We study the fate of the surface states of Bi$_2$Se$_3$ under disorder with strength larger than the bulk gap, caused by neon sputtering and nonmagnetic adsorbates. We find that neon sputtering introduces strong but dilute defects, which can be modeled by a unitary impurity distribution, whereas adsorbates, such as water vapor or carbon monoxide, are best described by Gaussian disorder. Remarkably, these two disorder types have a dramatically different effect on the surface states. Our soft x-ray ARPES measurements combined with numerical simulations show that unitary surface disorder pushes the Dirac state to inward quintuplet layers, burying it below an insulating surface layer. As a consequence, the surface spectral function becomes weaker, but retains its quasiparticle peak. This is in contrast to Gaussian disorder, which smears out the quasiparticle peak completely. At the surface of Bi$_2$Se$_3$, the effects of Gaussian disorder can be reduced by removing surface adsorbates using neon sputtering, which, however, introduces unitary scatterers. Since unitary disorder has a weaker effect than Gaussian disorder, the ARPES signal of the Dirac surface state becomes sharper upon sputtering.

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Introduction.— An important hallmark of three-dimensional topological insulators are their protected Dirac-cone surface states, which connect bulk valence and conduction bands [1–3]. Since these surface states arise due to a nontrivial wave function topology in the bulk [4], they are robust to nonmagnetic disorder with strength $\gamma$ smaller than the bulk gap $\Delta$ [5–11]. Moreover, their existence is independent of the surface orientation [12,13] and the local surface chemistry. Protected topological surface states have been experimentally observed in numerous topological insulators, such as Bi$_{1-x}$Sb$_x$ [14], Bi$_2$Se$_3$ [15], Bi$_2$Te$_3$ [16], by both angle-resolved photoemission spectroscopy (ARPES) [17,22] and scanning tunneling experiments [23,24]. State of the art spin-resolved ARPES has allowed to map out the predicted helical spin texture of the surface states [25,29]. However, in recent experiments only little attention has been given to the fundamental property that gave these materials their name, namely the topological protection of the surface states against disorder [30,31].

The surface of Bi$_2$Se$_3$ and other Bi-based topological insulators, adsorbs H$_2$O, H$_2$, or CO molecules upon exposure to air or vacuum rest gas [32,34]. This introduces a large number of impurity scatterers. Furthermore, as a result of chemical bonding between the adsorbates and the surface, the number of surface carriers is increased, which leads to band bending and the development of two-dimensional surface quantum well states [32,33]. Additional scattering centers on Bi$_2$Se$_3$ surfaces are caused by step edges and Se vacancies, which host impurity bound states with energies of the order or larger than the band gap [30,35]. While the adsorbates lead to a dense distribution of relatively weak impurities, the step edges and Se vacancies introduce a dilute distribution of very strong scatterers. The former is commonly called Gaussian disorder [36–39] and the latter is known as unitary disorder. Unitary scatterers can also be artificially created on the surface by sputtering it with neon or argon ions [35]. Due to their topological protection, the surface states are robust against both types of disorder as long as the disorder strength $\gamma$ is smaller than the bulk gap $\Delta$ [5,8,38,40], but not necessarily otherwise.

