Supplementary Information for:

Direct visualization of reversible dynamics in a Si\textsubscript{6} cluster embedded in a graphene pore

**Supplementary Figure S1.** 3D structure characterization. **a,c.** ADF images showing the oscillatory motion of a Si atom in the Si\textsubscript{6} cluster. **b,d.** Schematic of the Si\textsubscript{6} cluster where the oscillating Si atom is represented with a red circle, and its two nearest neighbors are represented with pale blue circles. The z-coordinate of the remaining three Si atoms, numbered and shown schematically in black, were determined using first-principles structural minimization methods. The 18 different configurations of the Si\textsubscript{6} cluster are classified by their relative energy and by the relative positions of the Si atoms (shown in black in the schematic) with respect to the graphene lattice, *i.e.* above the graphene lattice (Up) or below the graphene lattice (Down). The relative total energies are calculated with respect to that of the atomic structure with Up, Up, Up (UUU) coordinates. Scale bars: 0.2 nm.
Supplementary Figure S2. Experimental images overlaid with the fully relaxed Si$_6$ cluster with the lowest energy (UUU coordinates). a. The oscillating Si atom is at the Left site. b. The oscillating Si atom is at the Right site. The contrast level of the experimental images was adjusted to highlight the position of the Si atoms only.
Supplementary Figure S3. Sequential ADF images of the Si$_6$ cluster embedded in the graphene pore. L or R represents the position of the oscillating Si atom at the Left or Right site. The ratio of the Si atom at the Left and Right sites as observed in the ADF images is about two.
Supplementary Figure S4. The charge distribution in a charged cluster. 

**a.** The charge distribution of the highest-energy occupied state after an electron is added to the system. 

**b.** The charge distribution of lowest-energy empty state after an electron is removed from the system.
Supplementary Figure S5. The difference in the total charge of the system with and without the extra electron. Red corresponds to 70%, orange 30%, yellow 3% isosurface level of the density maxima. **a.** Δρ>0. **b.** Δρ<0. Net charge on the cluster is essentially zero because the positive and negative Δρ have almost identical distribution in the vicinity of the cluster (they are above and below the plane, respectively). The extra electron that we inserted manually into system is uniformly distributed as demonstrated by the yellow isosurface in Fig. S5 (a).
Supplementary Figure S6. The difference in the total charge of the system with and without the extra hole. The color scheme is the same as in Fig. S5, for the case of removing an electron from the system.