Structural and electrical transport properties of superconducting Au$_{0.7}$In$_{0.3}$ films:  
A random array of superconductor-normal metal-superconductor (SNS) Josephson junctions

Yu. Zadorozhny and Y. Liu

Department of Physics, The Pennsylvania State University, University Park, PA 16802  
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The structural and superconducting properties of Au$_{0.7}$In$_{0.3}$ films, grown by interdiffusion of alternating Au and In layers, have been studied. The films were found to consist of a uniform solid solution of Au$_{0.9}$In$_{0.1}$, with excess In precipitated in the form of In-rich grains of various Au-In phases (with distinct atomic compositions), including intermetallic compounds. As the temperature was lowered, these individual grains became superconducting at a particular transition temperature ($T_c$), determined primarily by the atomic composition of the grain, before a fully superconducting state of zero resistance was established. From the observed onset $T_c$, it was inferred that up to three different superconducting phases could have formed in these Au$_{0.7}$In$_{0.3}$ films, all of which were embedded in a uniform Au$_{0.9}$In$_{0.1}$ matrix. Among these phases, the $T_c$ of a particular one, 0.8 K, is higher than any previously reported for the Au-In system. The electrical transport properties were studied down to low temperatures. The transport results were found to be well correlated with those of the structural studies. The present work suggests that Au$_{0.7}$In$_{0.3}$ can be modeled as a random array of superconductor-normal metal-superconductor (SNS) Josephson junctions. The effect of disorder and the nature of the superconducting transition in these Au$_{0.7}$In$_{0.3}$ films are discussed.

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I. INTRODUCTION

The interplay between superconductivity and carrier localization has long been a focus of research on disordered superconductors. This question was initially raised in the study of granular superconducting films. Such films, in which individual grains are separated by insulating barriers, can be modeled as superconductor-insulator-superconductor (SIS) tunnel junction arrays. At lower temperatures, the film properties are dictated by the competition between the inter-grain Josephson coupling and the carrier confinement due to the charging energy of the junctions. For a superconducting grain, the number of Cooper pairs and the phase of the superconducting order parameter are conjugate variables subject to an uncertainty relation. As the fluctuation in the number of Cooper pairs is suppressed due to the charging energy, the fluctuation in the superconducting phase is enhanced. Josephson coupling between adjacent superconducting grains, if strong enough, will lead to global phase coherence.

The localization effect has also been examined for homogeneously disordered systems, such as ultrathin amorphous films, in which electrons in the normal state are strongly localized. In this case, superconductivity can also be suppressed, with a reduction in the superconducting transition temperature or a complete loss of superconductivity. The same phase-number uncertainty relation can be used to understand this phenomenon, since the fluctuation in the number of electrons is again suppressed, due to localization. In fact, a theoretical model based on this picture has been suggested. As the amount of disorder increases, a structurally and chemically homogeneous superconducting film will break down into superconducting droplets, linked by Josephson coupling. The superconducting state will survive only if the coupling is sufficiently strong. This model makes it possible to understand the interplay between superconductivity and localization in both granular and homogeneous films on the same footing.

A natural question to ask is whether it is possible to identify a system that can be disordered for superconductivity, but in which electrons or Cooper pairs are not particularly localized. An example of such a system could be a granular superconductor with a superconductor-normal metal-superconductor (SNS) type of intergrain Josephson coupling. In this regard, it is interesting to note that, while artificial proximity-coupled Josephson junction arrays have been examined in detail, including finite size effects and the effect of disorder in granular materials that can be modeled as an SNS junction array, such as niobium nitride films, have hardly been studied.

We have studied films of Au$_{0.7}$In$_{0.3}$. These films are strongly disordered as far as superconductivity is concerned due to a spatially varying local superconducting transition temperature, $T_{c0}$. (Phase separation in this binary alloy system is directly responsible for the spatially varying $T_{c0}$, as shown below.) On the other hand, Cooper pairs are not confined as in the case of conventional SIS granular films. We report several generic features of these Au$_{0.7}$In$_{0.3}$ films, grown at ambient temperature, in which the interdiffusion of Au and In led to the formation of several superconducting Au-In minority phases, including solid solutions and intermetallic compounds with distinct concentrations of Au and In. We show that these Au-In films may be regarded as ran-
The rest of the paper is organized as follows: in Sec. II we review various phases present in bulk Au-In alloy and summarize the results on superconductivity in the Au-In binary system previously reported in literature. In Sec. III we describe the techniques used in film growth and characterization. Sec. IV and V deal with the film conducting properties of these films. Finally, Sec. VIII summarizes the results on superconductivity in Au-In alloy.

