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Supplementary Materials for

Atomic-scale visualization of surface-assisted orbital order

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The PDF file includes:

Other Supplementary Material for this manuscript includes the following:
(available at advances.sciencemag.org/cgi/content/full/3/9/eaao0362/DC1)

- movie S1 (.mov format). Evolution of dumbbell structure by decreasing the TSD.

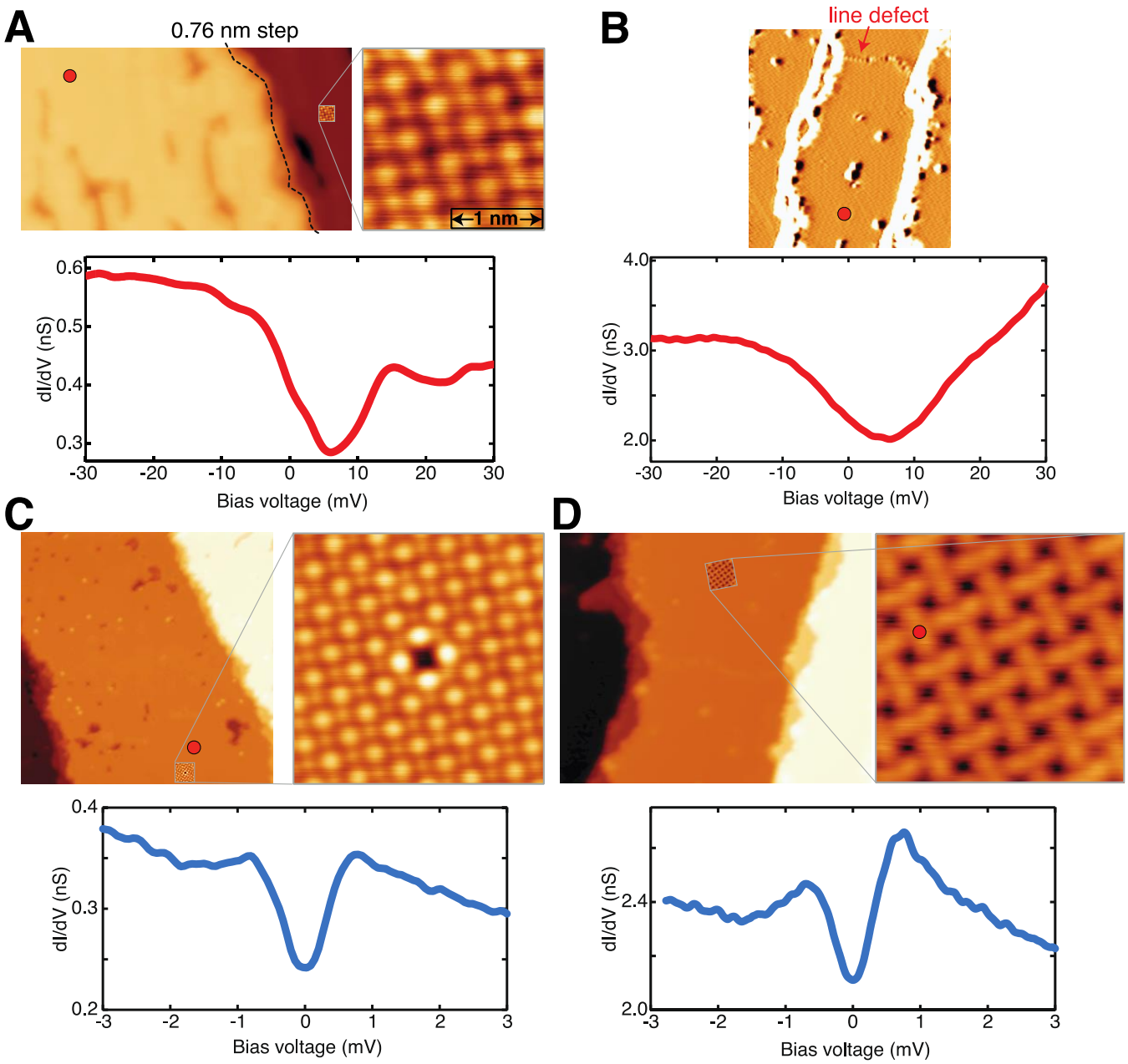

fig. S1. Tunneling spectra taken on CeIn- and Co-terminated surfaces and their locations. Red markers on the STM images indicate the positions where we obtained the corresponding spectra. (**A**) The spectrum taken on the surface where we can visualize Ce atom with small tip-sample distance. Spectrum shows a dip around 5 mV as in the case of CeIn planes reported in Ref. 18-20. (**B**) Spectrum taken on the surface where we can recognize a line defect explained in the main text as a domain boundary of the d_{yz}-d_{xz} orbital ordered structure on a Co terminated surface. The main feature of the spectrum, which has a dip around 5 mV, is similar with the one on the