



Electronic states and related orientational assembly in potassium-doped C₆₀ layer on SrTiO₃(001)

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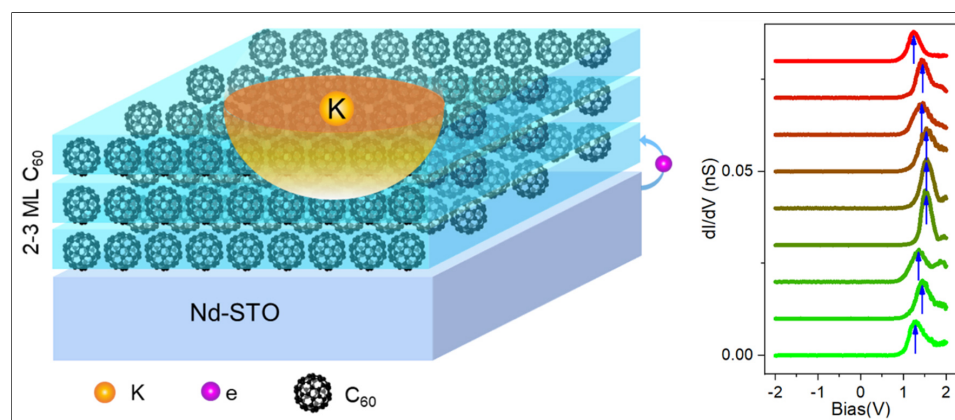
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Abstract

Two-dimensional (2D) fullerene (C₆₀) films grown on various metal and semiconductor substrates have been extensively studied but remain less explored on oxide substrates. By using cryogenic scanning tunneling microscopy and spectroscopy (STM/STS), we investigated the molecular orientation and electronic structure of C₆₀ films grown on a SrTiO₃(001) surface. Our STM/STS results display the variation of electronic energy levels of C₆₀ molecules as a function of both coordination and potassium doping. The orientation of each C₆₀ molecule can be identified through density-functional theory (DFT) calculations and STM simulations. The orientational assembly of the C₆₀ layer near a K-doped C₆₀ shows a chiral 2×2 superstructure of the hexagon-faced up (H) configuration. Intriguingly, the spatial distribution of the density of states (DOS) peak around the K-doped C₆₀ shows an electronic state in the highest occupied molecular orbital (HOMO)-lowest unoccupied molecular orbital (LUMO) gap that can be attributed to the charge-trapped state. The spatial dependence of this state indicates an upward-bending of the conduction band, induced by the screened electric field of a negatively charged center. Together, the band-bending behavior and the related chiral 2×2 superstructure suggest a possible scenario of a charge-trapped, orientational C₆₀ assembly.

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INTRODUCTION

Fullerene (C_{60}) molecules have been deposited on various metal and semiconductor surfaces, e.g., Au^[1,2], Ag^[3,4], Cu^[5] and Cd(0001)^[6]. Recently, the commensurate-incommensurate phase transition and unique strain relaxation modes, such as heptamer arrays, in C_{60} monolayers on Cd(0001) have been found by scanning tunneling microscopy (STM), highlighting the significant role of annealing temperature in determining the final superstructure^[7]. The interaction between C_{60} and the metal substrate is primarily dominated by the electronegativity of the substrate^[8]. In contrast, the interaction of C_{60} with semiconductor substrates is more complex. Indeed, it is quite strong on Si(111)^[9,10], Si(001)^[11,12] and SiC surfaces^[13,14] owing to the formation of strong Si-C covalent bonds, while relatively weak on layered inert materials such as graphene^[15,16] and hexagonal boron nitride (h-BN)^[17]. Moreover, the interaction of C_{60} with a GaAs surface was shown to vary with the doping type of the substrate, resulting in different band bending and thus different charge transfer^[18]. As a result, the charge-induced effect tends to dominate in hybridized systems consisting of C_{60} layers and semiconducting/insulating substrates. Notably, the growth and properties of C_{60} films on oxide substrates, particularly perovskite oxides such as $SrTiO_3$ (STO), have attracted significant interest. For example, a combined STM and low-energy electron diffraction (LEED) study revealed a close-packed 6×6 superstructure with specific molecular orientations for a C_{60} monolayer on STO(001)^[19]. Meanwhile, advancements in film growth techniques, such as the “two-step method”, have enabled the epitaxial growth of single-domain C_{60} films on Au(111)^[20].

On the other hand, a variety of novel physical phenomena have been discovered in perovskite oxide interfaces and/or surfaces, e.g., giant magnetoresistance effects^[21,22], metal-insulating phase transition^[23], interfacial superconductivity^[24], and surface-enhanced superconductivity^[25–27]. Previously, a K_3C_{60} film was shown to display fully-gapped strong coupling s-wave superconductivity with a relatively high critical temperature (T_c) of ~ 22 K^[28] on graphitized SiC(0001) substrates through controlled potassium (K) doping. The combination of a self-assembled C_{60} monolayer and the high dielectric constant semiconductor STO is anticipated to result in enhanced superconductivity due to the expected interface-induced charge transfer and enhanced electron-phonon coupling, similar to FeSe on STO(001) substrates^[27]. This expectation is further supported by the advances in novel low-dimensional carbon phases, such as van der Waals (vdW) heterostructures composed of C_{60} and B_{60} ^[29]. However, C_{60} layers grown on STO surfaces are rarely investigated^[30], especially with respect to the related electronic states induced by the STO substrate within the C_{60} layer.

In order to provide an in-depth understanding of the charge-related phenomena in K-doped C_{60} layers, including charge transfer, charge trapping and band bending, we synthesized C_{60} multilayers on two different STO(001) clean surfaces, namely pristine and Nb (0.7 wt%)-doped STO(001) surfaces, followed by controlled K doping.

MATERIALS AND METHODS

Preparation of STO substrates

Undoped and Nb (0.7 wt%)-doped STO(001) wafers measuring $10 \times 2 \times 0.5$ mm³ were degassed by direct current (DC) heating at 550 °C for more than 5 h in a molecular beam epitaxy (MBE) chamber. The STO wafer was heated at 800 °C for 1 h and then gradually heated up to 1,200 °C for another half an hour to obtain a TiO_2 -terminated STO(001) surface with 2×2 superstructure^[1,30]. The superstructure was confirmed by STM observations [Supplementary Figure 1]. C_{60} molecules were evaporated using a Knudsen cell. The monolayer C_{60} film was obtained by post-annealing at 220 °C for 5 min after 5-minute deposition of C_{60} on the STO surface kept at room temperature (RT), while multilayer C_{60} films were obtained without post-annealing after 8-minute deposition. Commercial alkali metal dispensers (SAES Getters, Italy) were

used and heated by a direct electric current ($I_K = 5.9$ A) to produce potassium atoms. The pressure during the deposition of potassium atoms was maintained at around 2×10^{-9} Torr.

