

Quasiperiodic arrays of finite-size C_{60} molecular chains induced by anisotropic growth

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Due to their diverse adsorption sites with distinct electronic, chemical, and geometrical properties, quasicrystal surfaces serve as a platform for growing novel overlayer structures. Here, we present the growth of C_{60} molecules on a twofold surface of the decagonal Al-Ni-Co quasicrystal, which possesses both periodic and quasiperiodic order. Scanning tunneling microscopy (STM) reveals that the deposited C_{60} preferentially grows along the periodic direction, producing chains of C_{60} molecules arranged in a Fibonacci sequence, a representative of one-dimensional quasicrystalline order. By comparing STM images at submonolayer coverage with the substrate model structure, we identified that C_{60} molecules preferentially adsorb at Ni/Co atomic sites matching the geometry of C_{60} molecules. The C_{60} molecules experience two competing processes: substrate-molecule interactions and molecule-molecule van der Waals interactions, resulting in chains of limited length. These findings provide insights into tailoring molecular assemblies on quasicrystalline templates, with potential applications in nanostructured materials and molecular electronics.

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I. INTRODUCTION

Quasicrystalline materials have no translational symmetry yet exhibit perfect long-range order [1], typically displaying rotational symmetries such as five-, eight-, and tenfold that are incompatible with periodicity. Quasicrystalline order has been observed in various materials, such as colloids [2,3], soft matter [4,5], perovskites [6,7], and, originally, as intermetallic alloys [1]. Intermetallic alloy quasicrystals can be divided into two families: icosahedral (*i*) and decagonal (*d*). Icosahedral quasicrystals are isotropically quasiperiodic, exhibiting quasicrystalline order in all three dimensions. In contrast, decagonal quasicrystals feature a structure characterized by a periodic stacking of quasicrystalline planes.

The surfaces of icosahedral and decagonal quasicrystals have been extensively studied using a variety of surface-sensitive techniques, as reviewed in the literature [8–11]. Under appropriate preparation conditions, these surfaces form atomically flat terraces at specific bulk planes. These surfaces have been used as substrates for growing unique overlayer structures of single elements and molecules. For example, three-dimensional quasicrystalline films of single elements [12] and quasicrystalline molecular overlayers have been reported [13–16].

The adsorption of molecular thin films on quasicrystal surfaces is of particular interest [13–16]. This is because quasicrystal surfaces exhibit a wide variety of adsorption sites

with distinct electronic, chemical, and geometrical properties, leading to selective and patterned adsorption. Owing to the site-specific adsorption behavior, molecules may also be used as chemical probes and can facilitate the development of functional thin-film materials [14].

In general, the growth of quasicrystalline molecular thin films is governed by a combination of several fundamental physical phenomena, including electron transfer between substrate atoms and molecules [13,14,16–18], symmetry matching between adsorption sites and molecular facets [13,19], molecular size compatibility with adsorption site spacing [16], and growth conditions including substrate temperature [15,20]. For example, electron-accepting C_{60} molecules form quasicrystalline films when deposited onto surfaces with electron-donating minority constituents, such as *i*-Al-Cu-Fe and *i*-Al-Pd-Mn [13,14,16]. The minority constituents not only provide an electronically attractive site but are also sparsely distributed at the surface, such that adjacent spacings are compatible with the size of the molecules, promoting long-range quasicrystalline order. Similar arguments have been made for electron-donating molecules atop electron-accepting constituents, such as pentacene on the *i*-Ag-In-Yb surfaces [14,17,18].

All previous studies on quasicrystalline molecular thin film growth have utilized substrates that are isotropically quasiperiodic, producing isotropically ordered or disordered films. However, the twofold surface of the decagonal quasicrystal family is anisotropically quasiperiodic: comprising both periodic and quasiperiodic directions. Therefore, this surface provides a unique opportunity to simultaneously investigate the influence of quasiperiodically and periodically ordered adsorption sites. Here, we present the first such investigation, reporting on the deposition of C_{60} molecules on the twofold surface of *d*-Al-Ni-Co. We find that, when dosed with the substrate held at elevated temperatures, the molecules

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adsorb preferentially along the periodic direction, forming finite-sized chains, arranged in a Fibonacci sequence, which appear to be dictated by the strong interaction between the molecule and the proposed adsorption sites. Here, the twofold surface converts an otherwise potentially isotropic molecular film into 1D finite chains, revealing how substrate symmetry can control the dimensionality of molecular assembly.

