A non-contact mutual inductance based measurement of an inhomogeneous topological insulating state in Bi$_2$Se$_3$ single crystals with defects

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**Abstract:** Pure Topological Insulating materials preserve a unique electronic state comprising of bulk insulating gap and conducting surface states. Here we use bulk Bi$_2$Se$_3$ single crystals possessing Se vacancy defects as a prototype topological insulator (TI) material for exploring the effect of non-magnetic disorder on the conducting properties of TI’s. We employ a sensitive, non-contact, mutual inductance based technique for measuring the surface and bulk contribution to electrical conductivity in the TI. We discern the different contributions, by observing that predominant surface electrical conduction produces linear frequency dependence of the pickup signal while bulk conductivity gives rise to quadratic frequency dependence. We also see an algebraic temperature dependent surface conductivity while an activated bulk conductivity. Using the above we uncover an interplay between surface and bulk contribution to electrical conductivity in the TI as a function of temperatures. In the Bi$_2$Se$_3$ crystals the transformation from surface to bulk dominated electrical transport is found to occur close to 70 K. This temperature range matches well with our results from activated bulk electrical transport results which shows an activation energy scale, $\Delta$ which is in the milli-eV range. The gap $\Delta$ is much less than the bulk band gap in Bi$_2$Se$_3$, and which we argue is associated with defect states in the TI material. To understand our results, we propose a model of a TI comprising of an inhomogeneous low electrically conducting medium (bulk) which is sandwiched between thin two high electrically conducting sheets (surface). The inhomogeneous TI state we argue is generated by Selenium vacancies defects in Bi$_2$Se$_3$, which is responsible for producing an interplay between bulk and surface conductivity.

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Introduction:

In recent times, new materials like three dimensional (3D) topological insulator, exhibit topologically protected bulk gapped state enclosed by conducting surface state [1,2,3,4,5,6]. Herein no continuous variation of the lattice parameter can change the unique character of the electronic states in these materials. These gapless surface states in a topological insulator (TI) exhibit, Dirac-like linear energy - momentum dispersion [7,8], chiral spin texture [5] and Landau level quantization [9]. Materials like Bi$_2$Se$_3$, Bi$_2$Te$_3$ and Bi$_{1-x}$Sb$_x$ are amongst the most well studied three dimensional TIs [2,7,10]. The characteristic TI properties have been confirmed via different techniques like angle-resolved photoemission spectroscopy (ARPES) [3,5,11,12], Shubnikov-de Haas (SdH) oscillations, [13,14] scanning tunneling microscopy (STM) and transport studies [2,15]. Spin momentum locking in TI causes chiral currents to flow which are unaffected by scattering from disorder [1,2,15,16]. Such properties make TI lucrative materials for spintronics [17] and quantum computation [18,19] applications. However, it is important to evaluate how the TI state is affected in the presence of material defects. While presence of magnetic impurities will lead to breaking time reversal symmetry and hence destroy the TI state, the issue of how non-magnetic disorder affects the TI state is worth investigating.

To study the effect of disorder the popular Bi$_2$Se$_3$ crystals are a suitable choice, as they intrinsically possess Se vacancies which is a source of disorder in the material. The TI nature of Bi$_2$Se$_3$ has been identified through magneto-transport studies which confirm the presence of conducting surface states through the observation of SdH oscillations and weak anti-localization effects [16,20,21,22,23,24]. In Bi$_2$Se$_3$ the bulk insulating gap has been estimated to be ~ 300 meV [7,10,25,26]. However the presence of Se vacancies electron dopes the material [27,28,29], thereby enhancing the bulk electrical conductivity. The resulting parallel conducting channels through the surface and bulk, makes it difficult to discern the individual bulk and surface state contributions to electrical conductivity in these TI materials [6,30,31]. In such materials, the surface states have been identified either by suppressing defect induced bulk conductivity through counter doping [7,15] or by comparing changes in the conducting properties as a function of the thickness of TI
thin films [20,21,22,32,33]. Without counter doping, the issue of investigating how Se vacancies affects the TI state in Bi$_2$Se$_3$ single crystals is none the less important. Studies show that increasing Se vacancy concentration weakens the SdH oscillations, which is a fingerprint of the conducting surface states characterizing a TI [20-23,32,33,34,35]. The SdH oscillations in magneto-transport are typically measurable at low temperature and high magnetic field regime. Hence, due to the limitations of the transport studies mentioned above, while the presence of disorder like Se vacancies seems to weaken the TI state. The details of how exactly the surface and bulk conductivities are affected by disorder and how these state are modified by Se vacancies still remains unclear. Furthermore, studies have shown that due to degradation of surface of Bi$_2$Se$_3$, for example due to oxidation of the surfaces, produces band bending of the surface states extending upto 20 nm into the bulk [36,37]. Hence, a need is strongly felt for a technique which distinguishes between bulk and surface contributions to electrical conductivity in a TI. We report here a non-contact measurement technique of shielding currents induced in a TI using a two coil mutual inductance setup. We study the frequency ($f$) and temperature ($T$) dependence of the pickup voltage, from five single crystals of different thickness of Bi$_2$Se$_3$ placed between the coils. The pickup voltage shows two distinct regimes of frequency dependence, viz., a quadratic and linear regime. We show that the quadratic frequency dependence is associated with bulk electrical conductivity in the TI while the linear frequency dependence regime is associated with surface conductivity in the TI. As a function of temperature we show that below 70 K, the frequency dependence of the pickup voltage is predominantly linear while it turns quadratic at higher $T$. Analysis of the data also shows that below 70 K we identify an algebraic temperature dependent electrical conductivity regime which saturates at low temperatures, while above 70 K electrical conductivity is of thermally activated type with a thermal activation energy scale ($\Delta$) of tens of meV. From our study we identify four distinct temperature regimes for the Bi$_2$Se$_3$ crystal, identifying the crossover between different surface and bulk dominated conductivity regimes. At high temperature above 180 K, we observe the unusual return of surface dominated conductivity coexisting with bulk conductivity in the material. To understand our results, we propose a simplified model of a TI, comprising of a low conducting bulk sandwiched between two high conducting surface sheets. Our simulations show surface and bulk conductivity values in the Bi$_2$Se$_3$ crystals to be of the order of $10^{11}$ S/m and $10^3$ S/m, respectively. While this minimal model explains the overall features of the data and offers a way to estimate of the surface and bulk conductivity.
electrical conductivities of the TI from the measurements, it doesn’t match the data exactly. A better agreement with the data is obtained by considering an inhomogeneous TI state. Inhomogeneous TI is modelled by incorporating in the minimal model, conducting channels threading the low conducting bulk medium. The model fits our experimental data by introducing by inhomogeneity 30 percent. The inhomogeneous TI state we argue is a result of disorder in the TI bulk, generated by Selenium vacancies. Excess charge carriers produced by Se vacancies produce the gap $\Delta$, which results in an interplay between bulk and surface conductivity.

**Experimental details: Transport measurement of Bi$_2$Se$_3$ single crystals.**

For our study we use single crystals of Bi$_2$Se$_3$ prepared by slow cooling stoichiometric melts of high purity bismuth (Bi) and selenium (Se) powders [for details see refs.34,35]. In this work we have investigated the surface and bulk conductivities across five different single crystals belonging to the same batch of Bi$_2$Se$_3$ crystals grown (with similar electrical transport characteristics, for example see Fig.1). The thickness (surface area) of the crystals are, 20 $\mu$m (2.4 mm × 1.9 mm) (hence forth referred to as S20 sample), 51 $\mu$m (S51) (2.8 mm × 2.5 mm), 69 $\mu$m (S69) (3.9 mm × 2.5 mm), 75 $\mu$m (S75) (3.7 mm × 2.6 mm) and 82 $\mu$m (S82) (3.2 mm × 2.8 mm). The electrical transport measurements shown in Fig.1 are on a Bi$_2$Se$_3$ single crystal with thickness of 70 $\mu$m. Note that as transport measurements involves making of electrical contacts on the sample which introduce irreversible physical changes in the sample, hence for our non contact measurements (which is the main topic of our manuscript) and transport measurements (here and refs.[34,35]), we are compelled to use different samples for the two measurements, albeit chosen from the same batch of single crystals grown with similar sample thickness. Figure 1(a) shows distinct SdH oscillation in longitudinal magneto-resistance ($R_{xx}$ vs magnetic field ($B$)) measurements at different temperature using standard Van der Pauw geometry. The low temperature SdH oscillations which weaken with increasing $T$, indicates the topological character of conducting surface states in TI [13,14,20,21,22,34,35] (more detailed analysis of the transport data is shown elsewhere [34,35]). Figure. 1(b) shows the Hall resistance ($R_{xy}$) measurement as a function of magnetic field ($B$) at different temperatures. This measurement establishes electrons as the charge carriers in these Bi$_2$Se$_3$ samples. Such electrical transport studies have shown that Se vacancies weaken the SdH oscillations [34,35]. Recent studies show a correlation between Positron annihilation lifetime with Se vacancy concentration in Bi$_2$Se$_3$. These studies, performed on this batch of samples show Se
vacancy concentration are in the range of 10^{17} \text{ cm}^3[34]. From these SdH measurements at low \( T \) and high \( B \) it is however difficult to determine the extent of surface and bulk contributions to conductivity, and hence study how it is affected by either \( T \) or disorder variation present either in on the surface or bulk of the material. As mentioned earlier the appearance of finite bulk conductivity in Bi$_2$Se$_3$ sample with Se vacancies mixes the surface and bulk contributions to electrical conductivity making it difficult to discern the individual contributions. In the next section, we discuss a non-contact measurement technique which allows us to distinguish between the two contributions to electrical conductivity.

\[
\Delta R_{xx} = a\sqrt{0.011B}\left(\frac{11.12}{\sinh(\frac{11.12}{B})}\right) e^{\frac{19.38}{B} (0.95)\cos \left[2\pi \left(\frac{F}{B} + \beta \right)\right]}.
\]

In this equation \( a = 0.0135 \Omega \) and \( F \) and \( \beta \) are the fitting parameters. A fit to the data (see black solid line in Fig. 1(a)), gives \( F = 46.95 \pm 0.25 \text{ T} \) and \( \beta = 0.43 \).

