

Impact of static disorder on quasiparticle spectra: Debye-Waller, mean free path, and potential fluctuation effects

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Angle-resolved photoemission spectroscopy (ARPES) is a widely used characterization technique in condensed matter physics, providing direct access to the single-electron spectral function of crystals, including their electronic band structure and Fermi surface. Measuring the band structure of novel quantum materials has been fundamentally important for determining, for example, nontrivial band topology, as in topological insulators, Weyl semimetals, and Dirac semimetals, or for identifying new classes of materials, such as altermagnets. A key challenge with these emerging quantum materials is that their initial crystalline quality is rarely optimized, which directly affects the spectra measured by ARPES. Here, we present a theoretical framework and experimental evidence addressing two common consequences of static disorder in photoemission experiments: the loss of coherent spectral weight and the broadening of spectral features. ARPES spectra can be understood as a sum of coherent and incoherent intensities, with their relative contributions controlled by atomic disorder and electron correlation effects. For disorder caused by phonons, the coherent intensity is exponentially suppressed as temperature increases, a phenomenon analogous to the Debye-Waller factor in diffraction, where Bragg peaks diminish in favor of diffuse scattering as disorder increases. In this work, we report a soft-x-ray study of the deliberately disordered (via Ar ion sputtering) InAs(110) surface, characterized by scanning tunneling microscopy, scanning tunneling spectroscopy, and low-energy electron diffraction. We introduce a framework that enables quantification of coherent photoemission intensity loss with increasing disorder, allowing both thermal and static disorder to be treated within a unified approach. Additionally, we identify a second major effect of disorder beyond lifetime broadening: inhomogeneous spectral broadening arising from local potential fluctuations. We show that such fluctuations increase the linewidths of the spectra of localized and delocalized states and contribute to the suppression of ARPES intensity from states near the Fermi level. The concepts and analysis methods presented should make ARPES useful for direct diagnosis of disorder effects on electronic states, for science as well as engineering.

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I. INTRODUCTION

Angle-resolved photoemission spectroscopy (ARPES) accesses a plethora of information about the occupied electronic states of crystalline solids, including single-electron spectral functions, band structure, and Fermi surface [1,2]. The surface/bulk sensitivity can be actively tuned by the photon energy, $h\nu$. In particular, the use of soft x-rays, where $h\nu$ spans from 300 eV up to 2000 eV, rather than extreme ultraviolet radiation, enhances the bulk sensitivity. This generally increases the inelastic mean free path (IMFP) of the photoexcited electrons in the final states of the photoemission

process, with typical values of 1–2 nm, limited primarily by electron-electron interactions [3]. The consequent increase of bulk sensitivity allows soft-x-ray ARPES (SX-ARPES) to optically probe the electronic structure of three-dimensional (3D) materials and buried interfaces [1,4–6].

Of special interest for both conventional and “quantum” technologies are heterostructures, which incorporate quantum wells [7], as well as metal-semiconductor [1] and superconductor-semiconductor interfaces [8–10] for a variety of applications. For example, the narrow-gap semiconductor InAs coated with superconducting aluminum has attracted attention as a host for Majorana zero modes, due to appreciable spin-orbit coupling and a large Landé g factor [11–13]. Disorder limits the robustness of these states [14], making it highly desirable to use ARPES to investigate such effects. More broadly, band-structure measurements of novel quantum materials are essential for establishing nontrivial band topology (as in topological insulators, Weyl semimetals, and Dirac semimetals [15,16]) or for identifying new classes of materials such as altermagnets [17]. A key challenge is that

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the initial crystalline quality of such emerging materials is rarely optimized, which directly affects the spectra measured by ARPES. In this work, we present a theoretical framework together with experimental evidence for the generic consequences of static disorder in photoemission experiments.

Generally, ARPES spectra can be decomposed into two components: (1) a sharp, momentum-dependent coherent component (I_{coh}) that originates from Bloch electrons in the unperturbed crystal structure and (2) a (usually) broad incoherent component (I_{inc}) that originates from thermal vibrations, elastic scattering from defects, and many-body physics. A theory that is well established in reproducing ARPES spectral intensity is the one-step model introduced by Pendry [18], Feibelman [19], Mahan [20], and Schaich and Ashcroft [21], and used to successfully model many condensed matter systems, with $h\nu$ ranging between a few eV and 10 keV [22]. In this framework, all electrons in the unperturbed crystal are described by Bloch waves and the states of the photoexcited electrons are identical to time-reversed low-energy electron diffraction (LEED) states [23]. Consequently, it is not surprising that quasiparticle peaks in ARPES spectra behave similarly to Bragg peaks in LEED. For example, thermal vibrations and uncorrelated defects in LEED transfer intensity from sharp diffraction spots to a background homogeneous in momentum space ($\mathbf{k}$ space) [24]. In ARPES, we expect a similar transfer from quasiparticle peaks in I_{coh} to the I_{inc} component. This has already been established for thermal disorder in the seminal work of Shevchik [25–27], who showed that the relation between I_{coh} and I_{inc} can be described in terms of the Debye-Waller factor (DWF), as discussed in Sec. III C 1. However, no progress has been made thus far in developing a similar intuitive description for static disorder. We show here that an extended version of the approach of Shevchik [25–27] to the DWF explains the impact of deliberately introduced static disorder in a model system, namely, InAs(110) damaged by sputtering.

Additionally, static disorder in the form of charged defects in insulators and weakly doped semiconductors induces spectral broadening through two distinct mechanisms: (1) elastic scattering of initial states by the disorder potential and (2) broadening caused by spatial variations in band bending [28], referred to as inhomogeneous broadening in this manuscript. The first mechanism, discussed in Sec. III C 2, is characterized by a Lorentzian lineshape of the quasiparticle peaks and depends on the spacing between defects, scaling as $c_d^{1/3}$ in 3D, where c_d is the defect concentration. The second mechanism, discussed in Sec. III C 3, becomes relevant in insulators and semiconductors when the density of compensated charged defects in the crystal is higher than the electron (or equivalently, hole) density. In this case, Coulomb defects are only partially screened, leading to potential fluctuations, also referred to as variable band bending. In particular, this broadening mechanism becomes significant when the screening length of the system is comparable to the interdefect distance. In such cases, the potential fluctuations extend throughout the whole crystal volume. When spectra are measured using a beam spot size several orders of magnitude larger than the screening length, the resulting spectra represent an average over all potential configurations. This mechanism, unlike (1), is char-

acterized by Gaussian lineshapes of the quasiparticle peaks and scales as $c_d^{1/2}$. These fluctuations lead to inhomogeneous broadening, affecting not only tightly bound core-level spectra but also the spectra of delocalized valence and conduction states along the energy coordinate. This mechanism has been suggested in previous theoretical studies [29] to explain the large spectral linewidths observed in high- T_c cuprates in the insulating phase, as measured using ARPES in Refs. [30–33]. While different explanations have been proposed for these observations, such as in Ref. [34], we provide compelling evidence in this paper for the universal nature of the inhomogeneous broadening mechanism in insulators and weakly doped semiconductors. In fact, our experimental data show that the linewidths of the valence bands (VBs) and the In 4d core levels exhibit a consistent square-root dependence on the disorder concentration. A direct consequence of the fluctuating potential in the presence of a finite electron density is that when the ratio between the bandwidth of the conduction electrons and the disorder strength is smaller than unity, electrons form disconnected puddles. These puddles, in the low-temperature regime, manifest in transport measurements with the typical conductivity of Efros-Shklovskii variable-range hopping [35,36]. This phenomenon has no intrinsic energy scale, as it depends on the aforementioned ratio. In particular, it is also highly relevant for the quantum applications mentioned above. In our experiment, variable band bending leads to a measurable decrease in the intensity of the conduction states.

Therefore, as shown schematically in Fig. 1, the two main impacts of static disorder on ARPES spectra measured on crystals are (1) the transfer of intensity from the coherent component, I_{coh} , to the incoherent component, I_{inc} , in exact analogy to what happens in crystallography, where Bragg peaks persist while losing intensity to diffuse scattering as disorder is increased, and (2) spectral broadening, resulting from a reduced lifetime of initial states due to electron-defect scattering, as well as from inhomogeneous broadening caused by spatial fluctuations in the potential.

II. METHODS

The ARPES measurements were performed at the soft-x-ray ARPES facility [5] of the ADRESS beamline [37,38] of the Swiss Light Source, Paul Scherrer Institut, Switzerland. The sample temperature was constant at $T = 12$ K, which minimizes the loss of spectral weight due to thermal disorder [39], and as discussed in Sec. III C 1, restricts the main visible effect to a mere prefactor weighting the coherent quasiparticle intensity.

We used vertically polarized light (p -polarized in the actual experimental geometry) at three different values of $h\nu$: 517, 815, and 1159 eV. The k_z component of the final wave vector can be calculated from the experimental geometry, the energy, and the momentum of the photons [38]. The above photon energies were selected so that the calculated k_z would correspond to the Γ point, at $k_x, k_y = 0$. Furthermore, the $h\nu$ and the (110) orientation of the InAs surface allowed us to measure the band structure along the Γ KX high-symmetry direction. This is the ideal direction to probe the valence bands due to the relatively large momentum-dependent separation

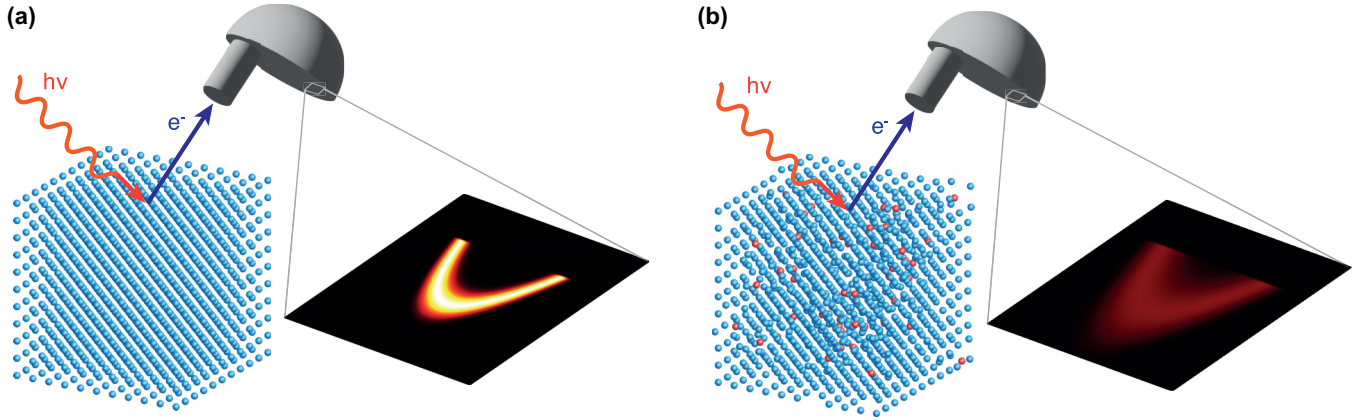


FIG. 1. (a) Schematic representation of an ARPES experiment performed on a well-ordered crystal (left), along with the corresponding schematic of a parabolic spectral function. (b) Schematic representation of an ARPES experiment performed on a disordered crystal, where the coherent intensity of the spectra is weakened and the lineshapes are broadened.

of the heavy-hole (HH), light-hole (LH), and split-off (SO) bands, as shown in Fig. 2(a).

The samples used for SX-ARPES, scanning tunneling microscopy (STM), scanning tunneling spectroscopy (STS), and LEED experiments, with dimensions of $4 \times 4 \text{ mm}^2$, were cut from an undoped InAs(110) wafer grown by Wafer Technology LTD. The samples were mounted on copper Omicron plates using epoxy (EPO-TEK[®] H21D) and subsequently cured at 150°C for 5 min. Next, a metal cleavage post was attached to the top surface of the samples using the same epoxy and curing procedure. After insertion into ultrahigh vacuum, the samples were thermalized at 10 K for the SX-ARPES experiment and 80 K for STM, STS, and LEED, and subsequently cleaved by removal of the cleavage post. To artificially introduce defects into the crystal structure, InAs(110) substrates were cumulatively sputtered at room temperature with Ar ions in steps of $\approx 20 \text{ s}$. The incident ion energy was 3 keV, and the partial pressure of Ar was $5 \times 10^{-8} \text{ mbar}$. Thermal annealing was not performed to ensure that the sputter-induced disorder persisted. After each sputter cycle, SX-ARPES measurements with equal integration time were carried out to record the evolution of the spectra as a function of the Ar^+ ion dose. To assess the impact of sputtering-induced disorder, we analyzed the lineshapes of the quasiparticle peaks in the valence band region. Our approach involved separation, as explained in Appendix F, of the coherent signal from the incoherent background. From the processed spectrum measured at 517 eV (815 eV, 1159 eV), we extracted 20 (15, 10) momentum distribution curve (MDCs) within the binding energy range of $[-4 \text{ eV}, -2 \text{ eV}]$ ($[-3.5 \text{ eV}, -2 \text{ eV}]$, $[-3.5 \text{ eV}, -2.5 \text{ eV}]$), using an integration window of $\pm 0.03 \text{ eV}$. Each coherent peak within these MDCs was fitted using the procedure described in Appendix G. This step was repeated for data obtained after each sputtering cycle, normalizing each peak area to the initial peak area before sputtering. The coherent residue for a given Ar^+ dose was calculated by averaging the normalized peak areas across all MDCs. In addition, as further detailed in Appendix G, we analyzed the FWHM of the peaks to evaluate the broadening effects. Another analysis was performed on the conduction band region measured at 517 eV and high-

lighted by the expanded boxes in Figs. 2(a)–2(c). The size of these boxes in k_x is $[-0.1 \text{ \AA}^{-1}, 0.1 \text{ \AA}^{-1}]$, while along E_b it is $[-0.2 \text{ eV}, 0.1 \text{ eV}]$. To extract the total coherent intensity from these regions, we integrated over the E_b coordinate, then subtracted a linear background, and finally fitted the remaining peak with a Gaussian.

