Bias-controlled multi-functional transport properties of InSe/BP van der Waals heterostructures

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Van der Waals (vdW) heterostructures, consisting of a variety of low-dimensional materials, have great potential use in the design of a wide range of functional devices thanks to their atomically thin body and strong electrostatic tunability. Here, we demonstrate multi-functional indium selenide (InSe)/black phosphorous (BP) heterostructures encapsulated by hexagonal boron nitride. At a positive drain bias ($V_D$), applied on the BP while the InSe is grounded, our heterostructures show an intermediate gate voltage ($V_{BG}$) regime where the current hardly changes, working as a ternary transistor. By contrast, at a negative $V_D$, the device shows strong negative differential transconductance characteristics; the peak current increases up to ~5 μA and the peak-to-valley current ratio reaches 1600 at $V_D = −2$ V. Four-terminal measurements were performed on each layer, allowing us to separate the contributions of contact resistances and channel resistance. Moreover, multiple devices with different device structures and contacts were investigated, providing insight into the operation principle and performance optimization. We systematically investigated the influence of contact resistances, heterojunction resistance, channel resistance, and the thickness of BP on the detailed operational characteristics at different $V_D$ and $V_{BG}$ regimes.

NDT is a characteristic in which an increase in gate voltage results in a decrease in drain current; the so-called anti-ambipolar characteristic, one type of NDT, refers to a situation in which current is generated in the middle gate voltage region but is not generated at both positively and negatively large gate voltages. Various heterostructures consisting of multiple different semiconductors have been shown to have NDT characteristics, including MoS₂/single-walled carbon nanotubes, MoS₂/WSe₂, MoS₂/Te₃-16, MoS₂/BP, ReS₂/BP, SnS₂/WSe₂, MoS₂/rubrene, InSe/BP, and heterostructures composed of n-type and p-type organic semiconductors. A strong anti-ambipolar behavior can be employed for multi-way switching. NDT devices also have potential use as oscillators, memories, and other low power logics.

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In this article, we present bias-controlled NDT and ternary transistors based on InSe/BP heterostructures encapsulated by hexagonal boron nitride (hBN). BP and InSe have mobilities notably superior than those of transition metal dichalcogenides\(^\text{25,26}\). They have relatively small effective masses, and the InSe conduction band and the BP valence band are close each other. The combination of InSe and BP could be, thus, promising for high-speed tunneling based devices. Several studies have demonstrated ternary inverters by using a NDT device and the part of the constituent semiconductor of the NDT device as a load resistor\(^\text{5,6,14,22,27,28}\). However, such approach is limited to inverting logics and cannot provide a versatile complementary metal–oxide–semiconductor (CMOS) circuit design strategy. Ternary transistors, where the drain current hardly changes within a specific gate voltage range (an intermediate logic state), are more essential to implement practical ternary-data-processing CMOS integrated circuits\(^\text{29}\).

InSe and BP yield different contact properties when contacted with Au/Ti contacts, and this asymmetry leads to asymmetric electrical characteristics depending on the polarity of drain bias (\(V_D\)). While ternary states are developed at positive \(V_D\), NDT characteristics are observed at negative \(V_D\). Four-terminal measurements were performed on each layer, where, in principle, the contributions of contact resistances and channel resistance can be identified separately. We fabricated multiple devices with different contacts and device structures and systematically investigated the influence of contact resistances, heterojunction resistance, channel resistances, and the thickness of BP on the detailed operational characteristics.

**Results and discussion**

Figure 1a describes the structure of our hBN/InSe/BP heterostructure device with Au/Ti electrodes (InSe-BP-Ti device), accompanied by its optical micrograph. The hBN/InSe/BP heterostructure was achieved using the conventional dry transfer method\(^\text{2,3}\). Individually exfoliated BP and InSe flakes were successively transferred on 300 nm-thick SiO\(_2\) followed by encapsulation with a pre-patterned hBN flake. InSe and BP are vulnerable to the air, but the hBN encapsulation allows improved stability and decent mobility\(^\text{26,30}\). Before the transfer, the hBN encapsulating layer was patterned using electron beam (e-beam) lithography and plasma etching to have eight openings. These allowed the InSe and BP to be partially exposed for metallization even after having the hBN cover on them. Four metal contacts on the InSe and BP were then defined by e-beam lithography, followed by Au(80 nm)/Ti(10 nm) deposition and lift-off. Figure 1b shows the atomic force microscopy (AFM) image,
InSe shows a nonlinear characteristic, indicating Schottky contacts. The dashed line represents \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}})\)^{-1}, which is comparable to the measured \(G_{\text{2pt}}\) of the InSe-BP-Ti device measured at a forward \(V_{BG}\) regime, which shows a distinct intermediate logic state with a small fluctuation. This Raman quenching is attributed to the weak but finite van der Waals coupling between the InSe and BP. Nearly consistent peak positions and the ratio of A1(LO) peak to E1 peak intensity suggest that the degree of strain and doping induced by having the heterostructure is negligible.

