Supporting Information for

Lateral Manipulation of Atomic Vacancies in Ultrathin Insulating Films

Zhe Li,[1] Hsin-Yi Tiffany Chen,[2] Koen Schouteden,[1] Koen Lauwaet,[1] Ewald Janssens,[1] Chris Van Haesendonck,[1] Gianfranco Pacchioni,[2,*] and Peter Lievens[1,†]

[1] *Laboratory of Solid-State Physics and Magnetism, KU Leuven, Celestijnenlaan 200d – box 2414, BE-3001 Leuven, Belgium*

[2]*Dipartimento di Scienza dei Materiali, Università di Milano-Bicocca, Via Cozzi 55, I-20125 Milano, Italy*

Corresponding authors: [*]gianfranco.pacchioni@unimib.it; [†]peter.lievens@fys.kuleuven.be

Contents

1. Formation of Nanostructures Composed of Multiple Vacancies

The controlled manipulation of multiple vacancies is illustrated in Fig. S1. Three [black dotted ellipse in Fig. S1(a)] and four [blue dashed box in Fig. S1(b)] vacancies are moved toward each other by the STM tip using the procedure described in the manuscript.

In Fig. S1(b) there are two vacancies that are outside the dashed box and that are very close to the four target vacancies within the dashed box. When we move the upper vacancy (labeled as vacancy 2 by the white arrow) within the dashed box by a local voltage pulse as described in the text, the neighboring vacancy 1 outside the box is also moved. Note that in between the first two images in Fig. S1(b), vacancy 3 is moved because manipulation (voltage pulses) of the four vacancies within the dashed box also affects vacancy 3. In our manipulation approach the tip is retracted relatively far away from the surface, which implies that the voltage pulse applied to a target vacancy has an intrinsic finite "radial extent" and hence can also affect vacancies that are located nearby.

However, we emphasize that this is a very local effect. In particular, when the vacancies are further separated from each other, *i.e.*, further than about 1.2 nm, there are no unintended movements as illustrated in Fig. 1 in the main text and in Fig. S1(a). On the other hand, we note that in between the first two images in Fig. S1(a), manipulations are not only performed for the three vacancies within the black dotted ellipse, but in separate steps also to other vacancies, *i.e.*, vacancies 1-3 indicated by the black arrows in the first image of Fig. S1(a). This is in this case the reason why the vacancies 1-3 outside of the dotted ellipse are moved as well when comparing the first two images in Fig. S1(a).