To directly characterize the nature and density of the sputter-induced defects responsible for the changes in the ARPES data and the disordered potential landscape, we performed STM, STS, and LEED measurements at the PEARL beamline of the Swiss Light Source, Paul Scherrer Institut. The STM and STS data were acquired using an Omicron low temperature STM. The PtIr tip (90% Pt and 10% Ir) was prepared by cutting a PtIr wire and subsequently conditioned in ultrahigh vacuum by repeated voltage pulses and controlled tip crashes into a clean Au(111) crystal surface, until stable, high-resolution images without tip artifacts were obtained and the surface-state step in the density of states of Au(111) near -0.5 V was observed. All STM and STS measurements were performed at 4.4 K. For the STS measurements, we recorded dI/dV curves on a 10×10 grid over a $100 \times 100 \text{ nm}^2$ area. All measured spectra as a function of bias V are shown together in Fig. 15 of Appendix J. If the work-function difference between the STM tip and the sample is nonzero, the tip can induce local band bending in the sample and create a so-called tip-induced quantum dot [40]. The energy (bias) position of the lowest peak corresponds to the ground-state energy of the tip-induced quantum dot, averaged over the spatial extent of the ground-state wave function centered at each measurement location [41]. This quantity is used to extract the coarse-grained potential landscape shown in Fig. 5(a).

III. RESULTS AND DISCUSSION

A. SX-ARPES measurements

Figure 2 presents the soft-x-ray angle-resolved photoemission spectroscopy data along the ΓKX high-symmetry line at $h\nu = 517 \text{ eV}$, with increasing Ar^+ fluences. In Fig. 2(a), we clearly identify the HH, LH, and SO valence bands. Figures 2(a)–2(c) show the ARPES data in ascending order of the Ar^+ dose, demonstrating that the coherent component of the

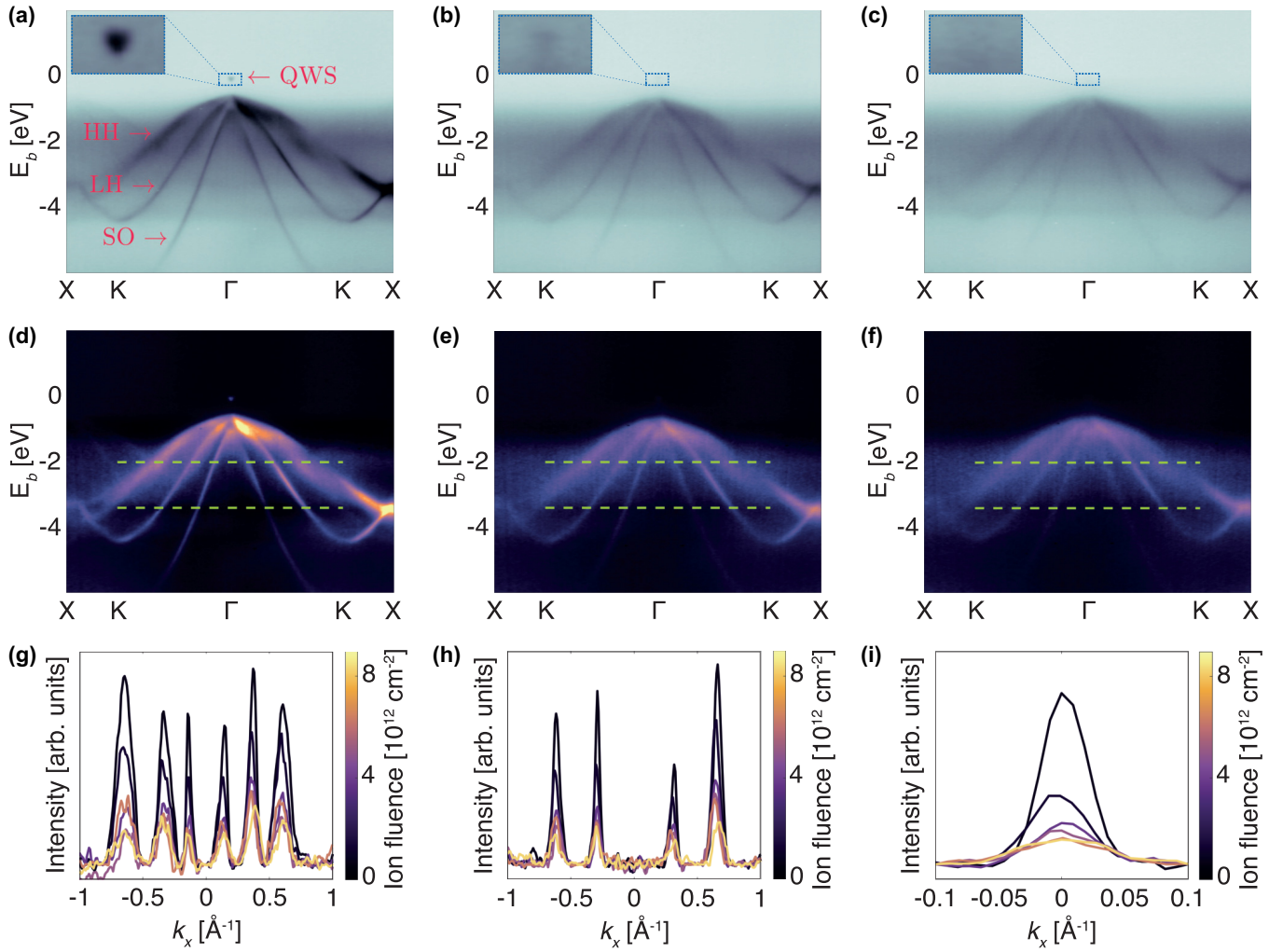


FIG. 2. (a)–(c) Band structure of InAs along Γ KX measured at $h\nu = 517 \text{ eV}$. (a)–(c) Total ARPES intensity shown in ascending order of Ar^+ fluence: $D = 0 \text{ cm}^{-2}$, $D = 4.5 \times 10^{12} \text{ cm}^{-2}$, and $D = 9 \times 10^{12} \text{ cm}^{-2}$. In the upper left corner, an expanded image of the quantum well states (QWSs) is shown for all three spectra. The HH, LH, SO, and QWS bands are individually identified in panel (a). (d)–(f) Same data as above, after subtraction of the k -integrated background. The colored lines indicate the positions where the MDCs shown in panels (g) and (h) have been extracted. (g)–(h) MDCs measured after increasing Ar^+ doses, as indicated by the color bar on the right. (i) Intensity of the expanded boxes in panels (a)–(c) after integration over E_b and subtraction of a linear background.

valence bands remains robust against sputtering, as HH, LH, and SO can still be resolved in Fig. 2(c).

However, with increasing Ar^+ doses, the relative intensity of the coherent component decreases, and a momentum-independent intensity emerges across the entire Brillouin zone. The interplay between coherent and incoherent components depends critically on the degree of static disorder introduced by ion bombardment. We explain this phenomenon by introducing the coherence residue F_C , as discussed in Sec. III C 1.

In Figs. 2(d)–2(f), we present the ARPES data after subtracting the k -integrated background. For the cleaved surface shown in Figs. 2(a) and 2(d), we observe a pocket occupied by quantum well states originating from the conduction band at the Γ point. These states disperse just below E_F and approximately 0.4 eV above the valence band maximum. They are induced by defects, which act like donors, creating a charge accumulation in the vicinity of the InAs(110) surface [42]. As shown in the expanded images in the upper left corner of

Figs. 2(a)–2(c), the QWS intensities appear to decrease at a faster rate with disorder compared to the valence band signal, which also displays a slight broadening with increased sputtering. Sections III D and III E present a quantitative analysis.

B. Spatial distribution of defects

Understanding the spatial distribution of sputter-induced defects is crucial for accurately interpreting the ARPES data. In this experiment, we employ a very low ion flux, several orders of magnitude lower than what is typically used for semiconductor surface preparation. This choice is motivated by the aim of preserving surface crystallinity, to investigate the effects of point defects on the momentum-resolved one-electron spectral function. Simultaneously, our method of introducing defects in a controlled manner also provides valuable insights into the ion sputtering technique itself, which is one of the most widely used surface preparation methods in semiconductor research and industry. The effects of individual

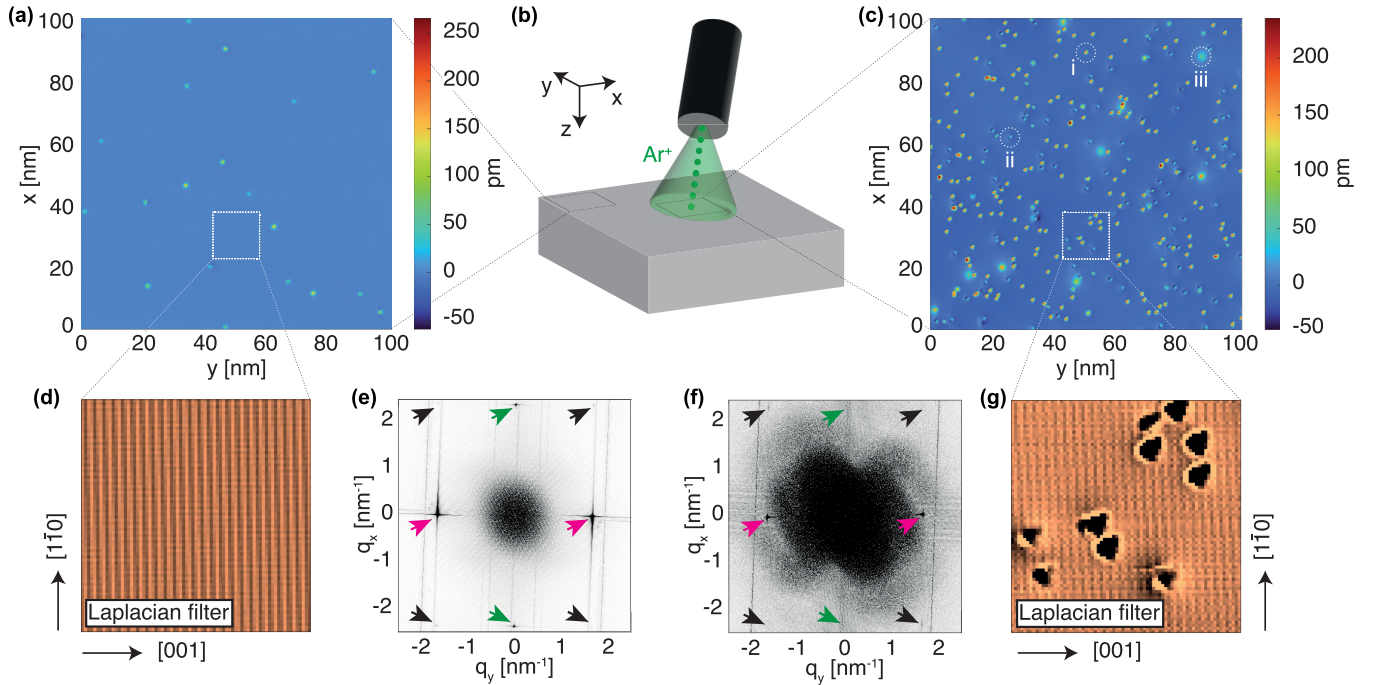


FIG. 3. Scanning tunneling microscope image of a $100 \times 100 \text{ nm}^2$ region of an InAs(110) surface (a) after cleavage and (c) after being subjected to a $D = 3 \times 10^{12} \text{ cm}^{-2}$ ion dose, acquired with $I = 100 \text{ pA}$ and $V = 0.4 \text{ V}$. (b) Schematic representation of an InAs crystal being sputtered with Ar^+ ions. (d), (g) Expanded images of the $15 \times 15 \text{ nm}^2$ squares shown in panels (a) and (c) and after applying a Laplacian filter to enhance edges. (e), (f) Absolute value of the Fourier transform of panels (a) and (c), respectively. The arrows indicate the Bragg peaks visible on both the pristine and disordered surfaces.

ions on semiconductor surfaces have been poorly explored experimentally, resulting in only a few published papers such as Refs. [43–45].

To investigate the in-plane distribution of defects, we performed both STM and LEED on *in situ* cleaved InAs(110) surfaces, before and after sputtering, to image defects in both real and momentum space. For the STM measurements, we examined a clean surface and the same surface after a total ion dose of $3 \times 10^{12} \text{ ions cm}^{-2}$, using the conditions described in Sec. II. The resultant topographic images, measured at $I = 100 \text{ pA}$ and $V = 0.4 \text{ V}$, are presented in Figs. 3(a) and 3(c). Figure 3(a) shows a $100 \times 100 \text{ nm}^2$ region of the freshly cleaved InAs(110) surface, revealing fewer than 20 defects. The origin of these defects is attributed to the cleavage procedure. Figure 3(c) illustrates the topography of the surface after sputtering. In particular, this image reveals a significantly higher concentration of defects than the nonsputtered surface. Most importantly, however, the STM image demonstrates that the underlying blue region, outlined by the $15 \times 15 \text{ nm}^2$ dashed square and more clearly visible in the magnified view in Fig. 3(g), preserves long-range lattice order. The absolute value of the Fourier transform of the STM image in Fig. 3 reveals Bragg peaks at $q_x = 2.37 \text{ nm}^{-1}$ and $q_y = 1.65 \text{ nm}^{-1}$, which correspond to the expected lattice constants $a_{(1\bar{1}0)} = a_{(001)}/\sqrt{2} \approx 0.42 \text{ nm}$, and $a_{(001)} \approx 0.61 \text{ nm}$, along the x and y directions, respectively.

