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Absence of superconductivity in topological metal $ScInAu_2$

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ABSTRACT

The Heusler compound $ScInAu_2$ was previously reported to have a superconducting ground state with a critical temperature of 3.0K. Recent high throughput calculations have also predicted that the material harbors a topologically non-trivial band structure similar to that reported for β-$PdBi_2$. In an effort to explore the interplay between the superconducting and topological properties properties, electrical resistance, magnetization, and x-ray diffraction measurements were performed on polycrystalline $ScInAu_2$. The data reveal that high-quality polycrystalline samples lack the superconducting transition present samples that have not been annealed. These results indicate the earlier reported superconductivity is non-intrinsic. Several compounds in the Au-In-Sc ternary phase space ($ScAu_2$, $ScIn_3$, and Sc_2InAu_2) were explored in an attempt to identify the secondary phase responsible for the non-intrinsic superconductivity. The results suggest that elemental In is responsible for the reported superconductivity in $ScInAu_2$.

1. Introduction

Many recent studies in condensed matter physics and materials science have been focused on the investigation of symmetry-protected topological states [1,2]. On top of the initial efforts to identify and classify different topological states, increasing efforts have been spent on exploring the interplay between these states and other electronic and magnetic phases [3,4]. One such avenue of particular interest is materials systems exhibiting both non-trivial topological states and superconductivity [5,6]. These compounds are candidates for being realized as true topological superconductors which are predicted to host Majorana fermions.

One such candidate, the 5.4K superconductor β-$PdBi_2$, attracted attention when it was found to have topologically non-trivial surface states [7]. Ensuing research of the compound revealed a variety of interesting properties including complex spin textures [8] and a possible spin-triplet order parameter [9,10]. Furthermore, spectroscopic measurements on thin films of β-$PdBi_2$ were claimed to have shown evidence of non-trivial superconductivity and Majorana fermions [11]. However, other measurements have shown that the topological surface states likely play no role in the compound's bulk superconductivity [12,13]. Clearly, it would be interesting to compare these results to those for a different compound with a similar combination of superconducting and topological properties.

The search for candidate materials with certain combinations of properties has recently been facilitated by the accessibility of new databases of both experimental and computationally predicted properties. In this case, we searched for materials that exhibited an intersection of two properties: 1. Previous experimental reports of superconductivity, and 2. Computational prediction of a topologically non-trivial band structure. The list of experimental T_c values was taken from the SuperCon database [14]. Topological classification for these compounds were obtained from the Topological Quantum Chemistry Project [15–17]. The compound $ScInAu_2$ was among a small number of materials that indicated superconductivity at readily accessible temperatures (above $\sim 2K$) and a "TI" (topological insulator) classification. This combination of properties lead us to investigate $ScInAu_2$ further. The topological classification "topological insulator - split electronic band representation" is the same as that for β-$PdBi_2$ [15–17].

Given the facts above, we thus sought to characterize the potential interplay of superconductivity and topological properties in $ScInAu_2$. Polycrystalline $ScInAu_2$ was synthesized via arc-melting. Annealing the samples yielded nearly single phase $ScInAu_2$ that displayed no superconducting transition down to 1.8K via electrical resistivity measurements and 2K via magnetization measurements. These results are in contrast to earlier work [18] which indicated superconductivity in

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$ScInAu_2$ with a critical temperature of 3K. Measurements reveal that only unannealed samples present the previously reported superconducting transition at 3K, though the shielding in the magnetic susceptibility is incomplete. These results indicate that $ScInAu_2$ is not superconducting down to 1.8K and that the previously reported critical temperature (T_c) of 3K is likely due to a secondary phase. Based on these results several other compounds in the Au-In-Sc system were probed in search of a potential superconducting phase that could explain the partial shielding of unannealed $ScInAu_2$ leading to the conclusion that elemental indium is responsible.