STM/STS measurements

All experiments were performed in a commercial ultrahigh vacuum (UHV) system (USM-1300s) consisting of a MBE chamber equipped with an insert chamber allowing *in situ* STM measurements at liquid-helium temperature. All STM measurements were performed in a constant current mode at 4.2 or 1 K, with a polycrystalline Pt/Ir tip and the bias voltage applied to the sample. Differential conductance (dI/dV) measurements were performed using a lock-in amplifier with a modulation frequency (f) of 271 Hz and a modulation amplitude (V) of 1–5 mV. STM images were analyzed using WsXM software^[31].

Computational details

Periodic density-functional theory (DFT) calculations were performed using the Vienna Ab initio Simulation Package (version 5.4.4)^[32,33]. Projector augmented wave (PAW)^[34,35] pseudopotentials were employed to describe electron-ion interactions. The exchange-correlation energy of the system was described by the Perdew-Burke-Ernzerhof (PBE) functional^[36]. Intermolecular forces were estimated using the vdW-D3 method developed by Grimme *et al.*^[37]. The cutoff energy for plane waves was set at 400 eV. The convergence criterion used for atomic forces in structural optimization calculations was 0.03 eV/Å. The convergence criterion for solving the electronic wave function is a 1×10^{-5} eV energy difference. The K-point setting was based on a $1 \times 1 \times 1$ Monkhorst-Pack grid centered at the Γ point. In this work, the simulated STM images, obtained using Hive program^[38], revealed atomic-scale surface features.

The gas-phase electronic structure calculations of isolated C_{60} were performed using the Gaussian 16 software package (version A. 03)^[39]. The geometrical structures of ground-state molecules were optimized by using the Becke three parameter Lee-Yang-Parr (B3LYP) hybrid functional combined with the 6-31G(d) basis set. Based on the optimized structures, the electronic structure of the ground-state molecule was obtained at the Becke-Lee-Yang-Parr (BLYP)/6-31G(d) level. The TiO_2 -surface of the $SrTiO_3(001)$ substrate was used for DFT calculation and STM simulation, as described in Ref.^[39].

RESULTS AND DISCUSSIONS

Morphologies and electronic properties of undoped and 0.7 wt% Nb-doped STO(001) surface

The flat and clean terraces of both undoped and Nb (0.7 wt%)-doped STO(001) substrates have been achieved, as shown in Figure 1. On the pristine STO(001) surface, randomly distributed surface atoms appear on the surface with a corrugation approximately 200–250 pm [Figure 1A]. In contrast, surface atoms become arranged in 2×2 order on the Nb-doped STO(001) surface [Figure 1B], indicating a well-known TiO_2 -terminated surface. The 2×2 reconstruction is further confirmed by the 2×2 fractional spots observed in the Fourier transform image in Supplementary Figure 1B. The height profile measured on the high-resolution image of the reconstructed Nb-doped-STO surface shows that the unit cell of the surface reconstruction is about 0.8 nm, which is twice the unit cell size of the pristine $SrTiO_3(001)$ surface (~ 0.4 nm), consistent with a 2×2 surface reconstruction. From the dI/dV spectra acquired on both surfaces, the Nb-doped $SrTiO_3(001)$ surface shows a semiconducting gap (~ 2.4 eV) with a conduction band minimum (CBM) at 0.7 eV and a valence band maximum (VBM) at -1.7 eV, while the undoped $SrTiO_3(001)$ surface shows a larger gap (~ 3.4 eV) with a CBM at 1.1 eV and a VBM at -2.3 eV [Figure 1C]. We note that the CBM and VBM are deduced by extrapolating the rising edges of the conduction/valence bands to their respective intersection with zero. Clearly, the Nb-doped $SrTiO_3(001)$ surface exhibits a smaller semiconducting gap with partially filled conduction and valence bands, which may introduce stronger interfacial charge transfer^[40,41], as demonstrated in Supplementary Figure 2 and Supplementary Table 1. The Nb-doped STO substrate can therefore provide a stronger charge transfer effect, due to interfacial oxygen vacancies^[42]. Bader

