Supplementary Information

Carbon Monoxide Stripe Motion Driven by Correlated Lateral Hopping in 1.4 × 1.4 Monolayer Phase on Cu(111)

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CO sub-monolayer coverages on Cu(111) at cryogenic temperatures in UHV

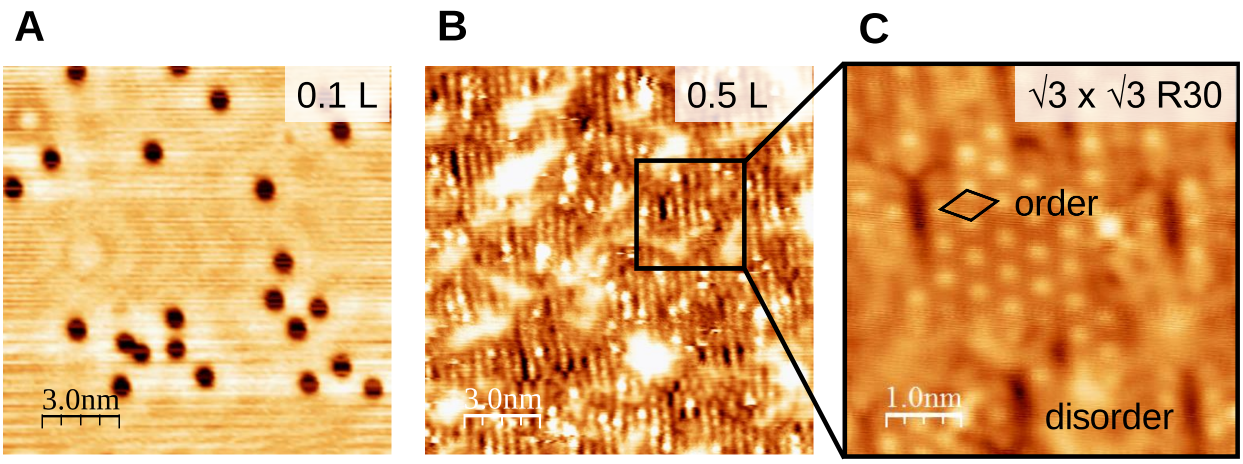

Figure S1. STM images (15 × 15 nm^2, V_s = –40 mV, I = 1 nA) of a Cu(111) surface with (A) 0.1 L and (B) 0.5 L CO adsorption in UHV at 20 K. All STM images were taken at 4.6 K. In (A), CO single molecules are observed as dark spots as a metal W tip was used to scan. In (B), ordered √3x√3 R30 domains and other disordered domains are mixed. (C) Enlarged image in (B).

Silver mean discussion

Fig. S2 shows STM topographic images of the CO rows on Cu(111). We can see the rows has a sequence of D S D S D S S D S S D S. The S -/D sequence is not periodic. Thus, once we thought this is just a random sequence. As shown in the FFT image in Fig .2 in the main text, the sub-main peak has the ration of 0.405, and this value is comparable to the irrational numbers of 0.414 = 1/(√2 + 1). These numbers have been known as factors of the silver mean value [1-3], which is related to a quasi-crystal-like behaviour [1]. Smith et al. discussed on the formation of a "silver mean" quasi-periodic Ag films on GaAs.

A quasi-crystal is a crystal that has a non-periodic sequence in the arrangement of the atoms or unit lattice. Non-periodicity requires a material parameter to follow a certain sequence related to the Fibonacci sequence or bronze-Fibonacci sequence, etc. These are sets of metallic mean or ratios found in shapes and structures of *natural occurrence*.

The silver mean is in analogy to the well-known golden mean or the Fibonacci sequence. A golden mean sequence is a sequence where each term is the sum of previous term and the term before that. i.e; $F_n = F_{n-1} + F_{n-2}$. The golden mean sequence ABAABABAAB…, can be formed by the iteration of $S_{n+1} = \{S_n, S_{n-1}\}$ from the seeds (building blocks) of $S_1 = A$, $S_2 = \{A, B\}$.

