# Supplementary Information

# Polarons in single-layer $MnBr_2$: properties and dependence on the substrate

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**CONTENTS**

**Supplementary note 1: Extended dataset for bias-dependent polaron imaging and mobility.**

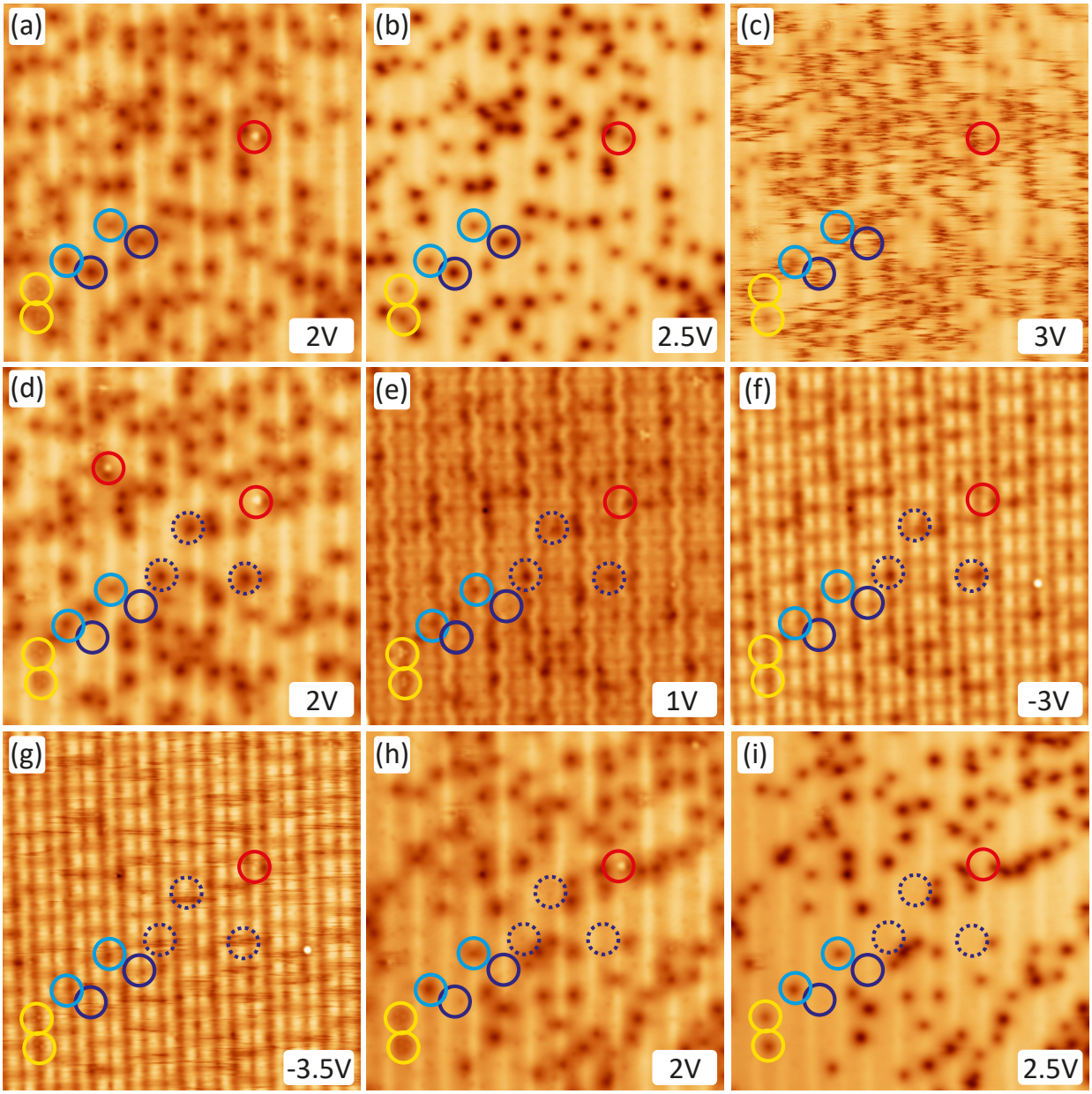


Figure S1. (a-i) Sequence of STM images of the same area acquired at varying $V_b$ ranging from $+3\,V$ and $-3.5\,V$, as indicated, and at $I_t = 50\,pA$. Selected polarons are encircled to track their position: dark blue (type-I mobile), light blue (type-I immobile), yellow (type-II 2x2), and red (type-II 1x1). From comparison of (d) and (h) it is obvious that type-I mobile polarons are also moved at $V_b = -3.5\,V$. Image sizes: $45\,nm \times 45\,nm$.