II. EXPERIMENTAL METHODS

The $d\text{-Al}_{71.8}\text{Ni}_{14.8}\text{Co}_{13.4}$ quasicrystal was grown using the Czochralski method and cut along the twofold (10000) surface. Decagonal Al-Ni-Co exists in several phases depending on the exact composition and temperature [22,23]. The sample used in this study corresponds to the type-I superstructure [24]. The surface was then polished with successively finer grades of diamond paste from 6 μm down to 0.25 μm and cleaned in an ultrasonic methanol bath before being inserted into an ultrahigh vacuum chamber with a base pressure of $\sim 1 \times 10^{-11}$ mbar. The surface was imaged using an Omicron VT-STM at room temperature. Surface preparation was performed with 30 min 2 keV Ar^+ sputter (drain current $\sim 4 \mu\text{A}$) and 2 h 700 $^\circ\text{C}$ anneal cycles—measured with a pyrometer set to 0.35 emissivity—until atomically flat terraces could be resolved with STM. STM bias voltage is typically -2 V and tunneling current -0.2 nA, unless otherwise stated in the text. C_{60} was thermally evaporated from a Pyrex tube wound with a tungsten filament. The source and substrate temperatures were monitored via a thermocouple. A substrate temperature during deposition of 250–300 $^\circ\text{C}$ facilitated an ordered thin film indicative of the substrate.

III. RESULTS AND DISCUSSION

A. Substrate structure

Figure 1 shows a bulk atomic model of $d\text{-Al-Ni-Co}$, where Al atoms are depicted by cyan spheres and Ni/Co atoms by red spheres. The bulk truncated tenfold (blue) and twofold (yellow) planes are outlined. The twofold (10000) surface under investigation is comprised of atoms from the periodically stacked [00001] direction and the orthogonal quasiperiodic [001 $\bar{1}0$] direction [25]. The periodic stacking of atomic planes along the [00001] direction is 0.2 nm [22].

Figure 2(a) shows an STM image of the (10000) surface after deposition of C_{60} . Bright circular features represent individual C_{60} molecules, while the vertical columns correspond to the substrate. The C_{60} coverage is 0.20 ± 0.02 monolayer (ML), determined by thresholding values above the flat surface until the substrate features were no longer visible. The columns of the substrate are highlighted by white and blue lines in a magnified image, shown in Fig. 2(b). The columns are separated by long ($L = 0.75 \pm 0.03$ nm) and short ($S = 0.47 \pm 0.02$ nm) distances, where $L = \tau S$, and $\tau = 1.618 \dots$ represents the golden mean, a distinctive feature of quasicrystalline structures. The separations form a Fibonacci sequence across the substrate: within Fig. 2(a), an unbroken Fibonacci sequence of 135 segments is observed, with the ratio of 84 L and 51 S close to τ , as expected for a perfect Fibonacci sequence. Although atomic resolution along the columns is limited, height profiles reveal a periodicity of

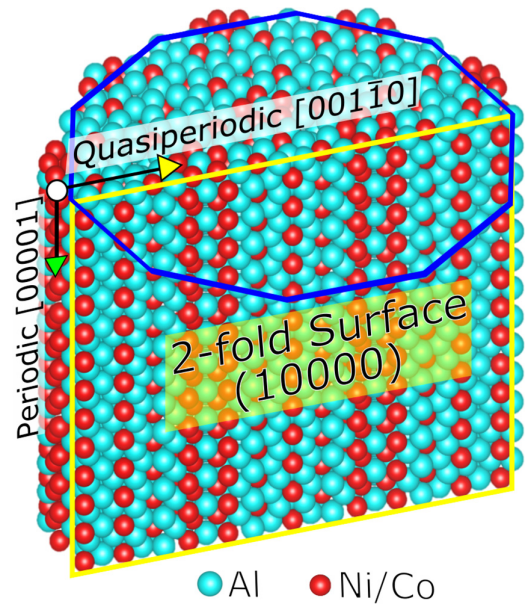


FIG. 1. The bulk structure model of decagonal Al-Ni-Co quasicrystal [21], displaying the tenfold (00001) (marked by a blue decagon) and twofold (10000) (marked by a yellow rectangle) planes. Periodic [00001] and quasiperiodic [001 $\bar{1}0$] directions are indicated with arrows. Al atoms are colored blue and Ni/Co atoms colored red.