However, for a silver mean sequence, each term is the sum of twice the previous term and the number before that. i.e; $F_n = 2F_{n-1} + F_{n-2}$. The silver mean sequence can be formed by the iteration $S_{n+1} = \{S_n,$

S_n, S_{n-1}}, with the same building blocks of A and B which result in the sequence ABAABABABAABABAA….

The real space arrangements of the CO chain sequence were found to be in good agreement with the silver mean sequence. From Fig. S2A, the sequence of the single and double rows are as follows; DSDSDSSDSSDS… The first half of the sequence DSDSDSSD follows the silver mean sequence of BABABAAB. The second half of the sequence SDSSDSD also follows the silver mean sequence ABAABAB. Analysis of a strip of the STM image shows that the sequence follows exactly of the silver mean until a certain coherence length. This would also occur in other strips of the STM image. These positions are where the silver mean sequence is broken (limited coherence length of less than 7 nm).

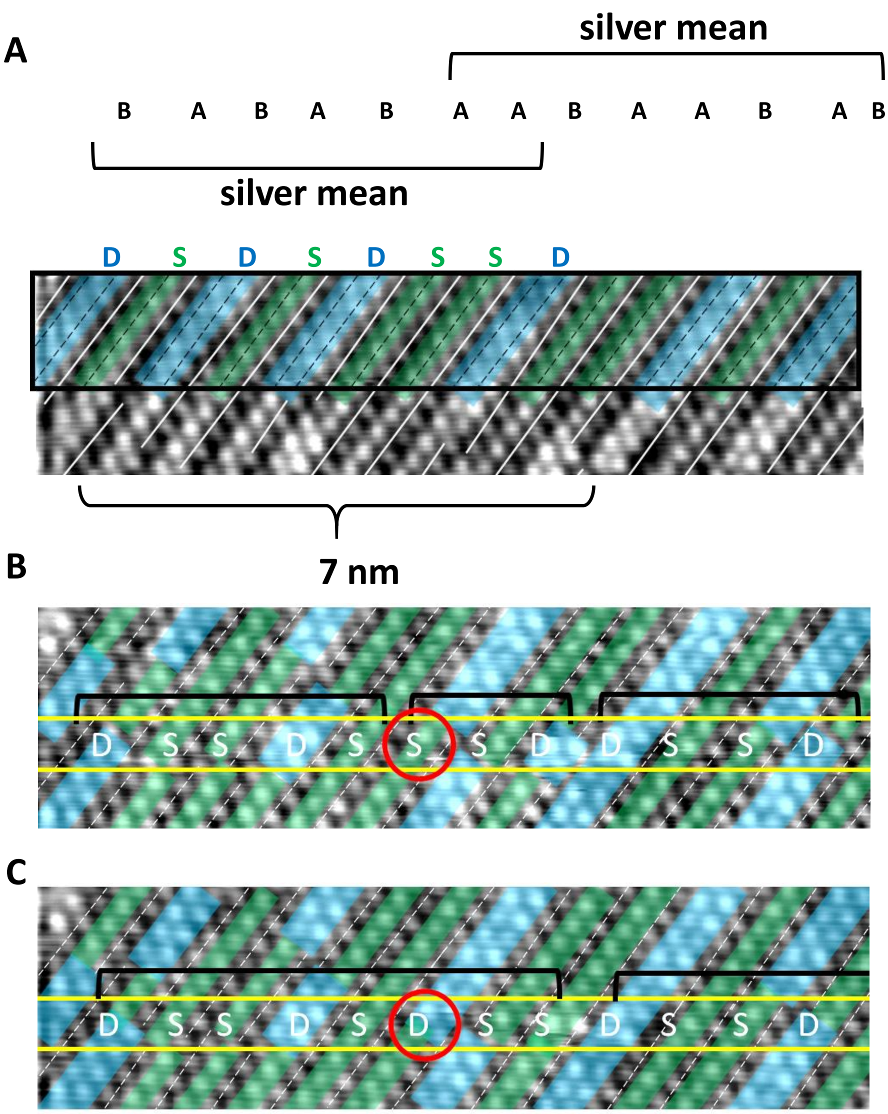

Figure S2. Possibility of "Silver mean" sequence in CO rows on Cu(111). (A) BABABAA or AABAABAB denote "Silver mean" sequence, which partially correspond to DSDSDSSD sequence in CO rows, whike the sequence ended within 7 nm. (B) and (C) denote before and after the hopping, where DSSDSSSDDSSD changed to DSSDSDSSDSSD.

However, this does not occur regularly and are different for some areas of the CO monolayer. This can be seen from Fig. S2B to Fig. S2C. To avoid confusion, we will discuss the strip marked with a yellow box. The first half sequence in Fig. S2B follows DSSDSS (silver mean). It is followed by a SSSDSSD. After scanning, the next image (Fig. S2C), the S changes to a D (red circle). This makes another silver mean sequence of DSSDSDSS in Fig. S2C. We concluded that, this occurrence does not provide enough evidence to support the argument that this is a silver mean sequence.

Note: Golden mean value = 1.618… Silver mean value = 2.414… = $\sqrt{2} + 1$. Bronze = 3. 302…

References

[1] Smith, A. R.; Chao, K.-J.; Niu, Q.; Shih, C.-K. Formation of Atomically Flat Silver Films on GaAs with a Silver Mean Quasi Periodicity. *Science* **1996**, 273, 226–8.

[2] Ali, M. K.; Gumbs, G. Quasiperiodic dynamics for a generalized third-order Fibonacci series. *Phys. Rev. B* **1988**, 38, 7091–7093.

[3] Gumbs, G.; Ali, M. K. Dynamical maps, Cantor spectra, and localization for Fibonacci and related quasiperiodic lattices. *Phys. Rev. Lett.* **1988**, 60, 1081–1084.

