Supplementary Information for
Bipolar giant unidirectional magnetoresistance

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S1. $E_{\text{in}}$ induced Zeeman-type spin splitting

Spin-orbit interaction (SOI) systems with intrinsic in-plane dipole moment allows for an out-of-plane spin-polarization when an electric field breaks the centrosymmetry of electronic wavefunctions. The electrified layers atop create an electric field ($E_{\text{in}}$) that is critical for the out-of-plane spin-polarization in the WSe$_2$.

In Fig. S3, the net dipole moment is along the plane. The bulk band structure registers the valence band maximum around $\Gamma$-point, in contrast to the reported K-point. The latter is a consequence of falsely optimized distance between layers from an inaccurate exchange-correlation function. The conduction band minimum appears at K-point, different from the reported intermediate point along $\Gamma$-K line.

We maintained the lattice constants from experimental work for the layered structure calculations, as the few-layered transport happens within the bulk. In Fig. S4, we glued together the band structures of monolayer (ML), bilayer (BL), triplelayer (TL), and quadlayer (QL) with electric field strengths of 0.0, 0.2, and 0.5 V nm$^{-1}$. For a single ML, the spin-splitting is on the order of $\sim$500 meV around K-point. Without or with electric field, $\Gamma$-point does not have spin polarization, which rules out the Rashba spin splitting. It seems that the splitting magnitude takes the order of $\text{ML} > \text{QL} > \text{BL} > \text{TL}$ for $E<0.5$ V nm$^{-1}$. That means the spin-polarization peaks for a single ML. The splitting of ML is an intrinsic property, which is not controllable by the E field. In contrast, we expect that for a stronger E, the splitting of BL could exceed that of ML but it probably requires a relatively higher E field that is beyond the scope of
ordinary experimental capability. We found that even number of layers exhibits a more pronounced spin-splitting in the band energies compared to odd number of layers (except for ML).

**S2. Magnetic-field enhanced recombination of photocarriers**

Actually, the field-enhanced recombination of photocarriers alone cannot cause such a giant PMR. But when this effect occurs in a CPC device, the role of magnetic field may be magnified. For the CPC effect, the generated photocarriers are stored in a nonvolatile metastable state, possibly resulting in a higher sensitivity to magnetic fields.

To understand the PMR effect, we plot the $I_j$-time curve of WSe$_2$/Q2DEG heterojunction with magnetic field applied at $V_B = 5$ V after optical charging in Fig. S6B. $I_j$ decreases rapidly at the beginning, resulting from drastic recombination of photocarriers stored at shallower regions of SCR. Then, the downward trend of $I_j$ slows down, because $V_B$ is not large enough to release the inert photocarriers stored at deeper region of SCR. To investigate the effect of magnetic field on the stored photocarriers, we apply the magnetic field onto the system at 1700 s with a rate of 0.02 T·s$^{-1}$. With $B$ increasing to $-6$ T, $I_j$ dramatically decreases by $10^6$ times. Later, with $B$ going along $-6 \rightarrow -9 \rightarrow 0$ T, $I_j$ remains almost zero without returning to the initial state. It seems that the photocarriers stored at deeper regions of SCR are further depleted at high field, implying that the magnetic field encourages the recombination of photocarriers.

Based on this understanding, we can clarify the role of magnetic field in the PMR. The application of bias voltage reduces the SCR width, and drives electrons and holes to meet and recombine at the interface. This process can be intensified by magnetic
field, causing the recombination of inert photocarriers. Therefore, the total photocarrier density is greatly decreased, resulting in a giant PMR. This physical process can be achieved only in the CPC device. By comparison, for the device with normal photoconductivity, all the photocarriers will recombine spontaneously at the end of their lifetime. There are no inert photocarriers to exist in the materials, so the role of magnetic field is not significant enough to cause a giant PMR.