II. Au-In ALLOY

The phase diagram of Au-In binary alloy is rich and relatively well studied. In the bulk form, the following alloy or intermetallic compound phases, stable at room temperature, have been reported: 1) face-centered cubic (fcc) solid solution with room temperature solubility ranging from 0 to approximately 10 atomic % of In (α phase); 2) the Nd- or Ni3Ti-type hexagonal α1 phase (11-13% of In). However, the existence of this phase as a stable one was questioned; 3) close-packed hexagonal (cph) solid solution (ζ, 10-16% of In); 4-6) solid solution phases of β1, c′ and γ′, with In concentrations around 22%, 25%, and 30%, respectively. The structure and precise stoichiometry of these phases are yet to be clarified.

Intermetallic compounds AuIn (triclinic) and AuIn2 (of fluorite-type crystal structure) with a negligible solubility of In or Au in these compounds. There is no appreciable solid solubility of Au in In. A schematic phase diagram around the room temperature is shown in Fig. 1, in which the α1 phase is marked with a dashed line to reflect the uncertainty associated with this phase. In thin-film samples prepared by interdiffusion of Au and In at room temperature, a technique employed in the present experiment, additional phases may be present.

Superconductivity in several phases of Au-In alloy has been reported, as summarized in Ref. 1. In the bulk form, pure In has a transition temperature of 3.40 K. The Tc of intermetallic compound AuIn was reported to be between 0.4 and 0.6 K. However, the measurements were done on a powder ingot; the phase composition of the sample was not independently verified. Measurements on AuIn2 ingots yielded values of Tc from 0.1 K to 0.2 K. For the α-phase, the Tc of Au0.9In0.1, close to the limit of solid solubility of In in Au, was 27 mK. It further decreased to 11 mK for Au0.94In0.06 and to 0.5 mK for Au0.95In0.05. The Tc of the ζ phase has been reported to be between 25 and 100 mK for a nominal composition of Au0.8In0.2.

From these previous studies it is clear that there exist numerous superconducting phases in the Au-In alloy system. In addition, a significant range of In concentrations does not correspond to any single equilibrium phase at room temperature. As a result, depending on the composition and the preparation technique, various phase-separated samples are expected. Since a wide range of superconducting transition temperatures are possible for these phases, the local Tc is expected to vary spatially as well.

The Au-In system has been of great interest for both fundamental research and practical applications. In particular, it has served as a model system for experimental studies of bulk and film solid diffusion and compound formation. The α-phase alloy attracted a large amount of attention in the pursuit of possible superconductivity in pure Au. Nuclear ferromagnetism was recently found to coexist with superconductivity in AuIn2 at micro-kelvin temperatures. We have previously reported observations of strongly enhanced magnetic fingerprints and of fractional-flux Little-Parks oscillations in disordered mesoscopic Au0.7In0.3 cylinders.

Au-In alloy is also important for practical applications. The wide range of superconducting transition temperatures in the same material system is useful for applications where a well targeted Tc is needed. Au-In films are easy to prepare, adhere well to most substrates, and are structurally stable. They do not oxidize significantly, either. AuIn2 is used in commercial superconducting quantum interference devices (SQUIDs).

III. SAMPLE PREPARATION AND MEASUREMENTS

Au-In films were grown in a high vacuum chamber by sequential deposition of 99.9999% pure Au and In. Glazed alumina or polished quartz was used as substrates. Three or five alternating layers were deposited, the top and the bottom layers being Au. The respective layer thicknesses for Au and In, not exceeding 10 nm each, were determined based on the desired stoichiometric ratio. During evaporation, the deposition rate was controlled using a quartz crystal thickness monitor. A typical film was 6 mm long and 0.5 mm wide, patterned for 4-point electrical measurements.

The initial mixing of Au and In apparently occurred during the evaporation at the ambient temperature. Subsequently, interdiffusion continued at room temperature. Based on a previous study, the film structure should have stabilized within several days. Depth profiles of Au0.7In0.3 films revealed signs of incomplete interdiffusion of Au and In right after the deposition. However, they disappeared after a few days (Sec. V). Resistance measurements showed no evidence for a superconducting transition at 3.4 K in any film, indicating that no macroscopic fraction of pure In was present.

