Observation of the Superconducting Proximity Effect from Surface States in SmB$_6$/YB$_6$
Thin Film Heterostructures via Terahertz Spectroscopy

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The ac conduction of epitaxially grown SmB$_6$ thin films and superconducting heterostructures of SmB$_6$/YB$_6$ are investigated via time-domain terahertz spectroscopy. A two-channel model of thickness-dependent bulk states and thickness-independent surface states accurately describes the measured conductance of bare SmB$_6$ thin films, demonstrating the presence of surface states in SmB$_6$. While the observed reductions in the simultaneously measured superconducting gap, transition temperature, and superfluid density of SmB$_6$/YB$_6$ heterostructures relative to bare YB$_6$ indicate the penetration of proximity-induced superconductivity into the SmB$_6$ overlayer; the corresponding SmB$_6$-thickness independence between different heterostructures indicates that the induced superconductivity is predominantly confined to the interface surface state of the SmB$_6$. This study demonstrates the ability of terahertz spectroscopy to probe proximity-induced superconductivity at an interface buried within a heterostructure, and our results show that SmB$_6$ behaves as a predominantly insulating bulk surrounded by conducting surface states in both the normal and induced-superconducting states in both terahertz and dc responses, which is consistent with the topological Kondo insulator picture.

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Introduction.—SmB$_6$ has long been identified as a mixed-valence Kondo insulator with an anomalous low-temperature resistance plateau that eluded explanation [1–4]. Following the discovery of topological insulators [5–8], it was proposed that this anomalous resistance plateau is due to topologically protected surface states, making SmB$_6$ the first topological Kondo insulator (TKI) [9–12]. Following this prediction, a flurry of experiments have investigated the basic features of such a TKI [13–29], yet despite the evidence in support of the TKI prediction, controversy has continued to surround SmB$_6$ [30–42]. Much recent work has therefore been dedicated to understanding experimental discrepancies and harmonizing results. Numerous studies have now highlighted common extrinsic issues with studies of bulk crystals, including subsurface cracks in polished bulk samples [41], aluminum inclusions in crystals grown by the aluminum flux method [43], residual bulk conduction attributed to one-dimensional crystalline dislocations [41,44–46], and localized metallic islands around sample impurities [47,48]. Furthermore, previous terahertz studies [49,50] of SmB$_6$ starkly diverged from dc transport by finding an anomalously large ac conductivity without evidence for surface states. These results created a confused picture of SmB$_6$ with radically different ac and dc behaviors that has been frequently invoked by both experimental and theoretical efforts [34,36,45,48]. However, these terahertz studies were performed using polished bulk crystals that may suffer from the confounding effects mentioned previously and may therefore be reporting extrinsic behaviors.

Whereas most experiments on SmB$_6$ have employed bulk crystals, it has recently become possible to grow high-quality epitaxial thin films of SmB$_6$ via sputtering [21,51,52], thereby avoiding the myriad extrinsic concerns with bulk crystals and circumventing issues [53] in comparing previous results achieved via the different bulk crystal growth methods. By forming thin-film heterostructures of SmB$_6$ with the isostructural BCS superconductor YB$_6$, perfect Andreev reflection has been observed at the surface of sufficiently thin SmB$_6$ overlayers via point-contact Andreev reflection (PCAR) spectroscopy [51]. These results indicate the presence of metallic surface states susceptible to the superconducting proximity effect in these epitaxially grown SmB$_6$ samples, and moreover indicate that these surface states are indeed topologically protected in accord with the TKI prediction. Such heterostructures are predicted to host topological superconducting states at the buried interface [8,54] and could be engineered to generate and manipulate Majorana modes to perform topological quantum computations [8,54]. However, such buried interface states are not accessible by standard
Notably, the conductance of the sample plateaus below 5 K in the sputtering chamber [51]. 100 nm layers of YB$_6$ is selected for the proximity-effect heterostructures. Altogether, we establish a straightforward and unified understanding of the intrinsic low-temperature conductance of SmB$_6$: in both the normal and induced-superconducting states, SmB$_6$ behaves as a predominantly insulating bulk surrounded by conducting surface states in both ac and dc, as expected under the TKI prediction.

**Results and discussion.**—Thin-film samples of SmB$_6$ are grown epitaxially on Si(001) substrates via sputtering [51]. In order to form a minimal-barrier interface with SmB$_6$, the isostructural BCS superconductor YB$_6$ is selected for the proximity-effect heterostructures. As the superconducting transition temperature $T_C$ of YB$_6$ is maximized in the case of mild boron deficiency [51], 100 nm layers of YB$_6$, are grown on Si(001) substrates via sputtering, which for convenience will be referred to as YB$_6$ throughout. Heterostructures of SmB$_6$/YB$_6$ are fabricated by growing a 20 nm or 100 nm SmB$_6$ overlayer sequentially atop 100 nm YB$_6$ samples in situ without breaking vacuum in the sputtering chamber [51].

