Recent advances in nanoscale lasers, amplifiers, and nonlinear optical converters have demonstrated the unprecedented potential of metal–insulator–semiconductor (MIS) structures as a versatile platform to realize integrated photonics at the nanoscale. While the electric field enhancement and confinement have been discussed intensively in MIS based plasmonic structures, little is known about the carrier redistribution across the heterojunction and photocurrent transport through the oxide. Herein, we investigate the photo-generated charge transport through a single CdSe microbelt-Al₂O₃-Ag heterojunction with oxide thickness varying from 3 nm to 5 nm. Combining photocurrent measurements with finite element simulations on electron (hole) redistribution across the heterojunction, we are able to explain the loss compensation observed in hybrid plasmonic waveguides at substantially reduced pump intensity based on MIS geometry compared to its photonic counterpart. We also demonstrate that the MIS configuration offers a low-dark-current photodetection scheme, which can be further exploited for photodetection applications.

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1. INTRODUCTION

Charge transport through a metal–insulator–semiconductor (MIS) heterogeneous junction holds the key to the performance of many classes of electronic devices, for example, MIS field-effect transistors and MIS capacitors [1]. In recent years, the MIS configuration has attracted great attention in the research community due to its novel applications in optical devices, in particular, nanoscale plasmonic devices such as hybrid plasmonic waveguides, hybrid plasmonic lasers [2–4], amplifiers [5], and nonlinear light conversion devices [6,7], in which the optical modes are tightly confined into a deep subwavelength scale of the interfaces of MIS, promoting strong light–matter interaction at the interfaces and bridging the size gap from a few nanometers of electronic components to a few micrometers of optical components.

For MIS based capacitors, the thickness of the insulator is usually larger than 5 nm to prevent any leakage current. For the application of ideal Schottky diodes, the insulator thickness is considered less than 1 nm. In photodetector applications, the insulator thickness is kept below 2 nm to allow an efficient carrier transport. In hybrid plasmonic structures, the thickness of the insulator is usually from 3 nm to 6 nm, allowing a strong electric field enhancement and a much reduced ohmic loss in the devices [8]. The interesting phenomenon of loss compensation in hybrid plasmonic waveguides at very low pump intensities was reported [5]. This demonstration opens up possibilities to significantly reduce the pump energy required to achieve gain-assisted signal modulation in plasmonic circuits. However, the mechanism is yet to be clarified. While the electric field enhancement and confinement have been discussed intensively in MIS based plasmonic structures, little is known about charge transport across the MIS heterojunction upon radiation of light, the redistribution of photo-generated carriers at the interfaces, their impacts on the optical gain/absorption of semiconductor material, and the potential application of MIS heterojunctions as photodetectors, particularly for oxide thickness ranging from 3 nm to 5 nm. Answers to above questions will also shed light on optically and electrically driven plasmonic signal modulation, which plays a pivotal role in integrated plasmonic circuits.
In this paper, we investigate the photocurrent transport through single CdSe microbelt (MB)-Al2O3-Ag interfaces with Al2O3 thickness varying from 3 nm to 5 nm. Peak photocurrent from a few nanoamperes to a few microamperes at peak incident laser intensity of 1 MW/cm² with single photon energy exceeding the bandgap energy of CdSe is detected at zero bias for Al2O3 thickness ~3–4 nm, which is demonstrated through finite element simulation to be sensitively dependent on the positions of the conduction band edge of the ultrathin oxide layer. We show that the requirement for the aligned Fermi level across the heterojunction leads to a charge redistribution across the heterojunction and high optical gain in the CdSe region at low pump intensity. As the oxide thickness increases to 5 nm, no photocurrent is obtained at zero bias upon exposure to light. Instead, we observe an abrupt photocurrent increase at a moderate bias voltage of ~1.5 V to ~4.2 V, which is attributed to the Coulomb blockade effect. The current gain compared to dark current can exceed 2 × 10⁷ at a bias voltage of ~2 V with a photoresponsivity of 0.03 A/W, which is of great importance for photodetection applications where low dark currents are highly desirable [9,10].