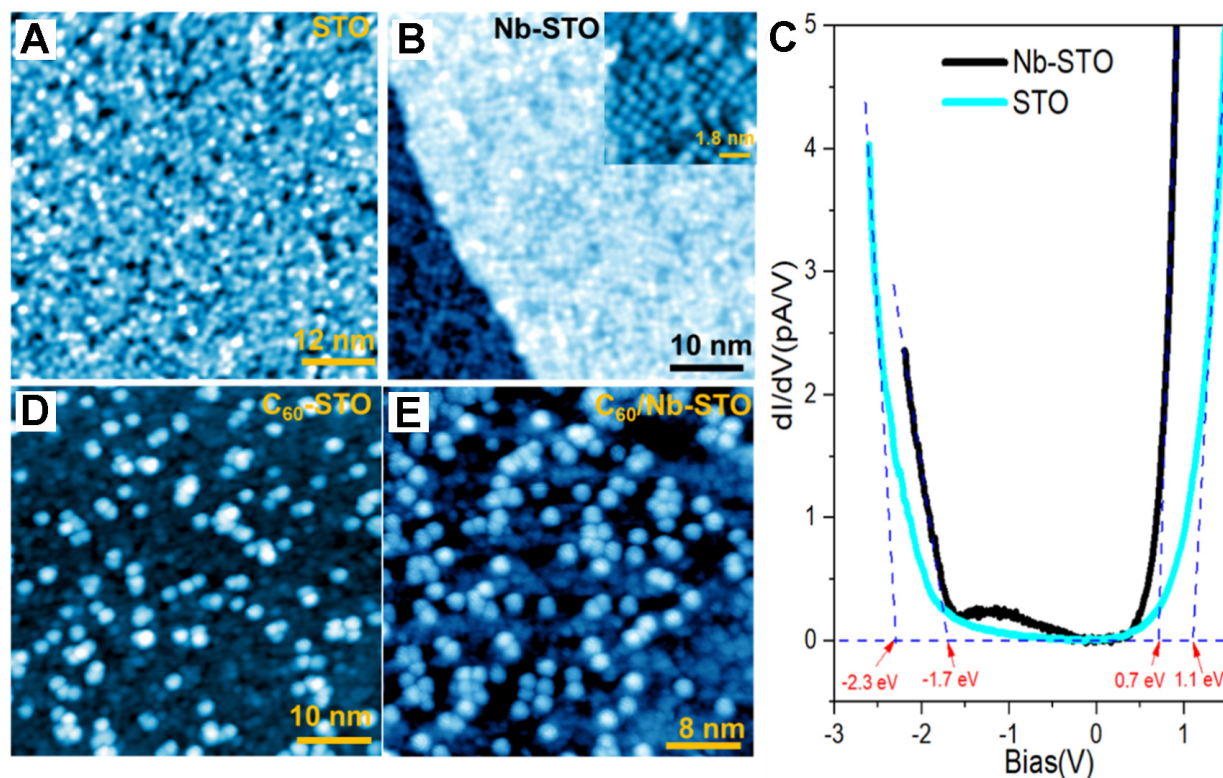


Figure 1. Images of pristine-STO and Nb-doped STO substrate and related dI/dV spectra. (A) Image of pristine STO surface ($60 \times 60 \text{ nm}^2$) with U_b : +1.0 V and I_T : 20 pA; (B) Image of Nb-doped-STO surface ($50 \times 50 \text{ nm}^2$) with U_b : +1.0 V and I_T : 50 pA. Inset: High resolution image of 2×2 reconstructed Nb-doped-STO surface ($14 \times 14 \text{ nm}^2$); (C) dI/dV curves taken on the clean surfaces of the pristine STO (cyan) and Nb-doped STO (black) substrate, respectively. All STS measured with V_b : 1.2 V, I_T : 10 pA and bias modulation of 20 mV at 4.2 K; (D) Image of C_{60} deposited on the pristine STO substrate ($50 \times 50 \text{ nm}^2$) with U_b : +1.0 V and I_T : 20 pA; (E) Image of C_{60} deposited on the Nb-doped STO substrate. Image size is $40 \times 40 \text{ nm}^2$ with U_b : +1.0 V and I_T : 20 pA. STO: SrTiO_3 ; C_{60} : fullerene.

charge calculations in [Supplementary Table 1](#) also show that the electron donation from STO to C_{60} . The inclusion of both undoped and Nb-doped samples demonstrates that the Nb-doped STO substrate significantly enhances the substrate's conductivity and ability to supply charge to adsorbed molecules, thus providing an effective method for engineering interfacial electronic properties of the C_{60} overlayer.

Small point defects exist on the flat terrace [Figure 1B], which may be attributed to TiO_x , a typical and unavoidable feature during thermal treatment. Such surface defects may influence the locally adsorbed C_{60} at short range, but have no long-range effect on the configuration and electronic properties of the C_{60} assembly layer. By further depositing C_{60} onto both undoped and Nb (0.7 wt%)-doped $\text{STO}(001)$ surfaces with an initial coverage of about 0.3 ML, C_{60} molecules tended to randomly distribute as monomers on both surfaces, instead of accumulating into islands [Figure 1D and E]. With further deposition of C_{60} , more fullerene molecules initially formed a less-ordered buffer layer, followed by the layer-by-layer growth of C_{60} films with self-assembled hexagonal lattices, as shown in STM images [Supplementary Figure 3]. New C_{60} layers formed on the existing C_{60} buffer layer initially nucleated in the form of trimers or tetramers, as marked by red circles in Supplementary Figure 4, indicating that the buffer layer enhances the interaction between C_{60} molecules and leads to a short-range ordered arrangement of C_{60} . The existence of a buffer layer indicates that the interaction between C_{60} and the $\text{STO}(001)$ substrate is relatively strong compared to the intralayer interaction within the C_{60} layer. Another piece of evidence for the strong interface interaction is the random adsorption of C_{60} observed in a typical zoom-in STM topography showing the internal structure of C_{60} [Figure 2A].