The surface of every Au-In film was studied by atomic force microscopy (AFM) and/or by scanning electron
microscopy (SEM). X-ray photo-electron spectroscopy (XPS) was used to probe the chemical composition of select samples. The relative concentrations of Au and In were inferred from the energy spectra of emitted electrons, averaged over a surface area of the order of 1 mm² and a depth of about 5 nm. After each scan, the surface layer was removed by Ar⁺ ion etching, exposing the underlying material. A depth profile of the chemical composition was generated by repeating this procedure until all of the film was etched away. Laterally resolved, depth-averaged composition analysis was carried out using back-scattered electron micrography (BSEM) and characteristic X-ray analysis. Crystalline structure of the films was also analyzed by X-ray diffractometry (XRD).

Electrical transport measurements of Au-In films were carried out in a 3He or a dilution refrigerator equipped with a superconducting magnet. All electrical leads entering the cryostat were filtered with the attenuation of 10 dB at 10 MHz and 50 dB at 300 MHz. The 4-point resistance of the films was measured using a d.c. current source and a digital nanovoltmeter. The measuring current was chosen in the ohmic regime of the current-voltage (I-V) characteristic (typically 1µA).

IV. FILM MORPHOLOGY

It is known that film morphology for a particular material system can vary widely depending on the preparation condition. The film morphology will, in general, affect the electrical transport and superconducting properties of the film. Therefore it is important to document the general morphological features of Au0.7In0.3 films. It was found that, depending on the evaporation rate (e) and the total thickness (t), two different types of Au0.7In0.3 films, marked by its distinct surface morphology, could be grown.

Films thinner than 30 nm were found to be smooth regardless of the evaporation rate (0.1 ≤ e ≤ 2 nm/s). Thick films (t ≥ 30 nm) were also smooth if deposited at about 0.5 nm/s or slower. However, micron-size clusters were found in thick films (t ≥ 30 nm) that had been deposited at 1-2 nm/s (Sec. IVA), resulting in bumpy films. The change in the film morphology for thicker films may be attributed to the higher atomic mobility during fast deposition. In thinner films, the formation of clusters was suppressed probably because of the reduced atom mobility originating from substrate effects.

AFM pictures of two Au0.7In0.3 films are shown in Fig. 2. Film #25, nominally 10 nm thick, represents the smooth films. The film was indeed uniform at large length scales, as seen in Fig. 2a. A close-up view (Fig. 2b) revealed the presence of grains with a typical size of 20 nm, larger than the nominal film thickness. This may indicate that the film was formed by a single layer of grains. The grains were most likely crystalline, according to XRD studies of thicker Au0.7In0.3 films.

A 40 nm thick sample, film #29 shown in Figs. 2c and d, is representative of bumpy films, was prepared by depositing at a rate faster than 1 nm/s. For these films, the variation in the surface height was comparable to the total film thickness. Distinct clusters, showing up as bright spots in the picture, protruded from the rest of the film surface by about 20 nm. The clusters, several microns apart, often appeared to have a somewhat rounded hexagonal shape, with a typical size of 1 µm. The clusters were surrounded by circular terraces, which were 4-5 µm in diameter and elevated above the rest of the film by a few nm.

A closer examination (Fig. 2d) revealed that the size of the grains in the clusters is approximately 60 nm, while the grains comprising surrounding areas were similar in size to those in uniform films. This probably was either a consequence of the larger film thickness at the protruding clusters, or an indication that these clusters were of a Au-In phase different from that of the surrounding area.

V. PHASE COMPOSITION

Another important set of questions is related to the spatial variation of the chemical composition of the Au0.7In0.3 films, and whether it correlates with the film morphology. To answer these questions, depth profiling of the chemical composition was carried out by XPS. The XPS analysis yielded direct information on the chemical composition in the vertical direction of the film. When combined with AFM imaging, the XPS depth profiling provided information on chemical composition in the lateral direction, as shown below.

A 100 nm thick sample (film #28) was prepared by alternating evaporation of six 10 nm thick layers of Au and five 8 nm thick layers of In onto a quartz substrate. An XPS depth profiling analysis was carried out immediately after the film deposition. Fig. 3 contains an energy spectrum of photo-electrons emitted from the film’s original surface. Au and In concentrations were calculated from the areas of Au 4p3/2 and In 3d5/2 lines, using standard sensitivity factors of 3.8 and 0.62 respectively. Traces of oxygen and carbon were found at the surface, likely due to post-deposition contamination in the air, but disappeared deeper into the film.