(b) Hall measurement of the transverse resistance, \( R_{xy} \) vs \( B \) at different temperatures. The sample used in the above measurements has thickness \( \sim 70 \mu\text{m} \).

**Two coil mutual inductance measurement setup: Non-contact measurement**
To characterize the surface and bulk states of TI we use a non-contact modified two coil mutual inductance measurement technique [38,39,40]. Note that whereas in a conventional electrical transport setup, Joule heating effects complicate temperature dependent conductivity measurements, here such issues are avoided through our non-contact two-coil mutual inductance measurement technique. Figure 2(a) shows the schematic of our setup where a crystal is placed between an excitation coil and a pickup coil. Both coils are designed with very closely matched coil parameters (for details on coil dimensions and other parameters see supplementary information, S1). The data for all our pickup measurements corresponds to AC current of amplitude 153 mA applied to the excitation coil (corresponding to a 1Volt drop across the coil) at frequency \( f \), and the real and imaginary components of the pickup voltage from the pickup coil is measured using a lock-in amplifier. Measurements at higher AC excitation current amplitude, yield similar results. In the supplementary section S1 we show the linear relationship between the excitation current in the coil and the pickup voltage for an S20 sample at 65 kHz excitation frequency. The excitation current generates a time-varying magnetic field, which induces currents inside the conducting sample, that in-turn leads to a time-varying magnetic field associated with the sample. This local field induces a voltage in the pickup coil, which is being measured. Effectively one considers that the presence of a sample between the two coils modifies the mutual inductance. Variations in the sample properties change the signal induced in the pickup coil. One may note that voltage is induced in the pickup coil not only from time-varying magnetic fields associated with the sample, but also the via the stray magnetic fields present outside the sample. These stray field induced pickup voltage signal can often lead to a large background signal which masks the signal from the sample if it's weak. In order to significantly reduce this stray field effect between the dipolar coil assembly, a 1.5 mm thick oxygen-free high thermal conductivity Cu (OFHC) sheet (resistivity \( \sim 1.7 \times 10^{-8} \, \Omega \cdot m \)) with a hole at its center (see Fig. 2(a)), is placed coaxially above the excitation coil. The hole diameter is chosen such that it doesn’t exceed the sample surface dimensions (the sample is placed above the Cu sheet covering the hole). Note that the OFHC Cu sheet thickness (1.5 mm) is larger than its skin depth, which at 60 kHz is 0.27 mm and at 5 kHz is 0.92 mm. The high conductivity thick OFHC Cu sheet shields the alternating magnetic field generated by the excitation coil, except over the hole. The Cu sheet reduces the stray flux linkage outside the sample, while the hole in the sheet concentrates the magnetic flux around the sample. Therefore, the Cu sheet with the hole effectively enhances the coupling of the
two coils via the sample in between and the effects of the stray field is minimized. In Figs. 2(b) and 2(c), the colored regions represent the simulated vertical (z) component of the AC magnetic field distribution ($B_z$) around excitation coil (for simulation details, see S2 section in supplementary information). Figure 2(c) shows the concentration of magnetic flux above the hole in the Cu plate. In Fig. 2(d), the simulated $B_z$ profile measured above the coil (black dashed line in Fig. 2(c)), shows the significant concentration of magnetic flux above the hole in the Cu sheet. The sample when placed above the Cu sheet experiences this concentrated oscillating magnetic field and its response is coupled to the pickup coil voltage. During the measurements with TI we balance out the imaginary part of the signal as it doesn’t change significantly during measurements. Note that for all our measurements we subtract the background voltages measured without a sample placed between the coils and the Cu sheet with a hole.

FIG. 2. (a) Schematic diagram of the two coil mutual inductance setup is shown. (b) & (c) Show the simulated normal component of the AC magnetic field ($B_z$) distribution for the two cases. The outline of the solid excitation coil bobbin is shown in yellow. In Fig. (b) only a solid Cu sheet (orange) is placed above the excitation coil and in Fig. (c) a Cu Sheet with a hole at the center is placed over the coil. The sample is placed above the hole in the Cu sheet. These simulations are performed using Comsol with 150 mA, AC current at 60 kHz sent through the excitation coil. (d)
Shows the simulated $B_z$ vs $r$ profile as measured across the black dashed line shown in Fig. (c) ($r = 0$ is the central (z) axis of the excitation coil bobbin), for three different cases: (i) without any Cu sheet above the pickup coil, (ii) Cu sheet with a hole, (iii) Cu sheet with no hole. The inset of Fig. (d) shows the schematic of the circular Cu sheet with a hole placed over the top of the excitation coil. (e) The superconducting transition of Iron Pnictide single crystal, BaFe$_{1.85}$Co$_{0.15}$As$_2$ is shown. The figure depicts the behavior of the real (left axis) and imaginary (right axis) component of the pickup voltage as a function of temperature. The BaFe$_{1.85}$Co$_{0.15}$As$_2$ crystal dimension is 3.2 mm × 2.4 mm × 0.5 mm. The $T_c$ is indicated as the green dashed line $\sim$ 24.7± 0.2 K. A similar $T_c$ value of this sample is observed through bulk magnetization measurement on a SQUID magnetometer.

We test the performance of the setup using a superconducting sample as its shielding response is well known. Figure 2(e) illustrates the performance of our setup by measuring the AC susceptibility response of a superconducting single crystal, viz., an optimally doped Iron Pnictide crystal (BaFe$_{1.85}$Co$_{0.15}$As$_2$) which is placed above the hole in the Cu sheet. Due to strong superconducting diamagnetic shielding of AC magnetic field, (Fig. 2(e)) we see the rapid drop in the in phase signal in the pickup coil ($V'$) at $T < T_c$. Near $T_c$, we also see the expected peak in the out of phase signal ($V''$). The superconducting transition temperature $T_c$ estimated from the peak position in $V''$ is 24.7 K which compares well with $T_c$ reported for this stoichiometry [41,42].

**Studying the shielding response of thin Bi$_2$Se$_3$ single crystal at different $f$ and $T$:**

Figure 3(a) shows the $V(f)$ for the Bi$_2$Se$_3$ sample (S20) at 140 K, where $V(f) = \sqrt{V'^2(f) + V''^2(f)} \approx V'(f)$, as for these TI crystals, the $V''(f) < V'(f)$. One can identify two regimes of behavior in $V(f)$, namely, one in the low frequency regime where $V(f) \propto f^2$ (see red dotted line through the data), and the other in the higher frequency regime where, $V(f) \propto f$ (see black solid line to the data). Inset of Fig. 3(a) shows the $V(f)$ data replotted in a normalized log-log scale, showing the change in slope from quadratic to linear behavior. In the inset, the slope of the red line (quadratic regime) is 1.76 ± 0.05 and black line (linear regime) is 0.88 ± 0.01. To understand the source of change in curvature of $V(f)$, we measure the pickup voltage response at fixed $f$ with varying $T$. Figure 3(b) shows the $V(T)$ behavior in S20 sample at two different frequencies of 5 kHz and 65 kHz (similar $V(T)$ behavior for S20 at other frequencies is shown in supplementary information (see S3 section in supplementary information)). The 65 kHz data
shows the pickup voltage is nearly constant upto 35 K after which decreases with increasing
temperature and at a higher temperature (> 180 K) the $V(T)$ again becomes nearly constant at ~
0.054 mV. However, at 5 kHz the voltage profile exhibits a completely different behavior
compared to 65 kHz data, the $V$ increases exponentially with $T$ and saturates beyond 90 K. The 5
kHz and 65 kHz data merge at a higher temperature. The frequency dependence of $V(f)$ at low $T$
in Fig. 3(c) is striking, it shows the $V(f) \propto f$ over the entire frequency range. Measurements at
other AC excitation current amplitude yield similar results (see S4 section in supplementary
information). Figure 3(d) shows that from 50 K, the nonlinear $V(f) \propto f^2$ dependence gradually
develops in the lower $f$ range while at a higher frequency range still a linear frequency
dependent regime is maintained. Therefore, below 50 K, $V(f)$ has a predominantly linear dependence on $f$
while at higher $T$ above 50 K $V(f)$ is an admixture of quadratic and linear dependence on $f$.