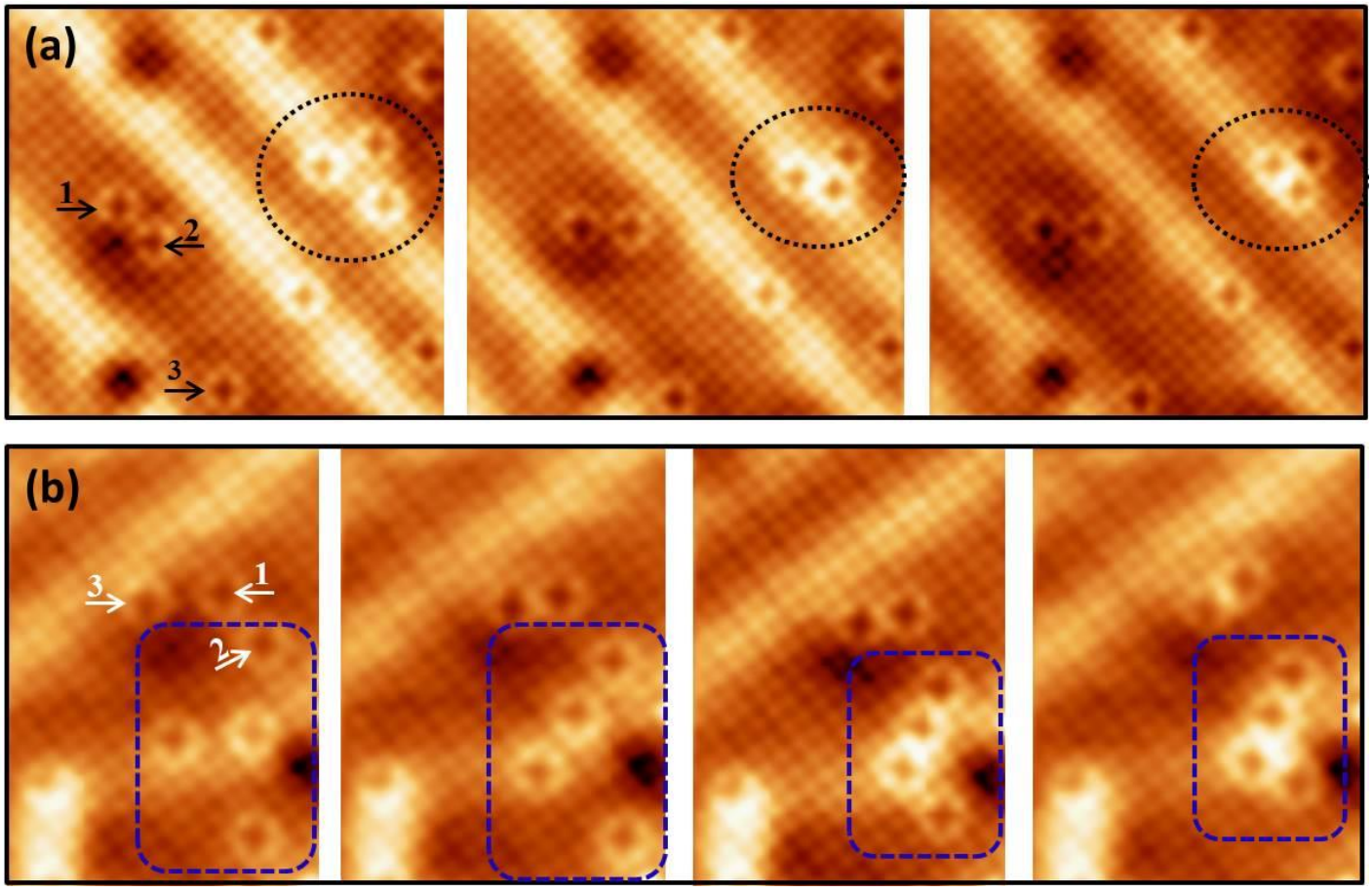

Figure S1. Controlled manipulation of Cl vacancies in 2L NaCl/Au(111). (a) Series of 10×9.5 nm^2 atomic-resolution STM topography images of Cl vacancies in 2L NaCl recorded at V = 1 V, I = 0.2 nA. The three vacancies indicated by a black dotted ellipse are moved towards each other by the STM tip as described in the manuscript. Intentional manipulations are also performed for the vacancies 1-3 indicated by the black arrows. (b) Series of 6×8 nm^2 atomic-resolution STM topography images of Cl vacancies on 2L NaCl recorded at V = 1 V, I = 0.2 nA. The four vacancies indicated by a blue dashed box are moved towards each other by the STM tip. White arrows indicate the vacancies referred to in the discussion in the text.

2. Height Profile of a Single Vacancy and a Vacancy Dimer at High Tunneling Voltage

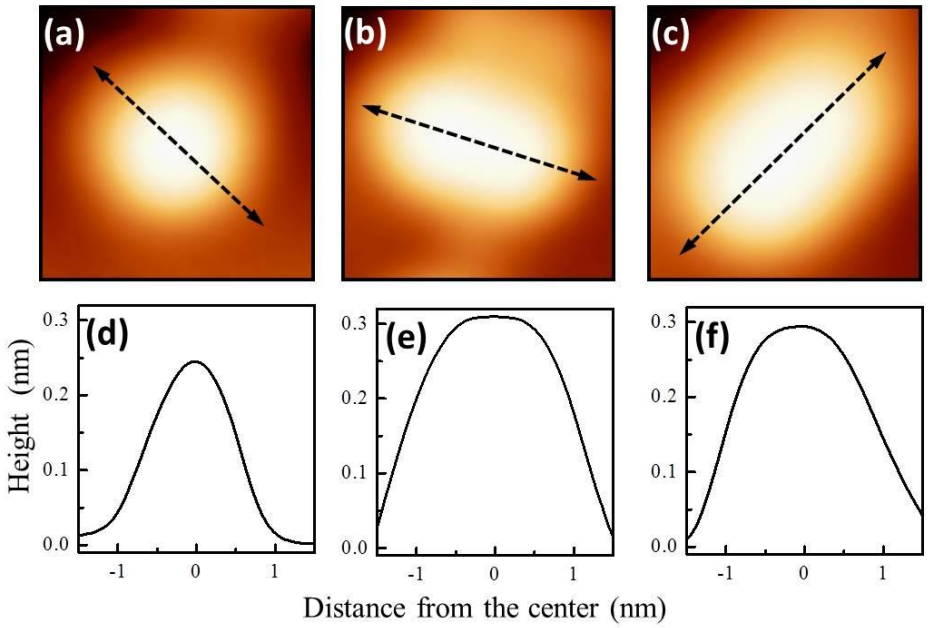

Figure S2. (a) 2.2×2.2 nm^2 STM topography image of a single Cl vacancy in 2L NaCl recorded at V = 3.35 V. (b) 3.0×3.0 nm^2 STM topography image of dimer I in 2L NaCl recorded at V = 3.30 V. (c) 2.2×2.2 nm^2 STM topography image of dimer II in 2L NaCl recorded at V = 3.15 V. [Same as in Figs. 3(b), (d), and (f) in the main manuscript.] (d)-(f) Topographic height profiles taken along the dashed arrows in (a)-(c), respectively.