In this Letter, we combine soft x-ray ARPES with numerical simulations to study how the surface states of Bi$_2$Se$_3$ are modified in the presence of strong disorder with $\gamma \gg \Delta$. We use UHV rest gas exposure to create Gaussian disorder and employ neon sputtering to introduce unitary disorder on the surface of Bi$_2$Se$_3$ (Fig. 1). Our numerical simulations show that the type of disorder matters for the topological insulator surface state. We find that Gaussian disorder with $\gamma \gg \Delta$ introduces a large number of impurity scatterers, which leads to a strong coupling between the bulk and surface states [40]. As a consequence, the momentum-space structure of the Dirac surface state is completely smeared out, leading to a surface spectral function that only exhibits broad features but no sharp quasiparticle peak (Fig. 2). Unitary disorder, on the other hand, creates a topologically trivial insulator at the surface, thereby pushing the Dirac state to inward quintuplet layers which are less disordered. Hence, unitary scatterers do not destroy the quasiparticle peak in the surface spectral function, but only reduce its sharpness and intensity. Our ARPES measurements of Bi$_2$Se$_3$ surfaces show that the spectral function becomes sharper upon sputtering (Fig. 3). This seemingly paradoxical observation is explained by taking into account the different disorder types of the surface adsorbates and the sputtering-induced impurities. That is, neon sputtering reduces the effects of Gaussian disorder by removing the surface...
adsorbates, which however comes at the expense of introducing unitary scatterers. Since unitary disorder has a weaker effect on the spectral function than Gaussian one, the quasiparticle peaks in the ARPES signal become more pronounced due to sputtering.

Numerical simulations.– To simulate the effects of disorder on the Bi$_2$Se$_3$ surface states, we employ a low-energy tight-binding Hamiltonian that describes the Bi-$p_z$ and Se-$p_z$ orbital bands close to the $Γ$ point of the Brillouin zone \([42]\).

The Hamiltonian can be conveniently expressed in terms of the spinor $\Phi_k = (|p_{z,k}^1, \uparrow\rangle, |p_{z,k}^2, \downarrow\rangle, |p_{z,k}^1, \downarrow\rangle, |p_{z,k}^2, \uparrow\rangle)$ as

$$\mathcal{H} = \frac{i}{2} \sum_k \Phi^*_k \mathbf{H}_k \Phi_k,$$

where $H_k$ is defined on a rhombohedral lattice with lattice constants $a = 4.08$ Å and $c = 29.8$ Å. \(\mathbf{H}_k\) contains a kinetic term $\epsilon_k = D_1[1 - \cos(k_y c)] + D_2[3 - 2 \cos(k_x \sqrt{3}a/2) \cos(k_y a/2) - \cos(k_y a)] - \mu$, a mass term $m_k = B_1[1 - \cos(k_y c)] + B_2[3 - 2 \cos(k_x \sqrt{3}a/2) \cos(k_y a/2) - \cos(k_y a)] + M$, and an interorbital coupling, which is parameterized by the vector $\mathbf{a}_k$ with the three components $a_{k}^0 = A_0 \sin(k_y c)$, $a_{k}^1 = A_1 \sqrt{3} \sin(k_x \sqrt{3}a/2) \cos(k_y a/2)$, and $a_{k}^2 = A_2 \cos(k_x \sqrt{3}a/2) \sin(k_y a/2) + \sin(k_y a)|.$

For the numerical calculations, the tight-binding parameters are determined by a fit of the energy spectrum of $H_k$ to that of ab initio DFT calculations \([42, 44]\). We observe that Hamiltonian \(\mathcal{H}\) satisfies time-reversal symmetry, but importantly breaks sublattice (chiral) symmetry \([11]\), which is in contrast to the model considered in Ref. \([40]\).

We implement surface disorder due to adsorbates and lattice defects by adding the potential $\delta \mathcal{H}$ to the Hamiltonian \(\mathcal{H}\), with

$$\delta \mathcal{H} = \sum_{\mathbf{k}, q} \sum_{a=G,U} \Phi^*_k \sigma_0 \otimes [V^m_{\alpha}(\mathbf{q}_\parallel)] \tau_3 + V^{\mu}_{\alpha}(\mathbf{q}_\parallel) \tau_0 \Phi_{k+q}$$.  