Regarding the type of defects induced by Ar sputtering, the red features marked by circle i in Fig. 3(c), and the half-dark, half-bright blue regions marked by circle ii in Fig. 3(c), indicate spots where Ar ions have penetrated the crystal surface.

These defects are more clearly visible in the magnified and Laplace-filtered image in Fig. 3(g). The difference between i and ii is attributed to the different charge states of the defects. It is also worth noting that these types of defects are absent on the pristine surface, confirming that they are related to the sputtering process. Additionally, the feature indicated by the dashed circle iii in Fig. 3(c) displays bright blue, blurred circular areas, which most likely correspond to sputter-induced charged defects located beneath the surface. Finally, as discussed in Sec. III C 1, the key to applying the Debye-Waller picture to static disorder is the notion that point defects, in this case generated by Ar^+ bombardment, induce distortions in the surrounding lattice. However, these displacements must occur on a length scale comparable to the wavelength of the final states in photoemission, approximately $\sim 0.1 \text{ \AA}$ in the soft-x-ray regime. This scale cannot be directly resolved in real space in the data shown in Fig. 3, although the reduction in Bragg peak intensity in the Fourier transform shown in Fig. 3(f), compared to Fig. 3(e), already indicates the static Debye-Waller behavior discussed in Sec. III C.

A direct technique to examine the impact of defects on the crystal lattice at the surface in momentum space is LEED. LEED patterns were measured on freshly cleaved InAs(110) surfaces before and after Ar^+ sputtering, using the procedure described in Sec. II. The results measured at an electron kinetic energy of 100 eV for three different defect concentrations are presented in Figs. 4(a)–4(c). Comparing the leftmost and rightmost patterns, we observe that the coherent diffraction peaks diminish in favor of an increasingly diffuse background. This behavior, briefly mentioned above, once

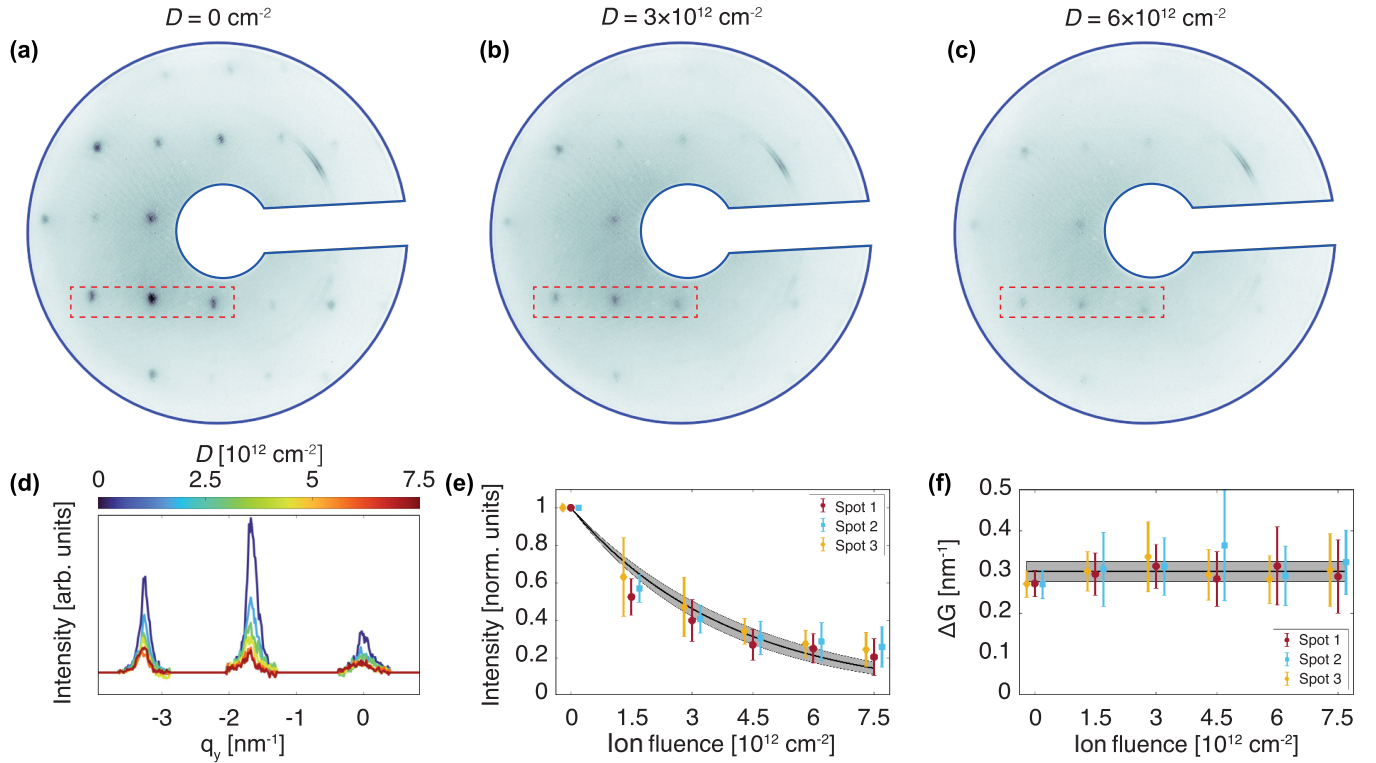


FIG. 4. LEED patterns measured at 100 eV kinetic energy on the same *in situ* cleaved InAs(110) surface: (a) after cleaving; (b) after Ar sputtering with a dose of $D = 3 \times 10^{12} \text{ cm}^{-2}$; and (c) after $D = 6 \times 10^{12} \text{ cm}^{-2}$. All intensities have been normalized to the maximum intensity of the freshly cleaved sample. (d) Intensity profiles extracted from the highlighted regions in panels (a)–(c), after linear background subtraction. (e), (f) Peak intensities and FWHM values of all clearly resolved peaks at 100 eV, obtained via Gaussian fitting. The three different colors indicate data points measured at equal disorder on three distinct spots of the surface, spatially separated by 500 μm .

again exemplifies the static Debye-Waller effect, which is a central focus of this work. From the same dataset, measured at six different defect concentrations, we extracted intensity profiles across all well-resolved peaks along the radial direction. After subtracting a linear background, we fitted Gaussian functions to extract both the peak area and the FWHM. Figure 4(e) displays the peak area normalized to the area before sputtering, while Fig. 4(f) shows ΔG , defined as the FWHM of the peaks in the radial direction. The peak intensity was fitted with an exponential function, while for ΔG we plot the mean value and the corresponding standard deviation. The mechanism responsible for the depletion of the coherent Bragg peak intensity, as previously mentioned and thoroughly discussed in Sec. III C 1, also accounts for the reduction in ARPES coherent intensity. Meanwhile, the peak linewidths show no clear trend with increasing disorder, with any broadening remaining below 0.05 nm^{-1} . Two mechanisms could explain a possible broadening: a reduction in long-range order and microstrain induced by defects, as schematically illustrated in Fig. 9. However, STM data and its Fourier transform, shown in Figs. 3(c) and 3(f), indicate that long-range coherence is preserved over at least 100 nm, implying a maximal contribution to broadening of $\sim 0.01 \text{ nm}^{-1}$, thereby ruling out this mechanism as dominant. The results in Fig. 4(f) therefore represent an upper bound for the displacement field around each point defect. The correlated atomic displacements give rise to a finite distribution of diffraction angles around the nominal Bragg condition, resulting in peak broadening. An

upper bound for the magnitude of the displacement field can be estimated from the difference between $a_{(001)}(\Delta G/G)$ and the corresponding quantity calculated using ΔG plus its standard deviation, yielding a maximal correlated displacement of approximately $0.1 \text{ \AA}$, consistent with the earlier discussion.

Finally, in Fig. 8 of Appendix C, we present simulations of the defect depth distribution based on the Stopping and Range of Ions in Matter (SRIM) software [46]. These simulations are used to estimate the effective defect concentration corresponding to the three different probing depths achieved with varying photon energies. This concentration, in turn, is used to quantify the magnitude of the atomic displacement field extracted from the ARPES experimental data.

C. Effects of disorder on ARPES spectra

There are three principal effects of disorder on the one-electron Green's function as measured by ARPES that also includes final state effects: (1) a reduction in the amplitude of the quasiparticle peaks due to elastic scattering of photoelectrons by defects and the atomic displacements associated with the disorder, (2) a broadening of the peaks in momentum space due to the finite mean free path introduced by disorder for the quasiparticles existing at equilibrium within the solid under investigation, and (3) inhomogeneous broadening in energy due to the unscreened Coulomb potential of the defects. In the remainder of this paper, we discuss (1) first, followed by (2) and (3) in turn.

1. The coherence residue

Effect (1) can be described by an extended version of DWF. Originally, this theory was developed to describe the effects of temperature on x-ray diffraction measurements [47], and was subsequently extended to photoemission [25–27]. At finite temperatures, the atoms that constitute the crystal vibrate around their ideal lattice positions. The fact that during the photoexcitation process the atoms are not localized at their perfect lattice sites creates a random phase mismatch between the initial and final states. This has severe consequences for the total coherent intensity I_{coh} . In fact, depending on both the temperature and photoelectron kinetic energy, the phase mismatch between the initial and final states can gain importance and eventually deplete a large portion of the coherent intensity. This depletion can be quantified via the DWF, which is normally interpreted as the fraction of coherent intensity that reaches the detector [48,49]:

$$\text{DWF} = e^{-\Delta K^2 \langle u_T^2 \rangle}. \quad (1)$$

In Eq. (1), ΔK is the difference between the wave vector of the initial and final states, and $\langle u_T^2 \rangle$ is the average square displacement of the atoms due to thermal vibrations. The former quantity, ΔK , depends on the photoelectron kinetic energy and, in turn, $h\nu$. In particular, the higher $h\nu$, the more important the phase mismatch becomes and, consequently, the smaller the portion of the coherent intensity. In the usual DebyeWaller picture, $\langle u_T^2 \rangle$ is proportional to temperature. In this paper, we extend the DWF to the case of static disorder. This step is also motivated by the successful treatment of thermal fluctuations within the one-step model [50], more precisely using the Coherent Potential Approximation [51] within the multiple scattering theory or the Korringa-Kohn-Rostoker formalism. Here, the thermal fluctuations of the atoms are approximated by a finite number of static displacement vectors. Furthermore, the sum of the squares of these vectors, weighted with the probability of such a displacement, is forced to be equal to $\langle u_T^2 \rangle$ [52]. An additional motivation comes from the schematics of a disordered crystal shown in Fig. 9 of Appendix D, corroborated by the LEED data shown in Fig. 4(i). When the defect concentration is high, certain atoms find themselves displaced from their ideal lattice positions due to strain generated by nearby defects. This scenario resembles the atomic displacements associated with thermal disorder, where all atoms are displaced (quasi)randomly from their ideal lattice sites. To understand how static disorder, which results from collision cascades of incident ions, affects the ARPES spectral intensity, we introduce the concept of an averaged square displacement, $\langle u_d^2 \rangle$. This measure is influenced by the defect concentration c_d , and is incorporated into the Debye-Waller framework.

We begin by demonstrating that the mere creation of vacancies or interstitials in the absence of a displacement field has minimal effects on the coherent photoemission intensity. In Appendix D, we consider a hypothetical scenario where M vacancies and M interstitials are introduced into an otherwise perfect crystal composed of N atoms. In particular, this means that the additional displacement of surrounding atoms is totally neglected. Our results reveal that in this idealized case, the coherent intensity is reduced by approximately

$|1 - M/N|^2$. This implies that even with a high vacancy concentration of 1%, the coherent intensity only decreases by about 2%. This observation underscores the importance of considering the secondary static atomic displacements caused by vacancies and interstitials, as depicted in Fig. 9, to account for the significant reduction in coherent intensity observed at defect concentrations on the order of 1% or less. To quantify these secondary displacements, we calculate the average squared displacement as $\langle u_d^2 \rangle = \frac{1}{N} \sum_{i=1}^N u_i^2 = \frac{1}{N} \sum_{i=1}^M u_{\text{sd}}^2 = c_d^0 u_{\text{sd}}^2$. Here, c_d^0 denotes the relative defect concentration, and $u_{\text{sd}}^2 = Z_{\text{nn}} u_{\text{nn}}^2 + Z_{\text{nnn}} u_{\text{nnn}}^2 + \dots$ represents the total squared displacement caused by a single defect (sd), equal to the sum of the squared displacements affecting nearest neighbors (Z_{nn}), next-nearest neighbors (Z_{nnn}), and so on.

$$F_C \equiv e^{-\Delta K^2 \langle u_d^2 \rangle} = e^{-\Delta K^2 c_d^0 u_{\text{sd}}^2}. \quad (2)$$

It is important to note that u_{sd}^2 varies depending on the material, serving as an indicator of the strength and reach of the displacement field around a defect. A higher u_{sd}^2 means atoms are more significantly displaced from their ideal positions, and the displacement field extends further from the defect. Consequently, a larger u_{sd}^2 results in greater depletion of coherent intensity when c_d^0 or momentum transfer ΔK , and thus photon energy $h\nu$, are increased. The spread and magnitude of the displacement field typically depend on the elastic modulus of the material, with materials possessing a high elastic modulus tending to confine the distortion more tightly around the defect. We therefore propose that Eq. (2) can be used to estimate the fraction of coherent intensity that originates from the static disorder within a crystalline sample. To some extent, F_C is equivalent to the DWF, where c_d^0 plays the role of an effective temperature. Moreover, the contributions of static and thermal disorder to the total mean displacement can be approximated according to $\langle u_{\text{tot}}^2 \rangle = \langle u_T^2 \rangle + \langle u_d^2 \rangle$. This is only possible if the thermal and static displacements are completely uncorrelated, for only then can we assume a vanishing cross term $\langle \mathbf{u}_T \cdot \mathbf{u}_d \rangle$. As a result, the total portion of coherent intensity is given by the product of F_C and DWF, namely, $F_C(c_d)\text{DWF}(T) = e^{-\Delta K^2 \langle u_d^2 + u_T^2 \rangle}$. Finally, we can decompose the total ARPES spectral intensity I as [48,49,53,54]

$$I(\mathbf{k}, E) = e^{-\Delta K^2 \langle u_d^2 + u_T^2 \rangle} I_{\text{coh}}(\mathbf{k}, E) + I_{\text{inc}}(\mathbf{k}, E, c_d, T), \quad (3)$$

where I_{coh} is the momentum-dependent coherent component that originates from Bloch electrons in the unperturbed crystal structure, I_{inc} is the incoherent-component-associated electron-phonon, electron-defect scattering and intensity that results from photoelectrons scattered by other electrons. At higher photon energies ($h\nu > 1000$ eV), the ARPES intensity often exhibits pronounced modulations along momentum that are not directly related to the intrinsic band dispersion. These arise from x-ray photoelectron diffraction, where photoelectrons scatter elastically from surrounding atoms and interfere, producing angular intensity variations [55]. Importantly, static disorder influences ARPES spectra in a manner similar to thermal disorder described in Ref. [55]: Both reduce the coherent fraction while enhancing the visibility of the diffraction pattern. Therefore, the analysis strategies discussed in Ref. [55] to separate coherent and diffraction-related contributions remain valid when considering static disorder as well.