Successive STM topographic images showing continuous hopping

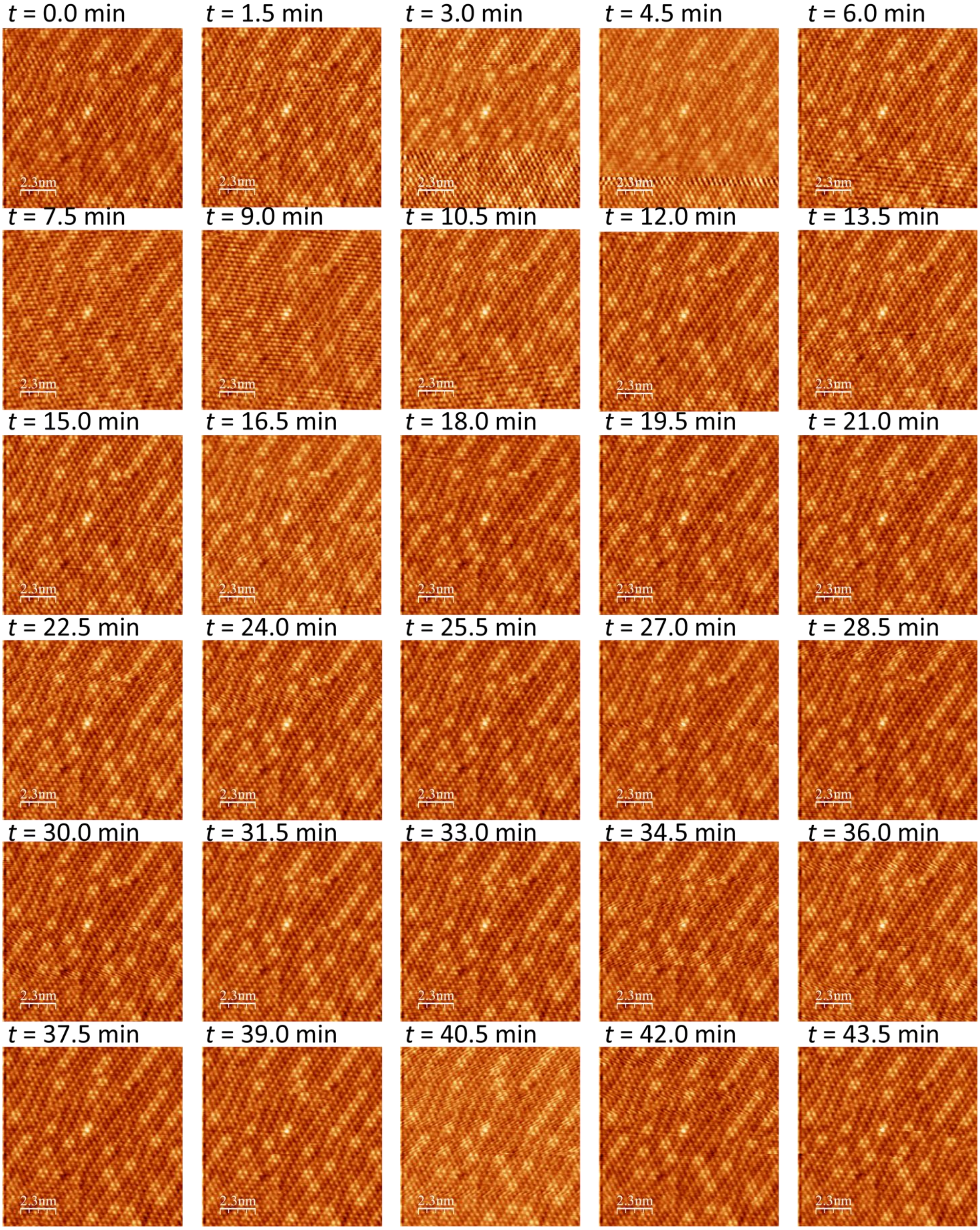

Figure S3. Successive 30 STM topographic images (t = 0 – 45 min) showing continuous correlated CO hopping on Cu(111) at 4.6 K in UHV.

