Supplementary Materials for

**Electrotunable artificial molecules based on van der Waals heterostructures**

Zhuo-Zhi Zhang, Xiang-Xiang Song, Gang Luo, Guang-Wei Deng, Vahid Mosallanejad, Takashi Taniguchi, Kenji Watanabe, Hai-Ou Li, Gang Cao, Guang-Can Guo, Franco Nori, Guo-Ping Guo

DOI: 10.1126/sciadv.1701699

**This PDF file includes:**

- Supplementary Text
- fig. S1. Source-drain current, $I_{SD}$, versus the global back-gate voltage, $V_{BG}$.
- fig. S2. Tunability of the gate DM over a wider range at $V_{BG} = 30$ V.
- fig. S3. Tunability of the gate DM in another similar sample.
- fig. S4. COMSOL simulation of the interdot barrier.
- fig. S5. COMSOL simulation on the potential well distribution for different values of $V_{DM}$.
- fig. S6. Gate controllability in the low-density regime.
- Reference (58)
1. **Source-Drain current $I_{SD}$ versus the global Back-Gate voltage $V_{BG}$**

A DC voltage was applied to the global back gate to tune the Fermi energy of the MoS$_2$ device without any local bottom gates voltage applied. The source-drain current ($I_{SD}$) was measured when sweeping the back-gate voltage ($V_{BG}$) at a fixed source-drain voltage ($V_{SD}$) of 5 mV. As shown in fig. S1, the device shows a typical n-type semiconductor behavior. The demonstration of the double quantum dot is presented above the turn-on threshold voltage, with an estimated field-effect mobility of ~ 300 cm$^2$/V·s. The estimated charge density at $V_{BG} = 25$ V and 30 V is $2.77 \times 10^{11}$ cm$^{-2}$ and $1.20 \times 10^{12}$ cm$^{-2}$, respectively.

![Graph](image)

**fig. S1. Source-drain current, $I_{SD}$, versus the global back-gate voltage, $V_{BG}$.**

Without any local bottom gates voltage applied, the source-drain current $I_{SD}$ was measured when sweeping the global back-gate voltage $V_{BG}$. The $I_{SD}$ versus $V_{BG}$ curve shows a typical n-type semiconductor behavior.
2. Tunability of the gate DM over a wider range

When tuning $V_{DM}$ to a negative range, varying from $-0.2$ V to $-1.2$ V, while all other gate voltages remain fixed, one large quantum-dot atom evolves into a double-quantum-dot molecule. Figure S2 shows a typical area of the charge stability diagram, which is much larger than that shown in Fig. 3. Figure S3 shows a similar phenomenon observed in another sample.

**fig. S2. Tunability of the gate DM over a wider range at $V_{BG} = 30$ V.** Current through the double quantum dot versus $V_{LB}$ and $V_{RB}$ applied to the gates LB and RB for $V_{BG} = 30$ V, $V_{LP} = V_{RP} = 0$ V, $V_{UM} = -2.1$ V, bias voltage at $V_{SD} = 100 \mu$V and $V_{DM} = -0.2$ V, $-1$ V, $-1.1$ V and $-1.2$ V for A to D, respectively.
**fig. S3. Tunability of the gate DM in another similar sample.** Current through the double quantum dot versus $V_{LP}$ and $V_{RP}$ applied to the gates LP and RP, for $V_{BG} = 40$ V, $V_{LB} = V_{RB} = -0.64$ V, $V_{UM} = -1.1$ V, bias voltage at $V_{SD} = 100$ μV and $V_{DM} = -0.4$, $-0.7$ V and $-0.9$ V for A to C, respectively. This data was measured from another sample with same structure.
3. Rough fitting of the fractional peak splitting $f$ versus the gate voltage $V_{DM}$

Consider the inter-dot potential barrier in a parabolic form, $U(x) = -\frac{1}{2}kx^2$, the inter-dot coupling strength which corresponds to the transmission coefficient $D$ can be determined (58) as $D = 1/(1 + e^{-2\pi\epsilon})$, where $\epsilon = (E/\hbar)\sqrt{m/k}$. We can roughly fit the fractional peak splitting $f$ versus the gate voltage $V_{DM}$ with a $1/(1 + e^{-kx})$ lineshape. It is worth noticing that in the strongly-coupled regime, the inter-dot potential barrier does not fit the quasi-classical model because of the high value of the transmission coefficient. So the fit is not suitable when the value of $V_{DM}$ is near 0.

fig. S4. COMSOL simulation of the interdot barrier.
4. COMSOL simulation for different values of $V_{DM}$

When tuning $V_{DM}$ to more negative values, the inter-dot potential barrier arises, leading to the reduction of the coupling strength between the dots (as shown in Fig. 3). A COMSOL simulation is used to calculate the change of potentials for different values of $V_{DM}$, while other gate voltages remain fixed, as shown in fig. S5. The inter-dot barrier increases when tuning $V_{DM}$ more negative. Meanwhile, the dot confinement potential remains almost unaffected.

The schematic diagram of the evolution of such double-dot to single-dot transition of the confining potential at the crossline in fig. S5 is shown in Fig. 3, C to E. Such simulation results agree well with the experiment results.

**fig. S5.** COMSOL simulation on the potential well distribution for different values of $V_{DM}$. COMSOL simulation on the potential-well distribution of the closed contours shown in Fig. 2D based on the designed pattern for $V_{BG} = 30$ V, $V_{LP} = V_{RP} = 0$ V, $V_{LB} = V_{RB} = -1.5$ V, $V_{UM} = -2.1$ V, and $V_{DM} = -0.3$ V, $-0.5$ V, $-1$ V and $-1.5$ V for A to D, respectively.
5. **Gate controllability in the low-density regime**

Because of the different values of $V_{BG}$, the formation of the double quantum dot was dominated by different mechanisms, as shown in Fig. 4, D and E. At a relatively low Fermi energy ($E_F$), the intrinsic and fabrication-induced impurities dominate the confining potentials of the transport behavior, which cannot be well controlled by electrostatic gating. The controllability of the electrostatic gates here is demonstrated in fig. S6, A to C. When tuning the value of $V_{DM}$ and $V_{UM}$ together over a wide range, the tunneling rate between the source/drain and the dot changes effectively, while the coupling strength of two quantum dots does not show any obvious signature of evolution.

![fig. S6. Gate controllability in the low-density regime.](image)

Current through the double quantum dot versus $V_{LB}$ and $V_{RB}$ applied to the gates LB and RB for $V_{BG} = 25$ V, $V_{LP} = V_{RP} = 0$ V, bias voltage at $V_{SD} = 100$ μV and $V_{DM} = V_{UM} = -0.1$ V, -0.5 V, and -0.9 V for A to C, respectively.