While the BP shows an ambipolar characteristic, the InSe shows a linear dependence at high carrier density and even at charge neutrality (at \(V_{BG} = 50\) V and 0 V), suggesting ohmic contacts, but the InSe shows a nonlinear characteristic, indicating Schottky contacts. The dashed line represents \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}})\)^{-1}, which is comparable to the measured \(G_{\text{2pt}}\) of the InSe-BP-Ti device measured at a backward \(V_{BG}\) regime, which shows a strong NDT behavior with PVCR reaching ~10^2 at \(V_{BG} = -1\) V. The dashed line represents \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}})\)^{-1}, which is comparable to the measured \(G_{\text{2pt}}\). \(G_{\text{2pt}}\) (\(R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}}\))−1, \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}})^{-1}\), \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}})^{-1}\), and \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{BP}})^{-1}\) for comparison with (b,c) data, respectively.

Figure 2. (a) Schematics describing the InSe-BP-Ti device (upper) and the four different equivalent circuits, consisting of \(R_{\text{InSe}}, R_{\text{BP}}, R_{\text{InSe}}\), and \(R_{\text{BP}}\). Depending on current direction, different contact resistance components are more effective. (b) \(G_{\text{2pt}}\) vs \(V_{BG}\) of the InSe-BP-Ti device measured at a forward \(V_{D}\) regime, which shows a distinct intermediate logic state with a small fluctuation. The dashed line represents \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}})\)^{-1}, which is comparable to the measured \(G_{\text{2pt}}\) of the InSe-BP-Ti device measured at a backward \(V_{D}\) regime, which shows a strong NDT behavior with PVCR reaching ~10^2 at \(V_{D} = -1\) V. The dashed line represents \((R_{\text{InSe}} + R_{\text{BP}} + R_{\text{InSe}})\)^{-1}, which is comparable to the measured \(G_{\text{2pt}}\).

Electrical characterization was performed on the BP and InSe individually via the two-terminal and four-terminal methodology, as illustrated in the schematic in Fig. 1a. Figure 1d shows the two-terminal conductances \(G_{\text{2pt}}\) of the InSe-BP-Ti device measured at a forward \(V_{BG}\) regime, which shows a distinct intermediate logic state with a small fluctuation. This Raman quenching is attributed to the weak but finite van der Waals coupling between the InSe and BP. Nearly consistent peak positions and the ratio of A1(LO) peak to E1 peak intensity suggest that the degree of strain and doping induced by having the heterostructure is negligible.

We note that the two-terminal field-effect mobilities of the InSe and BP are ~20 cm^2 V^{-1} s^{-1} and ~65 cm^2 V^{-1} s^{-1}, respectively, and the four-terminal field-effect mobilities of the InSe and BP are ~370 cm^2 V^{-1} s^{-1} and ~120 cm^2 V^{-1} s^{-1}, respectively (Figure S1). The two-terminal mobilities are obtained via \((1/(C(L_{2pt}/W)) (dG_{2pt}/dV_{BG}))\) and the four-terminal mobilities are obtained via \((1/(C(L_{2pt}/W)) (dG_{4pt}/dV_{BG}))\), where \(C\) is the back-gate capacitance and \(W\) is the channel width. The two-terminal mobilities are smaller than the four-terminal values due to contact resistances \(R_{C}\), as is particularly notable for the InSe.