0.39 ± 0.02 nm, consistent with the bulk periodicity along the [00001] direction.

The structure observed by STM can be compared to model planes perpendicular to the (10000) direction. Previous work determined that the surface can be considered as a bulk truncation through the center of high-density planes, consisting of both Al and Ni/Co atoms, separated by step-terrace heights of L and S [26–28]. Figure 2(d) illustrates an example of the atomic structure of one such plane, which consists of three layers of atoms, in both side and top views. For clarity, only the center slice of Al atoms are shown in the middle of Fig. 2(d), while all of the Ni/Co atoms are shown in the bottom region. Consistent with the STM results, the columns of Al atoms in the model are separated by long ($L = 0.750$ nm) and short ($S = 0.469$ nm) distances and form a Fibonacci sequence. As such, we overlay the model Al and Ni/Co atoms of an $LLSLSL$ section on top of the substrate columns in Fig. 2(c), such that we attribute the STM signals to Al atoms. The occurrence of brighter and darker columns can then be explained by both the topographical height and density of the atomic species in the model.

More specifically, in Fig. 2(c), we introduce a labeling system: M1 columns consist of singular columns of Al atoms which are separated by an inflated Fibonacci sequence (e.g., LSL and $LSLSL$ segments), while M2 columns consist of groups of singular Al columns which are individually separated by the $LSLSL$ sequences. As such, we find the Al atoms in M2 are more raised than those in M1 and are more densely arranged, such that we assign the M1 columns to the darker signals and the M2 as the brighter. These findings are in agreement with previous STM studies of the same surface [27,29,30] and theoretical calculations [31], which

