys with more than about 2.0-at. % Cd, T_c was linear in ϵ . This behavior is attributed to a change in the topology of the Fermi surface occurring at about 0.9-at. % Cd in In.

INTRODUCTION

"Whiskers" of In have an elastic limit an order of magnitude greater than bulk In, making it practical to measure the behavior of the superconducting transition temperature T_c under elastic strain. In addition, we have grown alloy whiskers of uniform alloy content and studied the effect of tension on T_c as a function of alloy content x. The effect of pressure p on T_c of Cd-doped In has been studied by Makarov and Volynskii,¹ hereafter referred to as MV. Although in their study T_c was a linear function of p for a given x, $\partial T_c / \partial p$ was a nonlinear function of x. Nonlinear behavior of this type has been connected with Fermi-surface (FS) topology changes.²⁻⁴ MV concluded from their data that the third-zone electron toroid was breaking up. A similar effect has been observed in T1⁵ and in Re.⁶

Since hydrostatic pressure and uniaxial tension are special cases of a general stress, one expects similar results for the two cases. However, uniaxial tension changes the crystal symmetry, whereas pressure does not. In these experiments we observe not only the nonlinear curves of $(\partial T_c/\partial \epsilon)_{\epsilon=0}$ vs x similar to those observed in pressure experiments, but also nonlinear T_c -vs- ϵ curves at fixed x near the region of the topology change. Similar data have been reported for pure Sn whiskers.⁷ In this paper we report these data for In alloyed with Sn, Cd, or Ag.

EXPERIMENTAL PROCEDURE

The samples were grown as whiskers by the squeeze technique.⁸ Indium (Indium Corporation of America, 99.999% pure) together with varying amounts of Sn, Cd, or Ag was vacuum deposited onto a stainless-steel strip. Washers 2.5 cm in diameter were punched from the strip, compressed by a nut on a bolt, and turned down on a lathe to 2.0 cm. Whiskers with diameters of about 1 μ m and lengths of about 2 mm grew within a week. All samples had axes parallel to the $\langle 101 \rangle$ crystallographic direction (in the face-centered tetragonal system) as determined from rotating crystal xray photographs. The T_c 's of whiskers grown from pure In were within 2 mK of each other. All transition widths (10-90%) of the resistance) at the currents used in these measurements $(1-10 \ \mu A)$ were less than 3 mK. This would indicate that the alloy was uniform along the whisker. While the alloy content of each whisker was always constant along its length, it varied randomly from whisker to whisker on a given growth up to the average plating concentration of that growth.

Strain was applied to the sample by mounting the



FIG. 1. Residual resistance vs transition temperature for the In alloys used. Also plotted are the results of Merriam (Ref. 13) and of Makarov and Volynskii (Ref. 1). The correlation between bulk and whisker results allows the alloy content of the whiskers to be determined.

3

3

3765



FIG. 2. Recorder tracings of the resistive transitions of three whiskers of different alloy content. The vertical axis is the voltage across the whisker at constant current on an arbitrary linear scale, starting at zero for each whisker. Strain is a parameter in each series of transitions, the transition temperature increasing with increasing strain. Strain was increased by approximately 0.03% between successive transitions. The nonlinearity of the transition temperature with strain is apparent.

whisker across a piezoelectric bimorph.⁹ The calibration of these bimorphs is particularly difficult. The motion is not linear in voltage over large voltages, and, moreover, it is considerably different at room and at helium temperatures. The procedure adopted was to use the strain dependence of the T_c of pure-In whiskers as a standard. This behavior was previously measured using a Mylar puller or quartz puller.¹⁰ The results on these two accurate, but inconvenient, pullers were consistent to within $\pm 5\%$.

Mechanical and electrical contacts to the whiskers were made with silver paint (DuPont No. 7713). The sample puller was placed in a small Dewar¹¹ which was lowered directly into the liquid-helium storage Dewar. Liquid helium was then sucked into the small Dewar. The temperature was controlled by regulating the pressure of the helium gas over the small Dewar bath. The temperature could be controlled to ± 0.1 mK. Magnetic fields were not shielded. Stray magnetic fields were about 50 μ T (0.5 G).

A CryoCal germanium resistor was used to monitor the temperature of the bath. The resistor was calibrated against a He⁴ vapor-pressure thermometer (1958 standard temperature scale) and also against a germanium resistor calibrated by Cryo-Cal. The two calibrations were consistent within ± 1 mK in absolute value in the temperature range of this experiment, and relative changes in temperature were consistent within ± 0.1 mK.

Resistance measurements were made using four sample contacts. The sample current was supplied from a battery and resistance network and was kept constant to within one part in 10^4 . Typical current

densities were 1 A/mm². The potential across the potential contacts was measured with a Rubicon six-dial potentiometer with a Keithley No. 147 null detector. The midpoint of the resistive transition, extrapolated to zero sample current, was used as T_c . Whisker cross sections were calculated from the room-temperature resistance and length of the sample between potential contacts, assuming a value¹² of 8.9 $\mu\Omega$ cm for the resistivity of In. Since the cross section was not critical, no compensation was made for the resistivity change with alloying.