2. Finite mean free path effects

With mean free path effects, we refer to initial state effects, meaning effects that are intrinsic to the quantum states within the crystal and do not depend on the photoemission process. The mean free path l , not to be confused with the IMFP of the final states, represents the average distance traveled by an electron in the crystal before undergoing a scattering event. In this subsection, we focus on elastic scattering caused by static defects within a crystalline medium. If the static potential in the crystal deviates from the perfect periodic potential, then Bloch states undergo $|n\mathbf{k}\rangle \rightarrow |m\mathbf{k}'\rangle$ transitions, where $E_n(\mathbf{k}) = E_m(\mathbf{k}')$ is ensured by the absence of a time dependence. Even though the scattering by defects is elastic, the finite probability of a scattering event implies a general reduction of the lifetime of $|n\mathbf{k}\rangle$, which in turn, through the Heisenberg uncertainty principle, determines a broadening of the energy levels according to $\Delta E \sim \hbar/\tau$, and a mean free path of $l = v_{n\mathbf{k}}\tau$. Concretely, the one-electron spectral function measured by ARPES can be written in terms of the self-energy $\Sigma(\mathbf{k}, E)$ as [2,56]

$$I(\mathbf{k}, E) \propto A(\mathbf{k}, E) = -\frac{1}{\pi} \frac{\Im \Sigma(\mathbf{k}, E)}{[E - E_0(\mathbf{k}) - \Re \Sigma(\mathbf{k}, E)]^2 + [\Im \Sigma(\mathbf{k}, E)]^2}. \quad (4)$$

The energy versus momentum dispersion relation is renormalized through the real part of the self-energy $\Re \Sigma(\mathbf{k}, E)$, while the imaginary part $\Im \Sigma(\mathbf{k}, E)$ gives some broadening to the spectral function. If, for practical purposes, we only consider static disorder, in the lowest-order Born approximation $\Re \Sigma(\mathbf{k}, E) \approx 0$ and $\Im \Sigma(\mathbf{k}, E) = 1/2\tau_{n\mathbf{k}}$ [57]. Here, we argue that the mean free path of the electrons is geometrically limited by the average distance between defects, which in d dimensions gives $l = c_d^{-1/d}$. As a consequence,

$$\tau_{n\mathbf{k}}^{-1} = c_d^{1/d} v_{n\mathbf{k}}. \quad (5)$$

Accordingly, the spectral function assumes a Lorentzian shape, with an FWHM given by $c_d^{1/d} v_{n\mathbf{k}}$. Importantly, this implies that the present broadening mechanism affects all bands equally along momentum, regardless of their average velocity, but leads to different broadening along the energy axis. In particular, bands with higher velocities appear broader in energy than bands with smaller velocities.

For a more quantitative description of this effect, one could adopt the approach of Popescu and Zunger [58]. Their method involves constructing a large supercell of the crystal, introducing and relaxing defects, computing the supercell eigenvalues under periodic boundary conditions, and unfolding these eigenvalues into the primitive Brillouin zone. This technique provides a direct way to quantify the finite mean free path effects described above.

3. Inhomogeneous broadening

Sputtering creates both interstitials and vacancies, which can act as dopants in InAs and, depending on the atomic species, become either positively or negatively charged [59]. Although an increased defect density can introduce additional free carriers [60], the associated Coulomb disorder can

localize a portion of those carriers and thus counteract this increase [61]. As a result, the carriers only partially screen the Coulomb potential generated by the defects. This leads to variable band bending [28], characterized by equipotential islands with a typical size given by the screening length r_s and overall potential energy fluctuations of order ΔV , as experimentally verified by STS in Fig. 5(a) and schematically illustrated in Fig. 5(b). The detailed shape of these islands depends on many factors and is discussed further in Ref. [62]. The STS data represent the ground-state energy of the tip-induced quantum-dot states and thus provide a local measure of the potential only up to the spatial extent of the ground-state wave function. Consequently, the measured ΔV is always smaller than the actual amplitude of the potential fluctuations, simply because of the spatial averaging introduced by the wave function. To estimate how ΔV depends on the defect concentration, we consider the potential generated by a screened defect, $\phi(r) = \pm \frac{e}{\kappa r} \exp(-\frac{r}{r_s})$. Assuming the defects are statistically distributed throughout the crystal, the mean-squared value of the potential energy, ΔV^2 , generated by a defect concentration c_d is given by [63]

$$\Delta V^2 = \int (e\phi(r))^2 c_d d^3r = \frac{2\pi e^4 c_d r_s}{\kappa} \propto r_s c_d. \quad (6)$$

Where the screening length r_s is much longer than the interdefect distance, the potential at each point in space is equal to the sum of the potentials generated by the individual defects. By the central limit theorem, one can finally assume a Gaussian distribution of the potential values across the crystal with an energy scale $\Delta V \propto c_d^{1/2}$. Here, we neglect the potential dependence of r_s on c_d , even though this should also vary as disorder is increased. Given that the beam spot size is several orders of magnitude larger than the typical screening lengths r_s , the photoemission spectra are the result of an average over all potential configurations, resulting in inhomogeneous broadening, which convolves the pristine spectra with a Gaussian with FWHM of $2\sqrt{2 \ln 2} \Delta V$ along the energy direction. This mechanism has been theoretically predicted by Chen *et al.* in Ref. [29], where Coulomb disorder in the form of charged defects in an insulator leads to a Gaussian broadening of the ARPES spectra. They also predict, in a first approximation, a square-root dependence of ΔV on the defect concentration.

Another relevant consequence of variable band bending, schematically shown in Fig. 5, is that when the strength of the potential fluctuations ΔV reaches the same order of magnitude as the bandwidth of the QWS dispersion, it is inevitable that in certain islands of the crystal, the bottom of the QWS dispersion will lie above E_F , and therefore the QWS pocket will not be locally occupied. As a consequence, the ARPES intensity of the QWS will be further depleted by this mechanism, as the regions where the QWS are absent do not contribute to the overall signal.

To quantify this effect, we again appeal to the normal distribution of the fluctuating potential and calculate that, for a given fluctuation strength of the potential ΔV and the bandwidth of the QWS E_{QWS} , the probability that at random spot on the surface the QWS states are not populated by electrons is given by $\Phi(E_{\text{QWS}}/\Delta V)$, where $\Phi(x) = \frac{1}{\sqrt{2\pi}} \int_x^\infty e^{-t^2/2} dt$ is the normal cumulative distribution function. Thus, the

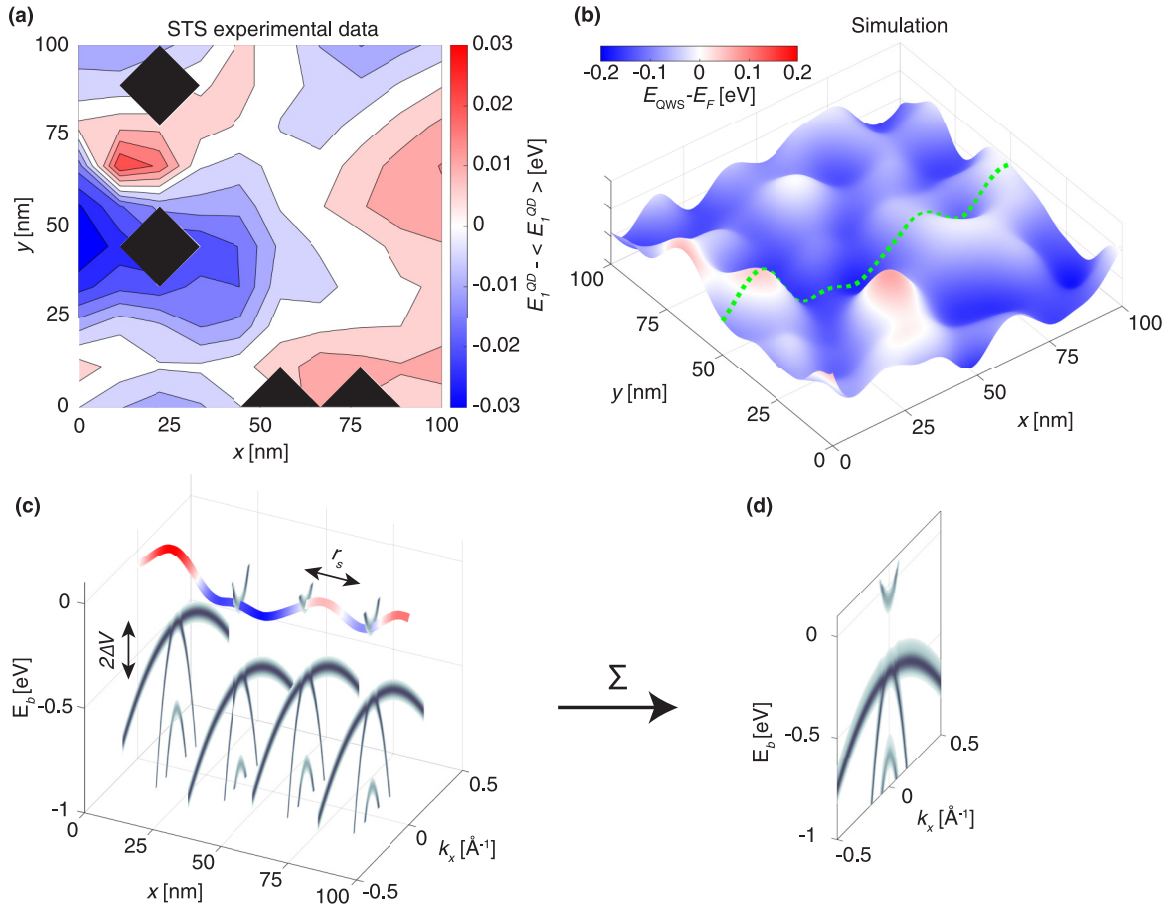


FIG. 5. (a) From the sputtered surface shown in Fig. 3(c), we measured dI/dV curves on a 10×10 grid extending over a $100 \times 100 \text{ nm}^2$ region. The ground-state energy of the tip-induced quantum-dot states E_1^{QD} , minus its mean value $\langle E_1^{QD} \rangle$, is shown as a contour map. Black squares indicate data points for which the ground-state energy could not be reliably extracted. (b) Simulated random potential, represented by the bottom of the QWS dispersion with $E_{QWS} = 50 \text{ meV}$, using a correlation length $r_s = 6 \text{ nm}$ and a potential fluctuation strength of $\Delta V = 150 \text{ meV}$. (c) Schematic representation of an inhomogeneous potential, illustrated by the minimum of the QWS dispersion [green dotted line in panel (b)] along the x direction. The band structure at selected points along x is shown, emphasizing the varying offsets in the binding energy coordinate. Averaging over a large area of the crystal surface produces the inhomogeneously broadened spectrum shown in panel (d).

attenuation of the ARPES intensity of the QWS due to static disorder and variable band bending is given by

$$I_{\text{coh}}^{\text{QWS}} \propto e^{-\Delta K^2 \langle u_d^2 \rangle} (1 - \Phi(E_{\text{QWS}}/\Delta V)). \quad (7)$$

This formalism is equivalent to stating that whenever, in a certain region of the surface, the bottom of the QWS dispersion is above E_F , the contribution from this region to the ARPES signal of the QWS is zero. Conversely, when it is below E_F , the contribution is unity, independent of the local electron density.

Finally, we emphasize that the disordered potential discussed here is at the origin of the metal-insulator transition. It can be imagined that at a certain value $E_{\text{QWS}}/\Delta V \lesssim 1$, the electrons are confined in nonconnected puddles, or in other words, they are below the percolation threshold, and their wave functions are exponentially localized. In this regime, conductivity at cryogenic temperatures follows the well-known variable-range hopping form of Efros-Shklovskii [35,36]. Also, as stated above, this effect is approximately scale-invariant, as it depends on the dimensionless parameter $E_{\text{QWS}}/\Delta V$. This

suggests that the effect also plays a crucial role at much lower electron densities and fluctuation strengths, implying that observations made at high disorder levels can provide insight into what occurs below metal-insulator transitions for the two-dimensional (2D) electron gases formed at surfaces.