2. Methods

Arc-melted samples were prepared by combining the raw elements (Au 99.95% metal basis, In 99.99% metals basis, and ultra high purity Ames Laboratory Sc) in stoichiometric ratios and melting on a water-cooled copper hearth under Ar atmosphere. Each sample was melted multiple times, and was flipped in between each melting to ensure homogeneity throughout the boule. Samples that did not contain In had negligible mass loss, whereas samples containing In showed mass losses around 3%. In order to compensate for this, extra In was added and the samples were arc-melted again until the mass of the sample indicated the correct stoichiometry had been reached. The samples were then annealed while wrapped in Ta foil under partial Ar atmospheres. The crystal structures were characterized with powder x-ray diffraction (XRD) using a Siemans D500 diffractometer or a Panalytical X'Pert Pro-diffractometer, and Rietveld refinements using GSAS-II [19] yielded lattice parameters consistent with those given in literature for each compound unless otherwise noted. Electrical transport and magnetization measurements were performed in Quantum Design PPMS and MPMS systems respectively, at temperatures down to $\sim 2K$.

3. Experimental results

3.1. $ScInAu_2$

Polycrystalline samples of $ScInAu_2$ were synthesized via arc-melting. Samples were measured both before and after annealing at 700°C for three days. Fig. 1 presents XRD data for both the annealed and unannealed samples. While the unannealed sample shows a mixture of phases, including $ScInAu_2$, $ScAu_2$, and In, the annealed data indicates nearly single phase $ScInAu_2$. The annealed sample presents a single unidentified impurity peak near 34°C (marked with an asterisk). The fit presented here indicates a cell parameter of 6.695øA which is in good agreement with literature values, and produces a residual of $\sim 8\%$. Electrical resistivity measurements performed on the annealed sample (Fig. 2) show metallic behavior from room temperature down to the base temperature of 1.8K with no indication of the superconductivity at 3K previously reported [18]. It should be noted that the earlier work did not mention if the samples were subjected to any annealing process. Therefore, we carried out additional measurements on the un-annealed multi-phase sample in order to confirm that the reported superconductivity comes from a secondary phase.

Fig. 3 shows the result of magnetic susceptibility measurements on unannealed $ScInAu_2$. The data show a clear drop in the susceptibility beginning slightly below 3K. At the base temperature of 2K the transition is still incomplete but has reached a shielding fraction of more than 50%. In order to estimate the shielding fraction, we included the demagnetization correction of the roughly spherical sample. The substantial shielding indicates that the secondary phase likely comprises a sizable fraction of the total sample volume. Hence, the XRD data suggests that either In or $ScAu_2$ is responsible. A measurement of the magnetization vs field at 2K (inset of Fig. 3) indicates $H_{c1} \sim 0.004T$ and complete flux expulsion by $\lesssim 0.015T$. The critical field of In at 2K is only 0.018T, which is roughly consistent with our observations [20]. The low critical field indicates that the superconducting impurity is almost certainly unreacted elemental indium ($T_c = 3.4K$). Though the T_c observed here is somewhat lower that that of indium ($\sim 3.0K$ from the onset in susceptibility), this could be caused by a combination of disorder, impurities, strain, and/or granularity. Nonetheless, we also tested several other compounds in the Au-In-Sc system (including $ScAu_2$) that had not previously been measured at low temperatures in order to determine if they could instead be responsible for the superconductivity observed in the unannealed sample.

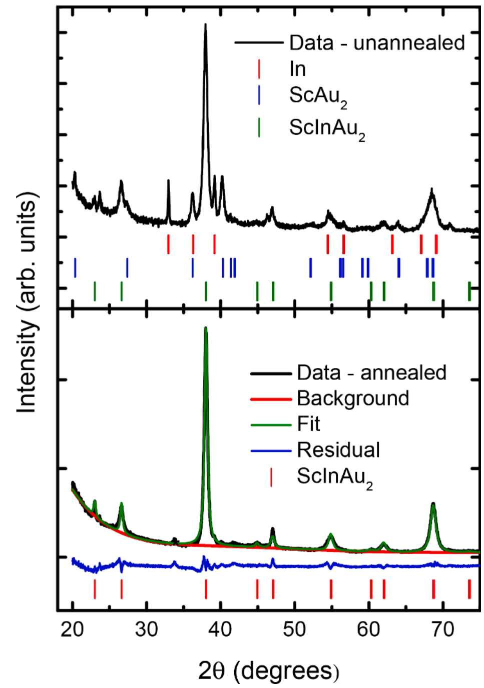

Fig. 1. Top: XRD pattern of unannealed $ScInAu_2$ with ticks indicating expected peaks of In, $ScAu_2$, and $ScInAu_2$. Bottom: XRD pattern of annealed $ScInAu_2$. The small residual indicates that a nearly single-phase sample of $ScInAu_2$ was grown. A small impurity peak is marked with an asterisk.