Shown in Fig. 3 are two depth profiles of film #28, obtained by taking XPS scans at approximately 4 nm steps. For the profile taken immediately after the deposition, remnants of the layers are evident near the surface but become less distinct as the substrate is approached. This can be understood by noting that the bottom layers, deposited first, were exposed to elevated temperatures during evaporation longer and therefore had inter-diffused more. In the depth profile obtained after the film had been aged, no evidence of any layered structure was found. The measured In concentration was approx-
imately 28% at the surface of the film and 10% deep in the film.

The above observation can be understood by taking into account the rate of ion etching for different alloy phases. In Fig. 2, we show a sequence of AFM scans of the film’s surface at different stages of profiling. The surface roughness increased significantly after etching, suggesting that during ion etching, the In-rich grains were preferentially sputtered away. Indeed, after two or three etching periods the XPS could only detect the Au$_{0.9}$In$_{0.1}$ component, in agreement with this preferential etching picture. Taking into account the bulk phase diagram described above, it is clear that the uniform matrix was the saturated solid solution, Au$_{0.9}$In$_{0.1}$ α phase. An XPS scan of the original film surface yielded In concentration close to the true average value of 28%. The difference between the measured In concentration and the expected In concentration of the film's surface at different stages of profiling. The surface layer immediately after the deposition, with Au being the ending layer.

The electrical transport measurements, discussed below, suggested that the In-rich grains found in the Au$_{0.7}$In$_{0.3}$ films might be made of different phases, including intermetallic compound AuIn. A previous study of the room temperature interdiffusion of Au and In in Au-In bilayer showed that for In fraction between 23 and 36%, both AuIn and AuIn$_2$ compounds were formed initially in the excess Au. However, after several days, and for months thereafter only AuIn could be found. If the same pattern of phase formation was applicable to our films as well, then the In-rich grains might be predominantly intermetallic compound AuIn.

The presence of protruding clusters in the bumpy films is very striking. Could the chemical composition of these clusters be different from the rest of the film? XPS studies of these bumpy films yielded depth profiles similar to those described above. Most importantly, the protruding clusters were not preferentially etched. Therefore these clusters were likely not the same In-rich phases found in the smooth films. The AFM images away from the clusters showed similar features (Fig. 3), suggesting that away from the clusters the film had similar compositional distribution as in the smooth films. While back-scattered electron micrography (BSEM), characteristic X-ray analysis, and X-ray diffractometry (XRD) were all attempted, the precise chemical composition of the protruding clusters remain undetermined, mainly due to the lack of reliable standards. To determine the precise phase of these clusters, considerable more work on crystallography of the Au-In system, including the preparation of pure phase samples to be used as standards, would be needed. These experiments, which are beyond the scope of the present study, were not carried out.

The picture emerging from these morphological and chemical studies is one in which multiple material phases were formed in the films. The predominate phase appears to be the saturated Au$_{0.9}$In$_{0.1}$ α phase. The others were various In-rich phases. The Au$_{0.9}$In$_{0.1}$ formed a matrix with In-rich regions embedded in it randomly, in both the position and the size.

#### VI. ELECTRICAL TRANSPORT PROPERTIES OF Au$_{0.7}$In$_{0.3}$

The characteristic features in film morphology and chemical composition are expected to dictate the electrical transport properties of the films. In Fig. 3, we show normalized resistances plotted against temperature for two sets of Au$_{0.7}$In$_{0.3}$ films of varying thickness. In each set, the films were grown under the same nominal conditions. The films in Fig. 3(a) were prepared by slow deposition, with all films exhibiting a smooth morphology, free of the clusters. The films shown in Fig. 3(b) were prepared by fast deposition. Protruding clusters such as those shown in Fig. 2c and d appeared clearly for films with thickness of 30 nm and larger.

For films of multiple phases, the onset of the superconducting transition usually marks the highest local $T_c$ for a particular phase. Films prepared by slow deposition shown in Fig. 3(a) had three onset superconducting transition temperatures, roughly 0.45K, 0.65K, and 0.8K, depending on film thickness. On the other hand, for films prepared by fast deposition shown in Fig. 3(b), the onset of superconductivity was found at 0.45 and 0.8K only, with a sharp drop in film resistance at 0.6 K. These onset temperatures probably correspond to the $T_c$ of different In-rich phases.

The temperature, 0.65K, is consistent with the highest $T_c$ reported for the intermetallic compound AuIn. Combining results obtained in morphological and compositional analyses presented above, it is reasonable to assume that this In-rich phase was AuIn. However, exactly which two phases were responsible for superconductivity at 0.45 and 0.8K are more difficult to determine. In this regard, it should be noted that 0.8K is higher than any superconducting transition temperature previously reported for the Au-In system. The $T_c$ reported for saturated Au$_{0.9}$In$_{0.1}$ α phase was 77 mK.