where $V^m_{\alpha}(\mathbf{q}_\parallel) = (1/\sqrt{N}) \sum_r v^m_{\alpha}(\mathbf{r}_r) e^{-i \mathbf{q}_\parallel \cdot \mathbf{r}_r}$ represents the Fourier transform of the uncorrelated random onsite potentials $v^m_{\alpha}(\mathbf{r}_r)$ at the surface sites $\mathbf{r}_r$. The disorder potential $\delta \mathcal{H}$ includes both local variations in the mass term ($a = m$) and in the chemical potential ($a = \mu$). For the surface adsorbrates we employ a Gaussian type disorder distribution ($a = G$), whereas for the lattice defects a unitary disorder distribution ($a = U$) is used. To realize the latter, we randomly choose $N_{\text{imp}}$ impurity sites $\mathbf{r}_i$ ($i = 1, 2, \ldots, N_{\text{imp}}$) at the surface, which all have the same large onsite potential $v^\mu_{\alpha}(\mathbf{r}_r) = 10^5$ eV. To implement the former, random potentials $v^G_{\alpha}(\mathbf{r}_r)$ at each lattice site $\mathbf{r}_r$ are drawn from a box distribution with width $\gamma_G$ and $p[v^G_{\alpha}(\mathbf{r}_r)] = 1/\gamma_G$ for $v^G_{\alpha}(\mathbf{r}_r) \in [-\gamma_G/2, +\gamma_G/2]$ \([36]\). Note that the strength of the Gaussian disorder is determined by the width $\gamma_G$ of the distribution, while the strength of the unitary disorlder is determined by the impurity density $\gamma_U = N_{\text{imp}}/N_{\text{tot}}$, where $N_{\text{tot}}$ is the total number of surface sites.

The effects of impurities on the Dirac surface state are best revealed by examining the momentum-resolved spectral function $A_l(\omega, \mathbf{k}_f)$ \([39, 40]\), which is directly proportional to the ARPES intensity. The spectral function in the $l$th quintuplet layer is given by

$$A_l(\omega, \mathbf{k}_f) = -\frac{\hbar}{4\pi} \Im \sum_{m, \nu} \frac{1}{\sqrt{N}} \sum_n \frac{|\Psi^m_{\nu,l}(\mathbf{r}_n) e^{-i \mathbf{k}_f \cdot \mathbf{r}_n}|^2}{\omega - E_m + i\eta},$$

where $\nu$ is the band index and $(E_m, \Psi^m_{\nu,l})$ represents the eigen-system of Hamiltonian \(\mathcal{H}\) with surface disorder \(\delta \mathcal{H}\). Using exact diagonalization algorithms \([43]\), we evaluate expression \(3\) for a (001) slab of dimension $25 \times 25 \times 25$ and an intrinsic broadening of $\eta = 0.02$ eV.

Figure 2 shows the spectral function for the first three outermost layers in the presence of unitary scatterers and Gaussian disorder with strength $\gamma_G = 1$ eV in panel (a) and $\gamma_G = 5$ eV in panel (b). We observe that Gaussian disorder with $\gamma_G = 1$ eV, which is of the order of the bulk gap $\Delta$, does not alter the surface states, apart from small broadening effects. That is, the Dirac cone surface state is clearly visible as a sharp quasiparticle peak in $A_l(\omega, \mathbf{k}_f)$, which decays exponentially into the bulk on the length scale of about three quintuplet layers [left column in Fig. 2(a)]. However, for Gaussian disorder with $\gamma_G = 5$ eV, the momentum-space structure of the Dirac state is completely destroyed. This is due to a large number of impurity bound states at the surface, which leads to a strong coupling between bulk and surface states. Thus, $A_l(\omega, \mathbf{k}_f)$ in the presence of very strong Gaussian disorder exhibits only broad humps, but no sharp features [left column in Fig. 2(b)]. It is important to note that even though the disorder potential completely breaks translation symmetry along the surface, the $\mathbb{Z}_2$ topological invariant in the
The density of surface defects is increased from $\gamma = 0\%$ in the left columns to $\gamma = 40\%$ in the right columns. The same color scale is used for all subpanels. The effects of surface adsorbates is simulated by Gaussian disorder with (a) $\gamma = 1\%$ and (b) $\gamma = 5\%$. To mimic the sputtering process, the density of surface defects is increased from $\gamma = 0\%$ in the left columns to $\gamma = 40\%$ in the right columns. (c) Energy-resolved distribution of the local density of states $P[\tilde{\gamma}(\omega)]$ for the surface and the first inward quintuplet layer as a function of Gaussian disorder strength. The solid line represents the maximum of the distribution, whereas the triangles and inverted triangles indicate the left and right standard deviation, respectively [41]. The insets show the probability distributions for the disorder strength $\gamma = 5\%$.