surface of fig. S1(**A**). The slight difference in shape is probably due to different tip status. (**C, D**) Superconducting gap spectra around E_F taken on the surface shown in Fig. 1(**B**) and the surface with the dumbbell structure, respectively. A superconducting gap with $2\Delta \sim 1$ mV is observed on both surfaces. Although the CeIn and Co surfaces were claimed distinguishable in Refs. 18 and 19 based on the spectral features, we do not find distinct difference in their spectra.

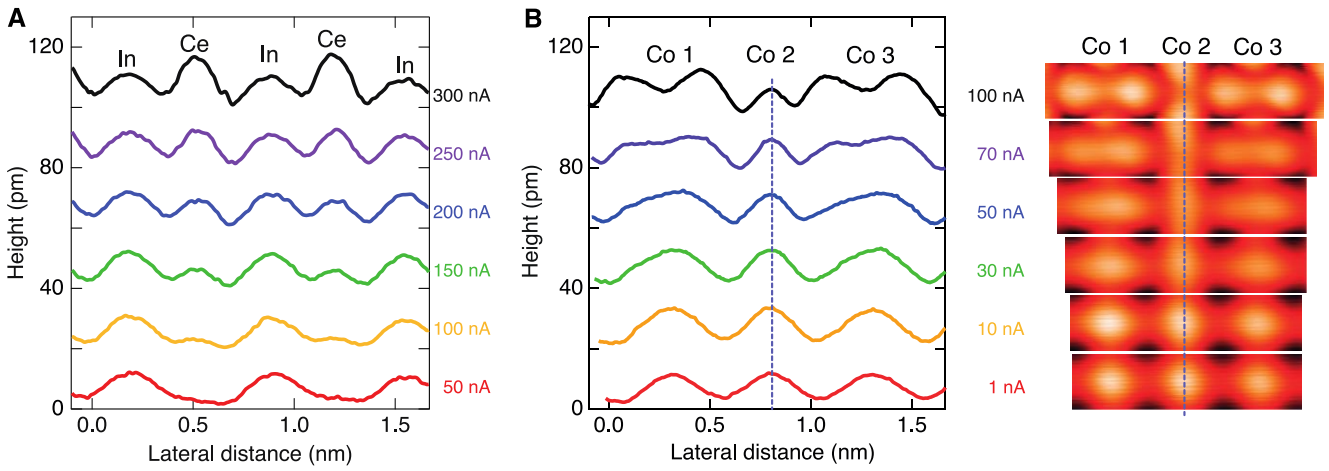

fig. S2. Current-dependent cross-sectional profiles of Ce-In and Co planes. (**A**) and those of Co atoms on a Co plane (**B**) with corresponding topographic images. **a,** Profiles at more than 100 nA are shifted upward for clarity. Enhancement of the Ce height is visible as the current set point is increased. The Ce height becomes even higher than those of In at 250 nA and 300 nA. **b,** Profiles at more than 10 nA are shifted upward for clarity. By increasing the current set point, Co atoms referred to as Co1 and Co3 elongate into two lobes while Co2 shrinks because of the elongation along the perpendicular direction.