FIG. 3. (a) Variation of pickup voltage ($V$) with frequency for Bi$_2$Se$_3$ single crystal (S20) at 140 K. The inset shows
$V/V_0$ vs $f/f'$ in log-log scale. For normalizing the signals, we use $V_0 = 1$ V which is the amplitude of the AC voltage
generated across the excitation coil and $f' = 65$ kHz. All data in the figures correspond to an excitation current of 153 mA in the excitation coil of the setup. Two lines show two distinct regions. Red dotted line is used for the bulk dominating fitting and black line is used for the surface part fitting. We have maintained the same color and style throughout the paper. (b) Variation of pickup voltage ($V$) with temperature ($T$) for S20 at two frequencies 5 kHz and 65 kHz. In 65 kHz data, $V \propto \frac{1}{(C' + D'T)}$ is fitted where $C' = 0.5884$ mV$^{-1}$, $D' = 0.772$ mV$^{-1}$K$^{-1}$ and $V \propto V_{b0} \exp \left( -\frac{\Delta}{K_bT} \right)$ is fitted in 5 kHz data where $\Delta = (5.77 \pm 0.45)$ meV and $V_{b0} = 0.15$ mV. (c) Frequency variation of $V$ is shown for two temperatures 15 K and 30 K for the same sample. Solid green line is the best fit line through the data points, which shows a linear behavior at low temperatures. (d) $V$-$f$ behavior is shown at higher temperature (50 K & 80 K). Here quadratic and linear fittings are shown with red dash line and black solid line respectively.

**Understanding the frequency dependence of the pickup signal:**

To understand the above frequency dependence of the pickup voltage, we recall that AC magnetic field produced from the excitation current in the primary induces screening currents extending upto different depths inside the conducting TI sample (skin depth). The magnetic field generated by these induced screening currents couple with the pickup coils to induce a pickup voltage. It is known that the depth upto which the currents are induced in the sample depends on the frequency of AC field. At low frequencies as the AC excitation penetrates deeper into the bulk of the sample (due to large skin depth), hence the bulk properties in the TI sample are probed with low $f$. The schematic of the distribution of the magnitude of induced screening current ($I_{\text{induced}}$) across the sample cross section is shown in Figs 4(a) and 4(b) for low frequency and high frequency respectively. The expression for skin depth ($\delta$) is $\frac{1}{\pi \sigma f \mu}$, where $\mu$ is permeability of Bi$_2$Se$_3$. At high frequency, screening currents circulate within the high conductivity surfaces (effective skin depth associated with high conductivity surface state at high frequencies is small, discussed later in the section on simulation of the mutual inductance behavior) hence the properties of the surfaces state of the TI is probed at high $f$. In Fig. 3(b) the temperature dependence of $V$ at 65 kHz is related to the $T$ dependence of screening currents induced in the surface of the TI, while the 5 kHz data corresponds to probing the temperature dependence of screening currents set up in the bulk of the sample.
FIG. 4. (a) Shows the schematic of the magnitude of the induced current distribution across the sample cross-section at a lower frequency. The induced current is shown to flow across the entire volume of the sample. The AC excitation magnetic field in the primary is shown in blue while the field induced in the pickup coils through the currents induced in the sample is shown in red. A schematic plot of the induced current profile across the sample cross-section at different \( f \) is shown. For the sake of simplifying the calculation, the current distribution across the bulk is considered to be uniformly distributed throughout the sample cross-section (b) Shows at a higher frequency, the nature of the induced current distribution which flows only in TI’s high conducting surface states. The nature of the excitation field in the excitation coil (blue) and the induced field in the pickup coil (red) are also shown.

To explain the pickup data, we use the following model: The AC current in the primary excitation coil produces an AC magnetic field \( B = B_0 e^{-i\omega t} \), where \( \omega \) is the angular frequency of the AC signal and \( B_0 \) is the amplitude of the AC magnetic field. For simplicity the sample is assumed to be cylindrical shape with radius \( r \). Due to this AC magnetic field, the current density induced in the sample is \( \vec{J}(r, z) = \sigma I e^{-i\omega t} \Phi(z) \phi \), where \( \sigma \) is total conductivity of the sample and \( \Phi(z) \) is the magnetic flux passing through the sample. The induced current will generate varying magnetic field sensed by the pickup coil. The pickup voltage is given by (see S5 section in supplementary information for detailed calculation)

\[
V_{\text{output}}(\omega) = \xi(r, z) \Phi \omega^2 e^{-i\omega t} \left[ \frac{\sigma_{\text{ov}}}{C + DT} + \frac{\sigma_{\text{ob}}}{e^{\Delta/\kappa T}} \right] \frac{1}{1 + i\omega \tau} \tag{1}
\]
where $\zeta(r,z)$ is a geometric factor which is a function of the pickup coil radius ($r$), the height between the two coils ($z$). To understand the frequency and temperature dependence of $V$, we use the parallel transport channel model [20,21] for TI, where the total electrical conductivity ($\sigma_t$) of the TI is $\sigma_t = \sigma_s + \sigma_b$, where $\sigma_s$ is the surface conductance and $\sigma_b$ is the thermally activated bulk conductance of the bulk. We use the known temperature dependence of the surface ($\sigma_s$) and bulk ($\sigma_b$) conductivities of TI, viz., $\sigma_t = \frac{1}{(C + DT)}$, where $C$ is related to static disorder scattering and $D$ to electron-phonon coupling strength and $\sigma_{b0}$ is the high temperature conductance of the bulk state. Assuming Drude’s AC conductivity [43] for a TI we calculate the frequency dependence of the pickup voltage, $V(f)$ in the low and high frequency regime (see supplementary information section S5 for details). In the low frequency limit of the AC excitation ($\omega = 2\pi f$), the alternating magnetic field penetrates the bulk of the sample. Therefore, the induced current ($I_{\text{induced}}$) is flowing throughout the sample volume (see schematic of the magnitude of $I_{\text{induced}}$ distribution across sample cross-section in Fig. 4(a)). For conduction through the bulk of the TI the $\tau$ (average collision time interval of electron with disorder site) is expected to be small due to large probability of collision, hence at low $\omega$ for small $\tau$ values, the approximate condition $\omega \tau \ll 1$ can be used. It can be shown that with this condition in Eq. 1 one gets (see supplementary S5 section for details), $V(f) \propto f^2$ for low frequencies (namely when response is associated with conduction through the bulk of the TI)

$$V_{\text{output}}(\omega) \approx \Phi_0 \zeta(r,z) \omega^2 e^{-\text{tot} \sigma_b}$$ [2]

In the high frequency ($\omega$) range as the AC field penetrates mainly the surface of the sample, the pickup signal is associated with the response from currents induced close to the surface of the TI. Therefore, induced current ($I_{\text{induced}}$) is flowing at the surface only (a schematic of the magnitude of the distribution of $I_{\text{induced}}$ across TI sample cross-section is shown in Fig. 4(b)). Recall the chiral spin nature in TI [44,45] leads to very small collision rates and hence a large $\tau$. Therefore for high $\omega$, one has a condition $\omega \tau \gg 1$, due to which from Eq. 1 we get $V(f) \propto f$ for the surface state of a TI, viz., (see supplementary S5 section for details)

$$V_{\text{output}}(\omega) \approx \Phi_0 \zeta(r,z) e^{-\text{tot} \sigma_s} \omega$$ [3]
Figure 3(b) shows Eq. 2 fits the 5 kHz $V(T)$ data (circles), by using a temperature dependence of the bulk conductivity of the form, $\sigma_b(T) = \sigma_{b0} \exp\left(-\frac{\Delta}{K_B T}\right)$. The best fit gives an activation barrier $\Delta \sim 5.77 \pm 0.45$ meV for S20. Figure 3(b) shows Eq. 3 fits the high $f$, 65 kHz $V(T)$ data (squares) by using a temperature dependent conductivity of the surface states of the form, $\sigma_s(T) = \frac{1}{(C + DT)}$. The linear frequency dependence of the pickup voltage in Eq. 3, suggest the features shown in Fig. 3(c) is dominated by high conducting surface state of TI. At high $T$ above 50 K, Fig. 3(d) shows an admixture of both surfaces ($V(f) \propto f$) and bulk ($V(f) \propto f^2$) response. It is interesting to note that only in an intermediate $T$ regime the $V(f)$ shows significant temperature dependence. The temperature independent $V(T)$ behavior below 40 K suggests that in this surface conducting regime ($V(f) \propto f$), $\sigma_s(T) = \frac{1}{(C + DT)}$ has $C >> D$ below 40 K. Similar saturation of surface conductivity at low temperatures have been reported in the past for transport measurements [20,21,30,32]. In Fig. 3(b) (5 kHz data) above 160 K, we see that rather than obeying the exponential thermally activated behavior (dashed red curve) the $V(T)$ behavior tends to saturate. At high $T$ as the electrical conductivity of the material is low combined with low frequency, the skin depth becomes large and becomes comparable to the sample thickness. In such a situation one measures only an averaged bulk response of the sample (this issue will be discussed in greater detail in the section on simulating the mutual inductance behavior).