S3. The theoretical model for highly-anisotropic NMR effect

Besides $E_{\text{in}}$, the application of a magnetic field can give rise to the Zeeman splitting of valence bands, generating the spin dependent Zeeman shift $\Delta E_Z = \pm 1/2g\mu_B B$, where $g$ is the Landé g-factor and $\mu_B$ is the Bohr magneton. The Zeeman effect determined by both the electric and magnetic fields should be responsible for the highly-anisotropic NMR effect. For Zeeman MR (see Ref. 48 in the main text), the electric current under magnetic field can be written as

$$I(B) = I_0[n_\uparrow\exp(mB) + n_\downarrow\exp(-mB)],$$

where $n_\uparrow$ and $n_\downarrow$ are respectively the percentages of spin-up and spin-down carriers in the total number, and $m$ is a fitting parameter determined by the degree of Zeeman effect. The out-of-plane spin polarization depends on the ratio of $n_\uparrow$ and $n_\downarrow$, which perpendicular to the WSe$_2$ surface. To simulate the UMR effect, $U_{\text{sp}}$ is used to estimate the degree of spin polarization instead of the difference between $n_\uparrow$ and $n_\downarrow$.

In mathematics, we can always make the following equations true:

$$n_\uparrow = n_0\exp(mU_{\text{sp}}),$$

$$n_\downarrow = n_0\exp(-mU_{\text{sp}}),$$

where $n_0$ is equal to $\sqrt{n_\uparrow n_\downarrow}$ and $U_{\text{sp}}$ is equal to $\frac{1}{2m}\ln\frac{n_\uparrow}{n_\downarrow}$. With $n_0 = 0.5$, the $B$ dependence of $I_j$ is written as
\[ I_j = I(B+U_{sp}) = I_0 \left\{ n_0 \exp \left[ m(B+U_{sp}) \right] + n_0 \exp \left[ -m(B+U_{sp}) \right] \right\}, \]  
where \( U_{sp} \) is fitted by the experimental data. For the fitting curves in Fig. 5D, \( m \) is about 0.07 T\(^{-1}\) and \( U_{sp} \) is 3.9 T. In addition, the theoretical model can also be used to explain the UMR effect. In the y-z plane, NMR is only determined by the component of magnetic field parallel to the z-axis. Thus, through theoretical and experimental analysis, the relation between \( I_j \) and \( \theta \) is expressed as

\[ I_j = I(B \cos \theta + U_{sp}) = I_0 \left\{ n_0 \exp \left[ m(B \cos \theta + U_{sp}) \right] + n_0 \exp \left[ -m(B \cos \theta + U_{sp}) \right] \right\}. \]

S4. The linear dependence of \( n \) on \( V_B \) and \( P_{opt} \)

According to our recent work, for the CPC device, the large SCR can be regarded as a container for photocarriers storage (see Ref. 46 in the main text). The photocarriers construct the photocarrier-induced pseudo SCR (PPSCR), and their recombination causes the enlargement of PPSCR. So we have the relation as (see Ref. 46 in the main text)

\[ \frac{dV_{bi}^{ph}}{dt} = \frac{R_f}{G t_e} I_{ph}, \]

where \( V_{bi}^{ph} \) is the photocarrier-induced pseudo built-in voltage, \( I_{ph} \) is the photocurrent, \( R_f \) is the recombination factor that is correlative to the distribution of photocarriers in ISCR, \( G \) is the generation rate of electron-hole pairs and \( t_e \) is the charging time. Here, the photocurrent actually reflects the recombination of photocarriers in SCR, which is expressed as

\[ I_{ph} = \frac{dN}{dt}, \]
where $N$ is the total number of stored photocarriers. Combing Equations [4] and [5], we have

$$dV_{bi}^{ph} = \frac{R_f}{Gt_c} dN,$$

where $N$ is the total number in SCR. This equation is available to simulate the I-V characteristics after optical charging. When the device is exposed to the continuous light illumination, Equation [6] needs to be changed. When the current is stable, the generated photocarriers per unit time should be equal to the consumed photocarriers per unit time in SCR. Here, $t_c$ is not a constant, and changes to be $\Delta t$. So we can write

$$\Delta V_{bi}^{ph} = \frac{R_f}{G} \frac{\Delta N}{\Delta t},$$

where $\frac{\Delta N}{\Delta t}$ can be regarded as the consumed photocarriers per unit time in SCR. Due to the continuous illumination, the conduction process occurs mainly in the transport layer. This implies that the higher the sheet carrier density is, the more the number of carriers happen to recombine. So $n_s$ should be proportional to $\frac{\Delta N}{\Delta t}$, that is $n_s \propto \frac{\Delta N}{\Delta t}$.