D. Depletion of coherent VB intensity

The experimental coherent intensity of the valence bands in the region $-4 \text{ eV} < E_b < -2 \text{ eV}$ as a function of disorder is obtained through the fitting procedure detailed in Sec. II and Appendix G. The procedure is based on fitting MDCs within a $\pm 0.03\text{-eV}$ integration window using pseudo-Voigt profiles, defined as a linear combination of a Gaussian and a Lorentzian function. For the valence bands, this approach provides direct access to the relationship between the coherence residue F_C and the fluence of Ar^+ ions D , as depicted in Fig. 6 for various photon energies. To model the dependence of F_C on D , we assume an average defect concentration that depends linearly on the Ar^+ dose $c_d^0 = \eta D$, where η represents the concentration rate constant. Utilizing the SRIM software, alongside the

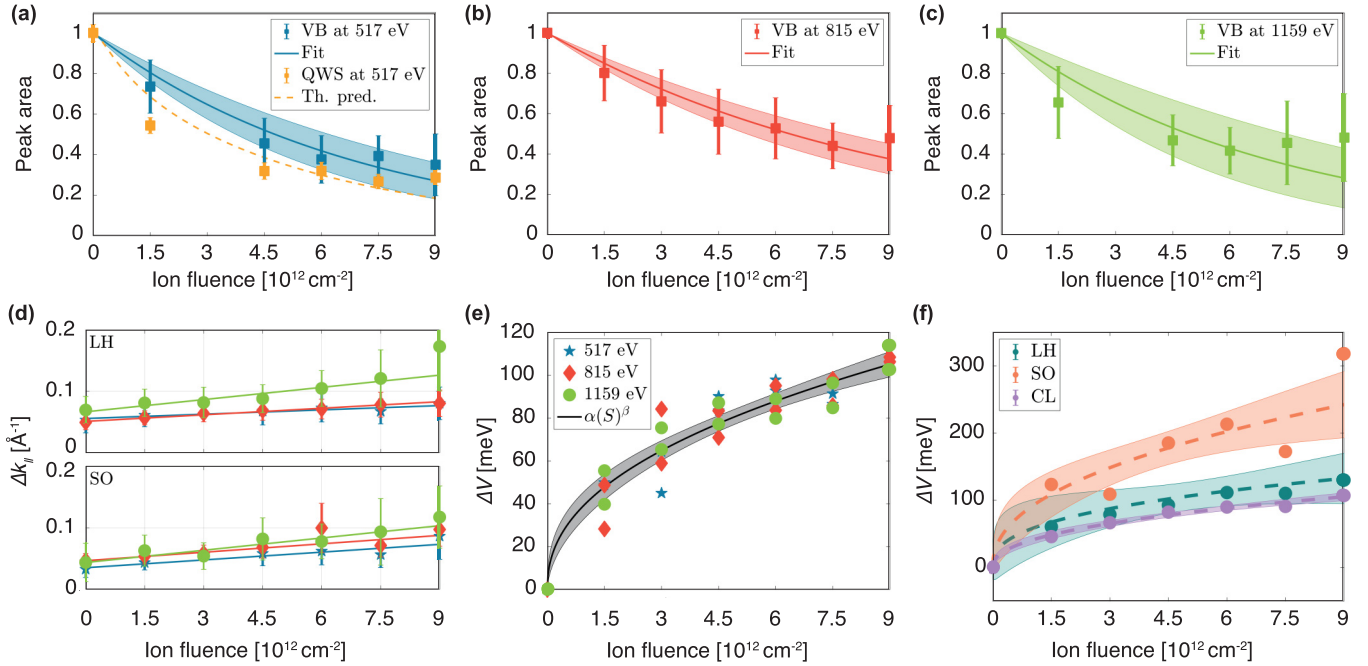


FIG. 6. (a)–(c) Normalized peak areas of the valence bands as a function of Ar⁺ dose at $h\nu = 517, 815$, and 1159 eV, respectively. The continuous lines correspond to the optimized fitting function expressed in Eq. (8), and the colored areas represent the 95% confidence intervals of the fitted parameters. Additionally, in panel (a), we show the intensity of the QWS extracted from the same spectrum measured at 517 eV, with the dashed line representing the theoretical prediction expressed in Eq. (7). (d) FWHM along $\Gamma K X$ after subtracting the instrumental broadening vs Ar⁺ dose for LH and SO bands, averaged over all MDCs for all three $h\nu$. (e) Change in the experimental FWHM of the In 4d core level, calculated through geometric subtraction of the FWHM at $D = 0$ ions cm⁻². The black line represents an optimized power-law fitting function. (f) Comparison between ΔV , obtained from the localized In 4d core states, and $\Delta k_{||}$ extracted from the delocalized valence states after geometrically subtracting both $\Delta k_{||}$ at $D = 0$ ions cm⁻², multiplying by $\frac{\partial E}{\partial k}$, and averaging across all $h\nu$.

methodology described in Appendix C, we derived estimates of η for each of the three photon energies. The determined values of η are listed in Table I. Incorporating this into Eq. (2), we derive the fitting function:

$$F_C = e^{-\Delta K^2 u_{sd}^2 c_d^0} = e^{-\Delta K^2 \gamma D}. \quad (8)$$

The parameter $\gamma \equiv \eta u_{sd}^2$ is optimized to align with the experimental observations. The optimal γ values are listed in Table I, found in Appendix G. We note that the γ values are slightly higher for spectra measured with lower $h\nu$. This may indicate that the effective defect concentration, higher near the surface, is more strongly probed at lower $h\nu$, which, due to the IMFP, is more surface sensitive. This hypothesis is supported by the γ value obtained from the exponential fit of the LEED Bragg peak area, which is significantly larger than all values measured with SX-ARPES.

TABLE I. Optimal values for the parameter γ expressed in Eq. (8) for the three photon energies used in the experiment.

Method	$h\nu/E_{kin}$ (eV)	γ (Å ⁴)	η (cm ²)	u_{sd}^2 (Å ²)
SX-ARPES	517	10.7 ± 2.7	1.20×10^{-15}	0.89 ± 0.23
SX-ARPES	815	5.1 ± 1.0	1.22×10^{-15}	0.42 ± 0.08
SX-ARPES	1159	4.6 ± 1.9	1.21×10^{-15}	0.38 ± 0.16
LEED	100	81.6 ± 9.5	–	–

Next, by multiplying γ by D , we obtain the magnitude of the atomic displacement field squared. For instance, taking $\gamma = 10.7$ (measured at $h\nu = 517$ eV) and $D = 1.5 \times 10^{12}$ ions cm⁻² yields $\langle u_d^2 \rangle \approx 0.002$ Å². Taking the square root gives an average displacement of 0.04 Å, which is below the independently obtained bound of ~ 0.1 Å from the LEED data. These displacement values are of the same order as the typical atomic displacement of 0.1 Å caused by thermal disorder at room temperature [25], further justifying the application of the Debye-Waller framework to this physical situation.

Finally, the simulated concentration rate constants η enabled us to calculate the total displacement squared u_{sd}^2 resulting from the generation of a single defect. The computed u_{sd}^2 values are also documented in Table I. Notably, the total squared displacement is more significant at $h\nu = 517$ eV compared to higher energies, which may again be attributed to the fact that photoelectrons excited with lower $h\nu$ predominantly probe regions of the crystal with the densest defect concentration, suggesting that the actual concentration rate constant η might differ from the value determined using the SRIM software. Nonetheless, by computing a weighted average across all photon energies, the total squared displacement is quantified as $u_{sd}^2 = 0.46 \pm 0.07$ Å².

E. VB spectral broadening

From the fitting procedure adopted to extract the area of the coherent peaks of the VB states, we could also determine the width of the MDC peaks for the SO and LH bands

TABLE II. Optimal values for the parameters a and b obtained by fitting $\Delta k_{\parallel}$ as a function of D using $\Delta k_{\parallel} = aD + b$, as shown in Figs. 6(e) and 6(f).

Band	$h\nu$ (eV)	a (Å)	b (10^{-3} Å $^{-1}$)
LH	517	23 ± 15	56 ± 8
	815	36 ± 7	51 ± 3
	1159	66 ± 40	67 ± 16
SO	517	42 ± 24	35 ± 10
	815	46 ± 23	46 ± 9
	1159	67 ± 39	43 ± 16

along the Γ KX direction as a function of the ion fluence D for all $h\nu$. We deconvolved the instrumental broadening from the FWHM of the optimized pseudo-Voigt profiles by geometric subtraction. The results displayed in Fig. 6(e) show $\Delta k_{\parallel}$, averaged over all MDCs, for the LH and SO bands as a function of D for all $h\nu$. We employed weighted linear regression to determine the slope of $\Delta k_{\parallel}$ versus D . The optimized parameters are listed in Table II in Appendix G. For both bands, we observed an increase in linewidths along the momentum direction with increasing disorder for all $h\nu$. Notably, the 95% confidence bounds are consistently positive, confirming the statistically significant trend of increasing linewidths with disorder for both LH and SO bands at all photon energies.

To corroborate the hypothesis that broadening due to the inhomogeneous potential plays a central role in this experiment, we fitted the In 4d core level measured under the same conditions using pseudo-Voigt functions. Appendix I introduces a model that explains when Gaussian broadening of the core levels is expected, and when, instead, multiple peaks associated with chemical shifts should appear, as discussed in Ref. [64]. In particular, when the screening length extends over multiple unit cells, as is the case in the present work [see Fig. 5(a)], Gaussian broadening is expected.

From the optimized functions, we extracted the FWHM. Since the determination of the Gaussian and Lorentzian contributions led to inconsistent results, we deconvolved the instrumental and lifetime broadening at $D = 0$ cm $^{-2}$ using the equation $2\sqrt{2 \ln 2} \Delta V = \sqrt{\text{FWHM}(D)^2 - \text{FWHM}(0)^2}$. The resulting ΔV versus D is shown in Fig. 6(e) for all $h\nu$. Based on the dependence of ΔV on c_d shown in Eq. (6), we fitted all data points using the function $\alpha(D)^{\beta}$, where both α and β have been optimized. The exponent $\beta_{\text{CL}} = 0.44 \pm 0.07$ is consistent with the expected square-root dependence, providing further evidence of the broadening mechanism discussed in Sec. III C 3. Finally, as mentioned in Sec. III C, to extract the magnitude of the variable band bending from the linewidths of the valence bands and compare it with that of core levels, we use $\Delta E = \frac{\partial E}{\partial k} \Delta k$, where Δk is obtained by geometrically subtracting the FWHM at $D = 0$. The comparison is shown in Fig. 6(f). From this figure, it is apparent that the broadening of the LH band, although slightly higher, quantitatively follows that of the In 4d core level. The SO band, on the other hand, has a significantly larger magnitude. In terms of the optimized parameters in the power-law fitting procedure, we obtained for the LH band $\beta_{\text{LH}} = 0.38 \pm 0.33$, while for the SO band, we get $\beta_{\text{SO}} = 0.45 \pm 0.19$ as documented in Table III. We expect both the LH and SO bands to exhibit

TABLE III. Optimal values for the power-law fitting of $\Delta V \propto (D)^{\beta}$.

Band	β
In 4d	0.44 ± 0.07
LH	0.38 ± 0.33
SO	0.45 ± 0.19

broader linewidths due to the finite mean free path mechanism discussed in Sec. III C 2, which is absent for the localized core states. In particular, we anticipate a larger discrepancy for the SO band due to its higher velocity, $v_{\text{SO}} = 8.53$ eV Å $\hbar^{-1}$, in the region of interest, compared to $v_{\text{LH}} = 4.49$ eV Å $\hbar^{-1}$. If, from the VB data shown in Fig. 6(f), we geometrically subtract the ΔV obtained from the core levels and divide the result by the respective velocities, the LH and SO data once again show good agreement, with a remnant broadening of the SO band with respect to LH. Furthermore, as shown in Fig. 7 of Appendix B, converting these results into an elastic mean free path yields very good agreement with the interdefect distance observed via STM in Fig. 3(c).

After subtracting the effects of the inhomogeneous potential and finite mean free path from the VB lineshapes, the remaining broadening of the SO states suggests that additional mechanisms may be involved, mechanisms that are not as pronounced for the LH band. A plausible explanation is that static disorder induces spatial inhomogeneities in the spin-orbit coupling across the sample. At the Γ point, and in the absence of disorder, the spin-orbit splitting is $\Delta_{\text{SO}} = 0.41$ eV. Without spin-orbit coupling, the HH, LH, and SO bands would be degenerate at Γ . This highlights how variations in spin-orbit coupling could produce substantial fluctuations in the energy position of the SO band, potentially explaining the remnant difference in linewidths.

We also make a final remark about the method used to extract inhomogeneous broadening. While extracting potential fluctuations from MDCs and then converting them into energy broadening may not seem the most direct approach, e.g., directly extracting the linewidths along energy at points in $\mathbf{k}$ space where the bands have zero velocity appears more straightforward, we argue that our method ensures greater robustness against artifacts. Given the increase in a density of states (DOS)-like background relative to the coherent component of the spectra, measuring the linewidths along momentum guarantees that any potential distortion of the lineshapes due to this effect is ruled out, since the DOS-like background remains a mere constant along $\mathbf{k}$. On the other hand, when measuring the linewidth of extrema under these conditions, the proper disentanglement of coherent and incoherent intensity becomes technically more challenging, and the risk of dealing with artifacts rather than physical phenomena increases. However, for completeness, we still show energy distribution curve (EDCs) measured at the Γ point in Fig. 10(a) of Appendix E, which corroborate both the inhomogeneous potential picture and the change in spin-orbit coupling upon increasing disorder.