**Studying the shielding response of thicker Bi$_2$Se$_3$ single crystal at different $f$ and $T$:**
FIG. 5. (a) Variation of pickup voltage ($V$) with temperature ($T$) for sample S69 at frequency 65 kHz. Red dotted line is the fitted line corresponding to bulk conductivity, viz., $V \propto V_{b0} \exp\left(-\Delta/\kappa_b T\right)$ and the black line is fitted with surface conductivity, i.e., $V \propto 1/(C' + D'T)$. The fitting parameters are $C' = 0.0052 \text{ mV}^{-1}$, $D' = 1.86 \text{ mV}^{-1}\text{K}^{-1}$, $V_{b0} = 18.7 \text{ mV}$ and $\Delta \approx 25.2 \pm 1.25 \text{ meV}$. (b) The behavior of pickup voltage with frequency is plotted at different low temperatures (15 K, 20 K, 25 K). (c) Shows $V(f)$ responses at higher temperatures, 150 K, 90 K, 80 K, 70 K (from top to bottom curves). The quadratic and linear regions are shown by the dotted red line and solid black line respectively. Insets show pickup response at 40 K and 220 K ($V$ is measured from $V(T)$ scan at different frequencies). For both 40 K and 220 K above 20 kHz, $V$ is increasing linearly with frequency. All data in the figures correspond to excitation current of 153 mA in the excitation coil.

We now investigate a thicker sample of Bi$_2$Se$_3$ with thickness 69 µm (S69). A comparison of 65 kHz, $V(T)$ data of S20 (Fig. 3(b)) with S69 (Fig. 5(a)), shows the effect of increasing bulk contribution to the overall conductivity in the thicker sample (S69). Figure 5(a) shows that $V(T)$ exhibits a saturated behavior at low $T$ upto 40 K (similar to Fig. 3(b) at 65 kHz). Between 40 K to 70 K the $V(T)$ data fits (solid black line) to Eq. 3 with $\sigma_s(T) = 1/(C + DT)$ while above 70 K to 170 K the data fits (red dashed line) to Eq. 2 with $\sigma_b(T) = \sigma_{b0} \exp\left(-\Delta/\kappa_b T\right)$, with $\Delta \approx 25.2 \pm 1.25 \text{ meV}$.

Unlike the thinner S20 sample at 65 kHz (Fig. 3(b)), where we see only surface conductivity over a wide $T$ regime, in the thicker S69 sample at the high $f$ (Fig. 5(a)), we see a dominating surface contribution below 70 K while bulk contribution takes over above 70 K. Figure 5(b) confirms $V(f)$ is linear over a wide frequency regime below 40 K (like Fig. 3(c)) indicating surface dominated transport at low $T$. Figure 5(c) confirms the bulk contribution to conductivity, as we begin to observe that at high $T > 50$ K a quadratic $V(f)$ dependence appears in the low $f$ regime which is absent at low $T$ in Fig. 5(b). The inset of Fig. 5(c) shows that at 40 K, $V(f)$ increases linearly with $f$ for the whole frequency range, whereas at 220 K $V(f)$ is not quadratic rather it is linear above 20 kHz. Figure 5(c) shows that at high $T$ and high $f$ we see the presence of a linear regime over a significant frequency range, which signifies the presence of surface conductivity at high $T$. The $V(T)$ profile showing identical features at different $f$ is shown in supplementary for S69 and for other samples as well (see supplementary sections S6, S7, S8 & S9).
Figure 6(a) shows the $V(T)$ measurements of Fig. 5(a) for the thick sample (S69). The data is fitted with the expression:

$$V(T) = P_{surface} \text{(Eq. 3)} + P_{bulk} \text{(Eq. 2)}$$

[4]

where, $P_{surface}$ and $P_{bulk}$ are mean fractions of the surface and bulk contribution to the pickup voltage with $P_{surface} + P_{bulk} = 1$. Note the $T$ dependence of $\sigma_s$ and $\sigma_b$ have already been shown by the black line and red dashed line in Fig. 5(a), respectively. The S69 data in Fig. 6(a) fits with $P_{surface} = 35\%$, $P_{bulk} = 65\%$. Note that at 65 kHz, the $P_{surface}$ is sensitive to the thin conducting regions within the sample where shielding currents are induced by the AC field. The minima in $V(T)$ at 70 K (Fig. 6(a)) suggests regions in the TI which exhibited predominantly surface conductivity response at low $T$ (Fig. 5(b)), with increasing $T$ begins to display bulk conductivity. The surface contribution is gradually degraded by transport channel being created in the bulk, as a result we see only an average 35% contribution to the total conductivity from the surface.
FIG. 6. (a) The $V(T)$ response for sample S69 is shown in red circles for 65 kHz frequency. The solid yellow line is the fit to the data, done using Eq. 4. Four distinct regions (I-IV) are shown with four different colors (see text for details). (b) A schematic showing the bulk conduction and valence bands as well as the surface states. The schematic on the right is an expanded portion of the left schematic. The violet curve in the right schematic shows upward band bending of the conducting band towards the Fermi energy of the surface state due to Se vacancies. The forward channel (Forward Ch.) is associated with electrons migrating from the bulk to the surface, the reverse channel (Reverse Ch.) is associated with electrons being thermally activated from the surface into the bulk. Black lines show the Dirac cone associated with the surface states. The Fermi energy is shown by red dotted line. The Fermi energy is shifted downwards due to electron doping due to Se vacancies. The green region represents the defect states produced by the Se vacancies. The activation energy gap ($\Delta$) is between the defect state and minima of the conduction band (c) Activation energy gap ($\Delta$) calculated from our model is plotted as a function of sample thickness, which is shown in left axis and bulk carrier ($n_{bulk}$) density is shown in the right axis. The $n_{bulk}$ is calculated using, $n_{bulk} = \frac{\sigma_{n}}{\mu(d)e}$.
where $\mu(d) = \frac{3000}{1 + 140/d}$ [22] is the thickness ($d$) dependent mobility and $\sigma_0 \sim 10^3$ S/m (based on the value used in the simulations above).

In the thinner S20 sample, surface contribution to conductivity measured at 65 kHz is more robust and it is degraded to a much lesser extent by bulk contribution to conductivity with increasing $T$. The fitting in Fig. 6(a) also shows that at high $T$ above 170 K there is a deviation from the Eq. 4 fit and the $V(T)$ becomes weakly temperature dependent. We have found in all the samples, the $V(f)$ at high temperatures exhibits linear and quadratic frequency dependence at high and low frequencies respectively (for eg. see 150 K data in Fig. 5(c)). This behavior suggests the presence of surface conductivity coexisting along with a bulk conducting regime in the sample even at high temperature. Based on our above discussions, in Fig. 6(a) we identified four different regimes: (i) Region I (below ~ 40 K the strong interaction dominated surface regime): In this regime, surface conductivity dominates in the TI and conductivity is nearly temperature independent due to strong electron-electron interaction effects. Enhanced electron-electron interactions producing large energy transfer scattering events at low $T$ [32,46], i.e., $C >> D$ in $\sigma_s(T) = \sqrt{1/(C + DT)}$  (ii) Region II (temperature dependent surface state): Here the surface state dominates the conductivity of the TI with a temperature dependence of the form $\sigma_s(T) = \sqrt{1/(C + DT)}$ due to the onset of electron-phonon scattering. As the temperature increases, the surface conductivity falls until it reaches close to the bulk conductivity value, which is close to the minima in $V(T)$. (iii) Region III (bulk dominated state): In this region, which extends about 70K to 170 K, Fig. 5(a) shows a good fit to bulk behavior (see dashed red curve in Fig. 5(a)). However, the fit doesn’t imply that in region III the surface contribution to conductivity is zero. Figure 5(a) shows that in regime III, the surface contribution to conductivity (solid black line) is much smaller than the bulk contribution, due to which the fit to bulk appears to be good and bulk dominated. Figure 5(c) shows that for temperatures in regime III (like 90 K and 150 K) the $V(f)$ has quadratic as well as linear regime, which shows the presence of both bulk and surface contributions to conductivity. Hence, region III is a regime with the bulk contribution to conductivity dominating over a surviving weak surface contribution. Here the response from the bulk conducting states in the TI begins to appear because of thermally activated conductivity behavior. The typical thermal activation energy scale is ~ 6 meV - 30 meV. Note that the activation energy gap $\Delta$ (shown Fig. 6(b)) we determine, increases
with sample thickness (see Fig. 6(c)). (iv) Region IV (high $T$, interaction dominated surface and bulk state regime): This is an unusual regime found at high temperatures, where $V(T)$ deviates from Eq. 4 fit. In this regime, we recall similar linear $V(f)$ behavior (inset Fig. 5(c) and Fig. 3). Just like the weakly $T$ dependent surface conductivity regime is present at low $T$, we propose the weakly temperature independent regime at high $T$ is where the effects of surface conductivity of a TI become important once again. We would like to reiterate here that surface and bulk, both contribute to the pickup voltage as the induced current are generated in sample both surface and bulk state due to AC magnetic field. The region III and region IV (Fig. 6(a)) are associated with a competition between bulk and surface state contributions to conductivity. Here within the temperature window III the bulk contribution to conductivity dominates over the surface contribution, while in region IV it is vice versa. We believe the surface state do not disappear at higher temperatures in the TI, instead they survives along with a bulk conducting state. In the next section based on the simulation results we develop the understanding of the measurements in Figs.3 and 5.