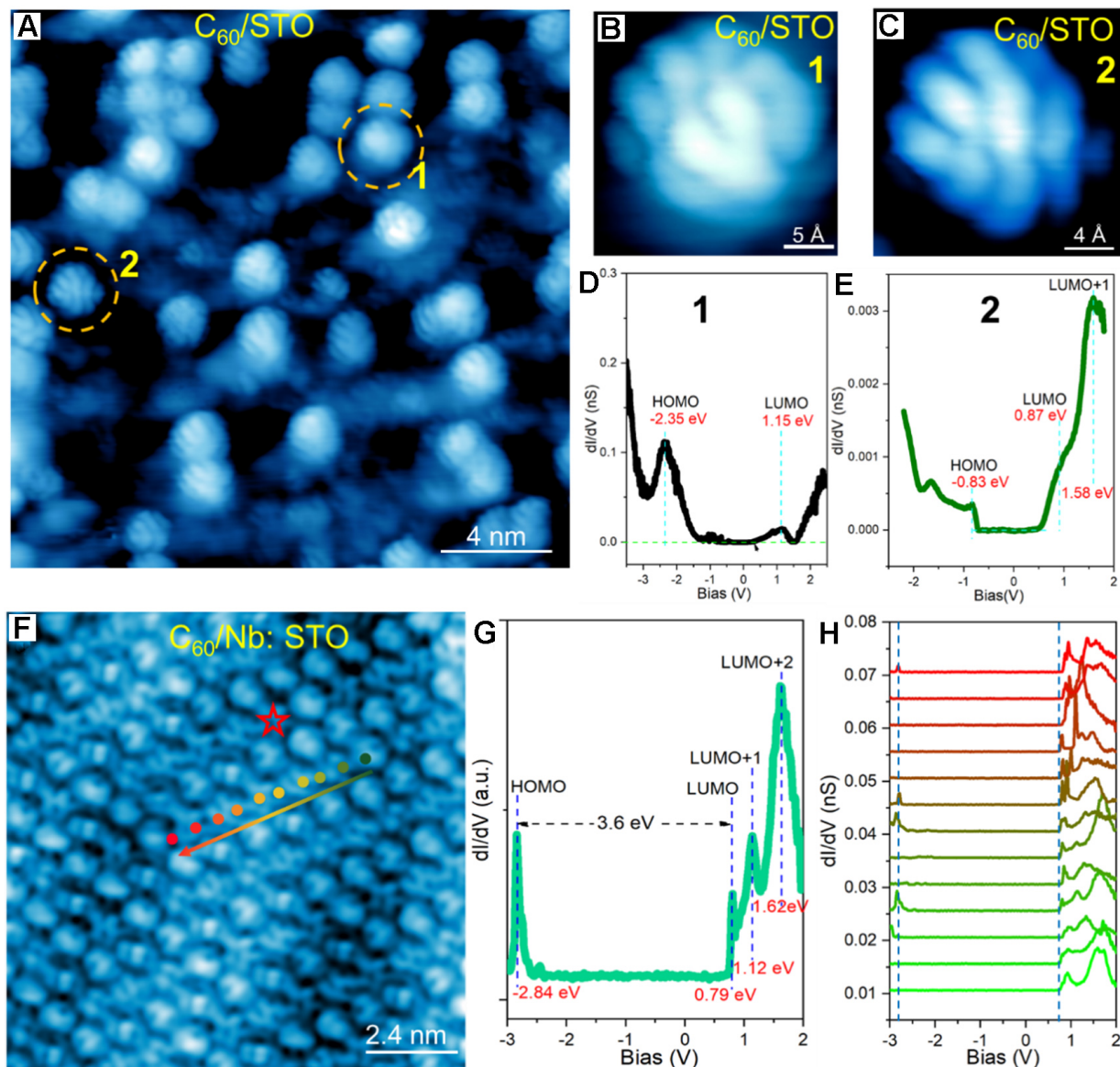


Figure 2. Topographic image and spectroscopy of C₆₀ on undoped STO and Nb-STO substrates. (A) STM image of C₆₀ monomers on undoped STO substrate. Image size is 20 × 20 nm² with U_b: +1.0 V and I_t: 10 pA; (B and C) High-resolution image of a single C₆₀ monomer with different internal structures. Image size is 2 × 2 nm² with U_b: +1.0 V and I_t: 10 pA; (D and E) Point STS measured on top of the C₆₀ monomer in panels (B) and (C), respectively. STS are taken by the setpoint of U_b: +1.5 V, I_t: 10 pA and bias modulation of 20 mV; (F) Orbital-resolved STM image of C₆₀ monolayer on Nb-STO substrate. Image size is 12 × 12 nm² with U_b: +1.5 V and I_t: 100 pA; (G) STS curve measured at the location marked by red star; (H) A linear spectroscopic survey taken along the dotted line in panel (F). STS are taken by the setpoint of U_b: +1.5 V, I_t: 100 pA and bias modulation of 20 mV. The color gradient from green to red corresponds directly to the gradient in the (H) panel, representing the one-to-one relationship between the atomic sites in the (F) panel and the STS data in the (H) panel. All STS are taken at 4.2 K. C₆₀: Fullerene; STO: SrTiO₃; STM: scanning tunneling microscopy; STS: scanning tunneling spectroscopy; HOMO: highest occupied molecular orbital; LUMO: lowest unoccupied molecular orbital.

HOMO-LUMO gap of C₆₀ monomers and 2D layers

By employing cryogenic scanning tunneling microscopy and spectroscopy (STM/STS), we found that fullerene molecules grow layer-by-layer after forming a relatively disordered wetting layer on the 2×2 reconstructed Nb-doped STO(001) surface. Furthermore, the monolayer of C₆₀ shows a gap of about 3.6 eV, which is close to the reported gaps for monolayer and multilayer C₆₀, measured to be ~4.1 and ~4.6 eV, respectively^[30]. At the initial stage of C₆₀ adsorption on the STO(001) surface, C₆₀ molecules exist as monomers [Figure 2A]. Measured at a cryogenic temperature of 4.2 K, due to relatively strong interaction

between C_{60} and the STO substrate, zoom-in STM images [Figure 2B and C] show the internal structure of C_{60} , either as “fish-scale”-like [type-I, Figure 2B] or “flower”-like [type-II, Figure 2C]. Such images indicate that the C_{60} is partially frozen with slight rotation^[43,44]. The corresponding dI/dV spectra for the two C_{60} configurations are shown in Figure 2D and E, respectively. The measured dI/dV curves display a small highest occupied molecular orbital (HOMO)-lowest unoccupied molecular orbital (LUMO) gap for C_{60} monomers, namely 3.5 eV for type I and 1.7 eV for type II, respectively. Previous work shows that the HOMO-LUMO gap of monolayer C_{60} on metal surfaces varies from 2.0 to 3.0 eV^[4,45], while that on graphene and h-BN sheets is about 3.5 eV^[17,46]. For isolated C_{60} in the gas phase, the difference of $\epsilon_{LUMO} - \epsilon_{HOMO}$ is ~ 1.6 eV^[15,18], which is in good agreement with our calculation [Supplementary Figure 5], and is usually assumed not to change significantly in the presence of a surface^[8,47]. The enlarged HOMO-LUMO gap (ΔE_{gap}) can be expressed as

$$\Delta E_{gap} = \epsilon_{LUMO} - \epsilon_{HOMO} + U$$

where $\epsilon_{LUMO} - \epsilon_{HOMO}$ is the molecular-orbital splitting of C_{60} in its ground state without considering the on-site Coulomb interaction (U)^[24].

The variation in the HOMO-LUMO gap of C_{60} monomers depends strongly on the adsorbed configuration of C_{60} . For type I-adsorbed C_{60} , the molecule is mostly surrounded by other C_{60} molecules, resulting in stronger intermolecular coupling; therefore, the measured HOMO-LUMO gap is about 3.5 eV, close to the 3.6 eV measured for the C_{60} monolayer. In contrast, for the type II-adsorbed C_{60} , the molecule is nearly isolated, and therefore the measured HOMO-LUMO gap is about 1.7 eV, close to that of an isolated C_{60} in the gas phase (1.6 eV).

The HOMO/LUMO gap of a C_{60} monomer on STO is about 1.7 eV; while the HOMO/LUMO gap of a C_{60} monolayer on STO is about 3.6 eV, with an uncertain contribution from the on-site U via neighboring C_{60} molecules [Supplementary Note 2]. When C_{60} molecules aggregate into a bulk or a film, U increases owing to enhanced Coulomb interaction from neighboring molecules and the substrate. Accordingly, the ΔE_{gap} value of bulk C_{60} increases to 3.7 eV, as detected by photoemission spectroscopy^[18].

A series of dI/dV spectra measured on the C_{60} layer in Figure 2F show a HOMO-LUMO gap of about 3.6 eV, as shown in Figure 2G and H. These spectra contain four distinct peaks corresponding to the HOMO, LUMO, LUMO + 1, and LUMO+2 of C_{60} at - 2.84, 0.79, 1.12, and 1.62 V, respectively. As seen in Figure 2H, although the HOMO energy varies with location, the LUMO energy remains unchanged, indicating enhanced Coulomb interaction for adding an electron due to neighboring molecules. As demonstrated in Supplementary Figure 6A-H, the density of states (DOS) of the C_{60} monolayer was calculated with various configurations. DFT calculations give a bandgap of about 3.6 eV with a DOS peak appearing inside the bandgap for all configurations. Such in-gap states could be induced by charge transfer between the C_{60} layer and the STO substrate, as shown in Supplementary Figures 2 and 6, and Supplementary Table 1. K-doped C_{60} induced a stronger charge transfer, as demonstrated by Bader charge calculations in Supplementary Table 1. More charge transfer occurs from STO to K-doped C_{60} , leading to electron accumulation around the K dopant and trapping charge within a radius of 3 nm.