Two smooth films, 30 and 40nm thick and prepared by fast deposition, showed two sharp drops in film resistance around 0.3 and 0.6K respectively (Fig. 3(a)). The drop at 0.6K should correspond to the superconducting transition of grains of compound AuIn. In principle, the second drop in resistance could correspond to the superconducting transition of a percolation path formed by the phase with a local $T_c$ around 0.45K as identified above, or it might be due to emergence of a global phase coherence among the AuIn grains, which were responsible for the resistance drop at 0.6K.

Experimentally, it was found that the temperature of this second drop could be increased significantly after low-temperature (< 400 K) annealing by exposing the sample to a heat lamp for one hour in vacuum (Fig. 3).
Based on the bulk phase diagram of Au-In alloy, such low-temperature annealing should not lead to the formation of a new stable phase. Indeed, the temperature corresponding to the first resistance drop remained essentially unchanged after annealing. On the other hand, the normal state resistance was reduced significantly. It is known the Josephson coupling in an SNS junction depends on the N-layer resistance. Therefore, it appears that the formation of a separate superconducting phase was not the cause for the resistance drop found at the lower temperature.

Two-step transitions were observed previously in proximity-coupled Josephson junction arrays. However, because the local $T_c$ of the superconducting islands is uniform and equal to the bulk $T_c$ of the superconductor, the resistance dropped sharply at this temperature. In the present study, because of the proximity effect, the In-rich grains embedded in the Au$_{0.9}$In$_{0.1}$ $\alpha$ phase matrix (which is a normal metal above its $T_c$ of 77mK) would have a local $T_c$ dependent on their size, and the exact local In concentration. Below the superconducting transition of these In-rich grains, but above that of Au$_{0.9}$In$_{0.1}$, the films should behave as random networks of SNS (proximity-coupled) Josephson junctions. As temperature is lowered, a sharper transition to a zero-resistance superconducting state was observed due to the Josephson coupling of all superconducting grains. This picture is fully consistent with the one inferred from the structural and compositional analyses.

VII. EFFECT OF DISORDER AND THE NATURE OF SUPERCONDUCTING TRANSITION

The normal-state resistivity of a Au$_{0.7}$In$_{0.3}$ film is a good measure of the degree of disorder that affects electron motion in the normal state. The normal-state resistivity can be controlled by the film thickness. In the present study, the underlying phases of films prepared by slow deposition may have been affected strongly by the details of the substrate conditions. On the other hand, the films prepared by fast deposition shown in Fig. revealed behavior as a function of film thickness more systematic than those prepared by slow deposition, and therefore will be the focus of this discussion.

The onset of the superconducting transitions of these films prepared by fast deposition was found to be at either 0.45K or 0.8K, corresponding to two possible Au-In phases, rather than three, as seen in the case of films prepared by the slow deposition. The zero-resistance $T_c$ corresponding to the onset of the global phase coherence was suppressed from about 0.6 K for the 35-40 nm thick films to about 0.2 K for the 8 nm thick film. For the 5 nm thick film, no superconductivity was found. However, even such a thin film remained metallic down to 0.3 K, indicating that the electrons in Au$_{0.7}$In$_{0.3}$ films are delocalized in the normal state.

The normal state sheet resistance, while changing from a few to a few dozen ohms (see Table 2), remained very low for all films studied. For thicker films, the values of the sheet resistance scaled with the film thickness, yielding a resistivity of approximately $12 \times 10^{-6}$ $\Omega \cdot \text{cm}$ at 1 K. Despite of the small normal-state resistivity, thin Au$_{0.7}$In$_{0.3}$ films had a broad transition regime ($> 0.2$ K), as seen in Fig. Such a broad transition is typically found in strongly disordered superconductors due to strong phase fluctuations. In artificially prepared SNS junction arrays, the transition width was found to increase with the increase of disorder. The broad superconducting transition is a clear indication that Cooper pairs experience substantial disorder despite the low normal-state resistance.