**Simulations of the mutual inductance behavior:**

A bulk gap in Bi$_2$Se$_3$ of 0.3 eV suggests that thermally activated conductivity should be seen at significantly elevated temperatures, but we see activated conductivity from $T \sim 70$ K onwards (viz., the region III in Fig. 6(a)). In Bi$_2$Se$_3$ samples it is known Se vacancies add defect states in the bulk. These defects states add excess electrons in the bulk due to which the conductivity of the bulk increases. We simulate the electromagnetic response of this topological insulator with defects using COMSOL multi-physics software. In Fig. 7(a) inset, we first model a defect free TI with high conducting thin metallic sheets (yellow) sandwiching a bulk low conducting slab (blue). We solve the Maxwell equation for a TI subjected to a time varying magnetic field applied using coil parameters identical to our experiment (see S2 section in supplementary information). We use an AC excitation current of $I = 150$ mA (similar to our experiment). First, the rate of change of induced flux ($\dot{\phi} = M \dot{I}$) in the pickup coil is calculated from which the mutual inductance (real and imaginary components) $M'$ and $M''$ are calculated (analog to our experiments, $M'' \ll M'$). The modelled TI has crystal thickness and surface area similar to S20 sample, with a thickness of 5 nm for the metallic high conducting sheets and 20 $\mu$m for the bulk insulating layer. The above is based on estimates indicating the thickness of conducting surface in Bi$_2$Se$_3$ is between 5 to 10 nm [47,48].
In Fig. 7(a) we see the data begins to look like the experimental $V(f)$ data when the bulk conductivity $\sigma_b \sim 10^3 \text{ S/m}$ and the conductivity of the metallic sheets $\sigma_s \sim 10^{11} \text{ S/m}$ (note OFHC triple nine purity copper has conductivity $\sim 10^8 \text{ S/m}$). Experimental data matches more accurately with simulation result in Fig.7(b). Earlier transport measurements have shown conductivity of surface states in Bi$_2$Se$_3$ $\sim 10^4 \text{ S/m}$ to $10^5 \text{ S/m}$ [21,49]. It may be mentioned here that in conventional transport measurements due to inability to distinguish between bulk and surface conducting channels, the measured surface conductivity via transport measurements could be much lower than the actual value.

To better understand the temperature and frequency dependent pickup signal behavior of Fig. 3 and Fig. 5, we recall the expression for the skin depth is $\frac{1}{\sqrt{\pi \sigma f \mu}}$. Temperature and frequency variation both lead to modifying the skin depth. At a constant frequency in a temperature dependent measurement, the electrical conductivity ($\sigma$) of the sample decreases with increasing $T$. Due to the decreasing conductivity of the sample with increasing temperature, the skin depth ($\delta \propto \frac{1}{\sqrt{\sigma}}$) increases until it becomes of the order of the sample thickness. Furthermore at low frequency $f$ as the skin depth is large (since $\delta \propto \frac{1}{\sqrt{f}}$), hence in a temperature dependent measurement at fixed low $f$ (like 5 Hz), only the average low bulk conductivity of the sample is probed. At a high $T$ above 100 K, using the low bulk conductivity value ($\sim 10^3 \text{ S/m}$, determined above) we estimate a skin depth to be $\sim 80,000 \mu\text{m}$ at 5 kHz in Bi$_2$Se$_3$ sample, which is far greater than the thickness of any of the sample we have investigated (at 65 kHz the skin depth above 100 K is still $\sim 22,000 \mu\text{m}$). In such a situation where the skin depth $>>$ sample thickness (at high $T$), one measures an averaged sample response. In this situation at high $T$, any appreciable difference in the pickup voltage at two different frequencies can be observed only if the materials intrinsic bulk electrical conductivity changes appreciably with frequency. Such changes in the intrinsic bulk electrical conductivity with frequency occur only at high frequencies, which are well above the frequency measurement range of our pickup coils. However, in our measurement above 100 K, the pickup voltage should have continued to exhibit the exponential temperature dependence of the bulk conductivity which is present below 100 K. Observation of a non-exponential, weak $T$ dependence
above 100 K (see Fig. 3(b)), suggest the re-emergence of weakly temperature dependent surface conductivity competing along with bulk conductivity at high $T$.

In a Topological insulator as the currents flow on the surface and our pickup measurement is sensitive to the currents induced on the conducting surface state, hence pickup signal is expected to scale with sample surface area if in our measurement we are primarily probing the surface conducting states of a TI at high frequency. In our measurements at low temperature and high frequency, as we picking up a response from these surface currents, hence it is expected that the net pickup signal will scale with the total surface area of the sample. In Fig. 7(c) we plot the pickup voltage versus sample surface area for all our five samples. Note that above 120 K at 5 kHz as the skin depth is greater than the thickness of all the samples, the pickup signal saturates to a value close to 1 mV for all the different samples. At high frequency ($f$) as the skin depth decreases (as $\delta \propto \frac{1}{\sqrt{f}}$) and at low $T$ as the conductivity enhances, the excitation signal primarily probes the high conducting surface states of the sample. In fact, at high frequency and low $T$ as the highly conducting surface states are effectively probed, the large conductivity of the surface states makes the skin depth smaller compared to that obtained using bulk electrical conductivity (which is much smaller than the surface conductivity). Due to this effect of the pickup signal at high frequency of 65 kHz and low $T$ probing the surface, Fig. 7(c) shows the pickup voltage scaling with the sample surface area as it is sensitive to the conducting surface states in the sample. Based on the above we understand that as the frequency of the pickup measurement is increased, the change from bulk to surface dominated conductivity is observed clearly. Figure 7(d) shows the variation of skin depth ($\delta$) as function of $f$ at room temperature using the high conductivity of the surface state in Bi$_2$Se$_3$ (viz., $\sigma_s \sim 10^{11}$ S/m). Note that in a TI a high electrically conducting sheath completely covers a much lower electrically conducting medium. An electromagnetic (EM) wave impinging on the TI will always first encounter the high conductivity surface sheath on the TI. Hence the attenuation and consequently the EM skin depth will be determined by the electrical conductivity of the high conducting surface sheet. We show in S10 in supplementary section a simulation showing the attenuation of an impinging EM signal is governed by the high conducting surface sheath in the TI material. In a frequency dependent measurement at moderately low $T$, at high frequencies as the skin depth is already smaller than the sample thickness, in this regime changing
the frequency at constant temperature (especially at low $T$), leads to variations in skin depth in the nanometer range. The inset of Fig. 7(d) we plot the behavior of the estimated decrease in skin depth ($\Delta \delta = \frac{df}{df} \Delta f$, for a $\Delta f = 500$ Hz increase in frequency) versus frequencies. The $\Delta \delta$ is seen to change in the range of a 100 nm and below above 20 kHz. Here were would like to mention that temperature dependent measurement of pickup voltage is not sensitive enough at high $T$ however a frequency dependent measurements at high $T$ of 150 K (see Fig. 5(c)) still observes a change in frequency dependent behavior. This change in curvature of the pickup voltage is seen in a frequency dependent measurement because the thin high conducting surface state regime begins contributing to the signal along with the comparatively lower conducting bulk.