Orientational assembly of the C_{60} layer and effect of K-doping

Five different orientations of C_{60} molecules were resolved with high-resolution imaging, e.g., C-atom faced-up (CA), hexagon-pentagon bond faced-up (H:P), hexagon-hexagon bond faced-up (H:H), pentagon faced-up and deflected (P:D) and hexagon faced-up (H). The intramolecular structures of assembled C_{60} in the layer can be visualized using cryogenic high-resolution STM imaging, as previously demonstrated by

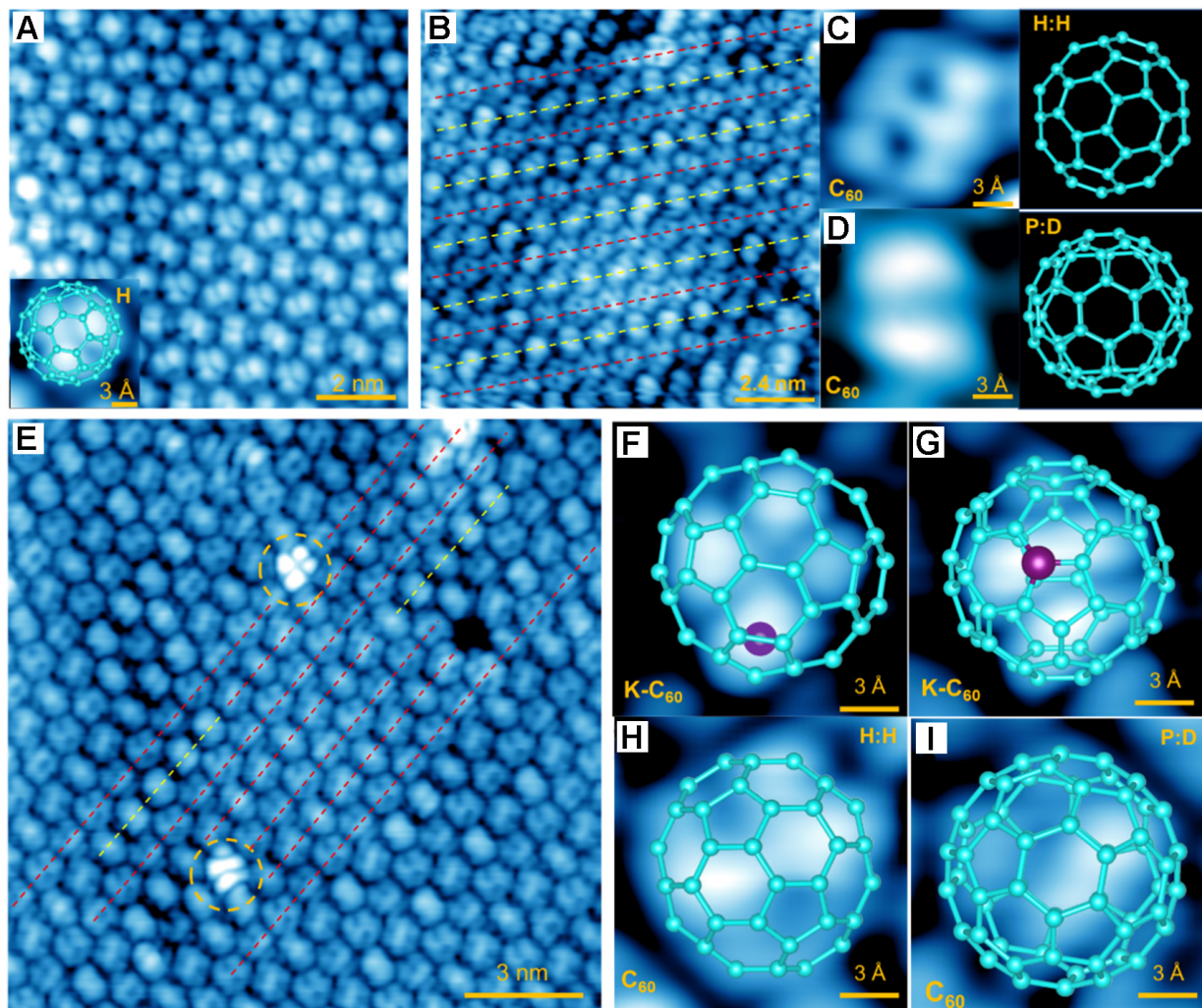


Figure 3. Topographic images of different orientational C_{60} assembly layers. (A) STM image of undoped C_{60} assembly layer showing a uniform orientation. Image size is $10 \times 10 \text{ nm}^2$ with U_b : +1.0 V and I_t : 150 pA. Inset: enlarged image of a single C_{60} molecule with a “clover”-shape, overlapped with a structure of C_{60} with H configuration. Image size is $1.5 \times 1.5 \text{ nm}^2$ with U_b : +1.0 V and I_t : 10 pA; (B) STM image of undoped C_{60} assembly layer showing a staggered orientation arrangement. Yellow and red dashed lines indicated two different oriented C_{60} chains, respectively. Image size is $12 \times 12 \text{ nm}^2$ with U_b : +1.2 V and I_t : 10 pA; (C and D) High resolution image showing different orbitals of C_{60} , corresponding to two distinct orientations of C_{60} , (“8”-shaped, H:H configuration) and (“two-lobe”, P:D configuration), respectively. [Right panels of (C) and (D)] The corresponding structure models of C_{60} ; (E) STM image of C_{60} layer with two K-dopants. Two bright spots, marked by orange-colored dashed circles, corresponding to two different K-doped C_{60} . Image size is $15 \times 15 \text{ nm}^2$ with U_b : +1.2 V and I_t : 50 pA; (F and G) High resolution STM images of two K-doped C_{60} , overlapped with proposed structures, (“Four leaf clover”, a K-atom embedded in a C_{60} with H:H configuration) and (“three-lobe”, a K-atom attached with a H:P configuration C_{60}), respectively; (H and I) Images for two oriented C_{60} , corresponding to the orientation of H:H configuration and hexagon facing up, respectively. Image size is $1.5 \times 1.5 \text{ nm}^2$ with U_b : +1.2 V and I_t : 50 pA. C_{60} : Fullerene; STM: scanning tunneling microscopy.

Hou *et al.*^[45]. Various molecular configurations were observed on the Nb-doped STO surface, as shown in Figure 3.