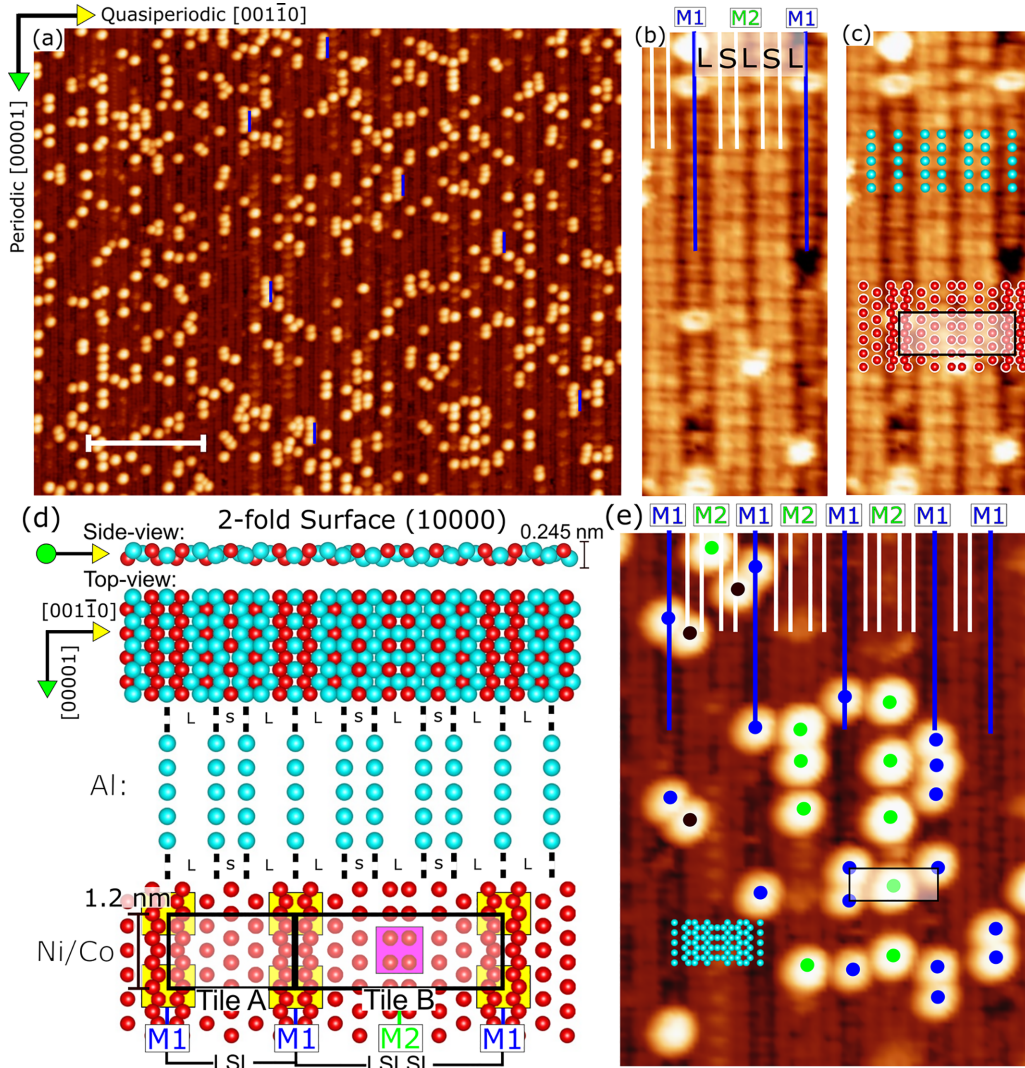


FIG. 2. (a) An STM image of the twofold (10000) surface of decagonal Al-Ni-Co quasicrystal after deposition of C_{60} . The quasiperiodic and periodic directions are indicated with arrows. Atomic columns form along the periodic [00001] direction. The atomic columns are separated by a Fibonacci sequence of short ($S = 0.47$ nm) and ($L = 0.75$ nm) segments. White scale bar = 20 nm. (b) Magnified STM image of the deposited surface, highlighting the substrate area and adsorption sites M1/M2 of C_{60} . (c) The same area as in (b), with the center slice of Al, which forms a Fibonacci sequence, and Ni/Co models overlaid. (d) The atomic structure of the twofold (10000) surface. A 0.245 nm slice of the surface is selected at the center of the high-density regions of the bulk. Center slice Al and Ni/Co atoms of the surface are shown separately. Al-atom columns are separated by short ($S = 0.469$ nm) and long ($L = 0.750$ nm) distances forming a Fibonacci sequence. M1 (blue) and M2 (green) indicate two different types of Ni/Co columns. (e) Magnified STM image showing M1 and M2 sites, indicated over adsorbed C_{60} molecules. Colored dots are placed over C_{60} molecules to show their adsorption sites. Blue dots = M1, green dots = M2, and black dots at remaining sites. A 0.2 nA tunneling current and a bias between ± 2 mV is used for STM.

demonstrated that the tunneling current in Al-based quasicrystals is predominantly contributed by the s - p electrons of Al atoms. As a result, Al atoms contribute more to the local density of states and appear as brighter contrasts compared to Ni/Co atoms in the STM images.

As a tool to aid our discussion of C_{60} adsorption, the atomic surface structure is mapped using two rectangular tiles, tile A and tile B, shown in Fig. 2(d). These tiles follow a Fibonacci sequence of long ($L = 0.75$ nm) and short ($S = 0.47$ nm) segments:

$$\text{tile A} = L + S + L = 1.97 \text{ nm}, \quad (1)$$

$$\text{tile B} = L + S + L + S + L = 3.19 \text{ nm} (\tau \times \text{tile A}). \quad (2)$$

Both tiles share a short edge of 1.2 nm along the periodic direction. The vertices of the tiles are decorated with squashed hexagonal motifs of Ni/Co atoms, labeled M1 sites [yellow in Fig. 2(d)], located along high-density Ni/Co columns. These M1 motifs repeat have a periodicity of 0.4 nm along the columns, enabling two motifs to fit along the short edge of a tile. Consequently, the tiling can be translated by 0.4 nm along M1 columns and still decorate a subset of M1 sites. The center of tile B contains a rectangular arrangement of Ni/Co atoms [purple in Fig. 2(d)], termed M2 sites. Unlike M1 sites, which include subsurface Ni/Co atoms [0.08 nm below the top layer; see Fig. 2(d) side view], M2 sites consist solely of top-layer atoms. Tile B's structure is overlaid on the