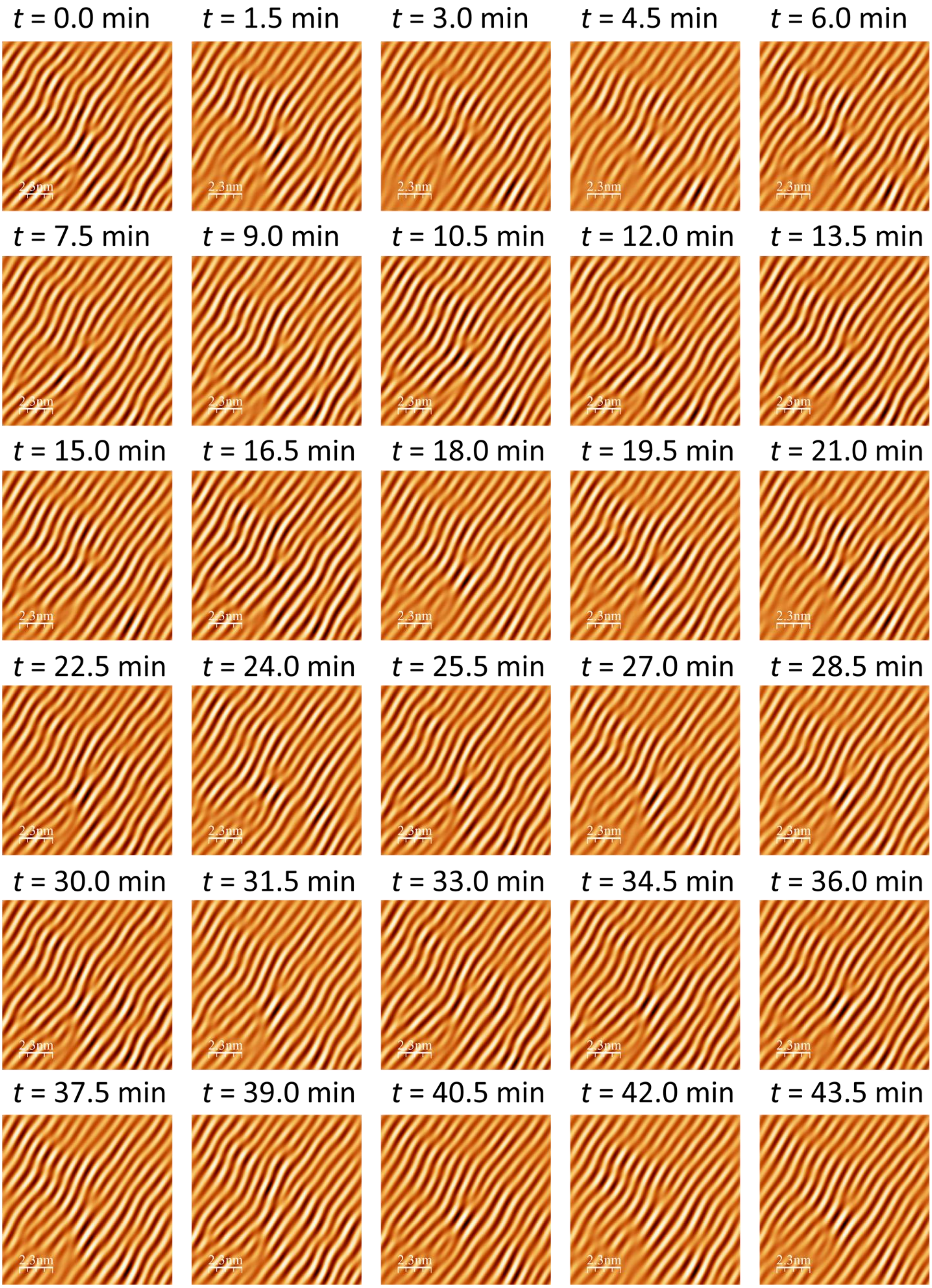

Figure S4. Successive 30 Inverse FFT images (t = 0 – 45 min) using the two sub-main peaks, clearly showing continuous CO stripe formation wave motion on Cu(111) at 4.6 K in UHV.

Spectroscopy measurements on CO rows on Cu(111)