Figure 3A shows a high-resolution image of a local area of the C_{60} layer, in which all C_{60} molecules exhibit a uniform configuration with a three-lobe clover-like shape, as shown in the inset figure. This image likely corresponds to the H configuration. STM imaging and simulations of C_{60} performed on the semiconducting CuSe monolayer seem to show a similar assignment of C_{60} configuration^[46]. Here, in order to account for the

substrate effect, we performed STM simulations for various oriented C_{60} on the TiO_2 -terminated surface of the $SrTiO_3(001)$ substrate [Supplementary Figure 7]. As shown in Supplementary Figure 7C and D, the simulated image of the H configuration displays a three-lobe shape. Figure 3B shows a high-resolution image for another local area of the C_{60} layer, in which C_{60} molecules exhibit a staggered orientational assembly. The red dashed line indicates a C_{60} molecule with an “8-shaped” pattern, corresponding to the H:H configuration, as shown in Figure 3C^[46]. The yellow dashed line indicates a C_{60} molecule with a “two-lobe” internal structure, corresponding to the P:D configuration, as shown in Figure 3D^[46]. However, because our simulated images do not show a clear “two-lobe” structure, we assign this feature to the P:D configuration based on Ref.^[46].

However, once K is doped into a preformed C_{60} layer, the doped C_{60} molecule becomes negatively charged due to the charge transfer from K atoms^[47]. This charge transfer causes the LUMO level of C_{60} to become partially occupied^[48]. When the sample is biased at +1.4 V and above, the tunnel current is contributed by (1) direct tunneling from the STM tip to STO; and (2) tunnelling via the LUMO of C_{60} . For a C_{60} molecule bonded with K, its LUMO level is partly occupied by electrons and hence it becomes more effective at conducting electrons when the sample is positively biased. This makes the doped C_{60} molecule appear brighter in comparison with undoped ones. Figure 3E shows a local area in the C_{60} layer with two prominent K-doped C_{60} molecules, marked by orange dashed circles. High-resolution images of two K-doped C_{60} molecules are shown as a “four-lobe” pattern in Figure 3F and a “three-lobe” pattern in Figure 3G, which may be assigned to a K-atom embedded in a C_{60} with H:H configuration and a K-atom attached to an H:P oriented C_{60} , respectively. DFT calculations of K-doped C_{60} were performed and simulations of the corresponding STM images are shown in Supplementary Figure 8. In addition, DFT calculations were carried out to analyze and explain the charge density difference for various C_{60} configurations in Supplementary Figure 9.

Between two K-dopants, the orientation of surrounding C_{60} becomes more random and disordered. As shown in Figure 3E, in the area between the dopants, most C_{60} molecules tend to adopt the H:H configuration, while some molecules occasionally adopt the P:D configuration.

A chiral 2×2 configuration around a K-doped C_{60}

C_{60} molecules tend to be either uniformly oriented or staggered in undoped C_{60} multilayer, and they form a chiral (2×2) superstructure in the area near the K-doped C_{60} dopant. In the region near a K-doped C_{60} molecule, Figure 4A shows a so-called chiral 2×2 superlattice. A high-resolution image of such K-doped C_{60} dopant [Figure 4B] shows a bright contrast with a three-lobe pattern, consistent with the proposed structural model of the configuration of “K-bond-C-atoms faced-up” (K:C) in Figure 4C. Three different C_{60} orientations can be observed in Figure 4A, i.e., round ball [Figure 4D], two-lobe [Figure 4E and F], and three-lobe [Figure 4G] shapes, with the latter two corresponding to the H:H and H configurations, respectively. For the C_{60} exhibiting the round ball shape [Figure 4D], it is well known that C_{60} molecules are free to rotate and flip on the surface. Therefore, the molecules do not exhibit a fixed orientation, and STM images of each molecule arise from a time-averaged signal^[49]. Focusing on the three-lobe molecules with H configuration, one may identify a local 2×2 arrangement, as indicated by the yellow triangles and red rhombus in Figure 4A. A similar 2×2 superstructure originating from the partially ordered arrangement of molecular orientations has been observed in the second C_{60} layer grown on the STO surface^[19], where H-oriented C_{60} molecules are well ordered, while others exhibit no long-range orientational order^[50]. Our results are consistent with observations from previous work. The only difference is that the chiral 2×2 superlattice here appears to be located near a K-doped C_{60} , rather than in the undoped C_{60} layer, which may be related to the enhanced charge transfer induced by K doping.