Note that the relation between $M'$ and induced voltage in the pickup coil is $V' \propto \omega M'$ (as $M'' < < M'$ or $V'' < < V'$, $M'(f) = \frac{V'(f)}{\omega I}$ where $I = 150$ mA, $\omega = 2\pi f$). Figure 7(a) shows simulated $M'$ as a function of frequency for different conductivities of the TI surface while keeping the bulk conductivity constant at $\sim 10^3$ S/m. The value $\sigma_b$ we use is based on estimates in literature [34]. The general behavior of $M'(f)$ is that, it increases linearly with frequency and it saturates at higher frequency. At lower frequency regime where $M'(f) \propto f$ (see dotted line drawn in Fig. 7(a)) corresponds the pickup voltage, $V(f) \propto f^2$. At higher $f$ where the $M'$ is weakly dependent on $f$, corresponds to $V(f) \propto f$. Figure 7(a) shows that the $M'$ begins to show saturating features as the surface state conductivity ($\sigma_s$) increases. This confirms that highly conducting surface states are responsible for the linear frequency dependence of pickup voltage $V(f)$ in the high frequency regime (see Fig. 3, Fig. 5 for S20 and S69 sample). Note that while the simulations in Fig. 7(a) shows the $M'(f)$ saturates above 40 kHz, however in our experiments (Fig. 3 and Fig. 5) showed that $V(f) \propto f$ regime (i.e., $M'(f) \sim$ constant) sets in above 20 kHz and not from 40 kHz.
FIG. 7. Inset (a) shows the schematic of an ideal TI. Blue color represents bulk state and yellow color represents high conducting surface state. The thickness of the high conducting surface state is taken as 5 nm (thickness of the yellow region), the other relevant dimensions are height = 20 μm, width = 1.9 mm, and length= 2.4 mm. The main panel shows the behavior of the simulated $M'(f)$ for different conductivity of surface state keeping bulk conductivity constant (~$10^3$ S/m). Red dash line shows the linear dependence of $M'$. It is clear that $M'$ seems to saturate beyond 40 kHz whereas the experimentally measured data shows saturation from 20 kHz onwards. (b) Shows the $M'$ is simulated as a function of $f$ for different inhomogeneity levels (0%, 20% and 30%) by incorporating conducting channels inside the bulk (see text for details and section S2 of supplementary information). Comparison of simulation and experimental result (scattered data) of $M'$ is shown for S20 sample at room temperature. (c) Shows the pickup voltage response with varying sample surface area at 5 kHz and 65 kHz. (d) Shows the skin depth at room temperature as a function of applied frequency. The inset shows an estimate of the decrease in skin depth ($\Delta \delta$) (which is $\Delta \delta = \frac{d \delta}{df} \Delta f$, $\Delta f = 500$ Hz) for a 500 Hz increase in frequency at different frequencies.
In order to match our simulations with the observed saturation of $M'(f)$ at relatively lower $f$ (from 20 kHz), without significantly enhancing the surface conductivity $\sigma_s$ (far beyond what have been reported in literature for these samples), we consider a situation where conducting channels exist in the bulk which connect to the high conducting surface states. We argue later these high conducting channels in the bulk are generated by Se vacancies. Such a state of the TI we refer to as an inhomogeneous topological insulator state. We incorporate inhomogeneity in our model for the TI by considering a distribution of solid high conductivity cylindrical regions threading through the bulk of the sample connecting the top and bottom high conducting surface sheets of the TI (see section S2 in the supplementary information for a schematic of the modelled inhomogeneous TI system). In our simulation the percentage inhomogeneity in the TI is varied by changing the density of cylinders and the conductivity of the cylinders is taken as $\sigma_s$. Figure 7(b) shows that the results of simulations match with our experimental results by, using $\sigma_s \sim 10^{11}$ S/m, $\sigma_b \sim 10^3$ S/m and 30% inhomogeneity. The introduction of the 30% inhomogeneity in our TI model causes the $M'(f)$ to saturate from lower frequencies of 20 kHz. The presence of 30% inhomogeneity in the sample would affect the net bulk electrical conductivity. However, estimating the extent of change in the net bulk electrical conductivity isn’t clear as yet as this would statistically depend on the fraction of high conductivity and low channels contributing to the electrical conduction paths when electric current flows through the bulk. We find that our above simulation results are not sensitive to the details of the spatial distribution of inhomogeneity introduced in the TI, but rather the result are quite sensitive on the inhomogeneity fraction in the TI (see section S2 in the supplementary information). Figure 7(b) shows the close match between the simulated $M'(f)$ (purple dashed line) and the measured voltage data (green scattered data) for S20 sample at 300 K using the inhomogeneous TI model. As mentioned earlier in context of Fig. 7(d), in the high frequency regime the changes in the skin depth is in the range of ~ 100 nm. Since in this high frequency regime our simulations suggest the emergence of inhomogeneity in the TI state, we suggest that these inhomogeneity are clustered over length scales which are $\leq$ 100 nm (which in our model for inhomogeneity in the TI, we take as the diameter of the cylinders).

**Role of disorder in Bi$_2$Se$_3$:**

The measured $\Delta$ values are almost an order of magnitude smaller than the estimates of bulk band gap in Bi$_2$Se$_3$ of about 0.3eV [7,10, 25,26]. We believe the $\Delta$ corresponds to a gap between the
defect states generated by Se vacancies and bottom of conduction band (see Fig. 6(b)). Figure 6(c) showed Δ increases with the sample thickness. It is known that Se vacancy causes an upward band bending (Fig. 6(b)) of the states near the bottom of the conduction band towards the surface state [50]. Analysis of the SdH oscillations in our Bi$_2$Se$_3$ (see Fig.1 and its caption for details) using the Lifshitz-Kosevich (LK) equation [34,51,52] gives a frequency of the SdH oscillations ($F$) = 46.95 ± 0.25 T. As $F = \left(4\pi^2 h n_s \right) / e$, where $n_s$ is the surface carrier density, we get $n_s = (2.268 \pm 0.012)$ cm$^{-2}$, which compares well with earlier estimates in this batch of samples [34]. Using $n_s$ we estimate the Fermi wave vector for the 2D surface states in our sample is, $k_F = \sqrt{2\pi n_s} = (0.0377 \pm 0.0027)$ Å$^{-1}$. By comparing this $k_F$ value with the ARPES spectrum measured for Bi$_2$Se$_3$ samples (see Fig.1 of Ref.[53]), we see that the Fermi energy ($E_f$) should be located approximately 30 meV above the Dirac point and about 100 meV below the bottom of the bulk conduction band (this comparison is illustrated in section S11 of supplementary information). In Topological insulators as there is a significant difference between the surface and bulk Fermi levels. The final location of the Fermi level depends on the charge transfer to make Fermi level equal everywhere in the sample. The above instances of $E_f$ lying within the bulk gap of our TI sample is not unusual and have been reported earlier in Bi$_2$Se$_3$ samples [34,54,55,56]. The process of equilibrating the Fermi level through charge transfer from the bulk to surface or vice-versa creates a potential difference which leads to shifting of bands or band bending effects. An upward or downward band bending depends strongly on the presence of defects in the material and in the present case the upward band bending is related to the presence of Selenium vacancies in the Bi$_2$Se$_3$. Degradation of surface of Bi$_2$Se$_3$, for example due to surface oxidation, also produces band bending of the surface states [36,37] which modifies surface conductivity. However, sample surface degradation induced band bending would lead to a modified surface conductivity compared to another sample with negligible surface degradation. However, surface degradation and associated band bending alone cannot explain the temperature dependent change in conductivity from surface to bulk dominated behavior as the temperature is changed from region II to region III in Fig. 6(a). The band bending favors the migration of electrons generated in the bulk (due to Se vacancies) to the surface. We refer to this as the forward channel (see Fig. 6(b)). Above 70 K the thermal energy is sufficient for thermal activation of charges from defects states into the bulk. We refer to this as the reverse channel. This reverse channel for charge migration begins at $T > \Delta / k_B$ (Fig. 6(b)). Recall, that in Bi$_2$Se$_3$ excess
charge carriers are produced by Se vacancies. In Fig. 6(c) we estimated the behavior of carrier density in the bulk \( (n_{\text{bulk}}) \) using a known form \([22]\), \( n_{\text{bulk}} = \frac{\sigma_b}{\mu(d)e} \), where \( \mu(d) = \frac{3000}{1 + \frac{140}{d}} \) is the thickness \( (d) \) dependent mobility and \( \sigma_b \sim 10^3 \text{ S/m} \). Figure 6(c) shows that \( n_{\text{bulk}} \) decreases with sample thickness. Thus the activation barrier \( \Delta \) is larger for smaller bulk carrier density. Akin to a tight binding like picture, we propose the hopping back and forth of charges between the defect states in the bulk and surface states enhances the average kinetic energy of charges leading to the formation of a broad defect band and this also helps to open a gap \( \Delta \) near the surface states. The thicker samples have a lower concentration of Se vacancies, and hence Coulombic interactions are strong due to lower screening effect. The \( \Delta \) increasing with samples thickness as seen in Fig. 6(c), could be a result of enhanced Coulombic interactions in these lower electron doped samples suppressing the kinetic energy and localizing the charges. At \( T < \Delta/k_B \) the gap protects the surface from the defect states present in bulk. At \( T > \Delta/k_B \) the bulk contribution to conductivity sets in and the TI becomes an inhomogeneous TI. The forward and reverse channel rates may not be identical leading to a collection of charges at the surface at high \( T \). Enhanced interaction between the electrons at high \( T \) once again competes against the effects of thermal fluctuations and surface conductivity begins competing with the bulk (viz., the region IV we see in Fig. 6(a). Enhanced interaction effects lead to the weak \( T \) dependence of conductivity not only at low \( T \) below 40 K but also at high \( T \) above 180 K (Fig. 6(a)). Thus Selenium vacancy induced electron doping of the bulk leads to an interplay between bulk and surface conductivity, leading to an inhomogeneous TI state in \( \text{Bi}_2\text{Se}_3 \). Based on the above discussion, we propose that in region III of Fig. 6(a) the bulk contribution to the conductivity dominating over the surface, due to thermal excitation of charge carrier in the bulk state. However, at higher temperature, the excess charge migration to surface state due to Se vacancies results in the contribution of the surface conductivity to enhance once again. It is due to this effect that we see the experimental data deviating from the fitting line in high temperature regime (region IV in Fig. 6(a)). We would like to mention here that earlier STM studies have primarily investigated the effect of surface defects in \( \text{Bi}_2\text{Se}_3 \). These studies imaged the presence of Se vacancies on the surface of \( \text{Bi}_2\text{Se}_3 \) which appear as triangular shaped defects \([57,58,59]\). They showed changes in local density of states near these surface impurities and the presence of low energy resonances near defects like Se vacancy on the surface which cause fluctuation in the Dirac point. The defects also lead to surface band bending effects. We believe
more detailed work using techniques like STM and ARPES, is required to probe the effect of defect states lying within the bulk and the resulting gap emerging in the TI.