Typical TDTS [55] data for the real conductance $G_1$ are shown for the 150 nm SmB$_6$ sample in Fig. 1(a). (Raw TDTS time trace data is provided in Fig. S1 of the Supplemental Material [56].) There are no pronounced spectral features across the reliable frequency range of $\sim$0.5–2.3 THz, though there is a mild Drude-like conductivity that decreases in prominence at lower temperatures. Notably, the conductance of the sample plateaus below 5 K across the entire spectral range. In order to compare the conductance between samples, the average of the spectrum is taken from 0.5 THz to 1.0 THz and shown in Fig. 1(c) for select temperatures. [See Fig. S4(a) of the Supplemental Material [56] for all temperatures.] At both 50 K and 280 K, the conductance increases linearly with sample thickness, consistent with bulk-dominated behavior, whereas the conductance is nearly independent of sample thickness at 2.7 K, consistent with surface-dominated behavior. The small amount of thickness dependence that remains at low temperature may be due in part to the limited number of samples available for study, but it may also result from a small residual bulk conductivity.

To assess the conductance across the temperature range and available sample thicknesses, we apply a two-channel model of the total conductance $G_{\text{tot}}$ [15,21]. One channel scales with thickness and is exponentially activated as a function of temperature, consistent with a bulk conductance $G_{\text{bulk}}$. The second is a temperature- and thickness-independent channel consistent with a surface conductance $G_{\text{surf}}$ resulting from both the upper and lower surface states. The two-channel model is thus given by

$$G_{\text{tot}}(T) = G_{\text{surf}} + G_{\text{bulk}}(T),$$

where $G_{\text{bulk}}(T)$ is the conductance at low temperature, $G_{\text{surf}}$ is the bulk conductivity at high temperature, $t_{\text{bulk}}$ is the thickness of the bulk conductance channel, $E^*_a$ is the characteristic activation energy of the bulk channel, $k_B$ is the Boltzmann constant, and $T_{HT}$ is the temperature at which the high-temperature conductivity is calculated. Since the measured low-temperature conductance is reasonably consistent across the thin films, in contrast to bulk...
The average fitted value of the bulk activation energy possibly due to the substrates being from different batches). Data above 50 K for the 50 nm sample proved unreliable. A strong fit to the data for the three samples (experimental Fig. 1(b), the two-channel conductance model provides a consistent fit to the data while extracting the conductance of superconducting behavior in the terahertz spectrum. By fitting structure are shown in Figs. 2(a), 2(b); in Figs. 2(c), 2(d); and in Figs. 2(e), 2(f), respectively, where the superconducting gap fitting until convergence, values of $\Delta_T \approx \Delta_0 \tanh(1.74\sqrt{T_C/T - 1})$. This provides strong evidence for surface conducting states in bare SmB$_6$ at low temperature and resolves the previous discrepancy between ac and dc conductance in SmB$_6$. Superconducting heterostructures of SmB$_6$/YB$_6$ are probed via the same TDTS method and compared to a thin-film sample of YB$_6$ ($T_C \approx 6.1$ K) with no overlayer of SmB$_6$. As all samples consist of 100 nm YB$_6$ and some thickness of SmB$_6$, each heterostructure is referred to by its SmB$_6$ thickness for convenience. Typical data for the bare YB$_6$, the 20 nm heterostructure, and the 100 nm heterostructure are shown in Figs. 2(a), 2(b); in Figs. 2(c), 2(d); and in Figs. 2(e), 2(f), respectively, where the superconducting low-temperature conductance $\tilde{G} = G_1 + iG_2$ is normalized by the normal-state conductance $G_N$ of the sample above $T_C$ at 10 K. Conductance data of this form are modeled by the Mattis-Bardeen formalism for the optical response of a BCS superconductor in the dirty limit below $T_C$ as the superconducting gap opens [57,58]. See the Supplemental Material for extended fitting details [56].

By simultaneously fitting the real and imaginary parts of the normalized conductance for a sample at a given temperature $T$, the superconducting gap $\Delta(T)$ at that temperature can be extracted for a given guess value of $T_C$. By taking an initial estimate of $T_C$ from the disappearance of superconducting behavior in the terahertz spectrum and repeating the simultaneous fitting for each temperature, as shown by the solid lines in Fig. 2, the temperature evolution of $\Delta(T)$ is extracted. For a BCS superconductor, this temperature evolution is approximated by [59]