To simulate the sputtering process, we increase the density of surface defects from $\gamma = 0\%$ to $\gamma = 40\%$. The right columns of Figs. 2(a) and 2(b) show that for $\gamma = 40\%$ the Dirac state is pushed to the second and third inward quintuplet layers, since the surface layer ($l = 1$) becomes more and more insulating. As a result the Dirac state reappears in the spectral function of the second and third layers as well-defined quasiparticle peaks.

**SX-ARPES measurements.**—In order to measure the effects of sputtering on the Dirac surface state using ARPES, it is necessary to employ large incident photon energies, such that the probing depth is considerably larger than one Bi$_2$Se$_3$ unit cell. Therefore, instead of conventional UV-ARPES, we used soft x-ray (SX) ARPES with a probing depth, defined as 3 mean free path lengths, of about 30 Å [47, 48], which corresponds to three Bi$_2$Se$_3$ quintuplet layers. The photoemission experiments were performed on in-situ cleaved single crystals of Bi$_2$Se$_3$ at the ADDRESS beamline of the Swiss Light Source using photon energies of $h\nu = 380\text{ eV and } h\nu = 725\text{ eV}$. To prevent freezing of the sputtering agent, the samples were cleaved and sputtered at room temperature. The SX-ARPES measurement, however, was carried out at the low temperature of 10 K, since otherwise the photoemission signal would be too blurry, due to the loss of spectral coherence as expressed by the large Debye-Waller factor [49, 50].

Our aim was to investigate how the surface spectral functions is changed as the density of unitary scatterers is increased, while the Gaussian disorder of the surface adsorbates is kept constant. For that purpose, it was crucial to keep the times betweencleaving, sputtering, cooling, and measuring the crystals fixed, such that all samples are exposed for the same duration to the UHV rest gas at a base pressure better than $5 \times 10^{-10}\text{ mbar}$. The time between cooling down the sample and measuring was of the order of half an hour, resulting in the adsorption of a significant amount of H$_2$, CO, and H$_2$O molecules. Different densities of unitary disorder were introduced by irradiating the Bi$_2$Se$_3$ surface for different time periods with Ne$^+$ ions with an energy of 0.7 keV at a pressure of $3 \cdot 10^{-6}\text{ mbar}$. For every sputtering cycle a new sample was cleaved under identical circumstances. After sputtering, the sample was immediately in-situ transferred to the measurement chamber to start the SX-ARPES experiment. To assure the reproducibility of the results the sample surface was scanned for homogeneity and the measurements, especially for the unsputtered and lightly sputtered samples, were repeated several times. All data shown here was obtained under identical conditions with respect to beamline and analyser settings, and integration time.

In Fig. 3 we present the SX-ARPES band maps and mo-
momentum distribution curves (MDCs) as a function of surface momentum along the $\Gamma$–$M$–$\Gamma$ direction of the (001) surface Brillouin zone. The soft x-ray photon energy was taken to be $h\nu = 380$ eV and the sputtering time was increased from 0 s in panel (a) to 112 s in panel (e). Additional SX-ARPES measurements with a photon energy of $h\nu = 725$ eV are presented in Fig. S1 of the SM [41].

