Probing Image Potential States on Topological Semimetal Antimony Surface

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A point charge near the surface of a topological insulator is predicted to generate an image magnetic charge in addition to an image electric charge as a result of electrostatic screening. We use scanning tunneling spectroscopy to study image potential states (IPS) of the topological semimetal Sb(111) surface. We observe five IPS with discrete energy levels well described by a one-dimensional model. Our investigation of the spatial variation of the IPS near surface step edges indicates an interaction between image charges. Our study of these IPS allows the exploration of the image charge geometry necessary to realize and manipulate a magnetic charge.

1 The surface states of topological material are spin-momentum locked, which reduces the local degrees of freedom and promotes unique electromagnetic responses. For example, when an external charge is located near the surface of a material hosting topological surface states, screening from surface charge will mimic an image electric charge [1]. As in a normal metal, the attractive potential between the real and image charges can give rise to bound states known as image potential states (IPS) [2]. However, if the material is topologically non-trivial, the magnetic and electric degrees of freedom are coupled by the topological magnetoelectric effect [3]. This effect opens a gap in the surface spectrum when time-reversal symmetry is broken, inducing a quantized Hall current and a magnetic monopole [4]. Thus the IPS in topological materials with broken time-reversal symmetry exhibit a combined image electric charge and image magnetic charge, which can be controlled by manipulating the real external electric charge near the surface. The manipulation of magnetic monopoles promotes applications such as circuitry based on magnetic charges [5].

2 Understanding the image electrical charge in topological materials is necessary to realize and control the image magnetic charge. The Coulomb potential between the external electric charge and its image results in a Rydberg series of energy levels below the vacuum energy level \( E_{\text{vac}} \)

\[
E_n = E_{\text{vac}} - \frac{0.85 \text{ eV}}{(n + a)^2}, \quad n = 1, 2, \ldots
\]  

(1)

where \( a \) is a correction factor of the crystal field [6]. Moreover, a strong electric field can alter \( E_n \) due to the Stark effect [7]. As a result, \( E_n \) is no longer a converging series below \( E_{\text{vac}} \), but the levels shift to higher energy and spread farther apart, as shown in Fig. 1(a).

3 IPS have been previously studied by \( z(V) \) spectroscopic measurements using scanning tunneling microscopy (STM) on surfaces of metals [8–11], but have yet to be reported for a topological material. Here we study single crystals of topological semimetal Sb, which were cleaved in cryogenic ultra-high vacuum and then loaded directly into the STM. All STM measurements were carried out at 5 K with mechanically cut PtIr tips cleaned by field emission on Au. The Sb crystals cleave on the (111) plane, which is atomically flat and free of defects in the 15 nm × 15 nm area shown in Fig. 2(a). Because of the electric field on the order of 1 V/nm applied between the tip and sample, IPS are all Stark-shifted. We use \( z(V) \) spectroscopy to measure the Stark-shifted IPS on Sb(111) surface and characterize the interaction of the charge with the topological surface states by the change in Stark shift with tip-sample junction setup conditions. Then we examine how states from distinct terraces evolve across a step edge, and we identify an extra novel state.

FIG. 1. Probing image potential states by STM. (a) Left: the Rydberg series of infinite discrete energy levels (left) approaches the vacuum \( E_{\text{vac}} \), as described by Eq. 1. Right: the quantized energy levels of image potential states can be shifted apart by stronger electric field (closer tip-sample distance, larger current), an effect known as the Stark shift. (b) A schematic drawing of the STM experimental setup. An electric image charge (red) is induced on a conducting sample when an external charge (blue) is placed above the surface. Together, these charges form a bound state (dashed oval). The measured tunneling current \( I \) is fed into a PID loop to adjust the tip-sample distance \( z \), by controlling the voltage applied on the \( z \) piezotube.
known, we define a reference tip-sample distance as the absolute tip-sample distance, manifested in a shift in the current leads to an abrupt retraction of the tip and a stepwise jump at IPS resonance voltages superimposed on a gradually increasing background. We observe two shifts of the $z(V)$ spectra as the current increases: the whole spectrum shifts down in the vertical axis; and the steps shift to the right in the horizontal axis. The shift in $z$ can be understood as the tip being pushed towards the sample to achieve a higher current at the same bias voltage. The shift in $V$ is the consequence of the Stark effect: with an increasing current (and thus decreasing $z$), the electric field increases and shifts the IPS to higher energies and farther apart.

6 In order to visualize IPS energy shifts more clearly, we numerically differentiate the $z(V)$ spectra and plot the $dz/dV(V)$ curves in Fig. 2(c). By counting the number of peaks, we observe five IPS up to 9.5 V. We fit each peak in Fig. 2(c) with a Lorentzian function and obtain $V_n$ from the centers of the Lorentzian peaks. Comparing Figs. 2(b) and (c), we extract $z_0$ relative to $z_0$ at each peak voltage $V_n$ and plot the values in Fig. 2(d). Fig. 2(d) shows that the Stark shift increases nonlinearly with increasing current and differs for each level index $n$. We also note that the peak width increases at higher $n$. The broadening of IPS peaks can be understood by elastic scattering of IPS electrons into the bulk continuum.