The relatively thick Au$_{0.7}$In$_{0.3}$ films, such as the 30nm and 35nm thick films, showed a distinctly different behavior. First, there was a gradual decrease in film resistance right below the onset $T_c = 0.8$ K. As the temperature was lowered, sharp drop to zero resistance was found at $T = 0.6$ K (slightly below the local $T_c$ of the compound AuIn, 0.65K). The temperature dependence of the resistance may be qualitatively explained using the following picture: The local $T_c$ of the protruding clusters is 0.8 K. These clusters induced superconductivity in their surrounding regions below 0.8 K due to proximity effect. The size of these superconducting regions increased as the decreasing temperature, resulting in a gradual drop in film resistance. Finally, slightly below 0.6K, a zero-resistance superconducting state was established due to either the emergence of global phase coherence in an SNS junction array formed by the protruding clusters, or alternatively by the formation of a percolation path of Au$_{0.7}$In$_{0.3}$ films with a sharp transition to global superconductivity may possess a high degree of disorder for Cooper pairs because of the inhomogeneity in the amplitude of superconducting order parameter, even though the normal state is a perfectly good metal, making these Au$_{0.7}$In$_{0.3}$ films a unique superconducting system.

In Fig. the results of more detailed resistance measurements are presented for 30 nm thick film. Perpendicular ($B_\perp$) and parallel ($B||$) magnetic fields were found to affect the superconducting transition of the film in very different ways. This was expected since $B_\perp$ created superconducting vortices in the film, which would induce a finite resistance if they were depinned and started to creep in the film. On the other hand, the effect of $B||$ was primarily to suppress superconductivity of each individual grain via the Zeeman effect, as in this case free vortices cannot form in the film. Correspondingly, a much lower $B_\perp$ than $B||$ was needed to suppress the global superconducting transition.

The motion of vortices in an SNS Josephson junction array is related directly to phase slips. Enhanced phase slips, and therefore phase fluctuations, will lead to a
wide superconducting transition regime and a suppressed
global $T_c$, as shown in Fig. 9. Similar behavior was found
when the film was made thinner, as discussed above. In
a parallel field, while the field does not introduce free
vortices to the film directly, suppressing superconductivity
in the grains with a sufficiently large $B_{||}$ will lead to
enhanced phase slip, which involves the unbinding of a
vortex-antivortex pair, again resulting in a wider super-
conducting transition regime. This effect is illustrated
clearly in Fig. 10 where the magnetic field dependence
for both on the global and the onset (local) $T_c$ is shown.

It is interesting to note that the resistance just above
the drop, $R_p$, corresponding to either the normal-state re-
sistance of the SNS junction array or the resistance of the
link or links that complete the percolating path, showed
an intriguing magnetic field dependence. It was found
that $R_p$ varied smoothly with the amplitude of the field,
independent of the field orientation, as shown in Fig. 10.
In addition, in small perpendicular magnetic fields, $R_p$
increased with field (Fig. 10). Such a variation may pro-
vide a hint as to how the global phase coherence emerges
in these bumpy films. To clarify such field dependence of
$R_p$, more theoretical and experimental work is needed.

VIII. CONCLUSION

In conclusion, we have presented results on the study of
superconducting films of Au$_{0.7}$In$_{0.3}$ alloy with varying
thickness. The films were grown by sequential de-
position and interdiffusion of Au and In. The structure
and the electrical transport studies of Au$_{0.7}$In$_{0.3}$
films, especially the observations of the onset supercon-
ducting transition temperatures of the films, suggest the
presence of several Au-In phases, including, most likely,
Au$_{0.9}$In$_{0.1}$ ($T_c = 77$ mK), two In-rich phases ($T_c = 0.45$
and 0.65 K, respectively), and the intermetallic com-
 pound AuIn ($T_c = 6.6$ K).

At temperatures below their onset superconducting
transition temperature, the In-rich regions underwent a
superconducting transition and became Josephson cou-
pled with one another, resulting in a random network
of Josephson junctions of SNS type, similar to previous
measurements of artificial arrays of SNS Josephson junc-
tions. The present work appears to have established a
unique system which is disordered as far as the Cooper
paris are concerned, while the normal state remains a
good metal.