**Discussion.**—We start by discussing the spectra of the unsputtered sample, Figs. 3(a) and 3(f). In the band map of Fig. 3(a) the bulk valence bands of the Bi- and Se-orbitals are clearly visible at $\sim 0.5$ eV and $\sim 1.2$ eV, respectively. Near the $\Gamma$ point these valence bands exhibit a characteristic “M-shape” dispersion, which agrees with ab initio DFT calculations [21, 42]. The Dirac surface state, however, is barely detectable, neither at $h\nu = 380$ eV nor at $h\nu = 725$ eV [41]. This is due to the large amount of surface disorder introduced by the adsorbates, which smears out the quasiparticle peak of the Dirac state. Indeed, we find that the ARPES band map of Fig. 3(a) is in good agreement with the simulated spectral function of the $\Gamma$–$M$–$\Gamma$ surface Brillouin zone. That is, the electron energies are chosen such to select a $k_z$, where the bulk conduction band is not observable. To our best knowledge this constitutes the first measurement of the surface state of a topological insulator at such high photon energies.

Sputtering the surface for a few seconds reduces Gaussian disorder by removing adsorbates, but creates local defects, which increases unitary disorder. As a net effect, we find that the quasiparticle peaks in the spectral function of the bulk and surface bands become considerably sharper [Figs. 3(b) and 3(g)]. The “V-shaped” dispersion and the Dirac point of the surface state, which is located at $\sim 0.3$ eV, are now clearly discernible. This observation corresponds well with the calculated spectral function of the middle column of Fig. 2(b), where the sputtering process was simulated by adding 20% surface vacancies. For longer sputtering times the concentration of surface defects is further increased, but the Dirac surface state remains visible, albeit with a broader quasiparticle peak [Figs. 3(c) and 3(d)]. This observation can be explained by noting that a large density of defects eventually leads to an insulating surface layer, thereby pushing the Dirac surface state to the second and third inward unit cells. Since SX-ARPES is less sensitive to the second and third unit cells, the spectral function intensity of the Dirac state becomes weaker. This interpretation is confirmed by our numerical simulations of Fig. 2 which show that for 40% defect density most of the spectral weight of the Dirac state is concentrated in the second and third quintuplet layers.

Finally, we find that sputtering for more than one minute radically changes the topology of the $\Gamma$–$M$–$\Gamma$ surface. That is, the entire crystal surface is cracked up into multiple tilted domains with sizes comparable to the synchrotron beam spot ($74 \times 30$ µm). As a result, the ARPES spectra contain contributions from several domains with relative shifts in surface momenta, since the normal photoemission angle now sensitively depends on the incident position of the synchrotron light [Fig. 3(e)]. While a broad signature of the bulk bands can still be observed, the Dirac surface state is completely absent in the spectra of the strongly sputtered samples. This indicates that the surface of $\text{Bi}_2\text{Se}_3$ has been rendered topologically trivial up to a depth of more than three unit cells, i.e., beyond the...
probing depth of SX-ARPES.

Conclusion.– In conclusion, we have investigating how topological insulator surface states are affected by disorder caused by neon sputtering and surface adsorbates. By comparing numerical simulations to SX-ARPES measurements of Bi$_2$Se$_3$, we have shown that the surface adsorbates correspond to Gaussian disorder, while the local defects introduced by sputtering represent unitary scatterers. We have demonstrated that the effects of Gaussian disorder can be reduced by sputtering, which removes surface adsorbates but at the same time introduces defects and vacancies. Since the latter have a weaker effect than Gaussian disorder, the ARPES signal of Bi$_2$Se$_3$ surfaces that have been exposed to air (or UHV rest gas) become sharper upon sputtering. Our findings demonstrate the extreme robustness of the Bi$_2$Se$_3$ surface state against any type of surface disorder, thereby confirming its topological origin.

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Supplemental Material for
“Sputtering induced re-emergence of the topological surface state in Bi$_2$Se$_3$”

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We present in this supplemental material additional SX-ARPES measurements and give the definition of the density of states distribution.