Figure S1 presents a complementary dataset to Figure 2 of the main text, extending the investigation to higher negative bias range. The behavior at positive $V_\mathrm{b}$ [Figure S1(a-e)] is consistent with our previous observations. The four types of polarons are distinguishable at $V_\mathrm{b} = 2\,\mathrm{V}$, and type-I mobile polarons change positions after increasing $V_\mathrm{b}$ to $3\,\mathrm{V}$, as tracked by the polarons encircled dark blue in Figure S1(a-d).

The tip-induced mobility is also observed at negative $V_\mathrm{b}$. If $V_\mathrm{b}$ is set to $-3.5\,\mathrm{V}$ in Figure S1(g), type-I mobile polarons are mobile as obvious from the comparison of Figures S1(d) and (h) (check position in dotted dark blue circles).

**Supplementary note 2: Mobility of type-I mobile polaron for lower positive $V_b$ at large tunneling current**

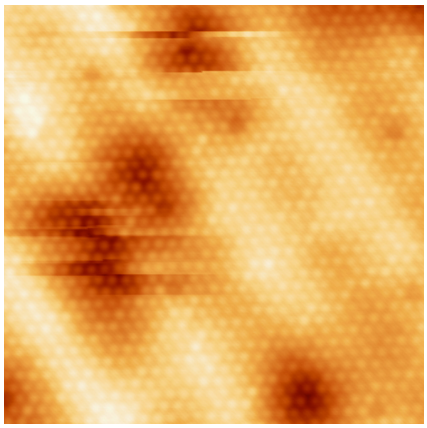

Figure S2. STM image acquired with $V_b = 2\,\mathrm{V}$, $I_t = 3\,\mathrm{nA}$. Image size: $15\,\mathrm{nm} \times 15\,\mathrm{nm}$.

**Supplementary Note 3: Overview topographs of single-layer $MnBr_2$ grown on different substrates.**

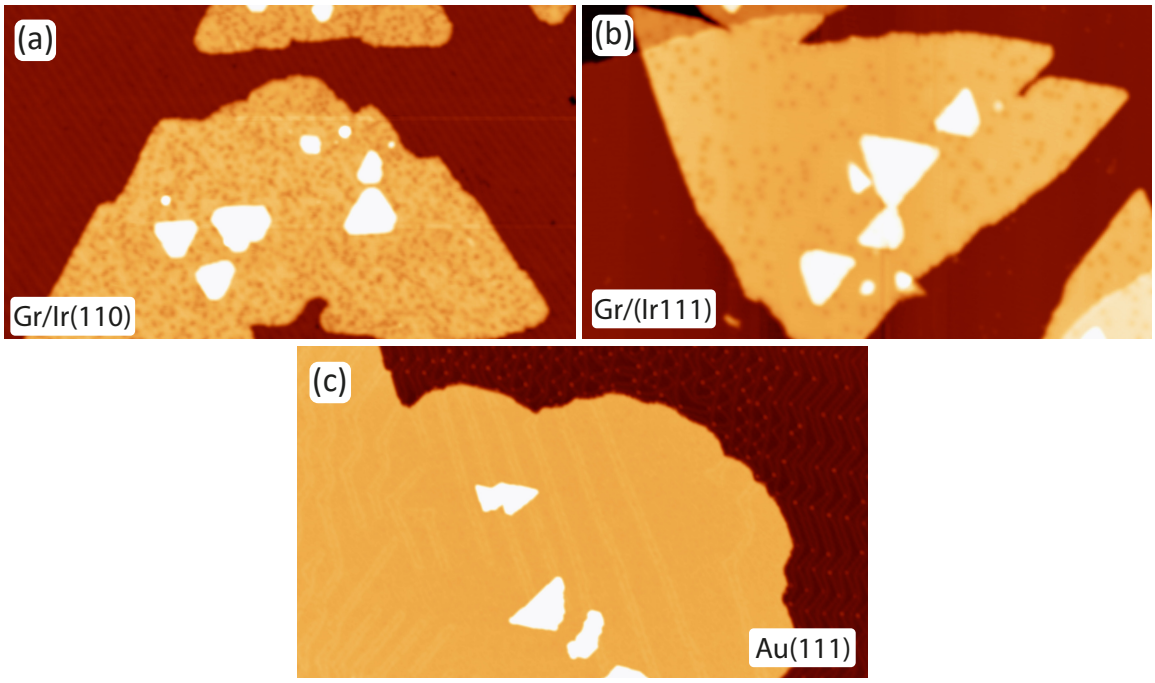


Figure S3. (a) Overview STM topographs of $MnBr_2$ on (a) Gr/Ir(110), (b) Gr/Ir(111) and (c) Au (111). STM Imaging Parameters: (a) $V_b = 3.5\,V$, $I_t = 20\,pA$, (b) $V_b = 3.5\,V$, $I_t = 20\,pA$, (c) $V_b = 2\,V$, $I_t = 50\,pA$. Image sizes: $240\,nm \times 140\,nm$.