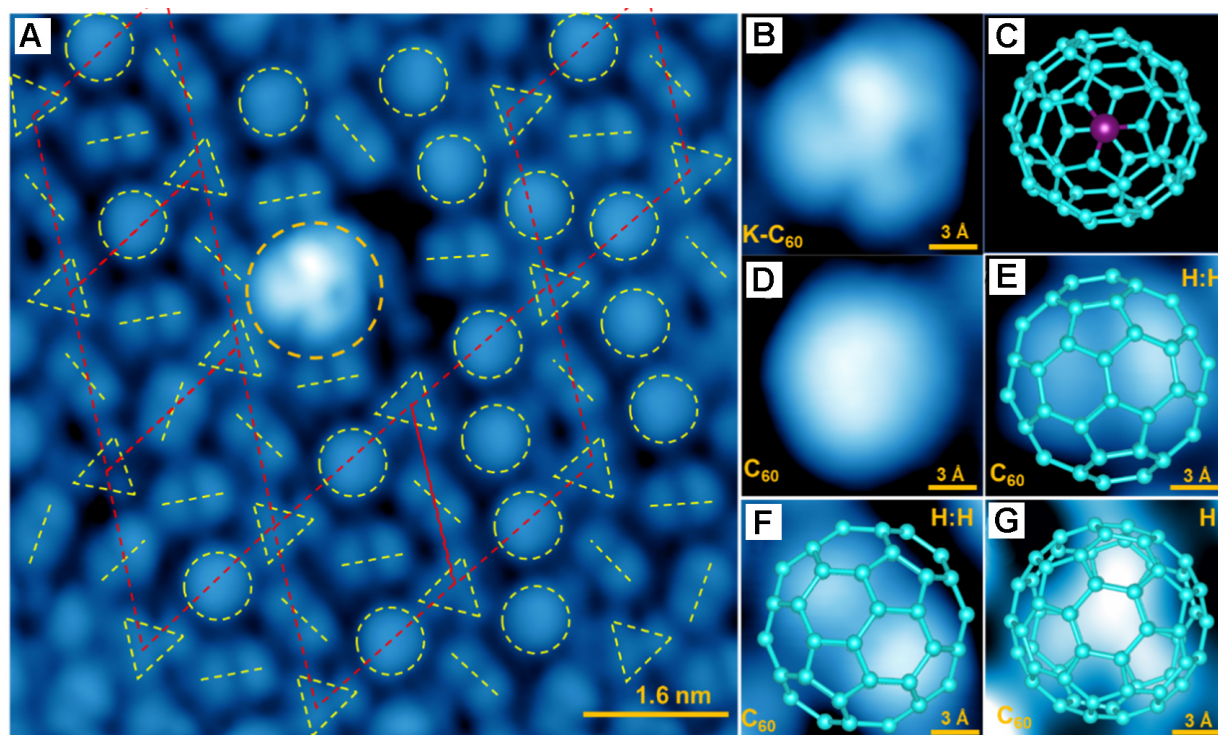


Figure 4. Topographic images of different oriented C_{60} assembled layers with a K-doped C_{60} molecule. (A) STM image of a K-doped C_{60} molecule surrounded by various oriented C_{60} molecules. Image size is $8 \times 8 \text{ nm}^2$ with U_B : +1.0 V and I_T : 50 pA. Yellow colored circles, dashed short lines and triangles, represent different oriented C_{60} molecules; (B and C) High-resolution STM image of a K-doped C_{60} molecule and the proposed structure with K atom replaced C and faced up; (D-G) Images of four different oriented C_{60} , overlapped with the corresponding structure models; Image size is $1.5 \times 1.5 \text{ nm}^2$ with U_B : +1.0 V and I_T : 50 pA; (D) Round ball shape, suggests a rolling C_{60} ; (E and F) Two-lobe shape, suggests H:H configuration; (G) Clover shape, suggests the H configuration. The triangles are arranged in a chiral 2×2 configuration, indicated by red rhombus, as reported previously [19]. C_{60} : Fullerene; STM: scanning tunneling microscopy.

The electronic effect of K-doping in the C_{60} multilayer

K-doping introduces an emergent charge-trapped state in C_{60} multilayer, which may be strongly related to the observed chiral (2×2) superstructure. A similar state has also been found as a charged quasiparticle formed by the coupling of excess charges (i.e., electrons or holes) with ionic vibrations from polarons in monolayer CoCl_2 on highly oriented pyrolytic graphite (HOPG) [50–52]. As shown in Supplementary Figure 10, a DOS peak appears in the STS of the C_{60} multilayer [see Supplementary Figures 11 and 12], located at 1.85–1.9 eV for the P:D and H:H configurations [Supplementary Figure 11], while the VBM lies below the measured spectroscopic range of -3 eV . However, after K-doping, the DOS peak of the K-doped C_{60} molecule shifts downward to 1.54 eV [Figure 5A]. A 0.3 eV energy downshift induced by K-doping reflects electron donation from C_{60} molecules to the K dopant, contrary to electron donation typically observed from K atoms in the artificial Kagome framework [53]. The calculated DOS of the K-doped C_{60} monolayer in Supplementary Figure 6F–H shows a slightly down-shifted central peak compared to the DOS of the non-doped C_{60} monolayer in Supplementary Figure 6A–E, indicating an enhanced electron transfer to K-doped C_{60} . More interestingly, an unexpected effect is revealed by a series of dI/dV spectra measured along the arrowed line crossing the K-doped C_{60} [Figure 5B]. Although the K-doped C_{60} shows a 0.3 eV downshift of LUMO, moving away from the K-doped C_{60} , the neighboring C_{60} molecules show the DOS peak shifted down to 1.24 eV, namely a 0.6 eV downshift compared to the 1.85 eV peak of undoped C_{60} molecules.

The plots for the peak energies as a function of distance to the K-dopant are shown in Figure 5C. The spatial dependence of the peak energy around the K-dopant can be expressed as $E_p = -\exp(r)$, rather than $1/r$ [Figure 5C], where r is the distance to the K-dopant. The peak energy variation may be due to the charge-trapped