I. Additional SX-ARPES spectra

Figure S1 shows the SX-ARPES band maps and momentum distribution curves with a photon energy of $h\nu = 725$ eV. This figure should be compared to the SX-ARPES spectra with $h\nu = 380$ eV, which are depicted in Fig. 3 of the main text. SX-ARPES with $h\nu = 725$ eV is sensitive to a different region of the bulk Brillouin zone than SX-ARPES with $h\nu = 380$ eV. This is the reason why the bulk spectrum in Fig. S1 is quite different from the one of Fig. 3. The Dirac surface state, however does not have any $k_z$ dispersion, and therefore its photoemission spectrum does not depend on photon energy $h\nu$. Moreover, we find that the dependence on surface sputtering is qualitatively similar to the one of Fig. 3.

![Figure S1](image_url)

**FIG. S1.** SX-ARPES measurements of the (001) Bi$_2$Se$_3$ surface with a photon energy of $h\nu = 725$ eV. The upper row [panels (a) to (e)] shows the band maps along the M–Γ–M crystallographic direction for different sputtering times. The ARPES intensity is indicated by the grey scale, with black and white corresponding to high and low intensity, respectively. The lower row [panels (f) to (j)] displays the momentum distribution curves (MDCs) for binding energies ranging form 0 meV to 442 meV with a spacing of 13 meV. For clarity each MDC is shifted vertically.

II. Distribution of local density of states

Information about the localization or delocalization properties of the Dirac surface state can be obtained by computing the distribution of the local density of states (LDOS) $P[\tilde{\rho}_l(\omega)]$. The probability distribution $P[\tilde{\rho}_l(\omega)]$ is defined in terms of the normalized LDOS on the $l$th quintuplet layer

$$\tilde{\rho}_l(\omega) = \frac{\rho_l(\omega)}{\langle \rho_l(\omega) \rangle},$$

(4)
where $\langle \tilde{\rho}_l(\omega) \rangle$ denotes the mean value of the LDOS. The distributions shown in Fig. 2(c) in the main text have been computed for an ensemble of one thousand disordered Hamiltonians with periodic boundary conditions along the $x$ and $y$ directions.

We note that a distribution $P[\tilde{\rho}_l(\omega)]$ centered at $\tilde{\rho}_l(\omega) = 1$ indicates that the majority of states are delocalized, while a distribution peaked at $\tilde{\rho}_l(\omega) = 1$ signals that most of the states are localized. The left (right) tails of the distribution give us insight, whether there exists a minority of localized (delocalized) states. To quantify the size of these tails we introduce the left and right standard deviations, $\sigma_L$ and $\sigma_R$, of $P[\tilde{\rho}_l]$, which are given by

$$\sigma_L^2 = \left( \sum_{\tilde{\rho}_l = 0}^{\rho_{\text{max}}} P[\tilde{\rho}_l] - P[\rho_{\text{max}}] \right)^{-1} \sum_{\tilde{\rho}_l = 0}^{\rho_{\text{max}}} 2 P[\tilde{\rho}_l] (\tilde{\rho}_l - \rho_{\text{max}})^2,$$

(5)

$$\sigma_R^2 = \left( \sum_{\tilde{\rho}_l = \rho_{\text{max}}}^{\infty} P[\tilde{\rho}_l] - P[\rho_{\text{max}}] \right)^{-1} \sum_{\tilde{\rho}_l = \rho_{\text{max}}}^{\infty} 2 P[\tilde{\rho}_l] (\tilde{\rho}_l - \rho_{\text{max}})^2.$$

(6)

In the above definitions we have treated $\tilde{\rho}_l > \rho_{\text{max}}$ and $\tilde{\rho}_l < \rho_{\text{max}}$ as independent distributions, where $\rho_{\text{max}}$ is the maximum of the distribution $P[\tilde{\rho}_l]$. The numerical results were obtained for 1000 disorder configurations of three dimensional lattices in real space, with $25^3$ lattice sites and periodic boundary conditions along the $(x, y)$ directions.