Figure 2a describes the measurement setup for the bias-controlled MLV and NDT properties of the InSe-BP-Ti device. While applying \(V_{D}\) to the BP and grounding the InSe, the two-terminal conductance between the source and drain \(G_{\text{2pt}}\) was measured in our device. Figure 2b,c show the measured \(G_{\text{2pt}}\) as a function of \(V_{BG}\) at positive and negative \(V_{D}\), respectively (Fig. 3a) also provides the corresponding \(I_D\) as a function of \(V_{BG}\).
In the positive $V_D$ regime, we note the particular region where the $G_{InSe-BP}$ hardly changes, creating an intermediate state "1/2" (at $-6 \text{ V} < V_{BG} < 0$), accompanied by a state "0" (at $V_{BG} < -20 \text{ V}$) and a state "1" (at $V_{BG} > 25 \text{ V}$). Several studies have demonstrated ternary inverters based on NDT\textsuperscript{5,6,14,22,27,28}, where the potential inside of the constituent semiconductor barely changes within the negative transconductance region. However, this methodology is limited to the inverting logic and indeed not suitable for versatile CMOS circuitry design. Ternary transistors are indispensable to design versatile ternary-data-processing CMOS integrated circuits. In the negative $V_D$ regime, a strong NDT behavior is observed, achieving a peak conductance ($G_{peak}$) of 140 nS and a peak-to-valley current ratio (PVC) of $\sim 10^2$ at $V_D = -1 \text{ V}$. Figure S2 shows transfer characteristics of our another InSe-BP-Ti device, which are qualitatively similar to those in Fig. 2b,c.

In the InSe-BP-Ti device, the total two-terminal resistance from source to drain can be approximately modeled as series connected resistors (Fig. 2a), which can include $R_{InSe}$, $R_{BP}$, the channel resistances of the InSe ($R_{InSe}$), and of the BP ($R_{BP}$). Each of these components is acquired via the four-terminal measurements at $V_D = 1 \text{ V}$, as discussed in Fig. 1c. $R_{InSe}$ ($R_{BP}$) corresponds to $\rho_{InSe}$ ($\rho_{BP}$) multiplied by the appropriate dimension of the InSe (BP) region of the InSe-BP-Ti device; Figure S3 provides schematics and a table to define the multiple parameters we introduced more specifically. We assume that most of the current flows along the InSe rather than the BP in the InSe/BP overlap region. The InSe region that sits above the BP is not effectively gate-controlled due to the asymmetric nature of Schottky barriers\textsuperscript{38,39}. In particular, the channel of our InSe-BP-Ti device consists of two different materials, InSe and BP, which leads to strong asymmetric characteristics, depending on the polarity of $V_D$. Figure 3. (a) $I_D$ versus $V_{BG}$ of the InSe-BP-Ti device at different $V_D$ values. Characteristic points (i), (ii), and (iii) are marked. (b) Schematic showing the band parameters and alignment between the InSe, BP, and both Au/Ti contacts. Previously reported Schottky barrier heights of the Au/Ti contacts for the InSe and BP, rather than metal work functions, are marked\textsuperscript{38,39}. (c-e) Band diagrams of the InSe-BP-Ti device at points (i), (ii), and (iii) and at (c) $V_D = 0$, (d) $V_D > 0$, and (e) $V_D < 0$. Red dots represent electrons, and blue dots represent holes. Corresponding equivalent circuits are illustrated in (d,e).
of $V_D$. Interestingly, the $(R_{C,\text{InSe}} + R_{\text{InSe}} + R_{\text{BP}})^{-1}$ is similar to the measured $G_{2pt,\text{InSe-BP}}$ at positive $V_D$, while the $(R_{\text{InSe}} + R_{\text{BP}} + R_{C,\text{InSe}})^{-1}$ is similar to the measured $G_{2pt,\text{InSe-BP}}$ at negative $V_D$. This is because when $V_D > 0$, electrons generally see a barrier at the junction between the Au/Ti contact and the InSe. By contrast, when $V_D < 0$, carriers generally see a barrier at the junction between the contact and the BP. The band diagrams further describe which side of the contact resistances, $R_{\text{InSe}}$ or $R_{\text{BP}}$, is more effective, depending on the polarity of $V_D$ (Fig. 3c–e).

Figure 3b represents the band parameters we used to build the band diagrams. The Schottky barrier for $\text{InSe-BP}$ of the InSe-Ti-BP-Ti device as a function of $V_{BG}$, without NDT behavior.