Besides, our recent work Ref.46 has demonstrated that $V_{bi}^{ph}$ shows a linear dependence on $V_B$ with $V_B > 2V$, thus deducing $\Delta V_{bi}^{ph} = V_B - V_0$. With $G \propto P_{opt}$, we obtain the relation available for the continuous illumination as

$$(V_B - V_0) \cdot P_{opt} \propto n_s,$$

where $V_0$ is the fitting parameter. It indicates that, for a given $n_s$, $V_B$ is inversely proportional to $P_{opt}$. Equation [8] is verified by the experimental results in the inset of Fig. 4D.
The GBU-PhMR effect originating from the competition between PMR and NMR

For the WSe$_2$/Q2DEG heterojunctions, the PMR effect is mainly caused by the magnetic-field enhanced recombination of photocarriers. So its junction resistance should increase monotonously with the increase of magnetic field value, regardless of the sign of magnetic field. By analyzing the experimental data, we can write an empirical equation as

$$\Delta R_{\text{PMR}}(B) = C_1 |B|^\alpha,$$

[9]

where $C_1$ and $\alpha$ are the fitting parameters.

From the theoretical model in section S4, it is known that the NMR effect originates from the Zeeman effect. For simplicity, from Equation [2], the field-induced decrease of junction resistance can be expressed as

$$\Delta R_{\text{NMR}}(B) = \frac{C_2}{\exp[m(B+U_{sp})]+\exp[-m(B+U_{sp})]},$$

[10]

where $C_2$ is a fitting parameter.

Actually, the observed GBU-PhMR effect results from the combined action of PMR and NMR mechanisms. The sign of magnetoresistance is determined by the competition between them. Therefore, the field-controlled overall change of junction resistance should be written as

$$\Delta R_{\text{GBU-PhMR}}(B) = \Delta R_{\text{PMR}}(B) + \Delta R_{\text{NMR}}(B) = C_1 |B|^\alpha + \frac{C_2}{\exp[m(B+U_{sp})]+\exp[-m(B+U_{sp})]}.$$

[11]

Here, the value of $C_1/C_2$ can describe the proportion of PMR and NMR roughly. A large value of $C_1/C_2$ indicates the dominant role of PMR in the magnetic transport, or otherwise the NMR dominates. Based on this model, we simulate the GBU-PhMR effect with different values of $C_1/C_2$ and $U_{sp}$ as shown in Fig. S8.
Figs. S8 A, B, C and D show the total resistance change of $\Delta R_{\text{GBU-PhMR}}$ that is the sum of $\Delta R_{\text{PMR}}$ and $\Delta R_{\text{NMR}}$. The magnetic field dependences of $\Delta R_{\text{PMR}}$ and $\Delta R_{\text{NMR}}$ are simulated with different values of $C_1/C_2$ and $U_{sp}$ in Figs. S8 E, F, G and H. With $C_1/C_2 = 0.1$ and $U_{sp} = 0.5$ T, the $\Delta R_{\text{GBU-PhMR}}$-$B$ curve shows the PMR effect with an approximate parity symmetry, where the NMR is covered by the PMR due to the large value of $C_1/C_2$. Such a $\Delta R_{\text{GBU-PhMR}}$-$B$ relation is applicable to the case for the sample with 11.3-nm WSe$_2$ layer in Fig. 2D. It is inferred that the thinner WSe$_2$ layer causes the smaller values of $C_1/C_2$ and $U_{sp}$. With the increase of WSe$_2$ thickness, the value of $C_1/C_2$ increases significantly, so the NMR becomes comparable to the PMR.

To investigate the effect of $U_{sp}$ on the GBU-PhMR effect, we have plotted several $\Delta R_{\text{GBU-PhMR}}$-$B$ curves with the different $U_{sp}$ values but the same $C_1/C_2$ value. In Figs. S8 B and F, with $C_1/C_2 = 10^{-3}$ and $U_{sp} = 4$ T, the $\Delta R_{\text{GBU-PhMR}}$ shows the similar dependence on the magnetic field to the samples with 30.2-nm and 40.3-nm WSe$_2$ layers shown in Fig. 2D. In this case, the NMR effect emerges only along the positive magnetic field, and the UMR behavior is just achieved, which is attributed to the large value of $U_{sp}$. In Figs. S8 C, D, G and H, with $U_{sp}$ decreasing to 0.5 T, the NMR effect can be achieved along the negative magnetic field, which explains the phenomenon in the 68.9-nm sample in Fig. 2D. In this case, both of the PMR and NMR effects play important roles in the magnetic transport. The Zeeman-effect induced NMR mainly works at low magnetic field, while the PMR is more pronounced at high magnetic field. Based on the above analysis, it is reasonable to conclude that the GBU-PhMR is the combined effect of PMR and NMR. As the WSe$_2$ thickness increases, the value of $C_1/C_2$ increases monotonously, but $U_{sp}$ increases first and then decreases. For a thicker WSe$_2$ layer, its property is closer to the bulk, thus causing a smaller value of $U_{sp}$. 