The sample mass (10^{-8} g) made it impossible to determine alloy content by chemical analysis. Alloy contents were indirectly inferred from the resistance ratio $\delta = R_{4,2}/(R_{300} - R_{4,2})$ and T_c . Figure 1 shows a plot of T_c vs δ for these whiskers. Also shown are the results of Merriam¹³ and of MV on bulk samples of known alloy content. The dependence of T_c on δ is the same for bulk samples and whiskers. Therefore, we assume that it is also true that the dependence of T_c (and δ) on alloy content is the same for whiskers as it is for bulk samples. This enables us to determine the alloy content from T_c , δ , and the work of Merriam and of MV. For our very small and pure samples, the resistance ratio must be corrected for increased resistance at 4.2 K due to surface scattering. This correction was determined from previous work on pure indium whiskers⁸ to be $\delta = \delta_m - 10^{-2}A^{-1/2}$, where A is the area of the whisker in μm^2 and δ_m is the measured ratio.

We do not know the effect of size on T_c . If one assumes that boundary scattering has the same effect as impurity scattering, the results of Marko-



FIG. 3. Shift in transition temperature T_c as a function of strain for Cd content up to 0.9 at. %. The transition temperature was taken to be the midpoint of the resistive transition. The two curves at 0.3 at. % represent the uncertainty in puller calibration and alloy content. One can see the change in initial slope and the departure from linearity of these curves as the Cd content is increased.

witz and Kadanoff¹⁴ then predict a depression of about 15 mK in T_c for a 1- μ m-diam sample (due to limiting of the electron mean free path by surface scattering). This correction should disappear rapidly as alloy scattering begins to predominate. Making such a correction would increase T_c for small alloy concentrations and would tend toward better agreement with MV and Merriam.

RESULTS

Figure 2 shows a recorder tracing in a typical experiment. As the sample is strained, T_c increases. Figures 3 and 4 show the results for several different alloy compositions in the In-Cd system. One can see that as the alloy content increases, the initial slope of the T_c -vs- ϵ curve decreases, and that the slope decreases at higher strains in alloys which have just less than 0.9-at. % Cd. At 0.9-at. % Cd, T_c is nearly independent of strain. As the fraction of Cd is increased above 0.9 at.%, the initial slope increases, and furthermore the slope now increases at higher strains. Finally, at about 2-at.% Cd, the value of the initial slope has nearly returned to the value obtained in pure In and the slope is again independent of strain. This is summarized in Fig. 5, which shows a plot of $(\partial T_c/\partial \epsilon)_{\epsilon=0}$ versus alloy content. The arrows indicate the sign of the curvature, arrows pointing down implying a decrease in slope with increased strain, and arrows pointing up an increase with increased strain. It is apparent that some change occurs in In at about 0.9-at.% Cd content.

When Sn was added to In, no changes in $\partial T_c/\partial \epsilon$ were observed up to 1.5-at.% Sn. When Ag was added, no change was observed up to the solubility level of Ag in In (<0.1%).



FIG. 4. Shift in transition temperature with strain for Cd contents greater than 0.9 at. %. The curve for 3.3-at. % Cd is linear, with nearly the same slope as the pure-In curve.



FIG. 5. Initial slope of the transition-temperaturevs-strain curve $(\partial T_c/\partial \epsilon)_{\epsilon=0}$ vs alloy content for whiskers. Also plotted is $\partial T_c/\partial p$ vs alloy content from the work of Makarov and Volynskii (Ref. 1). The minimum in both of these plots occurs at about 1-at.% Cd content. Our results have a somewhat sharper minimum, and the maximum relative change in $(\partial T_c/\partial \epsilon)_{\epsilon=0}$ is approximately three times larger than the change in $\partial T_c/\partial p$. The arrows attached to some of our points indicate that the slope changed with increasing strain, arrows pointing up representing samples whose slope increased, and arrows pointing down those that decreased.

CONCLUSION

We have observed a nonlinear variation of T_c with strain for In alloyed with Cd in the range 0 < x < 2at. % Cd. Such nonlinear behavior has been predicted theoretically when the FS topology changes. There is evidence of such a change in In as similar nonlinear behavior with x has been observed by Merriam¹³ on the change of T_c , by MV on the change of $\partial T_c/\partial P$, and by Ridley¹⁵ on the change in lattice constants. These nonlinear effects are consistent with a topology change in the third-zone toroids in the (001) plane. Higgins and Kaehn⁴ have calculated the effect of the addition of Cd on the FS of In. Their results agree with MV's proposal that the third-zone toroid first breaks into four cigar shaped and four small spherical pieces; second, at higher x, the four small spheres disappear. Each process leads to a nonlinearity. Our data evidently do not resolve these two processes, although a detailed analysis has not been done.

Ag has two less valence electrons than In and therefore should give about the same result as for Cd alloys at one-half the Ag concentration, assuming a rigid-band model. Unfortunately, the solubility of Ag in In at room temperature is quite small (0.1 at.%). At the highest Ag concentration we observed no change in $\partial T_c/\partial \epsilon$.