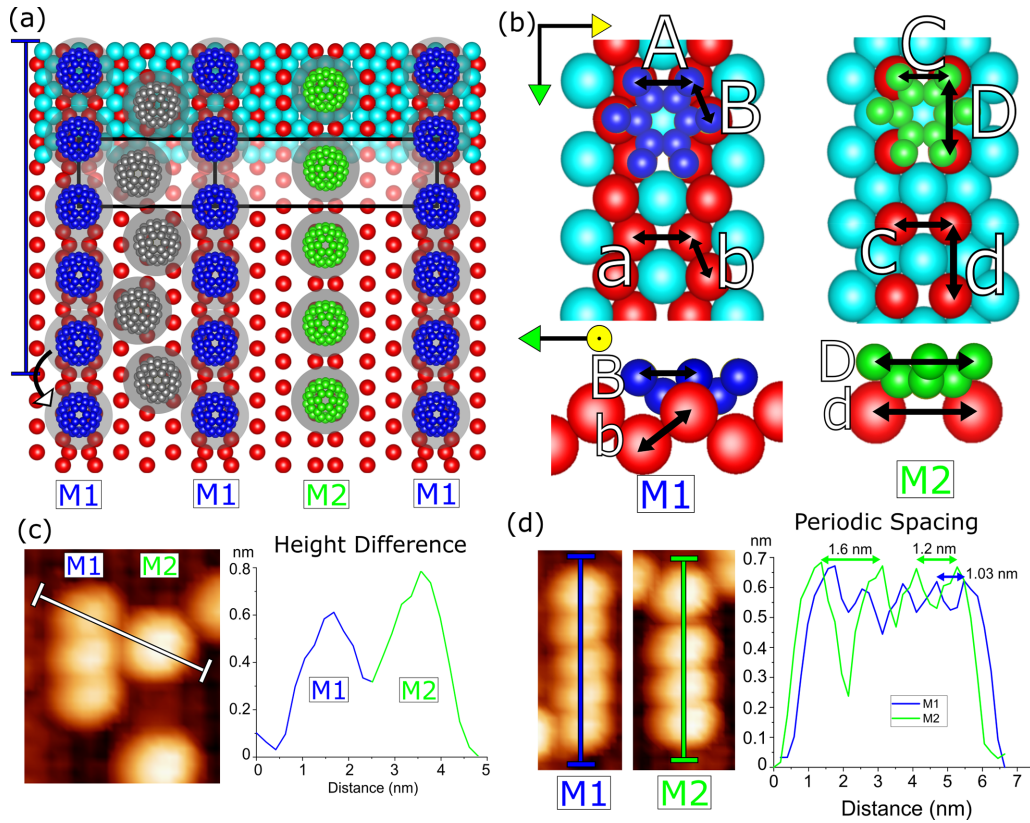


FIG. 3. (a) The model structure of the twofold *d*-Al-Ni-Co surface with C₆₀ molecules decorated. The adsorption sites are extracted from STM images, whereby blue, green, and black C₆₀ molecules represent molecules adsorbed at M1, M2, and elsewhere (black sites). Shaded circles indicate the van der Waals diameter of the molecules. The top rows include both Al (cyan) and Ni/Co (red) atoms and the remaining rows show only Ni/Co atoms. (b) A magnified view of the symmetrically parallel orientation of C₆₀ molecules with the substrate Ni/Co atoms at both the M1 and M2 sites. Side views are shown at the bottom. Distance between substrate atoms (a, b, c, and d), and C-C atom distances in C₆₀ (A, B, C, and D) are marked. (c) An example STM image with a height profile showing C₆₀ molecules atop M2 are measured, on average, 0.1–0.2 nm higher than those atop M1, consistent across the surface. (d) STM images with chains of C₆₀ at M1 (blue) and M2 (green) sites are shown with the minimum observed periodic spacing of 1.03 nm (M1) and 1.20 nm (M2), and the corresponding line profile.

Ni/Co model in Fig. 2(c) and correlated with C₆₀ adsorption in Fig. 2(e).

B. C₆₀ adsorption

1. Experimental Observations

Overlaying the substrate model on our data suggests that C₆₀ molecules adsorb at three distinct sites marked in Fig. 2(e): M1 columns (blue dots), M2 columns (green dots), and black sites (black dots). We see that C₆₀ molecules on M1 columns, separated by an average distance of 1.03 ± 0.06 nm, form finite chains, with a maximum observed chain length of five molecules (5.2 ± 0.2 nm). Similarly, C₆₀ molecules on M2 columns separated by 1.20 ± 0.04 nm also form finite chains, with a maximum observed chain length of three molecules (3.45 ± 0.2 nm). Typical spacing between C₆₀ molecules on M1 and M2 columns is shown in the line profiles in Fig. 3(d). There is a height difference observed such that C₆₀ molecules atop M1 sites consistently appear 0.1–0.2 nm lower than those atop M2 sites. This is illustrated in an STM image of C₆₀ molecules adsorbed on M1 and M2 sites, along with the corresponding height profile, shown in Fig. 3(c). The apparent height difference is attributed to topological