We investigated multiple InSe-BP devices with different contacts and structures: (1) an InSe/BP heterostructure device with few-layer graphene (FLG) contacts (InSe-BP-FLG device, Fig. 4a–d), and (2) another device where InSe and BP are serially connected via Au/Ti contacts (InSe-Ti-BP-Ti device, Fig. 4e–f). As described in Fig. 4a, two FLG contacts on the InSe and two others under the BP in the InSe-BP-FLG device. Figure 4b shows the $G_{2pt,\text{InSe-BP}}$ measured using the inner two contacts, where the InSe and BP are vertically overlapped over the whole measured region. It is notable that the $G_{2pt,\text{InSe-BP}}$ barely depends on $V_{BG}$, without NDT behavior. The InSe-BP-FLG device is also measured using the outer two contacts as source and drain (Fig. 4e). In contrast to the Fig. 4a setup, the individual non-overlapped InSe and BP regions are included in the Fig. 4e setup. Figure 4d shows the correspondingly measured $G_{2pt,\text{InSe-BP}}$ as a function of $V_{BG}$. Note the NDT behavior, similar to that observed in the InSe-BP-Ti device at a negative $V_D$ regime. This contrasts remarkably with the $G_{2pt,\text{InSe-BP}}$ in Fig. 4b, measured only in the InSe/BP overlap region. This clear distinction shows that the non-overlapped regions play a major role in inducing the NDT behavior. The measured $G_{2pt,\text{InSe-BP}}$ in Fig. 4d agrees well with $((G_{2pt,\text{InSe}})^{-1} + (G_{2pt,\text{BP}})^{-1})^{-1}$ (dashed), without considering the InSe/BP vdW junction resistance. The thicknesses of the BP and InSe layers used for the InSe-BP-FLG device are 4 nm and 15 nm (Figure S5), respectively, similar to those of the InSe-BP-Ti device. The $G_{2pt,\text{InSe-BP}}$ in Fig. 4b indeed agrees well with the $R_{\text{InSe}}^{-1}$ (dashed) estimated.

Figure 4. (a) Schematic describing the InSe-BP-FLG device and the measurement setup using the inner two electrodes. (b) $G_{2pt,\text{InSe-BP}}$ of the InSe-BP-FLG device measured as a function of $V_{BG}$ at different $V_D$ values, corresponding to the conductance of the InSe/BP overlap region. The inset shows an optical micrograph of the InSe-BP-FLG device (scale bar = 10 μm). (c) Schematic showing the measurement setup using the outer two electrodes of the InSe-BP-FLG device. (d) $G_{2pt,\text{InSe-BP}}$ of the InSe-BP-FLG device as a function of $V_{BG}$ at different $V_D$ values, corresponding to the conductance over the whole channel region, including the non-overlapped InSe and BP regions. (e) Schematic describing the InSe-Ti-BP-Ti device and corresponding measurement setup. (f) $G_{2pt,\text{InSe-BP}}$ of the InSe-Ti-BP-Ti device as a function of $V_{BG}$ at different $V_D$ values. The inset shows an optical micrograph of the InSe-Ti-BP-Ti device (scale bar = 10 μm).
Figure 5. (a) \(I_{\text{peak}}\) (b) \(I_{\text{valley}}\) (c) PVCR, and (d) SS of the InSe-Ti-BP-Ti (grey), InSe-BP-Ti (blue), and InSe-BP-FLG (red) devices as a function of \(V_D\).
The patterned hBN flake was annealed at 400℃ for an hour in the Ar/H₂ atmosphere to remove polymethyl carbonate (PPC) residue, which allowed the InSe and BP to be partially exposed for metallization even after having the hBN cover on them. InSe and BP flakes were mechanically exfoliated on polydimethylsiloxane (PDMS) film using cleanroom tape, while hBN was exfoliated on 300 nm-thick SiO₂/Si substrate. Once appropriate InSe and BP flakes with the desired thicknesses were identified, the PDMS films with the flakes were cut into 10 mm × 10 mm pieces and then attached to a glass slide for transfer to a designated location. The BP and InSe flakes on PDMS film were then successively transferred on 300 nm-thick SiO₂, thermally grown on a highly doped Si substrate, resulting in an InSe/BP stack. To minimize the surface contamination of the device, the exfoliation and stacking processes were performed entirely in a glovebox, keeping the oxygen level below 5 ppm. The separately exfoliated hBN flake suitable for top encapsulation was patterned before the transfer process.

The performance also depends on the mobility and the initial doping level of BP. A few studies have reported PVCR values higher than ours, but these either applied much higher doping levels45. Depending on the BP thickness ranging from 14 to 4 nm, the band gap of BP varies from 0.3 to 0.5 eV46. The BP band gap change from 0.3 to 0.5 eV leads to a notable impact on the off current and corresponding PVCR of the NDT. On the other hand, the band gap change of InSe of similar thickness (4–14 nm) is 1.3–1.4 eV46, and it does not cause much change in the off current because the band gap within that range is already quite large, compared to kT; k is the Boltzmann constant and T is the temperature. Further thicker InSe might lead to improved I_{peak} as the mobility increases for thicker InSe47. Multiple InSe-BP-Ti devices with different BP thicknesses ranging from 4 to 14 nm were examined. Figure 6b shows their PVCR values versus I_{peak} at different V_{D} along with previously reported PVCR values of other 2D material–based NDT devices5,6,13,14,17–19,23,28,48–51.