3. Atomic Scale Structure and Partial Charge Density of a Single Cl Vacancy (F^+ Center) and a Vacancy Dimer (M^{2+} Center)

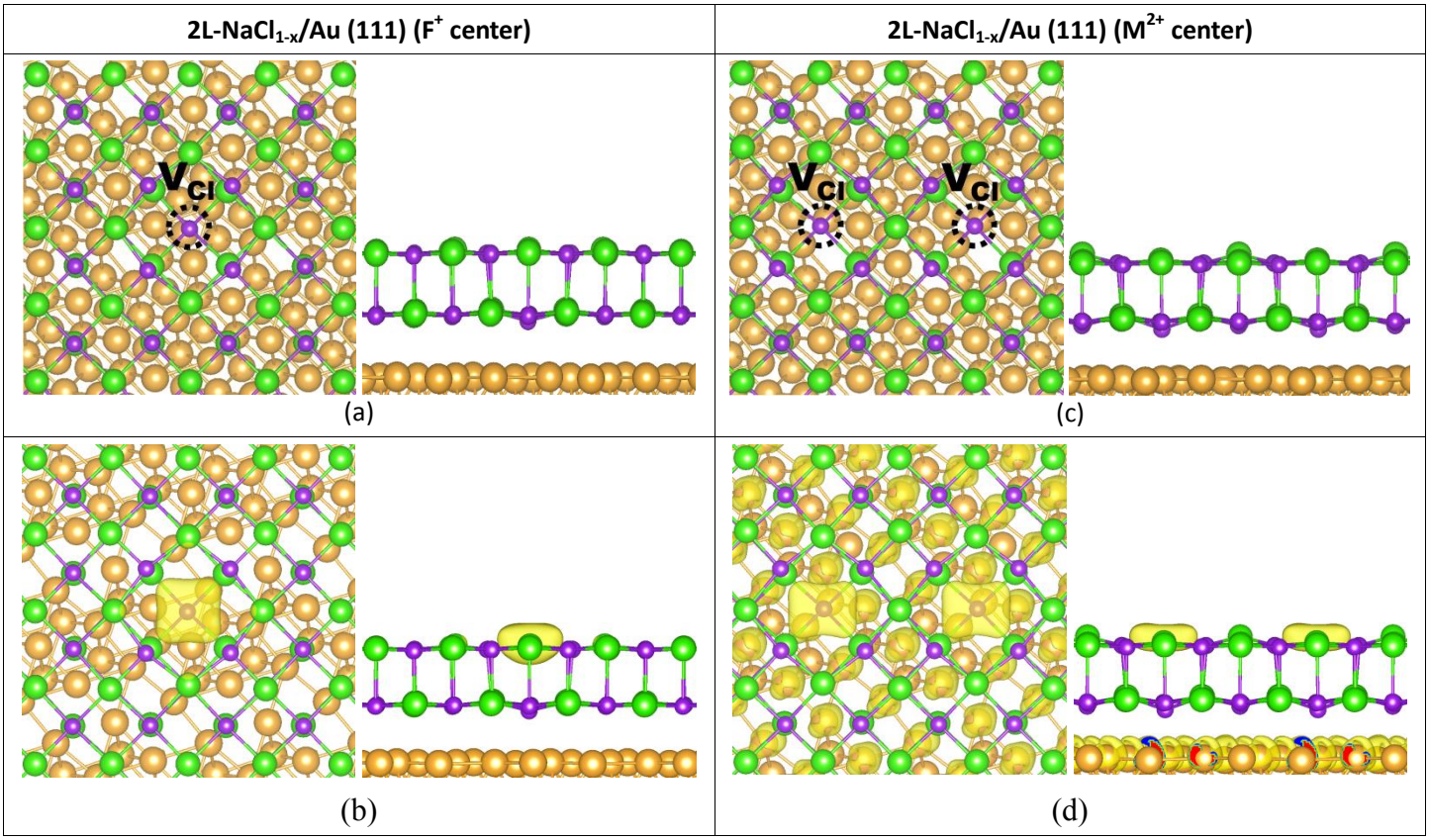

Figure S3. Left column: F^+ center in 2L NaCl/Au(111) with (a) top view and side view of the atomic scale structure (violet, green and gold spheres refer to Na, Cl and Au atoms, respectively) and (b) the partial charge density of the vacancy state. The isovalue for the isosurfaces of the charge density is 0.02 |e|/Å^3. Right column: (c) and (d) are the same as (a) and (b) but now for an M^{2+} center (vacancy dimer II) in 2L NaCl/Au(111).