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Anisotropic superconductivity in La(O,F)BiSeS crystals revealed by field-angle dependent Andreev reflection spectroscopy

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ABSTRACT

From field-angle dependent Andreev reflection spectroscopy on single crystals of La(O,F)BiSeS, which belongs to the recently discovered $BiCh_2$ (Ch = S, Se) based layered superconductors, we found that the superconductivity in La(O,F)BiSeS is highly anisotropic. We measured a superconducting energy gap of 0.61 meV for current injected along *c*-axis at 1.5 K. Detailed temperature and magnetic field dependent studies of the gap also reveal the presence of unconventional pairing in La(O,F)BiSeS. We show that the observed anisotropic superconducting properties can be attributed to the anisotropy in the superconducting order parameter with a complex symmetry in superconducting La(O,F)BiSeS.

1. Introduction

One of the key structural features that is common to almost all high temperature superconductors is the existence of alternating superconducting layers separated by insulating spacer layers [1–8]. The classic examples are the high T_c cuprates [1–4,9] and the ferro pnictides [10–14] where superconductivity occurs in the CuO_2 and the FeAs layers respectively. The nature of the spacer layer, in fact, plays a prominent role in deciding the important superconducting parameters. Therefore, by chemically tuning the spacer layer a large number of new high T_c superconductors have been derived out of the known layered high T_c superconductors [15]. In the context of the cuprate and the ferro pnictide superconductors it has also been observed that owing to their layered structures comprising of layers with varying electronic properties, the superconducting properties are highly anisotropic and the pairing mechanism is unconventional [16–18]. It is believed that detailed understanding of the superconducting parameters in such systems will eventually lead to the discovery of superconductors with higher critical temperatures (Table 1).

Recently, a new class of layered superconductors comprising of alternate stacks of $BiCh_2$ (Ch = S, Se) layers separated by a number of spacer layers have been discovered. Some of the members of this new family of superconductors display exceptional richness in their phase diagram where even apparently antagonistic phenomena like superconductivity and ferro magnetism coexist [19,20]. Transport measurements in presence of pressure reveal that the critical temperature of the BiS_2 based superconductors increase with pressure signifying the importance of the crystal structure for the occurrence of superconductivity [21–23]. Based on such measurements and pressure dependent structural analysis it has been thought that superconductivity in this system might be governed by lattice expansion. However, the nature of superconductivity in this family of superconductors is an outstanding issue which must be addressed. In this paper from field-angle dependent point contact Andreev reflection (PCAR) spectroscopy [24] measurements on single crystals of La(O,F)BiSeS [25] we show that the pairing in these class of superconductors is unconventional and highly anisotropic (Table 2).

2. Experimental results and discussion

The point contact Andreev reflection spectroscopy measurements were carried out in a liquid helium based cryostat equipped with a 3-axis vector magnet (6T-1T-1T). A conventional needle-anvil method was used for forming ballistic point contacts on single crystals of La(O,F)BiSeS for energy resolved spectroscopy. The differential conductance dI/dV vs. V spectra were recorded by a lock-in modulation technique at 1.7 kHz. It should be noted that when the point contact experiments are performed in the non-ballistic regime of transport, certain artefacts related to the role of critical current may appear. Such artefacts could be sharp conductance dips appearing at voltage values characteristic of the critical current of a given point contact or giant zero-bias enhancement [26]. All the spectra that we have presented in the manuscript are free from such artifacts (Table 3).

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Table 1
The fitting parameters for the spectra shown in Fig. 1(a).

Parameters	Δ (meV)	*Z*	*Γ* (meV)
T=1.51 K	0.61	0.550	0.190
T=1.71 K	0.61	0.572	0.190
T=1.88 K	0.59	0.590	0.174
T=2.07 K	0.52	0.645	0.154
T=2.33 K	0.44	0.720	0.110
T=2.68 K	0.38	0.790	0.037
T=2.90 K	0.28	0.950	0.0001

Table 2
The fitting parameters for the spectra shown in Fig. 2(a) and (b).

Parameters	Δ (meV)	*Z*	*Γ* (meV)
H (∥ c) = 0.0 kG	0.61	0.550	0.217
H (∥ c) = 2.0 kG	0.56	0.586	0.250
H (∥ c) = 3.5 kG	0.48	0.695	0.250
H (∥ c) = 5.0 kG	0.40	0.800	0.240
H (∥ c) = 8.0 kG	0.28	0.995	0.200
H (∥ a) = 0 kG	0.61	0.558	0.210
H (∥ a) = 4 kG	0.55	0.558	0.220
H (∥ a) = 6 kG	0.48	0.600	0.220
H (∥ a) = 8 kG	0.40	0.650	0.220
H (∥ a) = 10 kG	0.34	0.705	0.205

Table 3
The fitting parameters for the spectra shown in Fig. 3(a) and (b).