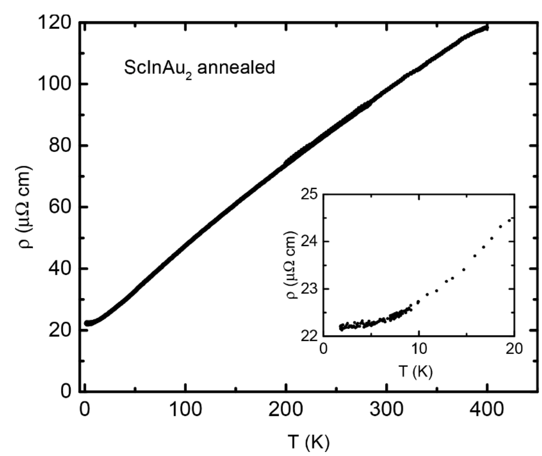

Fig. 2. Resistivity versus temperature of $ScInAu_2$ down to 1.8 K. No indication of superconductivity is observed.

3.2. $ScAu_2$

Arc melted and annealed samples of $ScAu_2$ show diffraction patterns that matched the expected $MoSi_2$-type structure [21]. Electrical resistivity measurements present metallic behavior with a residual resistivity ratio (RRR) of ~ 50. No evidence for superconductivity is

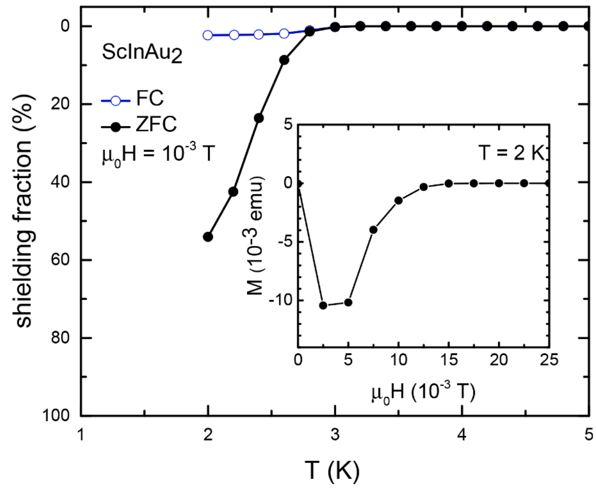

Fig. 3. Shielding percentage versus temperature on unannealed $ScInAu_2$. The incomplete shielding suggests that an impurity phase is responsible. The inset shows the magnetization as a function of applied field. Very small fields of order 0.01T are sufficient to suppress the superconductivity.

detected down to 1.8K (see Fig. 4). The weak upturn in resistivity below $\sim 10K$ could be due to a Kondo effect arising from magnetic impurities or due to grain boundary scattering in the polycrystalline sample.

3.3. $ScIn_3$

Single crystals of $ScIn_3$ were grown with the molten flux method: 80:20 at. % In:Sc were heated in an alumina crucible sealed in a quartz ampule under 70torr Ar gas to 1000°C and then cooled to 400°C over 240hours. After holding at this temperature of 8hours, the ampule was centrifuged to remove the flux. This revealed small, cubic crystals, confirmed by xray diffraction to be cubic $ScIn_3$ [22].

Magnetic measurements on $ScIn_3$ (see Fig. 5) yielded a diamagnetic signal with an onset of around 3K, but the shielding fraction of order 1%. Furthermore, a magnetic field of 0.05T removed this feature. Both of these facts indicate that the superconductivity is not intrinsic to the $ScIn_3$ but is due to droplets of In flux on the surfaces of the crystals. Superconducting transitions have been observed at 0.78K and 0.71K in YIn_3 and $LaIn_3$ respectively [23], suggesting that $ScIn_3$ probably becomes superconducting below 1K.

