Supporting Information

Significantly Enhanced Magnetoresistance in Monolayer WTe$_2$ via Heterojunction Engineering: A First-principles Study

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Figure S1. Charge density distributions of VBM (a) and CBM (b) of the graphene/WTe$_2$ heterojunction. For the charge density distribution iso-surface plot, the iso-value is 25 e/Bohr$^3$. The yellow and gray balls represent Te and W atoms, respectively.
Table S1: Effective mass $m^*$ ($m_0$ is the mass of an electron), elastic modulus $C$ (eV/Å²) and deformation potential constant $E_1$ (eV), and room temperature carrier mobility $\mu$ ($\times 10^3$ cm²V⁻¹s⁻¹) of the isolated WTe₂ monolayer and the WTe₂/graphene heterojunction.

<table>
<thead>
<tr>
<th>Direction</th>
<th>Carrier type</th>
<th>$m^*$</th>
<th>C</th>
<th>$E_1$</th>
<th>$\mu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>WTe₂ monolayer</td>
<td>a-axis</td>
<td>hole</td>
<td>0.88</td>
<td>7.70</td>
<td>-10.35</td>
</tr>
<tr>
<td></td>
<td></td>
<td>electron</td>
<td>0.31</td>
<td>7.70</td>
<td>-4.23</td>
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<tr>
<td></td>
<td>b-axis</td>
<td>hole</td>
<td>0.54</td>
<td>9.15</td>
<td>0.55</td>
</tr>
<tr>
<td></td>
<td></td>
<td>electron</td>
<td>0.25</td>
<td>9.15</td>
<td>-1.40</td>
</tr>
<tr>
<td>WTe₂/graphene</td>
<td>a-axis</td>
<td>hole</td>
<td>-</td>
<td>57.46</td>
<td>-3.66</td>
</tr>
<tr>
<td>heterostructure</td>
<td></td>
<td>electron</td>
<td>0.35</td>
<td>57.46</td>
<td>-6.78</td>
</tr>
<tr>
<td></td>
<td>b-axis</td>
<td>hole</td>
<td>-</td>
<td>58.31</td>
<td>-3.50</td>
</tr>
<tr>
<td></td>
<td></td>
<td>electron</td>
<td>0.30</td>
<td>58.31</td>
<td>-3.15</td>
</tr>
</tbody>
</table>
For the weak case, we should consider a parallel equivalent circuits of resistors made of graphene and WTe$_2$ to obtain the total MR resistance. Then, the total resistance ($R_t$) is related to that of graphene ($R_{gr}$) and WTe$_2$ ($R_{wt}$) as follows: $1/R_t = 1/R_{gr} + 1/R_{wt}$. And, the $MR = \frac{R(H) - R(0)}{R(0)}$, $R(0)$ is the resistance when the external magnetic field equal to 0. We use the MR of WTe$_2$ ($B^2$ for parabolic dispersion) and graphene ($B$ for a linear dispersion) and obtain:

\[
MR_{wt} = \frac{R_{wt}(H) - R_{wt}(0)}{R_{wt}(0)} = \mu_e^2 B^2, \quad (S1)
\]

\[
MR_{gr} = \frac{R_{gr}(H) - R_{gr}(0)}{R_{gr}(0)} = \mu_h B, \quad (S2)
\]

\[
MR_t = \frac{R_t(H) - R_t(0)}{R_t(0)}, \quad (S3)
\]

And,

\[
1 \frac{1}{R_t(0)} = \frac{1}{R_{wt}(0)} + \frac{1}{R_{gr}(0)}, \quad (S4)
\]

\[
1 \frac{1}{R_t(H)} = \frac{1}{R_{wt}(H)} + \frac{1}{R_{gr}(H)}, \quad (S5)
\]

Here we make an approximation that when there is no external magnetic field, $R_{wt}(0) \approx R_{gr}(0) = R$. Because both graphene and WTe$_2$ are semimetals, when there is no external magnetic field, their resistances are very small. From (S4), we can obtain $R_t(0) = R/2$. Then, we can rewrite (S1-3) as:

\[
R_{wt}(H) = (\mu_e^2 B^2 + 1)R, \quad (S6)
\]

\[
R_{gr}(H) = (\mu_h B + 1)R, \quad (S7)
\]

\[
R_t(H) = \frac{(MR_t + 1)R}{2}, \quad (S8)
\]

Combine equations of (S5-8), we obtain the total MR as:

\[
MR_t = \frac{2\mu_h \mu_e^2 B^3 + \mu_e^2 B^2 + \mu_h B}{\mu_e^2 B^2 + \mu_h B + 2}, \quad (S9)
\]
Figure S2. Top and side views of the geometric structure of the WTe$_2$/graphene heterojunction with different angle, $\theta$. (a)(b) and (c)(d) show the $\theta = 30^\circ$ (a $2 \times 7$ supercell of WTe$_2$ is used to match a $5 \times 6\sqrt{3}$ supercell of graphene) and $\theta = 10.8^\circ$ (a $2 \times 6$ supercell of WTe$_2$ is used to match a $2\sqrt{2} \times 2\sqrt{7}$ supercell of graphene) heterojunction for top and side view, respectively; $a$, $b$ and $c$ represent the lattice vectors. Brown, yellowish and gray balls represent C, Te and W atoms, respectively.
Table S2: Effective mass $m^*$ ($m_0$ is the mass of an electron), elastic modulus $C$ (eV/Å²) and deformation potential constant $E_1$ (eV), room temperature carrier mobility $\mu$ ($\times 10^3$ cm²V⁻¹s⁻¹) and MR when the external magnetic field $B=14.7$T of the WTe₂/graphene heterostructure with different angle $\theta$.

<table>
<thead>
<tr>
<th>$\theta$</th>
<th>Carrier type</th>
<th>$m^*$</th>
<th>C</th>
<th>$E_1$</th>
<th>$\mu$</th>
<th>$\mu_e \mu_h$</th>
<th>MR</th>
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<tbody>
<tr>
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<td>58.31</td>
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<td>-3.79</td>
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<tr>
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<td>115.18</td>
<td>299.46</td>
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<td>-6.77</td>
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