F. Depletion of coherent QWS intensity

As discussed above, the depletion of ARPES intensity originating from QWS due to disorder is a more complicated topic than the depletion of the VB ARPES intensity. While for the latter, the only mechanism responsible for the attenuation of the magnitude of the quasiparticle peaks is a Debye-Waller-like mechanism, for the QWS ARPES intensity, this effect is present along with additional inhomogeneous potential effects. The ARPES intensity from the QWS has been obtained as explained in Sec. II and is shown in Fig. 6(a), together with the intensity originating from the valence bands extracted from the same spectrum. This figure already demonstrates that the intensity of the QWS is significantly more suppressed than that of the VB.

Equation (7) is an expression that, apart from the coherence residue, depends on a single parameter, namely, the ratio $E_{\text{QWS}}/\Delta V$. ARPES directly accesses E_{QWS} , which in our case is approximately 50 meV. For ΔV , we use the fitting function optimized on the core-level data. The combination with the coherence residue extracted from the VB yields the dashed line in Fig. 6(a), which accurately describes the dependence of the QWS intensity on disorder. Finally, we point out that this analysis assumes that E_{QWS} is not affected by disorder. In principle, a corollary of the effect demonstrated here, and schematically shown in Fig. 5(b), is that the space available to electrons is reduced upon increase of the disorder. In particular, at constant electron density, this implies a smaller average distance between electrons and therefore larger Coulomb repulsion. This could result in a decrease in E_{QWS} . As a result, the ratio $E_{\text{QWS}}/\Delta V$ changes, and the 2D electron system would localize faster.

IV. CONCLUSIONS

We have developed a framework to explain two of the most common observations in semiconductor photoemission studies: low coherent intensity and broadened lineshapes. For the former, we extended the Debye-Waller factor, originally developed to describe thermal disorder in diffraction and photoemission experiments, to account for static disorder in ARPES measurements. Our experimental results, obtained on ion-bombarded InAs(110), exhibit quantitative agreement with the proposed theory and diffraction data, thereby reinforcing its validity in explaining the depletion of coherent intensity in the valence and conduction bands.

Our measurements show that suppression of the QWS intensity exceeds that of the valence bands, and that the linewidths of valence band quasiparticle peaks increase with disorder concentration. These observations support our description that charged defects generate regions of varying potential within the semiconductor. In the first case, disorder-induced variable band bending creates regions where QWS are unoccupied, leading to an overall reduction in signal from these states relative to the valence bands. In the second case, averaging over large regions of the crystal, namely, over lengths exceeding the screening length, results in Gaussian broadening of the lineshapes along the energy axis. This effect is further enhanced by the limited mean free path of the initial states, which contributes additional Lorentzian broadening.

For both localized core levels and delocalized valence states, we consistently extracted the magnitude of potential energy fluctuations and observed a disorder dependence consistent with theoretical predictions.

Overall, our observations on ion-bombarded InAs(110) provide valuable insights into the universal characteristics of ARPES intensity depletion and spectral broadening in semiconductors and insulators. Specifically, they clarify the role of the static Debye-Waller mechanism in suppressing coherent peaks and highlight the impact of potential fluctuations caused by charged defects, an effect that plays a crucial role in driving key phenomena such as the metal-insulator transition.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [65].

APPENDIX A: LEED

LEED patterns were measured on freshly cleaved InAs(110) surfaces before and after Ar^+ sputtering at an electron kinetic energy of 100 eV. Cleavage was performed at liquid nitrogen temperature, while sputtering was carried out using parameters equal to those described in Sec. II, namely, an Ar^+ ion kinetic energy of 3 keV and an Ar partial pressure of 5×10^{-8} mbar. Sputtering was conducted in cycles of 20 s each ($D = 1.5 \times 10^{12} \text{ cm}^{-2}$), until a total ion dose of $D = 7.5 \times 10^{12} \text{ cm}^{-2}$ was reached. The results for the three different defect concentrations are presented in Fig. 4.

APPENDIX B: DETERMINATION OF MEAN FREE PATH

The fact that both mean free path effects and inhomogeneous broadening contribute to the VB lineshapes, combined with the availability of inhomogeneous broadening estimates from the core levels, allows us to disentangle these two contributions and reconstruct the mean free path of the VB states. Specifically, we geometrically subtract the ΔV measured from the core levels from $\frac{\partial E}{\partial k} \Delta k$, where Δk is obtained by geometrically subtracting the FWHM at $D = 0$. According to Eq. (5), the result corresponds to the inverse lifetime of the VB states. To obtain the mean free path, we divide this quantity by the band velocity and take the reciprocal. In Fig. 7, we present the resulting mean free path extracted from both the LH and SO bands as a function of ion dose. When compared with the interdefect distance measured by STM in Fig. 2, the extracted values show excellent agreement for the SO band, while the LH band exhibits a discrepancy of approximately 30%. This discrepancy may arise from the fact that quasiparticle peaks are best described by Voigt functions, and using the geometric approximation to disentangle the Lorentzian and Gaussian

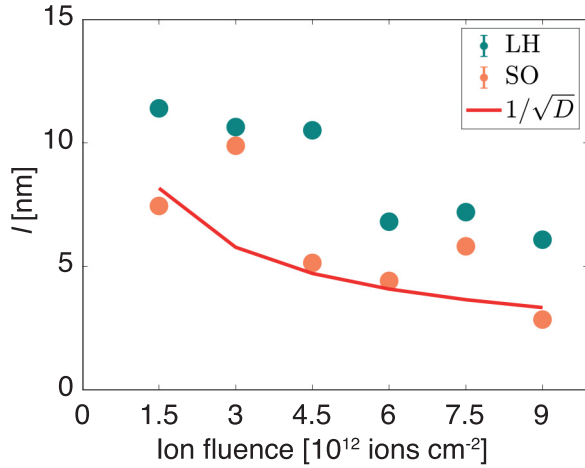


FIG. 7. Mean free path extracted from the valence bands after disentangling the contribution from the fluctuating potential. Green circles represent results from the LH band, orange circles correspond to the SO band, and the red line indicates the interdefect distance estimated from STM data.

components of the FWHM can introduce inaccuracies. An additional contributing factor could be slight variations in the sputtering process: Although the same parameters were used in both the STM and SX-ARPES experiments, the sputtering guns were different, potentially leading to a mismatch between the nominal ion doses and the effective doses experienced during SX-ARPES.

APPENDIX C: DEPTH DISTRIBUTION OF DEFECTS

Figure 8 shows the out-of-plane distribution of vacant In and As sites calculated using SRIM software [46]. Each of these displaced atoms stops at some interstitial site, effectively creating a second defect. For comparison purposes, the probing depth in the pristine sample, namely, three times the IMFP [1], is plotted on top of the defect distribution. Since, according to Ref. [66], the energy to displace an In atom

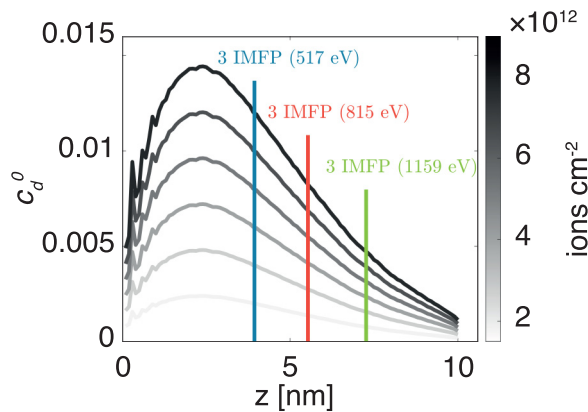


FIG. 8. Concentration of In and As vacancies as a function of depth, shown at different Ar^+ beam fluences. The colored vertical lines show the probing depths of the three photon energies in InAs. These results were obtained by running a simulation with the SRIM software package [46].

is slightly smaller compared to that of As, the In atoms are displaced more frequently during the bombardment process. Furthermore, the peak position at ~ 20 Å is explained by the cascade effect, generated by the Ar ions that displace target atoms, which in turn create more defects as they propagate away from the surface. Reference [67] has calculated that 63% of the sputter induce defects are canceled within a few picoseconds. Here, to take care of this significant effect we multiply the total number of displaced atom by 0.37. We point out that the simulation does not include temperature effects, which lead to self-annealing processes and therefore to a general reduction of the defects' concentration. Further, the simulation does not take into account the fact that atoms sitting at damaged areas of the crystal need less energy to be displaced. This leads to an underestimate of the actual concentration of defects. Finally, the quantity $c_d(z)$ shown in Fig. 8 allows us to estimate the effective concentration of vacancies perceived by photoelectrons with different probing depths. This is achieved through weighted averaging of c_d over the depth coordinate, with the weight being $\exp(-z/\text{IMFP}(h\nu))$.

APPENDIX D: ROLE OF VACANCIES AND INTERSTITIALS IN ARPES

Here, we demonstrate that the mere presence of vacancies and interstitials, in the absence of secondary displacements (as in the case in Fig. 9), cannot account for the significant depletion of coherent intensity observed experimentally. To

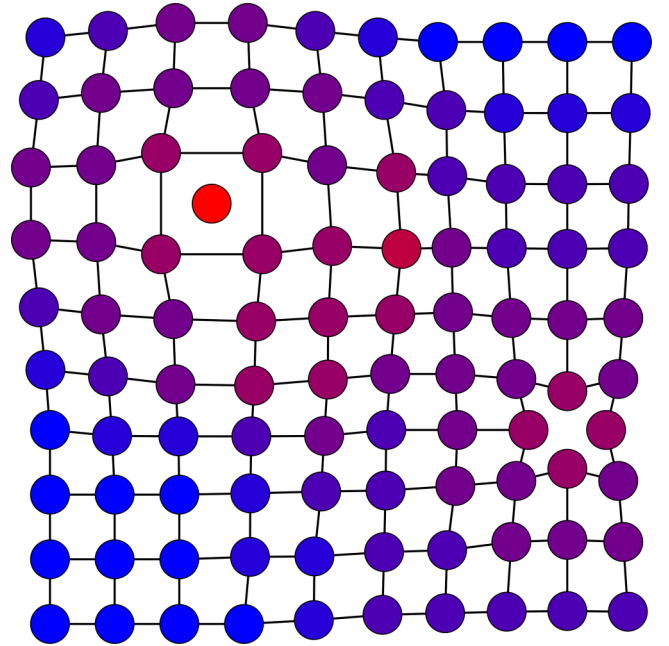


FIG. 9. Schematic representation of a crystal lattice following the creation of a vacancy and an interstitial defect. Atoms deviating from their equilibrium lattice positions are color coded to reflect the extent of their displacement, with a color gradient indicating the magnitude. Note that actual atomic displacements caused by point defects occur on a length scale typically 2 orders of magnitude smaller than the interatomic distance. This visualization aids in quantifying the mean square displacement, $\langle u_d^2 \rangle$, highlighting secondary displacement effects induced by defect formation.

elucidate this, we commence from the foundational principles, adopting an initial state $\Psi_{\mathbf{k}_i} = \sum_m e^{i\mathbf{k}_i \cdot \mathbf{R}_m} \phi_{\mathbf{k}_i}(\mathbf{r} - \mathbf{R}_m)$. We proceed by considering plane waves as final states $\Psi_{\mathbf{k}_f}(\mathbf{r}) = e^{i\mathbf{k}_f \cdot \mathbf{r}}$. It has been established that plane wave final states are not an indispensable prerequisite for achieving our ultimate result. This assertion is supported by the work of Shevchik, which utilized augmented plane waves instead. Consequently, the ARPES coherent intensity I_{coh} is expressed as

$$\begin{aligned} I_{\text{coh}} &= |\langle \Psi_{\mathbf{k}_f} | \mathbf{A} \cdot \mathbf{p} | \Psi_{\mathbf{k}_i} \rangle|^2 \propto \left| \int d^3r e^{-i\mathbf{k}_f \cdot \mathbf{r}} \Psi_{\mathbf{k}_i}(\mathbf{r}) \right|^2 \\ &= \left| \sum_m e^{i(\mathbf{k}_i - \mathbf{k}_f) \cdot \mathbf{R}_m} \int d^3r \phi_{\mathbf{k}_i}(\mathbf{r}) e^{i\mathbf{k}_f \cdot \mathbf{r}} \right|^2 \\ &= S(\mathbf{k}_i - \mathbf{k}_f) |\mathcal{F}(\phi_{\mathbf{k}_i}(\mathbf{k}))|^2, \end{aligned}$$

where $S(\mathbf{k}) = |\sum_m e^{i\mathbf{k} \cdot \mathbf{R}_m}|^2$. We proceed by making several assumptions regarding the static disorder within our crystal. It is presumed that atoms initially positioned at lattice sites $\mathbf{R}_{i_1}, \mathbf{R}_{i_2}, \dots, \mathbf{R}_{i_M}$ are permanently displaced from their ideal locations. The displacement vectors are denoted as $\mathbf{U}_{i_1}, \mathbf{U}_{i_2}, \dots, \mathbf{U}_{i_M}$. It is important to note that all other atoms $m \notin I = i_1, i_2, \dots, i_M$ remain at their ideal positions. We then endeavor to compute $S(\Delta\mathbf{k})$ for the disordered crystal, where $\Delta\mathbf{k} = \mathbf{k}_i - \mathbf{k}_f$ is used for brevity. For a specific configuration of disordered atoms I , corresponding to a disorder concentration $\frac{M}{N}$, where N represents the total number of atoms, $S(\Delta\mathbf{k}) = |\sum_{m \notin I} e^{i\Delta\mathbf{k} \cdot \mathbf{R}_m} + \sum_{n \in I} e^{i\Delta\mathbf{k}(\mathbf{R}_n + \mathbf{U}_n)}|^2$. In systems of considerable size, implying $N \gg 1$, Kohn and Luttinger [68] have illustrated that quantities such as $S(\Delta\mathbf{k})$ closely approximate their ensemble average for the majority of microscopic configurations. Thus, we can approximate $S(\Delta\mathbf{k}) \approx \langle S(\Delta\mathbf{k}) \rangle$ for sufficiently large values of N . Subsequently, we calculate the ensemble average as follows:

$$\begin{aligned} \langle S(\Delta\mathbf{k}) \rangle &= \sum_I P_I \int d^3U_{i_1} f(\mathbf{U}_{i_1}) \int d^3U_{i_2} f(\mathbf{U}_{i_2}) \dots \\ &\quad \times \int d^3U_{i_M} f(\mathbf{U}_{i_M}) \left| \sum_{m \notin I} e^{i\Delta\mathbf{k} \cdot \mathbf{R}_m} + \sum_{n \in I} e^{i\Delta\mathbf{k}(\mathbf{R}_n + \mathbf{U}_n)} \right|^2, \end{aligned} \quad (\text{D1})$$

where the initial summation encompasses all conceivable $\binom{N}{M}$ configurations, and the integrals $\int d^3U_i f(\mathbf{U}_i)$ span the entire volume of the crystal. The function $f(\mathbf{U}_i) d^3U_i$ signifies the probability of locating the displaced atom within the infinitesimal volume d^3U_i centered around the position $\mathbf{R}_i + \mathbf{U}_i$. We adopt the premise that the probability density function f is uniform for every atom and remains unaffected by the displacement of other atoms. The probability P_I for a single configuration equates to the reciprocal of the total number of configurations, expressed as $P_I = \binom{N}{M}^{-1}$. By expanding the ensemble average, we derive the following expression:

$$\langle S(\Delta\mathbf{k}) \rangle = \underbrace{\sum_I P_I \sum_{\substack{m \notin I \\ n \notin I}} e^{i\Delta\mathbf{k} \cdot (\mathbf{R}_m - \mathbf{R}_n)}}_I$$

