Absence of superconductivity in topological metal ScInAu$_2$

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(Dated: December 14, 2020)

The Heusler compound ScInAu$_2$ was previously reported to have a superconducting ground state with a critical temperature of 3.0 K. Recent high throughput calculations have also predicted that the material harbors a topologically non-trivial band structure similar to that reported for $\beta$-PdBi$_2$. In an effort to explore the interplay between the superconducting and topological 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$_3$InAu$_2$) 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$_2$.

I. 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 5.4 K superconductor $\beta$-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 $\beta$-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 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 $\sim 2$ K) 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 $\beta$-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 1.8 K via electrical resistivity and magnetization measurements. These results are in contrast to earlier work [18] which indicated superconductivity in ScInAu$_2$ with a critical temperature of 3 K. Measurements reveal that only unannealed samples present the previously reported superconducting transition at 3 K, though the shielding in the magnetic susceptibility is incomplete. These results indicate that ScInAu$_2$ is not superconducting down to 1.8 K and that the previously reported critical temperature ($T_c$) of 3 K 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.

II. METHODS

Arc-melted samples were prepared by combining the raw elements in stoichiometric ratios and melting on a water-cooled copper hearth under Ar atmosphere. Each...
Intensity (arb. units)

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.

The crystal structures were characterized with powder x-ray diffraction (XRD) using a Siemens 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 $\sim 2$ K.

III. EXPERIMENTAL RESULTS

A. 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. Figure 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° (marked with an asterisk). Electrical resistivity measurements performed on the annealed sample (Fig. 2) show metallic behavior from room temperature down to the base temperature of 1.8 K with no indication of the superconductivity at 3 K 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.

Figure 3 shows the result of magnetic susceptibility measurements on unannealed ScInAu$_2$. The data show a clear drop in the susceptibility beginning slightly below 3 K. At the base temperature of 2 K 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 2 K (inset of Fig. 3) indicates $H_{c1} \sim 40$ Oe and complete flux expulsion by $\lesssim 150$ Oe. The critical field of In at 2 K is only 180 Oe, which is roughly consistent with our observations [20]. The low critical field indicates that the superconducting impurity is almost certainly unreacted elemental indium ($T_c = 3.4$ K). Though the $T_c$ observed here is somewhat lower that that of indium ($\sim 3.0$ K 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.

B. 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 $\sim 50$. No evidence for superconductivity is detected down to 1.8 K (see Fig. 4). The weak upturn in resistivity below $\sim 10$ K could be due to a Kondo effect arising from magnetic impurities.
FIG. 2. Resistivity versus temperature of ScInAu$_2$ down to 1.8 K. No indication of superconductivity is observed.

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 100 Oe are sufficient to suppress the superconductivity.

C. ScIn$_3$

Single crystals of ScIn$_3$ were grown with the molten flux method: 80:20 atomic % In:Sc were heated in an alumina crucible sealed in a quartz ampule under 70 torr Ar gas to 1000 °C and then cooled to 400 °C over 240 hours. After holding at this temperature of 8 hours, the ampule was centrifuged to remove the flux. This revealed small, cubic crystals, confirmed by x-ray diffraction to be cubic ScIn$_3$ [22].

Magnetic measurements on samples yielded a diamagnetic signal with an onset of around 3 K, but the shielding fraction of order 1%. Furthermore, a magnetic field of 0.05 T 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 0.78 K and 0.71 K in YIn$_3$ and LaIn$_3$ respectively [23], suggesting that ScIn$_3$ probably becomes superconducting below 1 K.

D. Sc$_2$InAu$_2$

Samples of Sc$_2$InAu$_2$ were synthesized by arc melting. The tetragonal Mo$_2$FeB$_2$-type structure [24] was confirmed by x-ray diffraction, though some unidentified secondary phases were present. Nonetheless, magnetic susceptibility measurements from 2-300 K presented no evidence for superconductivity or any other anomalies.

IV. CONCLUSIONS

The previously reported superconducting behavior of ScInAu$_2$, a material that shares the same topological classification as $\beta$-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
$T_c \sim 3 \text{K}$ 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$_3$InAu$_2$ and found that they are all essentially non-magnetic non-superconducting metals with no anomalies in the resistivity or magnetic susceptibility down to 2 K.

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 $T_c = 3 \text{K}$ [18]. However, it is clear that there are a large number of known superconducting materials with non-trivial band structures awaiting further study.

**ACKNOWLEDGEMENTS**

Work on this project was supported, in part, by the National Science Foundation (NSF) via CAREER award DMR-1453752 (JJH), REU Program DMR-1852138 (GPM), DMR-1708410 (MWM), and DMR-1644779 (NHMFL), and the State of Florida. We thank G. R. Stewart for helpful conversations.

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