Conclusions:

In conclusion, our non-contact two coil mutual inductance measurements in Bi$_2$Se$_3$ single crystals suggest that the surface states are coupled to bulk state with filamentary high conducting structures. The inhomogeneous state gets precipitated in the bulk of the TI only above the threshold temperature, which is associated with the activation of charge between the surface and bulk. This leads to an interplay between bulk and surface dominated transport regimes at different $T$, which is produced as a result of disorder introduced in the system. While material disorder leads to interplay between bulk and surface conductivity in a TI material, it is also responsible for resurfacing of the surface conductivity at high temperature which is a potentially feature useful for applications.

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Fig. 1

(a) \(\Delta R_{xx} \times 10^{-3} \Omega\) vs. \(I/B \) for \(\text{Bi}_2\text{Se}_3\). The data points are for different temperatures: 3 K (circles), 4.2 K (green circles), 7 K (orange circles), and 10 K (magenta circles). The solid line represents the LK fitting.

(b) \(R_{xy} \) vs. \(B \) for \(\text{Bi}_2\text{Se}_3\). The plots show the temperature dependence at 3 K (empty square), 4.2 K (red circle), 7 K (green triangle), 10 K (blue triangle), and 25 K (magenta diamond). The graph includes a linear fit for the magnetic field dependence.
Fig. 2

(a) Diagram of the experimental setup showing the cryostat, pick-up coil, sample with Cu sheet, excitation coil, exchange gas cooling, and AC current from the lock-in amplifier.

(b) and (c) Images showing the distribution of the flux density $B_z$ and the Cu sheet.

(d) Graph showing the frequency response of $V'$ for different conditions: without Cu sheet, with Cu sheet, with Cu sheet and a hole, and with Cu sheet and a hole with exchange gas cooling. The frequency is 20 kHz, and the excitation coil current is 150 mA.

(e) Graph showing the temperature dependence of $V''$ for $\text{BaFe}_{1.85}\text{Co}_{0.15}\text{As}_2$. The Tc temperature is indicated.
Fig.3
Induced Bi$_2$Se$_3$
Fig. 5
Fig. 6

(a) Diagram showing the interaction dominated regions (I, II, III, IV) and the competing surface + bulk state.

(b) Schematic representation of Fermi energy bands and bulk gap (0.3 eV).

(c) Graph showing the change in $\Delta$ (meV) with respect to $d$ (µm) and $n_{bulk} (\times 10^{17})$.

Surface dominated

Competing surface + bulk state

Interaction dominated

Surface dominated

Defect bands

Fermi energy of the surface state

Bulk conduction Band

Defect Band

Reverse ch.

Forward ch.
A non-contact mutual inductance based measurement of an inhomogeneous topological insulating state in Bi$_2$Se$_3$ single crystal with defects

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**S1: Coil details.**

In our two coil mutual setup, both coils are dipole coils. The most important point in coil design is that the winding of the excitation and pickup coils should be as close as possible. The excitation coil has four layers and each layer have 36 turns while pickup coil has four layers having 32 turns. Figure (a) shows the schematic of the excitation coil. The bobbin and stand are made of an insulating materials macor to avoid any unwanted signals developed by eddy currents. The material of the bobbin is non-magnetic. Figure (b) and (c) show the inside of the two coil mutual setup. Figure d shows that pickup voltage as a function of applied current in the source coil at 65 kHz. Pickup voltage scales linearly with current in the excitation coil. This measurement is carried out at 140 K of S20 sample. The 1 volt drop across the excitation coil corresponds to 153 mA current in the excitation coil. Note that for our setup there is a linear dependence of the pickup signal with the excitation current in the coils shown below. This allows measurements to be done with higher currents in the coil for better sensitivity without changing any features in the frequency or temperature dependence of the measurements. The linear relationship between pickup voltage amplitude and current in excitation coil shows the measurements with different values of current in the excitation coil can be compared with each other due to the linear relationship.

**S2: COMSOL simulation.**

We have also verified our experimental data with simulation using a simple model. The simulation part has been performed using Comsol multiphysics software (AC-DC module). The simulation is done by the solving the standard Maxwell EM equations:
\[
\left(j\omega \sigma - \omega^2 \varepsilon_0 \varepsilon_r \right) \vec{A} + \vec{\nabla} \times \frac{\vec{B}}{\mu_0 \mu_r} - \sigma \vec{v} \times \vec{B} = \vec{J}_e
\]

\[
\vec{\nabla} \times \vec{A} = \vec{B}
\]

\[
J_e = \frac{N(V_{coil} + V_{in})}{R_{coil}}
\]

where \( \omega \) is the angular frequency of the applied AC signal, \( A \) is the magnetic vector potential, \( \sigma \) is the conductivity of the material, \( v \) is the velocity of the charge particle, \( N \) is the number of turns, \( R_{coil} \) is the resistance of the coil, \( J_e \) is the current density and \( V_{coil} \) is the applied AC voltage in the coil. Figure (a) shows the schematic of ideal topological insulator. It also shows dimension of our sample used for the simulation. The sample is 20 \( \mu \)m thick and the two surface states are 5 nm thick each. Figure (b) shows the schematic of inhomogeneous bulk state. The coupling channels are the cylinders having diameter 100 nm. Depending upon the inhomogeneity level, the conducting cylinder concentration is varied keeping the diameter fixed. Note we found no difference in mutual inductance for 30% inhomogeneity (see main text Fig. 7b) between an ordered and a disordered configuration of the conducting cylinders. Pickup voltage is sensitive to the fraction of the TI sample being inhomogeneous. Fig. (c) shows inhomogeneous bulk state which has disorder arrangement. For example, in Figs. (b) and (c) the inhomogeneity is 17%, as the number of cylindrical channels are equal in both cases. The overall schematic of the Comsol simulation is shown in Fig. (d).

S3: S20 sample response.
In Figs. (a) and (b) variation of pickup voltage ($V$) with temperature ($T$) is shown for S20 at two frequencies 2 kHz and 50 kHz. The 2 kHz data behavior is almost similar to the 5 kHz data and 50 kHz data behaves similar to the 65 kHz data. The red dash line is fitted to the eqn. $V \propto V_{00} \exp \left(-\frac{\Delta}{k_B T} \right)$ and black solid line is fitted with $V \propto \frac{1}{(C' + D'T)}$

| Frequency | 2 kHz   | 50 kHz        |
|-----------|---------|---------------|
| $V_{00}$  | 0.132 mV| $C'$          |
| $\Delta$  | 4.73 meV| $3.48 \text{ mV}^{-1}$ |
|           |         | $D'$          |
|           |         | $0.069 \text{ mV}^{-1}\text{K}^{-1}$ |

S4: S20 response, amplitude variation.
Frequency variation of $V$ is shown for different applied voltages in the excitation coil at 30 K. Solid lines are the best fit line through the data points, which shows a linear behavior at low temperatures. It also shows that $V(f)$ profiles are linear with applied voltage in the excitation coil.

**S5: Pickup voltage generated in the pickup coil.**

Let us consider an AC current across the primary coil to produce an AC magnetic field $B = B_0 \exp^{-io\omega t}$, where $\omega$ is the angular frequency of the ac signal and $B_0$ is the amplitude of the ac magnetic field. Due to this AC magnetic field, induced current is generated in the sample. From Maxwell equation $\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$, the total electric field ($\mathbf{E}(r,z,t)$) in the sample can be written as

$$\mathbf{E}(r,z,t) = \frac{i\omega \exp^{-i\omega t}}{2\pi r} \Phi(z) \hat{\phi}$$

where $r$ and $z$ are the sample position and $\Phi(z)$ is the magnetic flux passing through the circle of radius $r''$. We have used parallel conduction model in which total conductivity of the TI is given as $\sigma = (\sigma_s + \sigma_b)$, where $\sigma_s$ and $\sigma_b$ are the surface and bulk conductivities of the TI respectively.

The total induced current in the sample is

$$\mathbf{J} = \sigma_i \frac{i\omega \exp^{-i\omega t}}{2\pi r} \Phi(z) \hat{\phi}$$

The magnetic field is generated in the pickup coil from this induced current in the sample. So, the total output voltage in the pickup coil can be written using Faraday’s law:

$$V_{output}(\omega) = \frac{d}{dt} \int \int [\mathbf{B}(j',r',\theta',z) \cdot d\mathbf{S}']$$
\[
= \iiint \left[ \left( \frac{\mathbf{J} \times \mathbf{R}}{R^3} \right) \cdot d\mathbf{S} \right]
\text{where } \mathbf{R} = (r' - r'')r - z
\]

\[
=- \omega^2 e^{-i\omega t} \iint \left[ \left( \frac{\Phi'(z)\mathbf{\hat{J}}}{2\pi r} \times \frac{\mathbf{R}}{R^3} \right) \cdot d\mathbf{S} \right]
\]

\[
=- \omega^2 e^{-i\omega t} \sigma I \Phi \xi( r_0, z )
\]

where \( \mathbf{B}( j, r', \theta', z ) \) is the magnetic field at the point A in the pickup coil generated by the screening current flowing in the sample. The \( \xi( r_0, z ) \) is the geometry factor of the mutual coil setup between the sample and pickup voltage.

\[
V_{\text{output}}(\omega) = \xi( r, z ) \Phi \omega^2 e^{-i\omega t} \sigma \frac{1}{1 + i\omega \tau}
\]

Here we have used the Drude like ac conductivity for the surface as well as bulk state of the TI. Here we have assumed that the distance between the two coils is much larger than the sample thickness. If we consider the temperature dependent conductivity, the total output voltage becomes

\[
V_{\text{output}}(\omega) = \xi( r, z ) \Phi \omega^2 e^{-i\omega t} \sigma \frac{1}{1 + i\omega \tau} \left[ \frac{\sigma_{os}}{C + DT} + \frac{\sigma_{ob}}{e^{\Delta/kT}} \right]
\]

where \( C \) is the static disorder scattering and \( D \) introduces electron-phonon coupling, \( \Delta \) is the energy gap, and \( G_{bo} \) is the high temperature conductance of the bulk state.