In summary, we demonstrated bias-controlled MVL and NDT properties based on InSe/BP heterostructures. Due to the asymmetric nature of Schottky barriers at the junction between Au/Ti contact and InSe or BP, asymmetric electrical characteristics were developed depending on the polarity of V_{D}. At positive V_{D}, the InSe-BP-Ti devices worked as ternary transistors, but at negative V_{D}, the devices showed NDT characteristics. The contributions of contact resistances and channel resistance were identified separately via four-terminal measurements performed on each layer. Multiple devices with different contacts and device structures were investigated, and we systematically discussed the influence of contact resistances, heterojunction resistance, channel resistances, and the thickness of BP on the detailed operational characteristics of InSe/BP-based vdW heterostructures. These results provide insight into the operation principle and further performance optimization of general 2D material–based vdW heterostructures.

Experimental
Fabrication of the InSe-BP-Ti device. InSe and BP flakes were mechanically exfoliated on polydimethylsiloxane (PDMS) film using cleanroom tape, while hBN was exfoliated on 300 nm-thick SiO₂/Si substrate. Once appropriate InSe and BP flakes with the desired thicknesses were identified, the PDMS films with the flakes were cut into 10 mm × 10 mm pieces and then attached to a glass slide for transfer to a designated location. The BP and InSe flakes on PDMS film were then successively transferred on 300 nm-thick SiO₂, thermally grown on a highly doped Si substrate, resulting in an InSe/BP stack. To minimize the surface contamination of the device, the exfoliation and stacking processes were performed entirely in a glovebox, keeping the oxygen level below 5 ppm. The separately exfoliated hBN flake suitable for top encapsulation was patterned before the transfer using conventional e-beam lithography and reactive ion etching using SF₆ gas to have eight openings. These allowed the InSe and BP to be partially exposed for metallization even after having the hBN cover on them. The patterned hBN flake was annealed at 400℃ for an hour in the Ar/H₂ atmosphere to remove polymethyl methacrylate (PMMA) residue, which also allowed the hBN to be easily picked up using a polypropylene carbonate (PPC)/PDMS stamp. The patterned hBN was then transferred onto the InSe/BP stack by melting the PPC.

Figure 6. (a) I_{D} versus V_{BG} transfer characteristics of the InSe-BP-Ti devices (solid) and the BP (dashed) with different BP thicknesses, which are presented in parentheses. The V_{BG} is shifted relative to the minimum current point of the BP for each device. (b) PVCR versus I_{peak} characteristics of our multiple InSe-BP-Ti devices with different BP thicknesses, accompanied by previously reported PVCR values of other 2D material–based NDT devices for comparison.
at ~120 °C. Then the hBN/InSe/BP heterostructure was washed with acetone. Additional e-beam lithography was performed to define metal contacts for the InSe-BP-Ti device, followed by Au(80 nm)/Ti(10 nm) deposition via e-beam evaporation and lift-off. The device was annealed at 400°C for an hour in the Ar/H₂ atmosphere to remove the remaining polymer residues.

**Fabrication of the InSe-Ti-BP-Ti device.** The fabrication process for the InSe-Ti-BP-Ti device was similar to that of the InSe-BP-Ti device. One distinction was that the BP and InSe flakes were ~5 μm apart from each other without InSe/BP vdW heterojunction.

**Fabrication of the InSe-BP-FLG device.** FLG and hBN flakes were mechanically exfoliated on Si/SiO₂ substrates. They were annealed at 400°C for an hour in the Ar/H₂ atmosphere. Individually exfoliated BP and InSe flakes were successively transferred on the FLG exfoliated on SiO₂, similar to the transfer process employed for the InSe-BP-Ti device. Finally, hBN and top FLG flakes were successively picked up using a PPC/PDMS stamp. Then the hBN/FLG stack was transferred to the prepared InSe/BP/FLG heterostructure on SiO₂. Au/Ti contacts were deposited on a non-encapsulated region of the FLG, followed by annealing in the Ar/H₂ atmosphere.

**Material and electrical characterization.** The surface topography and material quality of the fabricated devices were examined via AFM (Park systems, XE-100) and Raman spectroscopy (Renishaw, 514 nm wavelength laser). The electrical measurements were performed using a semiconductor analyzer (Keithley, 4200A-SCS) at high vacuum (~10⁻⁶ bar).

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