7 To quantitatively understand the data in Fig. 2(d), we use a one-dimensional (1D) model to describe the electrical potential in the vacuum space between tip and sample, assuming that the radius of the tip is much larger than the absolute tip-sample distance. The 1D potential $\phi$, as plotted in Fig. 3(a), is the sum of the linear electrostatic potential from the bias of the STM tunnel junction, the image potential of the tip, and the tip potential of the sample.

\begin{equation}
\phi(\zeta) = \phi_t - (\phi_t - \phi_s + eV) \frac{\zeta}{z} = \frac{\alpha_0^2}{4\pi\epsilon_0} \left(\frac{1}{\zeta} + \frac{1}{z - \zeta}\right).
\end{equation}

In Eq. 2, the variable $\zeta$ is the 1D spatial coordinate, which has an origin at the surface of the tip ($\zeta = 0$), $\phi$ is expressed relative to Fermi level of the tip $E_{F,t}$, $\alpha = 1.15\ln 2$ is a factor that accounts for all image charges, and $\epsilon_0$ is the vacuum permittivity. The parameter $V$ is the applied bias voltage controlled in experiments. There are three unknown parameters: $z$ is the absolute tip-sample distance, $\phi_t$ and $\phi_s$ are work functions of the tip and the sample, respectively. We use the potential $\phi(\zeta)$ to solve the 1D Schrödinger equation numerically with the Numerov method and find the resonance condition where $E_n$ coincides with Fermi level of the tip $E_{F,t}$, i.e. $E_n = 0$. Figure 3(b) shows an example of seven derived eigenenergies and eigenwavefunctions with parameters $V = 8.2$ V, $z = 3.0$ nm. A resonance at the $n = 4$ IPS ($E_4 = 0$) indicates $V_4 = 8.2$ V, $z_4 = 3.0$ nm, which can be compared to our data points in Fig. 2(d) with an adjustable parameter $z_0$. We fit the...
data in Fig. 2(d) using the method of least squares for each pair of $\phi_s$ and $\phi_t$ in the grid shown in Fig. 3(d), to minimize the root-mean-square error (RMSE) in $z_n$ between all experimental data in Fig. 2(d) and the model result. The best fit with minimal RMSE shown in Fig. 3(c) gives fit parameters $\phi_1 = 4.79$ eV, $\phi_s = 4.08$ eV, and the offset $z_0 = 1.26$ nm. Our $\phi_1$ and $\phi_s$ show reasonable agreement with the work functions of Au 5.1 eV and Sb 4.55-4.7 eV [16], respectively.

We next investigate the influence of surface defects on the IPS. Figure 4 shows laterally resolved IPS along a line across three surface steps. We observe bi-atomic-layer steps of the height 4.0 Å, as shown in Fig. 4(a), consistent with previous reports [17–19]. We acquire $z(V)$ spectra at each point on a line [inset of Fig. 4(a)], and plot the $dz/dV(x,V)$ map in Fig. 4(b). The energies of all the IPS peaks are constant far from the step edges but bend to higher (lower) energy near a step edge on the higher (lower) terrace. Despite their different terrace widths, the bipolar bending appears identical near all three step edges in $dz/dV(x,V)$ map. Similar bipolar bending has been observed in nanostructures such as NaCl/Ag(100) [20], Co/Au(111) [21], Li/Cu(100) [22], and defects on InAs(111) [23], and it is attributed to the change of surface potential between different materials [20, 21, 24]. The fact that we see the bipolar bending of IPS on an elementary material Sb indicates a local variation of chemical potential near a step edge. This variation can be understood by Smoluchowski effect [25] as illustrated in Fig. 4(c), where positive and negative charge builds up on the upper and lower sides near a step edge, respectively. The charge redistribution gives rise to a local dipole moment, which effectively acts as a lateral perturbation to the model potential in Eq. 2 [26].

9 We extract the IPS peak voltages in the same way as in Fig. 2, and plot the peak voltages of different $n$ as a function of distance in the $z$-direction in Fig. 4(d). The bipolar bending follows approximately an exponential decay in $x$, with a decay length of 1.5 ∼ 2.0 nm for different $n$. We also notice near the step in Fig. 4(d) that each IPS peak splits into two. This splitting may stem from the Stark effect caused by the additional local dipole moment, or spilling of IPS electrons from one side.
of the step to the other [21].

More surprisingly, there exists an extra weak state that departs from the \( n \) state on the higher terrace towards the \( n + 1 \) state on the lower terrace. This transition state is most obvious for \( n = 1 \) in Fig. 4(b), and it spans 2.5 nm across the step as shown by green dots in Fig. 4(d). For higher \( n \), due to the increased width of the IPS peaks, it is difficult to distinguish between the transition states and the bending IPS. The transition states demonstrate the crosstalk between IPS electrons from both sides of the step, indicating the interaction between image charges. The existence of crosstalk between image charges provides the possibility to control the interaction between the induced magnetic charges near a step edge in a topological material and to design nonlinear “magnetotronic” circuitry.

In summary, we observed the first five IPS up to 9.5 V on the topological semimetal Sb(111) surface. We describe the Stark-shifted IPS levels with a 1D model. Additionally, laterally resolved IPS across surface steps show bipolar bending of the IPS levels and interaction between IPS. Our study of IPS not only enriches the understanding of image electrical charges on surfaces of topological materials but also paves the way to locally manipulate and control the interaction between magnetic monopoles.

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