On top of single-layer $MnBr_2$ islands also small second-layer $MnBr_2$ islands are visible. The overview STM image of $MnBr_2$ on Au(111) in Figure S3 shows the herringbone reconstruction in the island, which is only partly lifted by $MnBr_2$ overgrowth.

**Supplementary note 4: Polarons in $MnBr_2$ on Gr/Ir(111)**

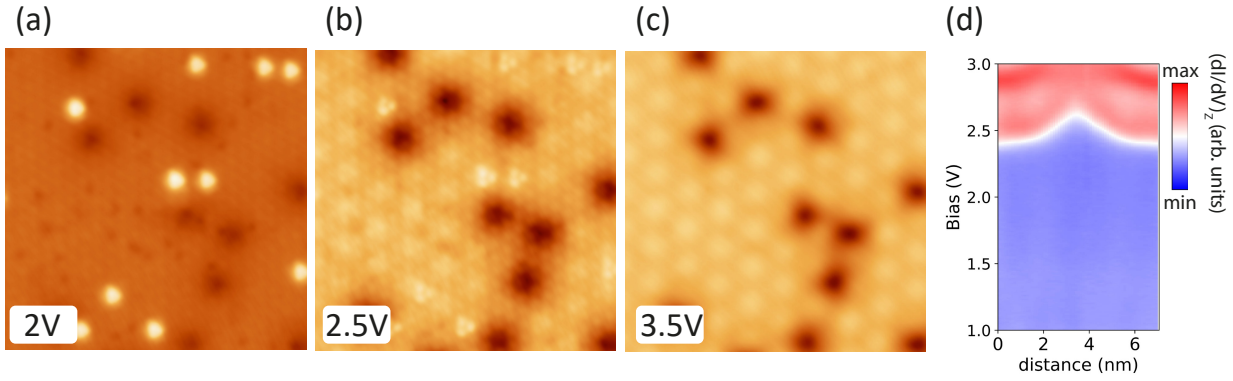


Figure S4. (a-c) Consecutive STM topographs of $MnBr_2$ on Gr/Ir(111) at different $V_b$ as indicated. (d) Constant current d$I$/d$V$ linescan map over the type-I immobile, Spectra are acquired with $V_{st}$ = +3 V, $I_{st}$ = 50 pA, $f_{mod}$ = 667 Hz, and $V_{mod}$ = 20 mV. STM imaging parameters: $I_t$ = 50 pA. Image sizes: 18 nm × 18 nm.

**Supplementary note 5: Tip induced conversions of type-II 1x1 polarons into type-I mobile polarons.**

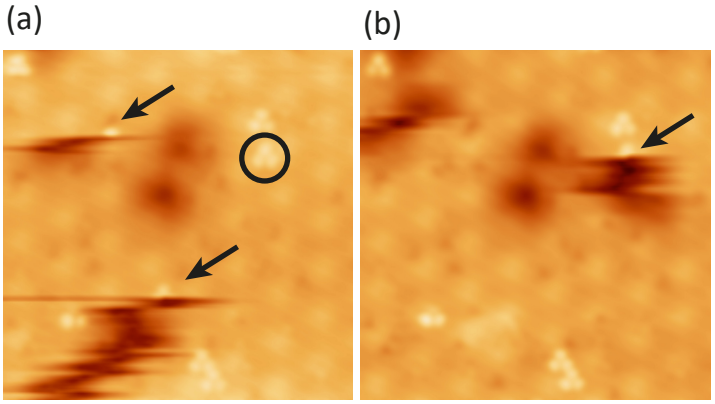


Figure S5. (a,b) Consecutive STM topographs of $MnBr_2$ on Gr/Ir(111). The black arrows in (a) indicate locations where the STM tip induces a conversion from a type-II 1x1 polaron to type-I mobile polaron. Additionally, also the type-II 1x1 polaron circled in (a) has converted to a type I mobile polaron in (b), as highlighted by the arrow in (b). STM imaging parameters: $V_\mathrm{b} = 2.5\,\mathrm{V}$, $I_\mathrm{t} = 50\,\mathrm{pA}$. Image sizes: $18\,\mathrm{nm} \times 18\,\mathrm{nm}$.