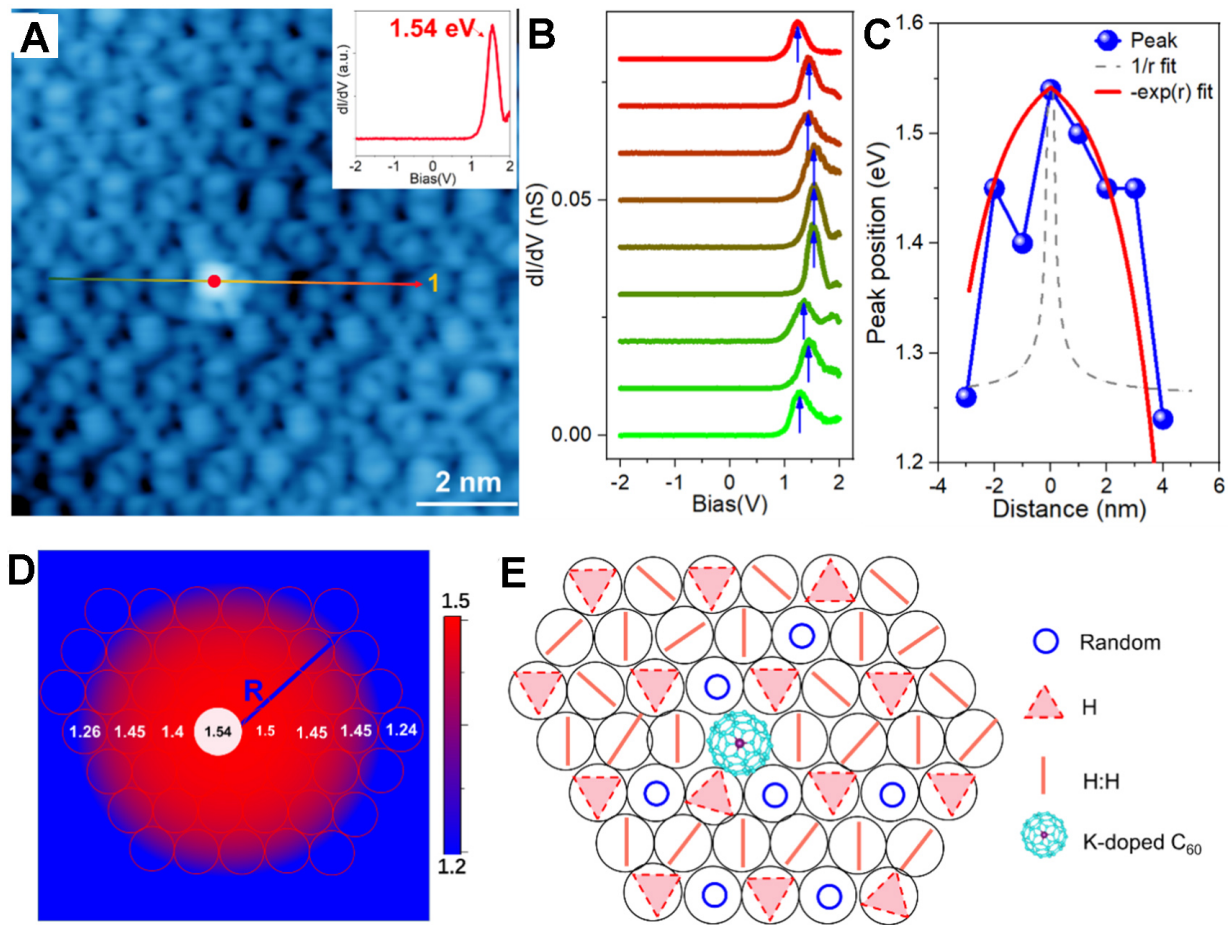


Figure 5. STS measured near K-doped C_{60} point defects. (A) Image of C_{60} layer with a point K-doped defect. (inset) dI/dV spectrum measured at the top of point defect. Image size is $10 \times 10 \text{ nm}^2$ with U_b : +1.6 V and I_T : 10 pA; (B) A linear STS survey measured along the colored arrowed lines in panel (A). The gradient from green to red matches the gradient color of the arrow in image (A), representing the one-to-one correspondence between the atomic sites in image (A) and the STS data in image (B). All STS measured with U_b : 1.6 V, I_T : 200 pA and bias modulation of 20 mV at 4.2 K; (C) Spatial dependence of peak energies measured along line STS survey (blue dots and lines). Grey dashed line is the fitting of $1/r$ and Red dashed line is the fitting of $-\exp(r)$, r is the distance to K-dopant; (D) Energies of peak at various spatial locations. Background is the calculated charge-trapped map based on the formula of $-\exp(r)$. R is the radius of the charge-trapped area, which is about 3 nm; (E) The orientations of each C_{60} molecule in layer. The circle, short bar and triangle represent the shape of round ball, two-lobe and three-lobe, respectively, corresponding to random, H:H and H configurations. The triangles (H configuration) show the quasi- (2×2) superstructure. STS: Scanning tunneling spectroscopy; C_{60} : fullerene.

center at the K-doped site. Such a charge center induces band-bending of the CBM, where the conduction band bends upward around the charged center. This behavior may originate from the screened electric field associated with a charged center^[51], suggesting that a negative charge is located at the center of the feature, consistent with the negatively charged nature of K-doped C_{60} .

The measured peak energies with spatial location of related C_{60} molecules were also plotted in Figure 5D. By comparing the data with the calculated charge-trapped map based on the $-\exp(r)$ expression in the background, the radius of the charge-trapped area (R) was determined to be about 3 nm. The orientational configurations of C_{60} around the K-doped C_{60} molecule were also plotted [Figure 5E]. As seen, the three-lobe configuration (H) forms the quasi- (2×2) superstructure. The trapped charge and related (2×2) superstructure may indicate a possible mechanism of polaron formation.

Excess charge (electron or hole) trapped by self-induced lattice distortion due to electron-phonon coupling interaction (EPI) is known as a polaron^[54]. Since the C_{60} layer is a wide-bandgap semiconductor with the

bandgap of about 4.5–4.6 eV, the induced charge from K doping extends across approximately one unit cell-length of the quasi-(2×2) superstructure (> 2 nm) [Figure 5D], which is indicative of a possible polaronic nature. The polaron scenario, demonstrated in monolayer CoCl_2 on HOPG^[51,52], indicates a composite quasiparticle evolved from the excess carrier trapped by local lattice distortion. The polaronic state (PS) appears inside the gap and is spatially localized^[52]. A significant upward bending of the conduction band near the polaron center has also been observed in monolayer CoCl_2 ^[51]. The energy downshift of the DOS peaks of K-dopant can also be attributed to the polaron formation energy, defined as the energy difference between a PS and the delocalized state at the conduction band edge^[52]. Here, the polaron formation energy of K-doped C_{60} in the surrounding chiral 2×2 superlattice can be estimated to be about -0.3 eV at the center of the polaron (the doping site), considering the CBM at 1.85–1.9 eV. This value is comparable to the -0.24 and -0.29 eV polaron formation energies reported for Co- and Cl-centered polarons in monolayer CoCl_2 on HOPG^[51,52]. In our experiment, such polaron formation is also accompanied by the occurrence of a short-ranged chiral 2×2 orientational C_{60} assembly near the K-dopant site. Excess electrons from doped K in the C_{60} layer, entangled with the oriented assembly of the quasi-(2×2) superstructure, may exert significant influence on the properties of C_{60} layers, such as lattice reconstructions^[55], carrier mobility^[56,57], ferromagnetic transition^[58–60], superconductivity^[61–63], and other many-body correlated states^[64–66].

CONCLUSIONS

In summary, the molecular orientation and electronic structure of potassium-doped C_{60} layers on $\text{SrTiO}_3(001)$ substrates were investigated, revealing key features such as local chiral superstructure, charge-trapped states, and conduction band bending. Fullerene multilayers were synthesized and grown on various $\text{SrTiO}_3(001)$ clean surfaces, followed by controlled K doping.

C_{60} molecules were found to grow layer-by-layer after forming a relatively disordered buffer layer on the $\text{SrTiO}_3(001)$ surface. An isolated C_{60} monomer showed a rotating orientation with a small HOMO-LUMO gap of ~ 1.7 eV, whereas the C_{60} monolayer had a wider HOMO-LUMO gap of ~ 3.65 eV. C_{60} molecules displaying different intramolecular structures were resolved using high-resolution imaging. C_{60} molecules tend to be uniformly oriented or staggered in undoped C_{60} multilayers, while forming a quasi-(2×2) superstructure near the K-doped C_{60} site. K-doped C_{60} molecules introduce a local shifting of DOS peaks and an enhanced imaging contrast, which may indicate charge-trapping effects and/or the formation of polarons. Potassium doping primarily induces a short-range, charge-trapped orientational C_{60} assembly near the K-dopant site and an upward bending of the conduction band due to the screened electric field of a negatively charged center. The underlying substrate acts as a template, influencing the fullerene overlayer through its geometric structure and electronic interaction at the interface. Tuning molecular orientation and electronic states in two-dimensional (2D) fullerene layers can be achieved by the cooperative effect of potassium doping and substrate selection.

These findings elucidate the profound interplay and cooperative regulation between charge, lattice, and molecular rotational degrees of freedom in such interfacial systems. This work provides new perspectives and experimental foundations for understanding and designing fullerene-based 2D functional materials.

DECLARATIONS

Authors' contributions

Conceived and designed the experiment: Pan, M.; Chi, L.

Grew the samples: You, S.; Wang, X.; Wang, J.; Zhang, M.

Performed the STM/STS experiments: You, S.; Wang, X.; Wang, J.; Zhang, M.

Performed the DFT calculations and STM simulations: Xie, M.

Pan, M. wrote the manuscript with input from You, S., Wang, X., Wang, J., Wang, Z., Zhang, M., Xie, M., and Chi, L.

Availability of data and materials

All data analyzed to evaluate the conclusions are available within the paper and its [Supplementary Materials](#). Further source data are available from the authors upon reasonable request.

AI and AI-assisted tools statement

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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Supplementary Materials

[Supplementary Materials](#)

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