height, electrical characteristics that caused the substrate M1 columns to appear darker than M2 columns, and bonding contributions from subsurface Ni/Co atoms, discussed further later. Finally, the molecules marked by black dots are typically adjacent to M1 molecules such that the distance between them is 1.00 ± 0.05 nm, close to the van der Waals separation of two C₆₀ molecules [15,32,33]. Among the 611 C₆₀ molecules observed in Fig. 2(a), 63% are adsorbed atop M1, 28% atop M2, and 9% on black sites. We note that a similar anisotropic distribution of C₆₀ molecules is observed at higher coverages (~ 0.5 ML), although the increased molecular density induces close-packing motifs between the chains; with increasing coverage the inter-molecular interactions begin to dominate as the preferential adsorption sites are already occupied.

2. Modeling adsorption

Almost all features of C₆₀ molecules observed by STM can be explained by an adsorption model, a section of which is shown in Fig. 3(a), with M1 sites marked in blue, those on M2 sites marked in green, and molecules adsorbed elsewhere (black sites) marked in black. The shaded circle around each molecule corresponds to its van der Waals radius. The model

is built in tandem between the predicted most-favored adsorption sites and experimentally observed values in STM. The squashed hexagons of Ni/Co atoms chosen for the vertices of tiles A and B now act as the most-favored site for the M1 motif. The hexagonal face of C₆₀ is oriented in contact with the M1 motif, with C atoms likely to bond with surface atoms shown magnified in Fig. 3(b). Two sets of finite chains with different intermolecular separations are observed, one set atop M1 and the other atop M2. The first finite chain is formed when C₆₀ is centered atop the M1 motif, indicated in yellow in Fig. 2(d), has nearest neighbors that are displaced from the next favored motif site, with a C₆₀-C₆₀ separation of 1.03 nm. Next, the rectangle of Ni/Co atoms at the center of tile B, indicated in purple in Fig. 2(d), represents the M2 motif. The second finite chain forms between C₆₀ molecules absorbed atop these motifs, with a separation of 1.2 nm. Tiles A and B then need to skew to accommodate M1 and M2 chains with up to 0.4 nm shifted motif centers. This 0.4 nm shift allows for chains to be centered around any M1 motif, agreeing with STM observations. Finally, C₆₀ molecules are absorbed atop black sites bonded to M1 molecules, with a distance consistent with that in the hexagonal close-packed structure of C₆₀, i.e., the C₆₀ - C₆₀ distance is equal to the van der Waals radius [34]. The finite length of these chains is discussed in detail later.

The expected coverage frequency of a single monolayer of C₆₀ at each adsorption site can be calculated by generating an extended model (single layer, 26.3 nm × 29 nm) of Fig. 3(a), including M1 (blue), M2 (green), and the remaining (black) sites, with 513 molecules. In this model, molecules are placed with no overlap: starting with occupying the M1 and M2 sites first, then black sites which populate at least after M1 sites are occupied. The model frequencies are M1 = 54.6%, M2 = 24.4%, and black sites = 21%. The model distribution supports our experimental observations at 0.2 ML coverage (M1 = 63%, M2 = 28%, black = 9%), with a similar $\frac{M1_{\text{model}}}{M2_{\text{model}}} = 2.25$ and $\frac{M1_{\text{exp}}}{M2_{\text{exp}}} = 2.24$. Additionally, the lower proportion of black sites observed at 0.2 ML coverage supports a stepwise adsorption at black sites after M1 sites populate.

The proposed adsorption model is further supported by electronic bonding, high-temperature adsorption, and local geometry. The apparent STM height difference of raised M2 molecules compared to M1 molecules previously discussed can be attributed, in part, to subsurface Ni/Co atoms only present along M1. There is a 0.08 nm height difference of these subsurface Ni/Co atoms relative to the topmost layer. These subsurface Ni/Co atoms act as additional electron donors, bonding and pulling the C₆₀ molecule closer, increasing the strength of the bond between the C₆₀ molecule and substrate. A stronger covalently bonded C₆₀ molecule will also appear dimmer due to a lower tunneling current between the surface and STM tip. M1 sites must therefore be rich in Ni/Co electron donors. A combination of these factors leads to C₆₀ molecules appearing darker when absorbed along M1, compared to M2, as seen with STM observations. This observation is supported by high-temperature growth promoting the adsorption of C₆₀ molecules onto the subsurface Ni/Co atoms. While subsurface atoms are not readily accessible at room temperature, annealing the substrate lowers the energy