The addition of Sn up to x=1.5 at. % apparently produces no topology change. Sn has one more valence electron than In and so would tend to make the FS larger. This small increase evidently produces no topology change. *Research supported by the Air Force Office of Scientific Research under Grant No. 68-1548.

[†]Present address: Department of Natural Science, State College, Orangeburg, S. C. 29115.

¹V. I. Makarov and I. Y. Volynskii, Zh. Eksperim. i

Teor. Fiz. Pis'ma v Redaktsiyu <u>4</u>, 369 (1966) [Sov. Phys. JETP Letters 4, 249 (1966)].

²I. M. Lifshitz, Zh. Eksperim. i Teor. Fiz. <u>38</u>, 1569 (1960) [Sov. Phys. JETP <u>11</u>, 1130 (1960)].

³V. I. Makarov and V. G. Bar'yakhtor, Zh. Eksperim. i Teor. Fiz. <u>48</u>, 1717 (1965) [Sov. Phys. JETP <u>21</u>, 1151 (1965)].

⁴R. J. Higgins and H. D. Kaehn, Phys. Rev. <u>182</u>, 649 (1969).

⁵B. G. Lazarev, L. S. Lazareva, T. A. Ignat'eva,

and V. I. Makarov, Dokl. Akad. Nauk SSSR 163, 73

(1965) [Sov. Phys. Doklady 10, 620 (1966)].

⁶C. W. Chu, T. F. Smith, and W. E. Gardner, Phys.

Rev. Letters <u>20</u>, 198 (1968).

 7 J. H. Davis, M. J. Skove, and E. P. Stillwell, Solid State Commun. <u>4</u>, 597 (1966).

⁸R. M. Fisher, L. S. Darken, and K. G. Carroll,

Acta Met. 2, 368 (1954).

⁹E. P. Stillwell, M. J. Skove, and J. H. Davis, Rev. Sci. Instr. 39, 155 (1968).

¹⁰D. R. Overcash, M. J. Skove, and E. P. Stillwell, Phys. Rev. <u>187</u>, 570 (1969).

¹¹E. P. Stillwell, R. L. Gardner, and H. T. Littlejohn, Am. J. Phys. <u>35</u>, 502 (1967).

¹²G. K. White and S. B. Woods, Rev. Sci. Instr. <u>28</u>, 638 (1957).

¹³M. F. Merriam, Phys. Rev. <u>144</u>, 300 (1966).

¹⁴D. Markowitz and L. P. Kadanoff, Phys. Rev. <u>131</u>, 563 (1963).

¹⁵N. Ridley, J. Phys. D 1, 955 (1968).

PHYSICAL REVIEW B

VOLUME 3, NUMBER 11

1 JUNE 1971

Onset of Superconductivity in One-Dimensional Systems*

J. R. Tucker[†]

Department of Physics, Harvard University, Cambridge, Massachusetts 02138

and

B. I. Halperin[‡]

Bell Telephone Laboratories, Murray Hill, New Jersey 07974 (Received 8 January 1971)

Time-dependent Ginzburg-Landau theory is used to investigate the resistive transition in a "one-dimensional" superconductor as a function of temperature and current through the wire. A diagrammatic expansion, in powers of the interaction between fluctuations, is described for the electrical conductivity. The conductivity is then calculated using a Hartree-Fock approximation for the interaction. When large currents flow through the wire, an unstable region is found near the depressed critical temperature, suggesting possible hysteresis effects which may have been observed in recent experiments.

I. INTRODUCTION

There has been a great deal of interest in investigating fluctuation effects in superconductors, and recently some of this interest has centered on attempts to obtain a detailed understanding of the resistive transition in samples which are effectively one dimensional. Such systems consist of tiny whisker crystals of superconducting material with cross-sectional dimension d much smaller than $\xi(T)$, the Ginzburg-Landau coherence length.

The original microscopic calculations by Aslamazov and Larkin¹ described the additional conductivity due to the presence of fluctuating pairs at temperatures $T > T_c$, and in the limit of small electric field, for systems of one, two, and three dimensions. Later several authors² obtained essentially the same results from linearized timedependent Ginzburg-Landau (TDGL) theory, and succeeded in generalizing expressions for the excess conductivity to include the case of finite electric field.³ All of these calculations were intended to describe the effects of thermally fluctuating pairs on the properties of the normal state at temperatures sufficiently above T_c that interaction between fluctuations could be neglected. The TDGL equation in this case is linear in the order parameter.

A theory of intrinsic resistance in one-dimensional systems below the superconducting transition, in which the (repulsive) interaction between Cooper pairs plays a dominant role, was proposed by Langer and Ambegaokar⁴ (LA) and developed in detail by McCumber and Halperin⁵ (MH). These theories assume the system to be initially in one of the metastable current-carrying states obtained as solutions to the equilibrium Ginzburg-Landau equations. Resistance is then due to thermal fluctuations which cause a transition from this metastable state to one with a smaller current, and depends primarily on the free-energy barrier for