When the bulk conductivity is dominated over surface conductivity \( (\sigma_{os} \ll \sigma_{ob}) \) and also in the low frequency regime \( \omega \tau \ll 1 \), the output voltage becomes

\[
V_{\text{output}}(\omega) \approx \Phi \xi( r, z ) \omega^2 e^{-i\omega t} \sigma_b
\]

\[
V_{\text{output}}(\omega) \propto \omega^2 \sigma_b
\]

The surface state of the TI is free from back scattering due to chiral nature of the surface state. If the surface state is dominated in current conduction and in the higher frequency regime \( \omega \tau >> 1 \), the output voltage becomes

\[
V_{\text{output}}(\omega) = \Phi \xi( r, z ) \omega^2 e^{-i\omega t} \sigma_r \frac{1}{1 + i\omega \tau}
\]

\[
V_{\text{output}}(\omega) = \Phi \xi( r, z ) \omega^2 e^{-i\omega t} \sigma_r \frac{1 - i\omega \tau}{1 + \omega^2 \tau^2}
\]

Using \( \omega \tau >> 1 \)

\[
V_{\text{output}}(\omega) \approx \Phi \xi( r, z ) e^{-i\omega t} \sigma_r \frac{\omega}{\tau}
\]
\[ V_{\text{output}}(\omega) \propto \sigma_i \omega \]

S6: S69 sample response.

Variation of pickup voltage \( V \) with temperature \( T \) is shown for S69 at two frequencies 30 kHz and 70 kHz. The 30 kHz and 70 kHz profiles almost behave similarly with the 65 kHz data (Fig. 5(a) in main manuscript). The red dash line is fitted with \( V \propto V_{bo} \exp\left(-\Delta/k_B T\right) \) and the black solid line fitted with \( V \propto \frac{1}{(C' + D'T)} \). The fitting parameters are shown in the table.

| Frequency | 30 kHz | 70 kHz |
|-----------|--------|--------|
| \( V_{bo} \) | 7.45 mV | 37.5 mV |
| \( \Delta \) | 9.8 meV | 25.6 meV |

S7: S75 sample response.

| Frequency | 30 kHz | 70 kHz |
|-----------|--------|--------|
| \( C' \) | 0.0282 mV\(^{-1}\) | 0.0062 mV\(^{-1}\) |
| \( D' \) | 0.082 mV\(^{-1}\)K\(^{-1}\) | 0.199 mV\(^{-1}\)K\(^{-1}\) |
Variation of pickup voltage \( (V) \) with temperature \( (T) \) is shown for S75 at two frequencies 5 kHz and 70 kHz. The 70 kHz profile is almost similar to the 65 kHz data (Fig. 5(a) in main manuscript). The red dash line is fitted with, \( V \propto \frac{\nu_0 \exp\left(-\frac{\nu}{k_B T}\right)}{\sqrt{C^* + D^* T}} \) and the black solid line is fitted with \( V \propto \frac{\nu}{\sqrt{C^* + D^* T}} \). The fitting parameters are shown in the table.

| Frequency | 5 kHz  | 70 kHz |
|-----------|--------|--------|
| \( \nu_0 \) | 0.948 mV | 97.9 mV |
| \( \Delta \) | 11.5 meV | 28.6 meV |

S8: S51 sample response.

Variation of pickup voltage \( (V) \) with temperature \( (T) \) is plotted for S51 at two frequencies 5 kHz and 30 kHz. The 30 kHz profile shows almost similar behavior with 65 kHz data (Fig. 5(a) in main manuscript). The red dash line is fitted with, \( V \propto \frac{\nu_0 \exp\left(-\frac{\nu}{k_B T}\right)}{\sqrt{C^* + D^* T}} \) and black solid line is fitted with \( V \propto \frac{\nu}{\sqrt{C^* + D^* T}} \). The fitting parameters are shown in the table.

| Frequency | 30 kHz |
|-----------|--------|
| \( \nu_0 \) | 0.19 mV | 106.8 mV |
| \( \Delta \) | 8.52 meV | 24.12 meV |

| Frequency | 30 kHz |
|-----------|--------|
| \( C' \) | 0.00052 mV\(^{-1}\) |
| \( D' \) | 0.026 mV\(^{-1}\)K\(^{-1}\) |
S9: S82 response.

Variation of pickup voltage ($V$) with temperature ($T$) is shown for S69 at two frequencies 5 kHz and 60 kHz. The 60 kHz profile is almost similar to the 65 kHz data (Fig. 5(a) in main manuscript). The red dash line is fitted with $V \propto V_{b0} \exp(-\Delta / k_B T)$ and black solid line is fitted with $V \propto 1/(C' + D' T)$.

The fitting parameters are shown in the table.

| Frequency | 5 kHz | 60 kHz |
|-----------|-------|--------|
| $V_{b0}$  | 0.675 mV | 121 mV |
| $\Delta$  | 10.2 meV | 32.8 meV |

S10: Bz distribution of the sample.

Figure (a) and (b) are shown the B$_z$ distributions for two different frequency applied in the source coil. The current applied across the source coil is 150mA which is similar to our experiment. In the simulation, we have used the idle topological insulator model without encounter any inhomogeneous fraction in bulk. The schematic of the simulation is shown in fig. c (the cross-section of the sample is shown in fig. c, yellow color shows the surface state with conductivity $10^{11}$ S/m and blue color shows the bulk state with conductivity $6 \times 10^3$ S/m. Bulk state is 20 μm thick and surface state is 10 nm thick). At low frequency, the magnitude of the B$_z$ has much higher value above the sample (Fig. (a)) due to the large skin depth but at a higher frequency the B$_z$ value decreases to ~200 Oe at 50 kHz above the sample surface (fig. b). Now we replace the Bi$_2$Se$_3$ sample with another normal sample which has same conductivity in bulk and surface ($6 \times 10^3$ S/m). Figure (d) and (e) are shown the B$_z$ distributions for two different frequency applied in the source coil. From these two figures, we can notice that the B$_z$ distributions are same for these two cases as the skin depths are mm range in these frequency range. From these simulation, we can notice

\[
C' = 0.0058 \text{ mV}^{-1} \\
D' = 0.0016 \text{ mV}^{-1}\text{K}^{-1}
\]
that due to the presence of micron thickness sample, the $B_z$ distribution changes with frequencies in spite of the bulk skin depth in mm range. So, the high conducting surface state which is extended in nm rang, has strong contributions in the $B_z$ distribution.

S11: Position of the Fermi energy.

We use Lifshitz-Kosevich (LK) equation is to analyse the SdH oscillation seen in our Bi$_2$Se$_3$ sample. From the oscillation period of the SdH oscillations seen in the transport data (Fig. 1a in revised manuscript and see the figure caption), the measured surface carrier density per area is found to be $n_s = (2.268 \pm 0.012) \times 10^{12}$ cm$^{-2}$, which corresponds to a Fermi wave vector for the 2D surface state to be $k_F = \sqrt{2\pi n_s} = (0.0377\pm0.0027)$ Å$^{-1}$. By placing the location of $k_F$ on the ARPES spectrum of Bi$_2$Se$_3$ (see $k_F$ marked by a vertical yellow line in the adjoining figure which is Fig.1 ARPES data of Bi2Se3 as published by M. Bianchi et al., Nature Commun. 1, 128 (2010)), we see that the Fermi energy is approximately 30 meV above the Dirac point and about 100 meV below the bottom of the bulk conduction band. Such instances of $E_f$ lying within the bulk
gap is not unusual and have been reported earlier in Bi$_2$Se$_3$, for example, J.G. Analytis, R.D. McDonald, S.C. Riggs, J.-H. Chu, G.S. Boebinger, I.R. Fisher, Nat. Phys. 6, 960 (2010); J.G. Analytis, J.-H. Chu, Y. Chen, F. Corredor, R.D. McDonald, Z.X. Shen, I.R. Fisher, Phys. Rev. B: 81, 205407 (2010); M. Brahlek, N. Koirala, M. Salehi, N. Bansal, S. Oh, Phys. Rev. Lett. 113 (2014) 026801, our Ref. 34, T. R. Devidas et al *Euro Phys. Lett.* 108, 67008 (2014). In Topological insulators there, is a significant difference between the surface and bulk Fermi levels. The final location of the Fermi level depends on the charge transfer to make Fermi level equal everywhere. The charge transfers from the bulk to surface or vice-versa creates a potential difference which leads to shifting of bands or band bending effects. An upward or downward band bending depends strongly on the presence of defects in the material and in the present case the upward band bending is related to the presence of Selenium vacancies in the Bi$_2$Se$_3$ samples as suggested by the study in Ref.34.