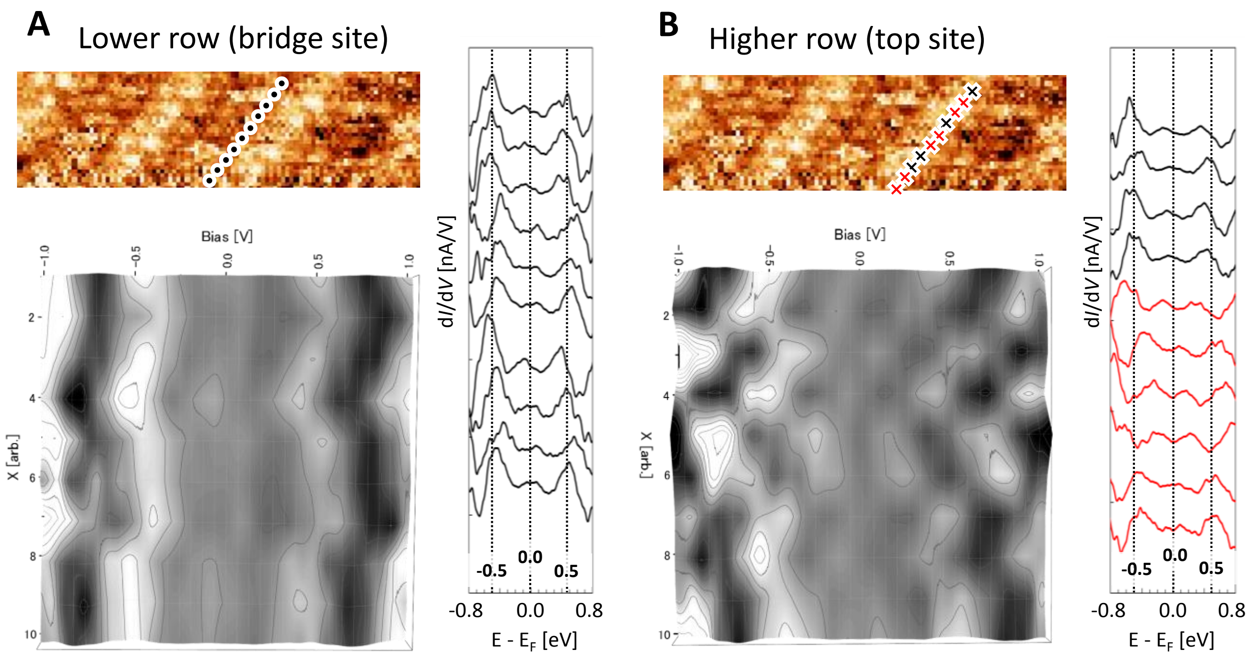

Figure S5. STS grid mode results obtained on (A) lower rows and (B) higher rows. Experimentally obtained scanning tunneling spectroscopy d*I*/d*V* curves (setpoint: $V_s = -40$ mV, $I = 1$ nA), which are proportional to sample LDOS. Upper panels denote STM topographic image and d*I*/d*V* curves (right panels) were measured at dotted points. Lower panels show d*I*/d*V* plots along the row. x and y axes denote the sample bias voltage and position on the row respectively.

Schematic diagram of the CO unit lattice on Cu(111)