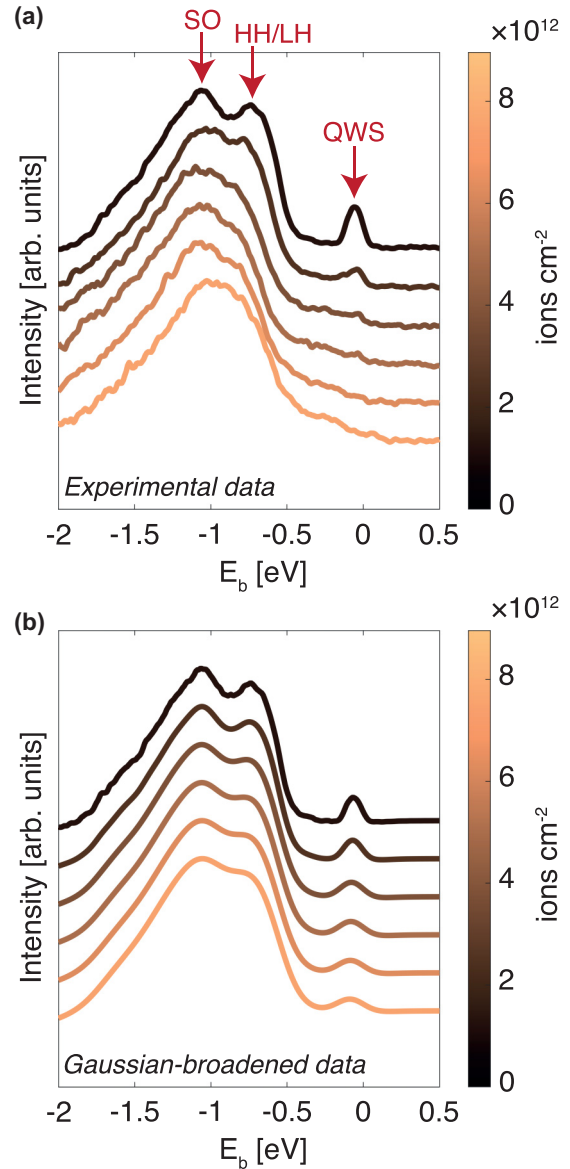


FIG. 10. (a) Normalized EDC extracted at the Γ point with an integration window of $\pm 0.03 \text{ \AA}^{-1}$ as a function of Ar^+ ion fluence, after subtracting an EDC extracted from the same spectrum at the K point, and plotted with a vertical offset at the zero position of the intensity. In the figure, we also identify the SO band, the HH and LH bands, which are degenerate at Γ , and the QWS. (b) The EDC shown in black in panel (a) is Gaussian broadened using ΔV measured from the In 4d core levels.

$$\begin{aligned} &+ 2\Re \left[\underbrace{f(\Delta\mathbf{k}) \sum_I P_I \sum_{\substack{m \notin I \\ n \notin I}} e^{i\Delta\mathbf{k} \cdot (\mathbf{R}_m - \mathbf{R}_n)}}_{\text{II}} \right] \\ &+ \underbrace{|f(\Delta\mathbf{k})|^2 \sum_I P_I \sum_{\substack{m \in I \\ n \in I}} e^{i\Delta\mathbf{k} \cdot (\mathbf{R}_m - \mathbf{R}_n)}}_{\text{III}}, \end{aligned} \quad (\text{D2})$$

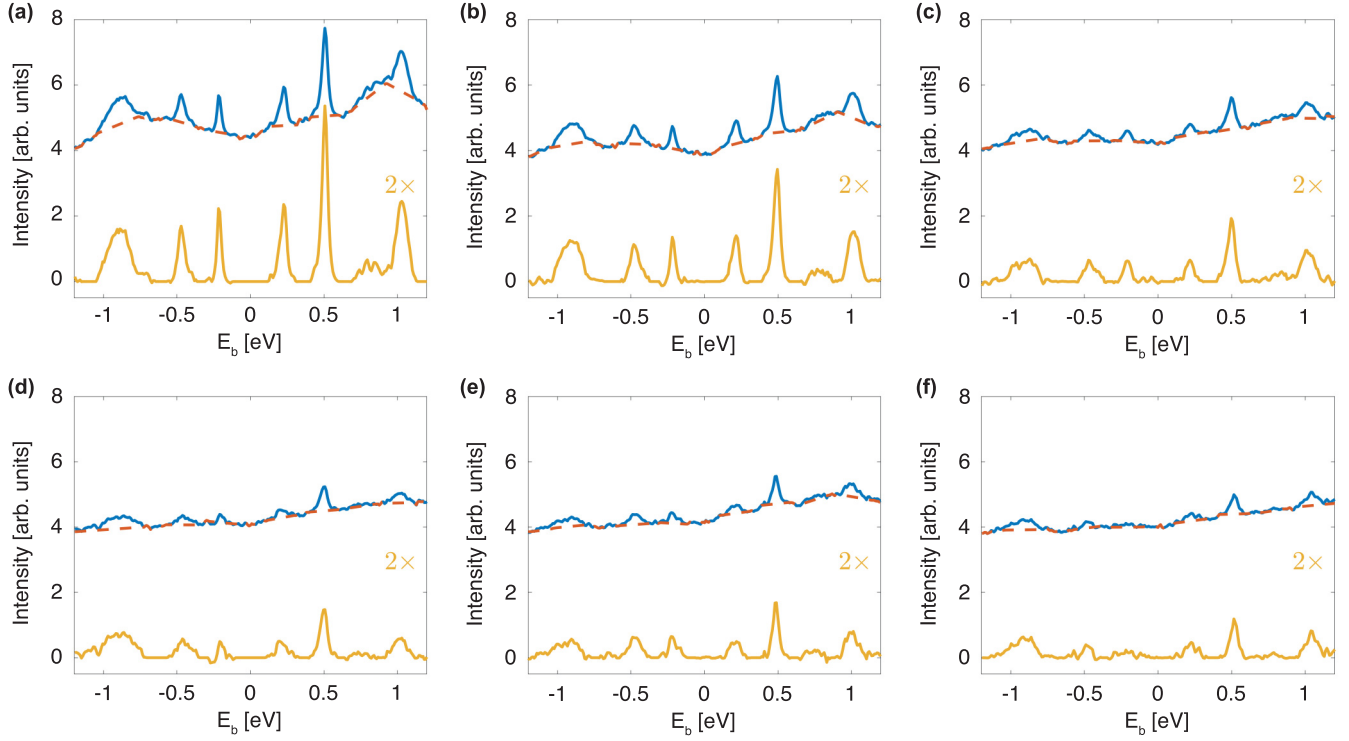


FIG. 11. Separation of the coherent and incoherent signal from the dataset measured at $h\nu = 517$ eV. Panels (a)–(f) show the MDC extracted at $E_b = -2.63$ eV for all ion doses in increasing order. For each figure, the blue solid line represents the total intensity, the red dashed line is the background, and the yellow solid line is the coherent intensity.

where $f(\Delta\mathbf{k}) = \int d^3U f(\mathbf{U}) e^{i\Delta\mathbf{k}\cdot\mathbf{U}}$ represents the Fourier transform of the probability density function f . Ultimately, it is feasible to demonstrate that the term I can be reformulated into $\frac{(N-M)^2}{N^2} \sum_{m,n} e^{i\Delta\mathbf{k}\cdot(\mathbf{R}_m - \mathbf{R}_n)} = \frac{(N-M)^2}{N^2} |\sum_m e^{i\Delta\mathbf{k}\cdot\mathbf{R}_m}|^2$. Similarly, it can be shown that term II equates to $2\Re(f) \frac{(N-M)M}{N^2} |\sum_m e^{i\Delta\mathbf{k}\cdot\mathbf{R}_m}|$, and term III corresponds to $|f|^2 \frac{M^2}{N^2} |\sum_m e^{i\Delta\mathbf{k}\cdot\mathbf{R}_m}|^2$. By amalgamating these terms, the entire expression can be simplified to

$$\langle S(\Delta\mathbf{k}) \rangle = \left| 1 - (1 - f(\Delta\mathbf{k})) \frac{M}{N} \right|^2 \left| \sum_m e^{i\Delta\mathbf{k}\cdot\mathbf{R}_m} \right|^2. \quad (\text{D3})$$

For homogeneous probability distributions f , $\langle S(\Delta\mathbf{k}) \rangle$ is proportional to the interference function of the perfect crystal $S(\Delta\mathbf{k})$, with the factor being $|1 - \frac{M}{N}|^2 \leq 1$.

APPENDIX E: INHOMOGENEOUS BROADENING AT THE Γ POINT

To further corroborate the inhomogeneous picture described in this paper, we extracted EDCs at the Γ point with an integration window of $\pm 0.03 \text{ \AA}^{-1}$, after subtracting an EDC extracted from the same spectrum at the K point, and plotted them as a function of Ar^+ fluence in Fig. 10(a). The subtraction of the EDC at the K point allows us to remove the DOS-like incoherent background. In this figure, we also identify the three main peaks, which correspond to the SO band, separated from the degenerate HH and LH bands by 0.41 eV, and the QWS. The figure shows that with increasing disorder, the splitting between the SO and HH/LH bands becomes less and less visible. This corroborates the inhomogeneous broadening picture. We also observe the merging of the two peaks, which suggests that the spin-orbit interaction is likely

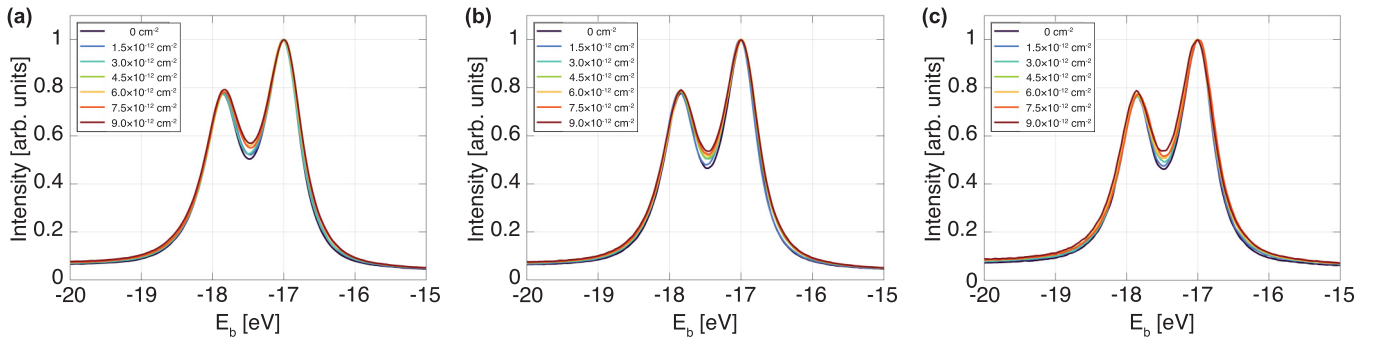


FIG. 12. In 4d core levels measured at (a) 517 eV, (b) 815 eV, and (c) 1159 eV for all Ar^+ fluences.