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1 G. Deutscher, Y. Imry, and L. Gunther, Phys. Rev. B 10,
4598 (1974); D. C. Dynes, J. P. Garno, and J. M. Rowell,
Phys. Rev. Lett. 40, 479 (1978); B. G. Orr, H. M. Jaeger,
and A. M. Goldman, Phys. Rev. B 32, 7586 (1985).
2 see, for example, Percolation, Localization, and Super-
conductivity, edited by A. M. Goldman and A. A. Wolf
(Plenum Press, New York, 1984).
3 M. Tinkham, Introduction to Superconductivity, 2nd Ed.,
(McGraw-Hill, Inc., New York, 1996). P. 52.
4 B. G. Orr, H. M. Jaeger, A. M. Goldman, and C. G.
Kuper, Phys. Rev. Lett. 56, 378 (1986); H. M. Jaeger,
D. B. Haviland, B. G. Orr, and A. M. Goldman, Phys.
Rev. B 40, 182 (1989).
5 For a recent review, see, for example, A. M. Goldman and
N. Markovic, Physis Today 51, November issue, 39 (1998),
and references cited therein.
6 F. Zhou, and B. Spivak, Phys. Rev. Lett. 80, 5647 (1998).
7 For a recent review, see R. S. Newrock, C. J. Lobb,
U. Geigenmüller, and M. Octavio in Solid State Physics,
vol. 54 (Academic Press, San Diego, 2000), p. 263.
8 S. T. Herbert, Y. Jun, R. S. Newrock, C. J. Lobb,
K. Ravindran, H.-K. Shin, D. B. Mast, and S. Elhamri,
Phys. Rev. B 57, 1154 (1998).
9 D. C. Harris, S. T. Herbert, D. Stroud, and J. C. Garland,
Phys. Rev. Lett. 67, 3606 (1991).
10 M. W. Johnson and A. M. Kadin, Phys. Rev. B 57, 3593
(1998).
11 Phase Diagrams of Binary Gold Alloys, edited by
H. Okamoto and T. B. Massalski (ASM International,
Materials Park, Ohio, 1987), p. 142.
12 C. H. Hamilton, Ch. J. Raub, B. T. Matthias, E. Coren-
zwit, and G. W. Hull, Jr., J. Phys. Chem. Solids 26, 665
(1965).
13 J. H. Wernick, A. Menth, T. H. Geballe, G. Hull, and
J. P. Maita, J. Phys. Chem. Solids, 30, 1949 (1969).
14 S. Rehmann, T. Herrmannsdörfer, and F. Pobell,
Phys. Rev. Lett. 78, 1122 (1997).
15 Ch. Buchal, R. F. Mueller, F. Pobell, M. Kubota, and
H. R. Folle, Solid State Commun. 42, 43 (1982).
16 R. F. Hoyt and A. C. Mota, Solid State Commun. 18, 139
(1976).
17 H. L. Luo and K. Andres, Phys. Rev. B 1, 3002 (1970).
18 T. G. Finstad, T. Andreassen, and T. Olsen, Thin Solid
Films 29, 145 (1975).
19 V. Simić and Ž. Marinković, Thin Solid Films 41, 57
(1977).
20 M. Seyffert, A. Siber, and P. Ziemann, Phys. Rev. Lett.
67, 3792 (1991); Thin Solid Films 208, 197 (1992).
21 M. Millares, B. Pieraggi, and E. Lelievre, Solid State Ionics
63, 575 (1993).
22 T. Herrmannsdörfer, P. Smeibid, B. Schröder-Smeibid,
and F. Pobell, Phys. Rev. Lett. 74, 1665 (1995).
TABLE I. Summary of the sample characteristics of \( \text{Au}_{0.7}\text{In}_{0.3} \) films discussed in this paper, including morphology ("s" for smooth films, "b" for bumpy films with protruding clusters), nominal film thickness \( d \), evaporation rate \( e \), normal-state sheet resistance \( R_N \), onset temperature for superconductivity \( T_{\text{onset}} \), and zero-resistance transition temperature \( T_c \). Actual thickness, estimated from AFM edge profiles for some films, was larger than \( d \) by several percent. \( T_{\text{onset}} \) and \( T_c \) were evaluated at the 0.99 \( R_N \) and 0.01 \( R_N \) resistance level, respectively. Electrical transport measurements were not carried out for Films #28, 29, and 32, which were prepared for structural and compositional studies.