$$\Delta(T) \approx \Delta_0 \tanh(1.74\sqrt{T_C/T - 1}). \quad (4)$$

By fitting $\Delta_0$ and $T_C$ to the extracted values of $\Delta(T)$, the guess value of $T_C$ can be updated. Thus, by iteratively performing the simultaneous Mattis-Bardeen fitting and BCS gap fitting until convergence, values of $\Delta_0$ and $T_C$ for each sample are extracted from the data. As $\Delta_0$ varies on both sides of the interface of proximity-effect heterostructures.
the heterostructure. However, the reductions observed in the heterostructures here vary only slightly, despite the thickness of the metallic layer for thin films, where the sample thickness to the proximity effect depends strongly on the thickness of the structures. The weak SmB$_6$ length, which was previously determined to be the value of $T_C$ expected for bare 100 nm YB$_6$ is fitted to permit measurement of the superfluid spectral weight by extrapolation of the fit down to zero frequency. The iterative method results in a high-quality fit, as shown for bare 100 nm YB$_6$. This result therefore implies that the dominant contribution to the conductivity is being induced in some portion of the SmB$_6$ heterostructures concords with the model of SmB$_6$ as consisting of metallic surface states surrounding an insulating bulk.

The measurement of the complex conductance in the superconducting heterostructures also affords a measurement of the superfluid spectral weight, indicating the temperature evolution of the superfluid density in the samples. The superfluid spectral weight can be extracted by two methods, which we will call the extrapolation and integration methods. The extrapolation method makes use of the fact that the superfluid spectral weight is given by

$$S_{\text{extr}}(T) = \lim_{\omega \to 0} \omega G_2^{\text{SC}}(\omega, T),$$

where $G_2^{\text{SC}}$ is the imaginary conductivity in the superconducting state. Extracting values of $S_{\text{extr}}$ for each temperature is accomplished by fitting to the linear portion of $\omega G_2^{\text{SC}}(\omega, T)$, as shown in Fig. 3(b), and extrapolating to zero frequency. The integration method directly calculates the loss of spectral weight when passing below $T_C$ according to

$$S_{\text{int}}(T) = \int_0^{\infty} d\omega (G_1^{\text{N}}(\omega) - G_1^{\text{SC}}(\omega, T)),$$

where $G_1^{\text{N}}(\omega)$ and $G_1^{\text{SC}}(\omega, T)$ are the real conductivity in the normal state and superconducting states, respectively.
Given the convergence of $G_1$ at high frequency, the upper limit of integration can be reasonably truncated to the limit of reliable data, as shown in Fig. 3(c), introducing only minor error. Figure 3(d) shows that while the integration method consistently yields a slightly larger value for the superfluid spectral weight, the two methods show reasonable agreement across the temperature range. There is a minor error. Figure 3(d) shows that while the integration of reliable data, as shown in Fig. 3(c), introducing only the limit of integration can be reasonably truncated to the limit of integration.

The temperature dependence of the superfluid spectral weight is given by [59,62]

$$S(T) = S(0) \frac{\Delta(T)}{\Delta_0} \tanh \left( \frac{\Delta(T)}{2k_B T} \right).$$  

Simultaneous fits of the data for both the extrapolation and integration methods are shown in Fig. 3(d), showing strong agreement across the temperature range. There is a clear decrease in the superfluid spectral weight between each sample. The decrease from bare YB$_6$ to the heterostructure is expected as a result of the superconducting proximity effect. However, whereas $\Delta_0$ is quite comparable between the heterostructures and shows a difference of just 5%, $S(0)$ shows a more significant decrease of 12% The minimal difference in $\Delta_0$ indicates that the proximity effect is predominantly confined to the same volume in both heterostructures, namely the surface states as identified above. The further reduction in $S(0)$ with increased SmB$_6$ thickness, however, may be attributable to very weak conducting states existing in the bulk [41,44–46]. As the superfluid spectral weight is not yet thoroughly explored in the literature, further work is warranted to understand the significance of this behavior.

To summarize, these results provide a simple and unified picture in concord with the TKI prediction: SmB$_6$ behaves as a predominantly insulating bulk surrounded by conducting surface states in both the normal and induced-superconducting states in both ac and dc conduction. Experimental explanations and theoretical speculations that invoked the previous anomalous ac response may need to be reconsidered in light of these findings. While a topologically trivial explanation for this behavior cannot be ruled out by measurements presented here, the previous observation of perfect Andreev reflection [51] in similar SmB$_6$/YB$_6$ heterostructures supports the topological origin.

Furthermore, the measurements presented here demonstrate that TDTS can provide an effective probe of superconducting states at the buried interface of these important superconductor–topological insulator heterostructures, providing a powerful new tool for the investigation of engineered topological superconducting systems. Looking forward, our methods can extend to other topological superconducting heterostructures with bulk-insulating topological insulators such as Bi$_2$Se$_3$ [63–65] and Sb$_2$Te$_3$ [66], where the proximity effect does not reach the sample surface yet remains active in the buried interface.

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