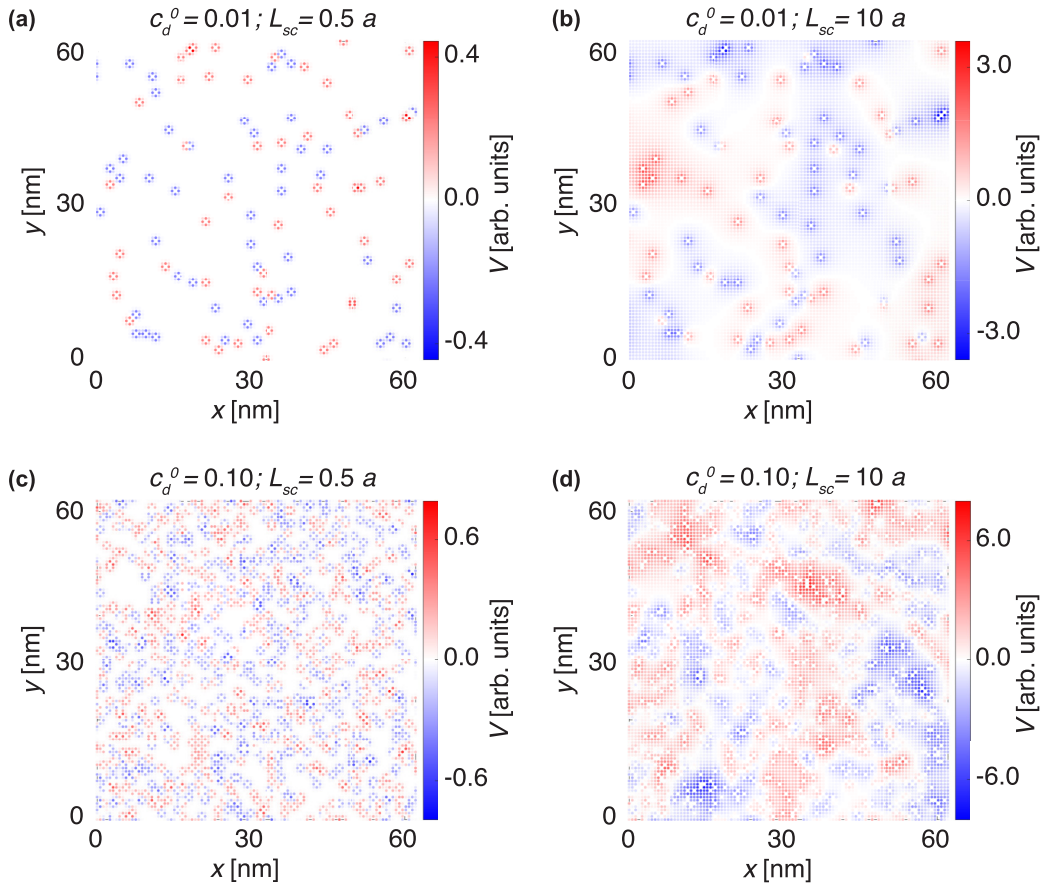


FIG. 13. Simulated electrostatic potential of a 2D square lattice with lattice constant $a = 0.6$ nm for two charged-defect concentrations, $c_d^0 \in \{0.01, 0.1\}$, and two screening lengths, $L_{sc} \in \{0.5a, 10a\}$. The charged defects have random signs (± 1). Each panel displays the central 100×100 region of the lattice.

weakened by the disorder. In Fig. 10(b), we take the EDC at $D = 0$ cm⁻¹ and Gaussian broaden it using ΔV obtained from the In 4d core-level fitting. This shows the expected decrease in resolution between the two peaks; however, it also suggests that the spin-orbit coupling likely needs to change to fully reproduce the evolution shown in Fig. 10(a).

APPENDIX F: SEPARATION OF COHERENT AND INCOHERENT INTENSITY

To separate the coherent peaks from the incoherent background we smoothed the spectral data along the momentum ($\mathbf{k}$) axis using Gaussian convolution. Following this, we calculated the second derivative of the spectrum along $\mathbf{k}$, identifying the peak bases at points where the second derivative exceeded a specific threshold. To capture the full extent of the coherent peaks, including their tails, we expanded the peak base by a fixed constant. We then employed linear interpolation across the peak bases to isolate the coherent intensity from the incoherent background within the peak region. The results of this procedure are shown in Fig. 11 for six different MDCs.

APPENDIX G: EXTRACTION OF THE COHERENCE RESIDUE AND LINEWIDTHS FROM ARPES DATA

In the Appendix, we explain the process of extracting the coherence residue and linewidths from ARPES data. Initially,

we extract the coherent intensity from the ARPES data using the background subtraction method outlined in Appendix F. For each dataset measured at three different photon energies, we analyze ten MDCs with a ± 0.03 -eV integration window in the valence band region -3.5 eV $< E_b < -2.5$ eV. We fit the four peaks, corresponding to the LH and SO bands, with pseudo-Voigt profiles, defined as a linear combination of Gaussian and Lorentzian components. The fit parameters include the peak height, FWHM, mixing ratio between Gaussian and Lorentzian components, and the k_x position. Each peak's area is normalized to its area at zero Ar⁺ dose for comparison.

Subsequently, we calculate the average normalized peak areas of the LH and SO bands across all binding energies for each Ar⁺ beam fluence. This average provides the data point for each dose, with the standard deviation serving as the uncertainty estimate for each point. The final step involves presenting the averaged intensities and FWHM values for the LH and SO bands as a function of Ar⁺ dose, as depicted in Fig. 6. Our method allows us to systematically assess the impact of sputtering-induced disorder on the ARPES-measured electronic structure.

The optimal values γ , obtained by fitting F_C versus D with Eq. (8), are displayed in the third column of Table I. The fourth column shows the concentration rate constant η calculated with the SRIM software, while the fifth column contains u_{sd}^2 .

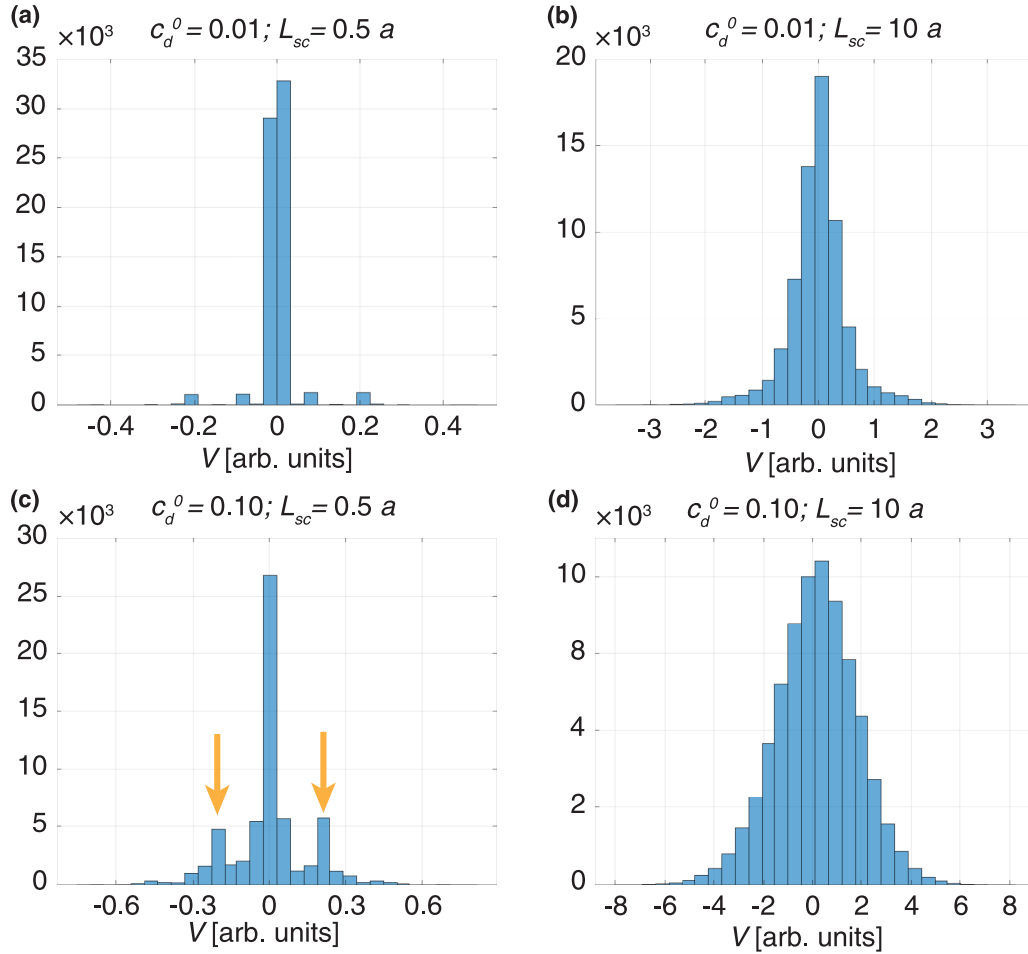


FIG. 14. The distributions of the electrostatic potential evaluated at each atomic site are shown as histograms, where panels (a)–(d) correspond to those depicted in Figs. 13(a)–13(d). The histograms include the potential values of all atomic sites within the central 260×260 region of the lattice. The 20 outermost atoms on all sides are excluded to avoid spurious edge effects.

APPENDIX H: XPS FITTING

To extract the FWHM for the In $4d$ core states, we first normalized the data to the height of the In $4d_{5/2}$ peak. The peaks are shown for all three $h\nu$ in Fig. 12. All spectra are fitted using pseudo-Voigt functions. Since a priori knowledge of how disorder affects the spin-orbit states is unavailable, we opted for a parameter-independent fitting procedure for the two peaks shown in Fig. 12. More precisely, we did not constrain the relative height, the distance, the mixing ratio, or the FWHM of the two peaks. These were all treated as independent parameters to be optimized. At the same time, we account for the effect of inelastic electron scattering by performing a Shirley background subtraction [69], which was optimized self-consistently with the peak fitting. By observing the intensity between the peaks, it is already possible to deduce an increasing trend in the FWHM.

APPENDIX I: BROADENING DUE TO COULOMB CHARGES

To investigate the effects of broadening on the core levels due to the presence of Coulomb charges, we simulate a

300×300 square lattice with lattice constant $a = 0.6$ nm, as appropriate for InAs. At random positions of this 2D lattice, we introduce Coulomb charges with random sign ± 1 . For each atomic site $\mathbf{R}_m$, we then compute the electrostatic potential generated by these charges, assuming a screening length L_{sc} . The resulting potential is of Yukawa type and is given by $V(\mathbf{R}_m) = \sum_i \frac{q_i}{|\mathbf{R}_m - \mathbf{r}_i|} \exp(-\frac{|\mathbf{R}_m - \mathbf{r}_i|}{L_{sc}})$, with $q_i = \pm 1$. In Fig. 13, we show examples of the resulting potential landscapes for two different charged-defect concentrations, $c_d^0 \in \{0.01, 0.10\}$, and two screening lengths, $L_{sc} \in \{0.5a, 10a\}$. To quantify the distribution of on-site energies, and thereby estimate the energy broadening of tightly bound states such as core levels, we histogram the potential values at all atomic sites within the central 260×260 region of the lattice. The 20 outermost atoms on all sides are excluded in order to avoid spurious edge effects. The resulting distributions are shown in Fig. 14. We observe that the behavior of the energy distributions depends strongly on the interplay between the defect concentration and the screening length. For relatively large screening lengths ($L_{sc} = 10a$, corresponding to ≈ 6 nm in InAs), the potential distribution is Gaussian, as discussed in the main text, and is largely independent of the overall charged-defect concentration. In contrast, for short screening

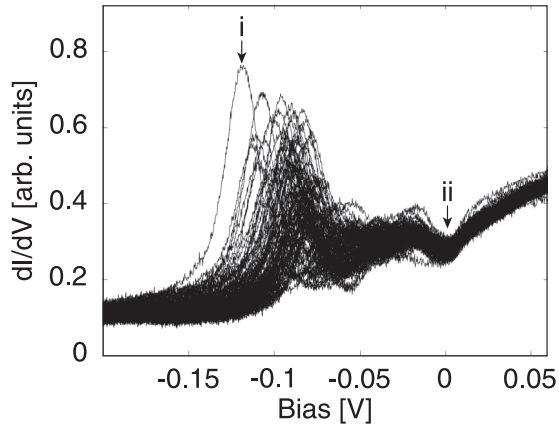


FIG. 15. From the sputtered surface shown in Fig. 3(c), we measured dI/dV curves on a 10×10 grid covering a $100 \times 100 \text{ nm}^2$ region. All dI/dV curves are displayed together here. The ground-state energy of the tip-induced quantum-dot state for one representative curve is marked by “i”, while “ii” indicates a dip in the density of states that appears in all curves.

lengths ($L_{sc} \lesssim a$), qualitatively different behaviors emerge. At low defect concentrations (e.g., $c_d^0 = 0.01$), the majority of atoms are essentially unaffected by the charged defects, as seen in Fig. 14(a), and only a negligible fraction of sites experience significant perturbations. In this regime, the effect on the core-level spectrum is minimal, since the contribution of perturbed atoms to the overall photoemission signal is negligibly small compared to that of the unperturbed lattice.

However, for higher defect concentrations (e.g., $c_d^0 \sim 0.1$), the number of perturbed atoms becomes comparable to the number of unperturbed ones. As shown in Fig. 14(c), this results in the appearance of well-defined additional peaks (indicated by arrows) in the potential distribution. In a core-level spectrum, these peaks correspond to distinct chemical shifts. In this regime, the simple inhomogeneous broadening picture discussed above is no longer applicable.

APPENDIX J: POTENTIAL LANDSCAPE FROM STS DATA

The STS data have been measured on the same surface as the one where the STM data shown in Fig. 3(c) have been acquired. We measured dI/dV curves on a 10×10 grid over a $100 \times 100 \text{ nm}^2$ surface. The measured spectra, as a function of bias V are all shown in one figure in Fig. 15. The bias position of the lowest peak indicates the ground-state energy of the tip-induced quantum-dot state at that precise location. We highlight the ground state of one arbitrary curve by an arrow and a “i”. For each point measured on the 10×10 grid, we extracted the bias position of the lowest peak, and used this to illustrate the potential landscape shown in Fig. 5(a). Another interesting feature that is visible from the data representation in Fig. 15 and highlighted as “ii” is the apparent onset of a pseudogap at the Fermi level. Further analysis of this phenomenon goes beyond the scope of the present paper, but this matches the expectations of a gapped phase when below the metal-insulator transition, and could be an indication of closeness to such a phase transition.

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