Parameters	Δ (meV)	*Z*	*Γ* (meV)
$\theta = 0°$	0.59	0.572	0.262
$\theta = 30°$	0.58	0.565	0.220
$\theta = 60°$	0.57	0.545	0.174
$\theta = 90°$	0.56	0.540	0.174
$\theta = 120°$	0.57	0.558	0.225
$\theta = 150°$	0.58	0.582	0.265
$\theta = 180°$	0.59	0.576	0.263
$\phi = 0°$	0.59	0.550	0.222
$\phi = 30°$	0.59	0.548	0.214
$\phi = 60°$	0.59	0.560	0.245
$\phi = 90°$	0.59	0.555	0.230
$\phi = 120°$	0.59	0.550	0.220
$\phi = 150°$	0.59	0.552	0.230
$\phi = 180°$	0.59	0.552	0.230

In Fig. 1(a) we show temperature dependent PCAR spectrum between La(O,F)BiSeS and a metallic tip of Ag (dotted lines). Theoretical fits to the spectra using modified Blonder-Tinkham-Klapwijk (BTK) theory [27] are also shown as solid lines. Modification to the BTK theory [28] was done to incorporate an inelastic broadening parameter (*Γ*) that takes care of broadening of the spectra.

For lower temperature spectra, as it is seen, with a single gap BTK formula the lower bias part of the spectra fit well while a deviation is seen at higher bias. This deviation could be attributed to the existence of low-bias phonon modes [29] or multiple superconducting gaps [30–35]. In fact, additional spectral features are observed on superconductors like MgB_2 [36,37] where access to both gaps is possible by injecting current along a specific crystallographic direction. The qualitative shape of such spectral features are different from the features we observe here. On the other hand, at relatively higher temperatures, the additional features disappear and the spectra fit very well with the modified BTK theory. This unique temperature dependence is not observed for MgB_2 where the features associated with both the gaps evolve smoothly before disappearing at the critical temperature.

In Fig. 1(b) we show the temperature dependence of the superconducting energy gap as extracted from the data presented in

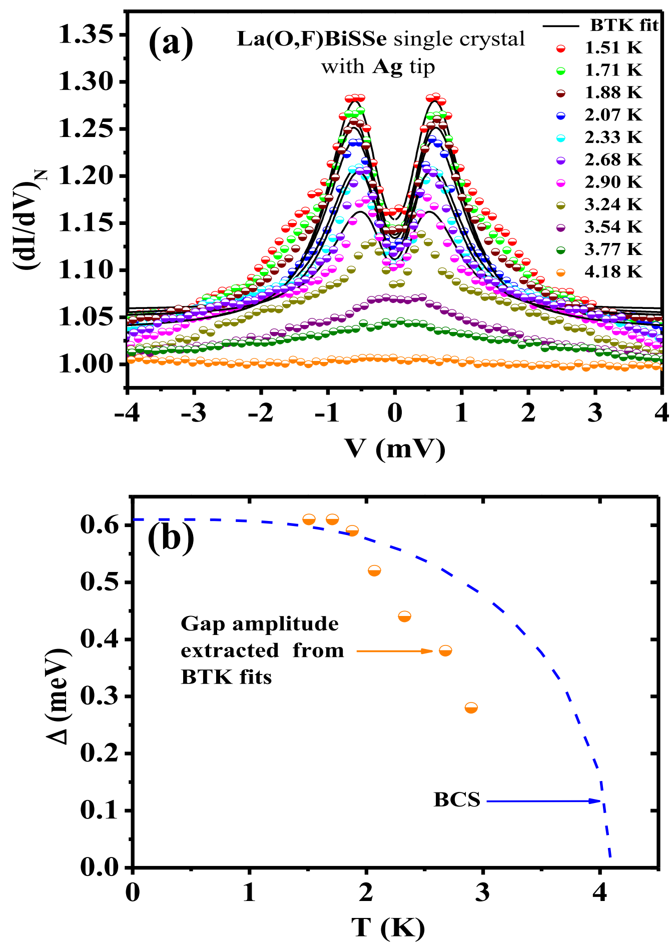

Fig. 1. (Color online) (a) Point contact spectra (dots) with BTK fit (solid line) at different temperature below T_c = 4.2 K obtained on La(O,F)BiSeS single crystal using silver (Ag) tip. Observed double peak structure symmetric about V = 0 is a hallmark of Andreev reflection. The superconducting energy gap amplitude (Δ) extracted by BTK fit is 0.61 meV. (b) The temperature dependence of superconducting gap (orange dots) extracted from BTK fit do not follow conventional BCS behaviour.

Fig. 1(a). The dotted blue line in Fig. 1(b) shows the expected temperature dependence for a conventional BCS superconductor. It is clearly seen that the experimentally measured temperature dependence deviates significantly from the BCS prediction [38]. Therefore, it can be concluded that the superconductivity in La(O,F)BiSeS is unconventional in nature. Such observation of an unconventional pairing is consistent with the recent theoretical prediction of the possibility of unconventional superconductivity in BiS_2-based superconductors. [39]

In order to gain further understanding of the nature of superconductivity in La(O,F)BiSeS we have performed Andreev reflection spectroscopy at different magnetic fields applied along different crystallographic directions. Fig. 2(a) shows the magnetic field dependence of the Andreev reflection spectra when the magnetic field is applied along the *c*-axis ($H \parallel c$) of the crystal. The dotted lines show the experimental data points while the solid lines are the theoretical fits. In contrast to the temperature dependent measurements it is seen that at finite magnetic fields, the spectra fit very well with the modified BTK theory. This indicates that the deviation of the data from the fit seen in the zero-field spectrum might be due to the presence of additional superconducting gaps which get suppressed in small magnetic fields. The superconducting energy gap systematically decreases with increasing magnetic field. Though the gap is not clearly resolved, the features associated with superconductivity survive at higher fields up to 3 Tesla (in order to keep the panel uncluttered we have not shown the spectra above 8 kG) which can be considered to be the critical field of the