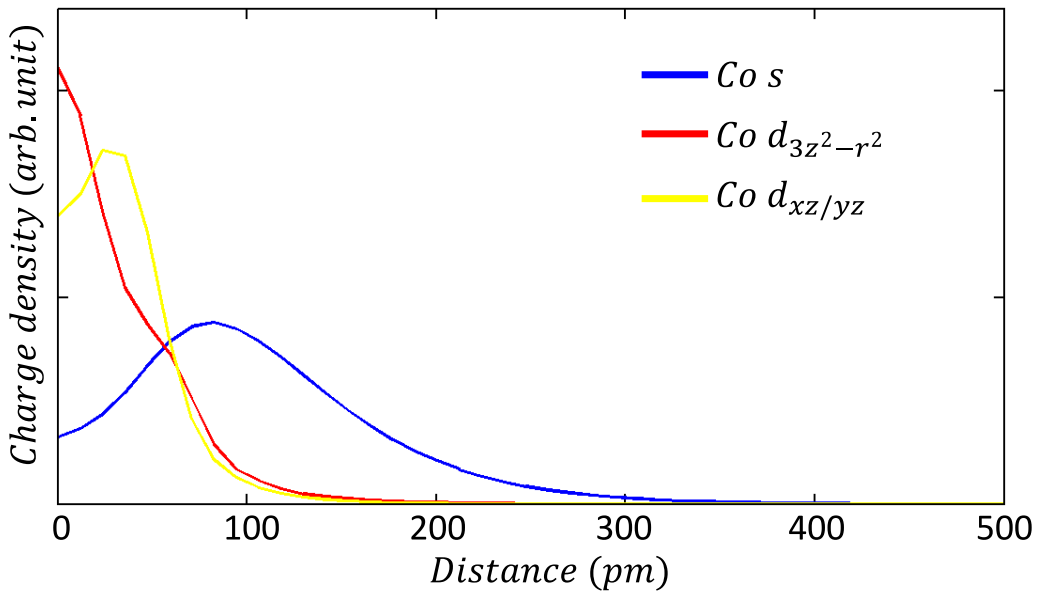

fig. S3. Charge density profiles of the Co atom orbitals on the Co termination. The distance on the horizontal axis corresponds to the out-of-the-plane direction. They are obtained from slab calculations, through projection on each spherical harmonics Y_{lm} centered at the atom at hand. The orbital wave functions are extended to a few hundred pm. When the STM tip approaches the Co plane, the *d* orbitals show up only when the STM tip is very close to the Co atom.

note S1. Excluding the possibility of tip-induced artifacts in dumbbell formation.

We have excluded possible experimental artifacts that may affect our observations. As described in the main text, we measured I_T as a function of tip-sample distance (TSD) to ensure that the observations were performed in the tunneling regime. We observed the characteristic exponential dependence of the current up to $I_T \sim 160$ nA on the Co surface (Fig. 2C). We reproduced identical results by repeating the experiment with three mechanically sharpened PtIr tips and many microscopic tips and on two independently cleaved surfaces. Electric fields caused by small TSDs are negligible because V_S is only 10 mV. These experimental facts rule out the possibilities of any artifacts caused by strong interactions between the tip and sample. Moreover, a detailed investigation of the line defects on the terraces also supports that the dumbbells are not tip-induced artifacts. Since the defects are visible even when the tip is far away from the surface (Fig. 1C), the ordered structure is clearly not a result of the tip-induced artifact.

note S2. Topographic similarity between our dumbbell ordered structure and the images in the study of Takahashi *et al.* (*14*) and their difference in underlying physics.

Accessibility to orbital selective tunneling was recently reported on several systems with changing TSD in STM (*12-14*), as mentioned in the abstract. Most recently, Takahashi *et al*. (*14*) reported a topographic evolution from a dot array to an alternating dumbbell array with decreasing TSD on a monoatomic layer of Fe_4N on Cu (001), which resembles the structure we observed on a Co plane of $CeCoIn_5$. However, the physical origins behind the two structures are totally different, although their STM images and tendency in TSD are similar at a glance. In Ref. 14, the alternating dumbbell structure originates from dimerized d_{z2} orbitals of Fe atoms, which are hybridized through the orbitals of N atom. While in $CeCoIn_5$, it stems from the alternatingly selected d_{xz} (d_{yz}) and d_{yz} (d_{xz}) orbitals of Co atoms on the Co plane, which is due to the lifted degeneracy by on-site Coulomb potential enhanced at a surface.

movie S1. Evolution of dumbbell structure by decreasing the TSD. The movie demonstrates evolution of the dumbbell ordered structure depending on tip-sample distance with the tunnel resistance between 10 MΩ and 91 kΩ.