Figure S5 demonstrates conversions of type-II 1x1 polarons to type-I mobile polarons induced during STM scanning. After these conversions, the type-I mobile polarons move with the tip, appearing as fuzzy and continuous dark features in the topographs.

**Supplementary note 6: Creation of a type-I 1x1 mobile polaron in $MnBr_2$ on Gr/Ir(111).**

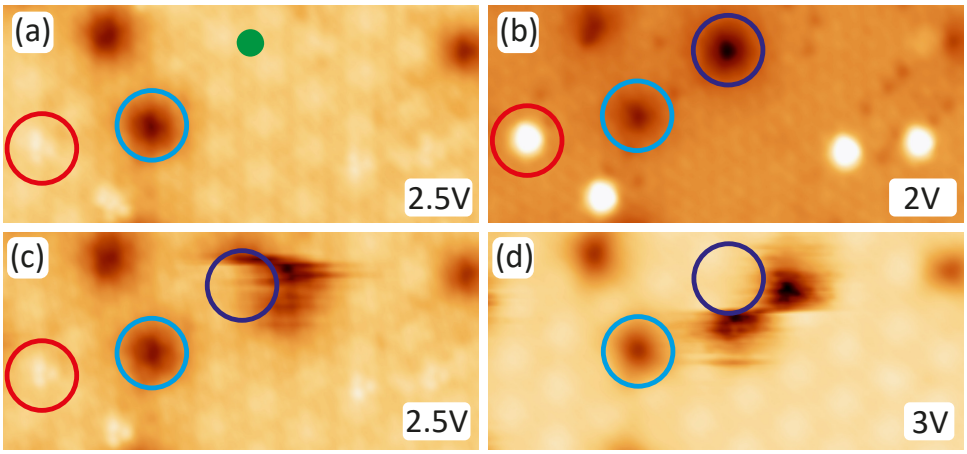


Figure S6. Sequence of STM images illustrating the creation of a type-I mobile polaron in $MnBr_2$ on Gr/Ir(111) via bias pulsing. (a) STM topograph before bias pulsing with pre-existing type-II 1x1 and type-I immobile polarons (circled red and light blue, respectively). (b) STM topograph after bias pulse with $V_b = +3.5\,V$ and $I_t = 500\,pA$ at green dot in (a). (c,d) Subsequent imaging at higher biases shows the mobile polaron moving under the influence of the STM tip. $I_t = 50\,pA$. Image sizes: $22\,nm \times 10\,nm$.

Figure S6 shows the creation of a type-I mobile polaron. A bias pulse ($V_b = +3.5\,V$, $I_t = 500\,pA$) is applied at the green spot in Figure S6 (a). Subsequent imaging at $V_b = +2\,V$, as shown in Figure S6(b), reveals a type-I mobile polaron (encircled purple) at pulse location along with pre-existing type-I immobile (encircled blue) and type-II 1x1 polarons (encircled red). In subsequent imaging at $V_b = +2.5\,V$ and $+3\,V$ in Figure S6(c) and (d) the observed mobility of the polaron justifies its classification as type-I mobile polaron.

**Supplementary note 7: Bias dependent absolute tip heights above Gr/Ir(110) and $MnBr_2$/Gr/Ir(110).**

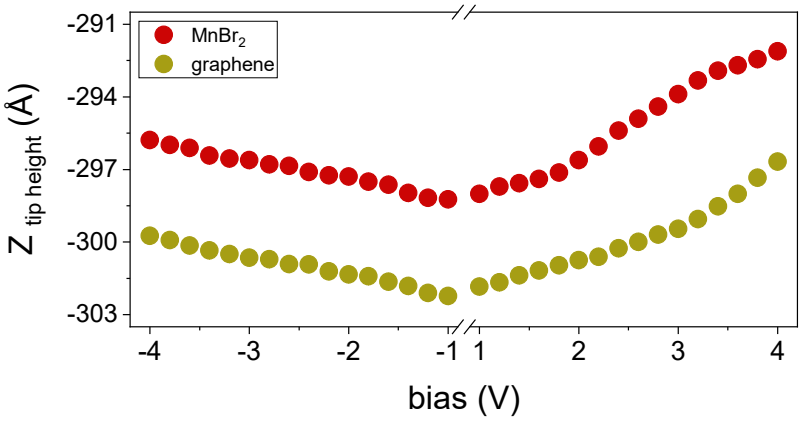


Figure S7. Absolute heights of the STM tip over Gr/Ir(110) (red squares, right $y$-axis) and $MnBr_2$/Gr/Ir(110) (black squares, left $y$-axis) for $I_t = 20\,pA$ as a function of $V_b$.