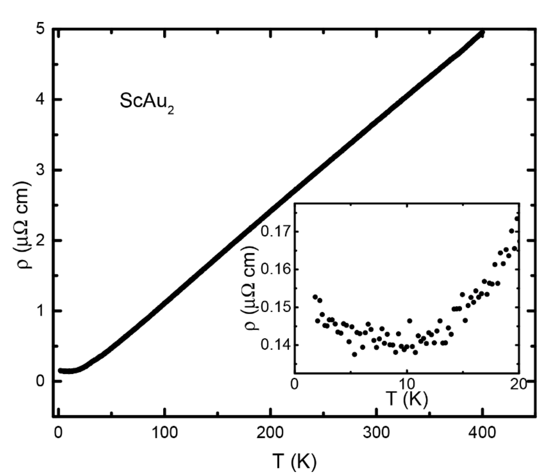

Fig. 4. Electrical resistivity versus temperature for $ScAu_2$ measured from 1.8 to 400K. The sample is non superconducting in this temperature range.

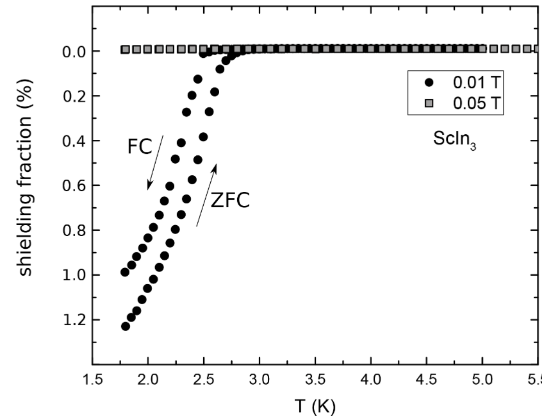

Fig. 5. Magnetic susceptibility vs temperature for $ScIn_3$. The onset of a diamagnetic signal near 2.8K at 0.01T (100Oe), together with the tiny shielding fraction near 1% is consistent with superconductivity deriving from small amounts of indium secondary phase.

3.4. Sc_2InAu_2

Samples of Sc_2InAu_2 were synthesized by arc melting. The tetragonal Mo_2FeB_2-type structure [24] was confirmed by x-ray diffraction, though unidentified secondary phases were present. Nonetheless, magnetic susceptibility measurements from 2-300K presented no evidence for superconductivity or any other anomalies.

4. Conclusions

The previously reported superconducting behavior of $ScInAu_2$, a material that shares the same topological classification as β-$PdBi_2$, has been re-analyzed. These measurements suggest that $ScInAu_2$ is not intrinsically superconducting, but that unannealed samples can exhibit partial superconducting shielding in the magnetic susceptibility due to a secondary phase - most likely unreacted indium. We also investigated the possibility that another phase is responsible for the superconductivity in unannealed samples of $ScInAu_2$. Queries were performed with the Materials Platform for Data Science [25] and the Superconducting Material Database [14] to search for compounds in the Au-In-Sc family that are reported to be superconducting. However, no other phases with reports of $T_c \sim 3K$ were found. Several compounds in this ternary phases space had not previously been characterized at low temperature, so we also screened $ScAu_2$, $ScIn_3$, and Sc_2InAu_2 and found that they are all essentially non-magnetic non-superconducting metals with no anomalies in the resistivity or magnetic susceptibility down to $\sim 2K$.

With the existence of large databases of experimental and computational properties, the search for materials with certain combinations of properties is now straightforward. In this case we identified an inaccuracy in the record - $ScInAu_2$ is non-superconducting, though it had previously been reported to have $T_c = 3K$ [18]. However, it is clear that there are a large number of known superconducting materials with non-trivial band structures awaiting further study.

CRediT authorship contribution statement

J.M. DeStefano: Investigation, Writing – original draft, Writing – review & editing. **G.P. Marciaga:** Investigation, Writing – original draft. **J.B. Flahavan:** Investigation. **U.S. Shah:** Investigation, Software. **T.A. Elmslie:** Investigation. **M.W. Meisel:** Supervision, Writing – review & editing. **J.J. Hamlin:** Supervision, Conceptualization, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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