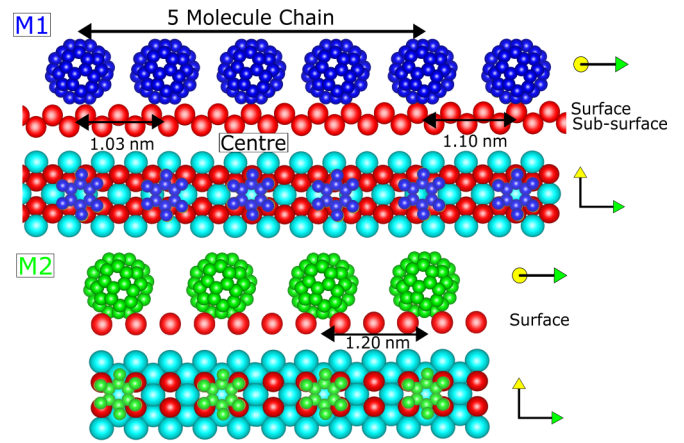


FIG. 4. A model chain of C₆₀ molecules at the M1 (blue), and molecules decorated at M2 (green) sites are shown in side and top views. The distance between M1 molecules in chains is 1.03 ± 0.06 nm, determined from STM data, which is close to the van der Waals separation between molecules. Chains of up to five molecules are observed with STM. The next molecule in the chain (top right blue) shifts to the next most favorable M1 state away from the van der Waals separation. Only the contact C atoms with the surface Ni/Co are shown in the top view.

barrier, facilitating bonding with Ni/Co. We note that an ordered film is only found when the substrate is annealed between 250–300°C during deposition, a disordered film is produced at room temperature across all coverages. These findings are consistent with previous works on adsorption of C₆₀ molecules on the fivefold surface of various icosahedral quasicrystals [13,14]. M2 columns are less rich in Ni/Co donors but still facilitate a high Ni/Co density at M2 motif sites for a strong C₆₀-substrate interaction. Finally, the local symmetry and dimensions of M1 and M2 motif sites match those of C₆₀ molecules. For example, the distances between Ni/Co atoms in M1 sites ($a = 0.289$ nm and $b = 0.241$ nm) are close to the nearest-neighbor distances of C atoms in the hexagonal faces of C₆₀ ($A = 0.3$ nm and $B = 0.239$ nm), as shown in Fig. 3(b). Similarly, at M2 sites, the Ni/Co-Ni/Co distances ($c = 0.287$ nm and $d = 0.4$ nm) match the second-nearest-neighbor distances of C atoms in the hexagonal face of C₆₀ ($C = 0.3$ nm and $D = 0.466$ nm).

3. Finite-chain analysis

Finite chains along M1 can be explained by a compounded translational mismatch, caused by the very strong substrate-C₆₀ bonding, and C₆₀-C₆₀ interactions. First, the observed C₆₀-C₆₀ separation within chains along M1 of 1.03 nm is close to the C₆₀ van der Waals separation of ≈ 1.00 nm [15,32,33], suggesting these molecules are bonded with each other. To explain finite length of C₆₀ chains, we propose the model illustrated in Fig. 4. The chain begins with a molecule where six C atoms are aligned with the M1 motif, corresponding to the hexagon of Ni/Co atoms (marked as the center). Additional C₆₀ molecules are bidirectionally added along the periodic direction along the column. The distance between C₆₀ molecules is set to the value determined from STM measurements. Tiles A and B accommodate the minimum van der

Waals separation of 1.00 nm and standard M1 motif spacing of 0.4 nm, leading to the available separation between M1 motifs in a chain being 1.2 nm. Given the available separation between M1 motifs equals 1.2 nm, and C_{60} – C_{60} separation equals 1.03 nm, and the mismatch between the first site is $\delta = 0.17$ nm. As the chain grows, the C atoms in subsequent molecules gradually displace further out of alignment with the M1 motif until a critical displacement length Δ_{crit} is reached, where the hexagonal face of C_{60} no longer matches the M1 motif hexagonal. Δ_{crit} corresponds to half the available M1 motif separation between the vertices of Tile A and B, $\Delta_{\text{crit}} = \frac{1.2}{2} = 0.6$ nm. If the chain is displaced by greater than 0.6 nm then the next molecule grown would interact too weakly with the local environment and would therefore prefer to shift away from the chain and to the next available M1 motif site. We are able to predict the number of molecules in a chain with a cumulative displacement equation that models the mismatch between number of molecules n , δ , and Δ_{crit} :

$$n_{\text{crit}} = \frac{\Delta_{\text{crit}}}{\delta} = \frac{0.6 \text{ nm}}{0.17 \text{ nm}} \approx 3 \text{ molecules.} \quad (3)$$