| Film | s/b | \( d \), nm | \( e \), nm/s | \( R_N^2 \Omega \) | \( T_{\text{onset}} \), K | \( T_c \), K |
|------|-----|-------------|-------------|----------------|----------------|--------|
| #8   | b   | 30          | 1.2         | 4.2            | 0.734           | 0.586  |
| #9   | s   | 20          | 0.9         | 11.5           | 0.669           | 0.378  |
| #10  | b   | 35          | 1.6         | 3.9            | 0.804           | 0.561  |
| #11  | s   | 25          | 1.0         | 6.4            | 0.767           | 0.561  |
| #13  | s   | 28          | 1.8         | 6.6            | 0.631           | 0.375  |
| #16  | s   | 15          | 1.0         | 34.4           | 0.423           | 0.333  |
| #18  | s   | 8           | 0.5         | 89.1           | 0.431           | 0.229  |
| #19  | s   | 5           | 3000        | -              | -               | -      |
| #21  | s   | 40          | 0.4         | 3.1            | 0.624           | 0.321  |
| #22  | s   | 30          | 0.4         | 4.7            | 0.653           | 0.340  |
| #23  | s   | 20          | 0.5         | 8.2            | 0.791           | 0.374  |
| #24  | s   | 10          | 0.3         | 25.0           | 0.456           | 0.300  |
| #25  | s   | 8           | 0.3         | 42.6           | 0.416           | 0.273  |
| #28  | b   | 100         | 0.6         | -              | -               | -      |
| #29  | b   | 100         | 1.3         | -              | -               | -      |
| #32  | b   | 100         | 1.5         | -              | -               | -      |

FIG. 1. Schematic Au-In phase diagram, after Ref. 11. Phases reported to be superconducting include pure In \( (T_c = 3.4 \text{ K}) \), AuIn\(_2\) \( (T_c = 0.1 - 0.2 \text{ K}) \), AuIn \( (T_c = 0.4 - 0.6 \text{ K}) \), \( \zeta \) \( (T_c \approx 0.1 \text{ K}) \), and \( \alpha \) \( (T_c = 0 - 77 \text{ mK}) \).

FIG. 2. (a-d) AFM images of two Au\(_{0.7}\)In\(_{0.3}\) films. Brighter areas correspond to more elevated regions. (a,b) are for 10 nm thick sample, film #25. Image areas are 20 \( \times \) 20 and 1 \( \times \) 1 \( \mu \text{m}^2 \) respectively. Image depth, the surface height variation corresponding to the entire gray scale, is 5 nm for both pictures. (c,d) are for 40 nm thick (film #29). Image areas 20 \( \times \) 20 and 2 \( \times \) 2 \( \mu \text{m}^2 \), with an image depth of 100 nm.

FIG. 3. (a) Photoelectron spectrum of the surface layer of a 100 nm thick Au\(_{0.7}\)In\(_{0.3}\) sample, film #28. Major lines are identified. (b) Depth profiles of the relative concentrations of Au and In for film #28. The concentrations are plotted as functions of the etching time (see text), which is proportional to the depth. Open circles – immediately after deposition; solid circles – 2 months after the initial analysis.

FIG. 4. AFM images of film #28 at different stages of depth profiling: (a) before ion etching, (b) with \( \approx 5 \text{ nm} \) removed, and (c) with \( \approx 50 \text{ nm} \) removed. All pictures are 10 \( \times \) 10 \( \mu \text{m}^2 \), with an image depth of 100 nm. Preferential sputtering of the In-rich phase is evidenced by the surface roughening in the process.

FIG. 5. Normalized resistance plotted as a function of temperature for film #22 before (left curve) and after annealing (right curve).

FIG. 6. Resistance plotted as a function of temperature for film #22 before (left curve) and after annealing (right curve).

FIG. 7. Normal-state resistance, taken at \( T = 1 \text{ K} \), plotted as a function of inverse of the film thickness for films shown in Fig. 5. The filled circles are for films grown by slow deposition, while the empty circles are for films prepared by fast deposition.
FIG. 8. Resistance of 30 nm thick bumpy film (film #8), plotted as a function of temperature for different values of (a) perpendicular and (b) parallel magnetic fields, and as a function of (c) perpendicular and (d) parallel field at different temperatures. For (a), from top to bottom, $B_\perp = 300, 170, 160, 150, 120, 110, 100, 75, 50, 25, 10, \text{ and } 0 \text{ G}$. For (b), from top to bottom, $B_\parallel = 4900, 3920, 2940, 2450, 2960, 1500, 980, \text{ and } 0 \text{ G}$. For (c), from top to bottom, $T = 0.65, 0.625, 0.60, 0.55, 0.50, 0.40, \text{ and } 0.33 \text{ K}$. For (d), from top to bottom, $T = 0.60, 0.55, 0.50, 0.425, \text{ and } 0.33 \text{ K}$. Measurements were carried out during two separate cool-downs. Temperatures below 0.3 K were not available for these measurements.

FIG. 9. Values of the onset and the global $T_c$ plotted as a function of magnetic field in (a) perpendicular and (b) parallel directions, for film #8.

FIG. 10. Critical resistance, $R_p$, plotted as a function of magnetic field in perpendicular (open circles) and parallel (filled circles) directions for film #8.