The PMR and NMR mechanisms may be separated as two independent processes in the space charge region (SCR). Due to the large width of SCR of WSe$_2$/Q2DEG, the magnetic transport near the interface may be different from the region far from the interface. It seems that the NMR occurs at the location close to the interface due to the Anderson localized states, while the PMR may reflect the natures of WSe$_2$ and Q2DEG, so it occurs at the position further from the interface.
**Supplementary Information Figures**

**Fig. S1.** Schematic of fabrication process for WSe$_2$/Q2DEG heterostructures. Few-layered WSe$_2$ is mechanically exfoliated from a piece of bulk single crystal onto STO substrates. More than half of the WSe$_2$ were covered with photoresist by lithographic, and the remaining part was exposed to air. Then, an Au electrode was deposited on the sample by magnetron sputtering. After that, the lithographic technique was used again to cover the part of WSe$_2$ flake contacting the Au electrode, and to leave the area containing bare STO and WSe$_2$. The sample was irradiated for 3 minutes by an Ar$^+$ ion beam with a beam voltage of 400 V. A water-cooled sample holder was used during the etching process. Using the above AIBA method, the WSe$_2$/Q2DEG heterojunctions were successfully fabricated after removing photoresist with acetone.
Fig. S2. Raman measurement. (A) Raman spectrum of the multilayer WSe$_2$, showing the typical Raman peaks of $E^{1}_{2g}$ and $A^{1}_{1g}$. (B) Raman mapping of the WSe$_2$/Q2DEG heterojunction.
**Fig. S3.** Lattice and band structures of bulk 2H-WSe$_2$. (A), (B) Top and side view of crystal structure of 2H-WSe$_2$. The net dipole along the mirror plane of a single mono-layer WSe$_2$ is formed by charged cation W$^{4+}$ and anion Se$^{2-}$, which is critical for the spin-polarization (see (B)). (C) Brillouin zone of 2H-WSe$_2$. (D) Electric band structure of bulk 2H-WSe$_2$. 
Fig. S4. Thickness dependences of electric-field induced Zeeman-type spin splitting. Electronic band structures of the monolayer (ML), bilayer (BL), trilayer (TL) and quad-layer (QL) of WSe$_2$ under the electric fields of 0, 0.2 and 0.5 V·nm$^{-1}$, respectively. Blue and red emphasize the bands of out-of-plane spin-up and spin-down polarizations, respectively.
Fig. S5. Magnetic hysteresis loops of Ar+-ion-etched STO substrate at 5 K.
Fig. S6. Impact of magnetic field on optical charging and discharging. (A) I-V loops with different magnetic fields applied after full optical charging. (B) $I_t$-time curve with the magnetic field changing in darkness after full optical charging, with $B$ going along 0→-9→0 T during 1700 ~ 2600 s.
Fig. S7. Wavelength dependence of BGMR effect. $R_J$-$B$ curves under continuous optical illuminations at 10 K in the wavelength range of 405-808 nm. The power densities are adjusted at different wavelengths, to ensure the same number of incident photons per unit time.
**Fig. S8.** Numerical simulation of BGU-PhMR. $\Delta R_{\text{BGU-PhMR}}$, $\Delta R_{\text{PMR}}$ and $\Delta R_{\text{NMR}}$ as a function of magnetic field, respectively. $C_1/C_2 = 0.1$ and $U_{sp} = 0.5$ T for (A) and (E), $C_1/C_2 = 10^{-3}$ and $U_{sp} = 4$ T for (B) and (F), $C_1/C_2 = 10^{-3}$ and $U_{sp} = 3$ T for (C) and (G) and $C_1/C_2 = 10^{-3}$ and $U_{sp} = 0.5$ T for (D) and (H). Here, $\alpha$ is 3 for all these curves.
Fig. S9. MR hysteresis. $R_j$-$B$ loops under continuous optical illuminations at 10 K with $P_{\text{opt}} = 0.07, 0.15, 0.67$ and $2.1 \text{ mW cm}^{-2}$, respectively.