Therefore, our model predicts that bidirectionally only three molecules may grow along a chain, with a total chain length of five molecules (assuming the center molecule is centered on an M1 motif site). The total critical length of a chain of C_{60} molecules along M1 is also 4.12 nm (1.03 nm $\times$ 4).

Experimentally, the critical length is determined to be equal to five molecules, with a center-center length of 4.12 ± 0.15 nm, derived from STM data [some chains indicated by blue lines in Fig. 2(a)]. Chains of up to five molecules are also observed in the high-coverage data. No chain longer than five molecules with the minimum separation is observed at any coverage. The observed critical length of five molecules agrees with the determined n_{crit} growing bidirectionally from a molecule centered on an M1 motif site. Once the chain reaches this critical length, the next molecule is dominated by molecule-substrate interactions rather than C_{60} – C_{60} Van der Waals forces. This pulls it to the next M1 motif with a separation from the chain of 1.10 ± 0.05 nm, represented by the rightmost blue molecule in Fig. 4. The critical threshold arises due to the cumulative translational displacement between the molecules and the substrate.

C_{60} absorbed on M2 motifs and black sites can also be interpreted in terms of the substrate composition. Molecules of C_{60} absorbed along M2 are determined to be dominated by substrate– C_{60} interactions. The molecular separation along M2 closely matches the separation between the centers of tile B (≈ 1.2 nm). This can be explained by the density of Ni/Co atoms only being sufficient at M2 motifs. Without the contribution of subsurface Ni/Co between M2 motifs (found along M1), substrate– C_{60} bonding dominates. This in turn leads to C_{60} absorbed along M2 to primarily reside atop the M2 motif. The only examples of dominating C_{60} – C_{60} interactions occur at the black sites [black molecules shown in Figs. 2(d) and 3(a)] that are bonded to M1 molecules via van der Waals interactions. This is because they exist in a metastable environment without a high-density Ni/Co motif but are sufficiently satisfied to bond with both C_{60} and the

substrate. If the black site C_{60} – C_{60} interaction was as strong as the substrate interaction, we would see longer chains.

IV. CONCLUSION

We have presented the growth of C_{60} molecules on the twofold surface of a decagonal Al-Ni-Co quasicrystal, studied using STM. This surface features both periodic and quasiperiodic directions, and we found that C_{60} preferentially grows along the periodic direction, forming chains of C_{60} molecules separated by a Fibonacci sequence. The C_{60} adsorbs on Ni/Co sites, where the symmetry of these sites closely matches that of the hexagonal faces of C_{60} . Additionally, the separation of Ni/Co atoms was comparable to the C–C distance in the hexagonal face of C_{60} . The growth of these chains was governed by two competing interactions: substrate-molecule and molecule-molecule van der Waals forces, which resulted in chains of finite length.

The self-limited nature of these chains is unique among other molecular epitaxy studies conducted on quasiperiodic surfaces, and our finding that the effect of quasiperiodic anisotropy quantizes the C_{60} assembly length has relevance for molecular electronics and templated nanostructures. As our chains can effectively be considered as a set of molecular resonators coupled to a metallic substrate, it would therefore be of further interest to probe their electronic states to compare to similar low-dimensional molecular systems [35,36], for example. Similarly, our chains represent an almost completely inverse analog to a recent study which produced 1D chains of quasiperiodically distributed copper boride grown on periodic Cu(110) [37]. Comparing and contrasting the properties of these two systems may provide further insight into 1D quasicrystals, highlighting how quasiperiodicity can arise either from the overlayer or from the substrate, and how both routes embody the same fundamental principles of frustrated epitaxy and dimensional confinement. Finally, we note that C_{60} has previously been explored from a molecular bearing perspective in which the molecule may act as a frictional lubricant at a surface [38,39]. Therefore, it would be of interest to assess how the nanotribological properties of the twofold surface are affected by the molecular overlayer, particularly as this facet displays a high friction anisotropy, where friction is eight times higher along the periodic direction than compared to the aperiodic direction [40].

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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