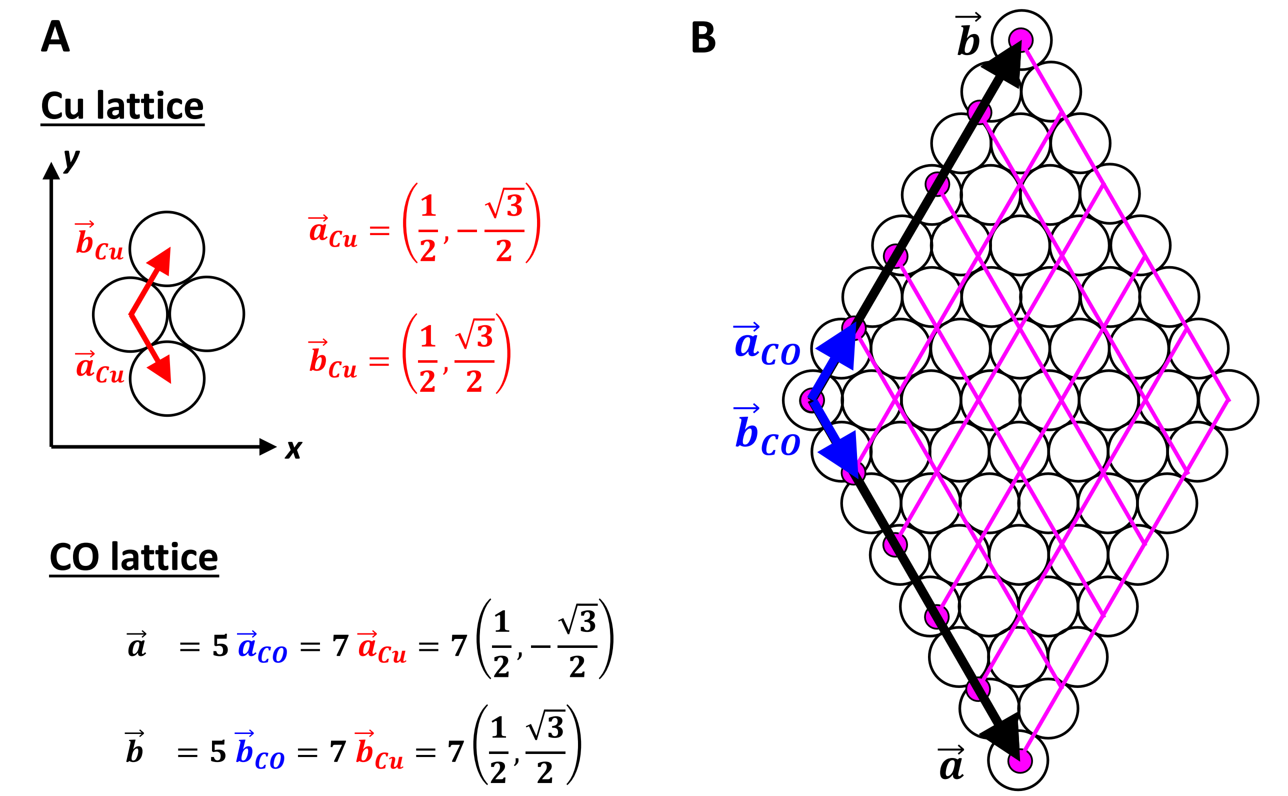

Figure S6. Schematic diagram depicting the Cu lattice unit vectors and the corresponding CO unit lattice.

Scanning tip effect: comparison between forward and backward scan images

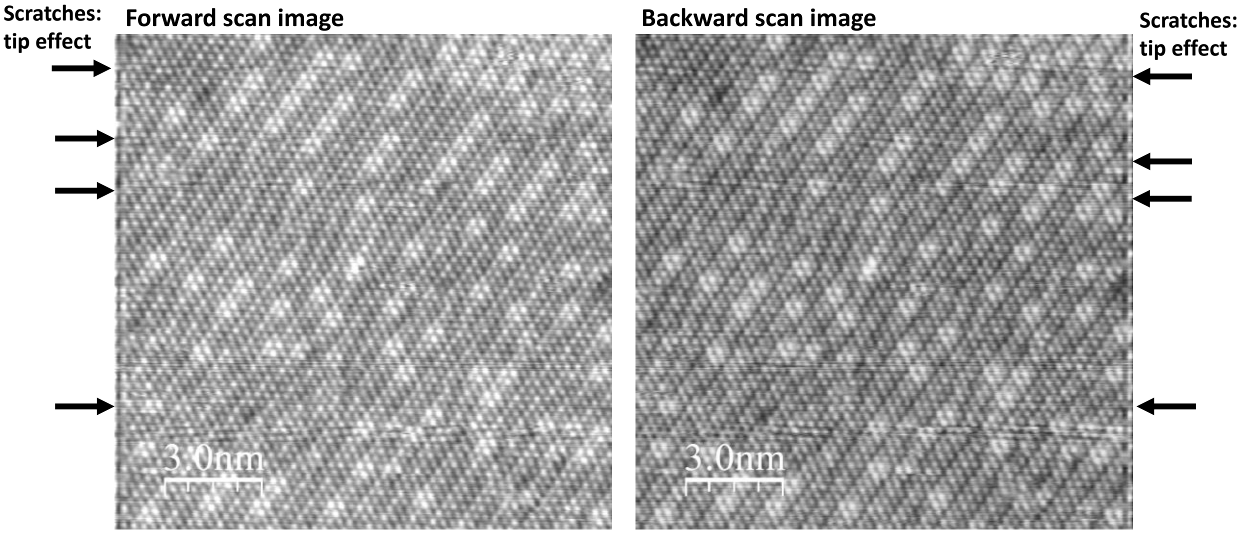

Figure S7. STM topographic images obtained on the CO 1.4 × 1.4 array at 4.6 K (V_s = 40 mV, I = 0.73 nA). Right panel images were obtained when the STM tip scanned from left to right direction (forward scan). Left panel images were obtained when the STM tip scanned from right to left direction (backward scan). Scratches are marked by the arrows.