2. SAMPLE FABRICATION AND DARK CURRENT–VOLTAGE CHARACTERIZATION

To fabricate the heterojunction, CdSe MBs grown by chemical vapor deposition are deposited on the glass coverslip first. The Au electrode is then deposited on one side of the chosen CdSe MB, followed by ultrathin Al2O3 layer growth via atomic layer deposition. Finally, the Ag electrode is formed on the other side of the CdSe MB (see Appendix A for details). One example of the final devices is given as an inset of Fig. 1(a). Before the photocurrent measurements are conducted, current–voltage (I-V) characteristics of the devices are obtained first without light. Figure 1(a) shows the typical dark I-V characteristics obtained on three devices with Al2O3 thickness ~3–4 nm. As the current through individual CdSe MB is extremely small (tens of picoamperes), a lock-in technique is used to obtain the current at ambient conditions and room temperature. In the measurements, the Ag electrode is kept at zero bias, while the voltage applied to the Au electrode varies (see Appendix A for details). It is clear from Fig. 1(a) that the absolute current is significantly larger for the negative bias voltage, behaving as a diode where the forward direction corresponds to the negative voltage. The absolute value of the current varies from one device to another, sensitively dependent on the thickness of CdSe MB, the contact area with the Au and Ag electrodes, and the lateral distance between the Au and Ag electrodes (see Appendix B).

Out of 10 samples we measured with Al2O3 thickness ~3–4 nm, 70% of them showed measurable dark current. For devices with Al2O3 thickness ~5 nm, no dark current is detected within the bias range of ~9 V to 9 V, meaning that the current is below the detection limit of our setup (~3 pA).

The I-V characteristics of the devices observed experimentally can be reproduced using the finite element simulation package COMSOL Multiphysics 5.5 semiconductor module. The simulated I-V curves on a 500 nm thick CdSe MB with an Al2O3 thickness of 3 nm are plotted in Fig. 1(b). In the simulations, in addition to the drift-diffusion equations [1], tunneling of electrons through the Al2O3 layer and Au-CdSe Schottky barrier [11] is also included to properly reproduce the experimental I-V characteristics (see Appendices A and B for details). Here the work functions of Ag and Au are set at 4.6 eV [12] and 5.1 to 5.13 eV [13] while the conduction band and valence band of the CdSe are set to 4.5 eV and 6.23 eV [14], respectively. The band positions and bandgap of the ultrathin Al2O3 layer, on the other hand, have been reported with very different values in the literature, with the conduction band edge varying from 1.27 eV [15] to 3.8–4.7 eV [16,17], sensitively dependent on the thickness of the oxide, the growth

![Fig. 1. Current–voltage characteristics across Ag-Al2O3-CdSe MB-Au heterojunction. (a) I-V characteristics of three samples of Al2O3 thickness ~3–4 nm. Data from sample 2 are multiplied by a factor of 10 for clarity. The onset voltages for six samples (three extra samples shown in Appendix C) are summarized into the histogram in the inset. The error bar indicates the standard deviation from all samples. The positive and negative onset voltages are the intersection voltages with the zero current axis obtained by linear fits to the data points in the corresponding voltage regions. The wide field optical image shows the heterojunction of sample 1 with the scale bar 5 μm. (b) COMSOL simulations on the I-V characteristics across Ag-Al2O3 (3 nm) -CdSe MB (500 nm)-Au heterojunction. In the simulations, the Ag work function and CdSe energy levels are kept the same while the conduction band of Al2O3 and Au work function are allowed to vary. The results demonstrate that the I-V can change significantly with even a small change in Al2O3 conduction band and Au work function. The inset shows the logarithm of electron density distribution, log(n), at zero bias across the entire heterojunction with the conduction band of Al2O3 at 3.74 eV and Au work function of 5.13 eV. Here the unit of n is m⁻³.]
temperature, and fabrication methods. Our simulations show that a small change in the conduction band position of the Al₂O₃/work function of Au can lead to a large change in the tunneling current at the positive/negative voltage, as illustrated in Fig. 1(b).

One important result from the simulation is that even at zero bias, the electron density within the CdSe is much larger than the hole density within the CdSe due to the presence of Ag (see Appendix D for details). Since the conduction band edge of CdSe is very close to the work function of Ag, if a common Fermi level is assumed across the Ag-Al₂O₃-CdSe heterojunction at equilibrium, extra electrons are transferred from the Ag electrode to the CdSe region. This means that even at zero bias, the CdSe MB is negatively charged and the highest electron density within CdSe is located at the region closest to the Ag electrode. The electron density across the entire heterojunction at zero bias with Al₂O₃ electron affinity at 3.74 eV and Au work function at 5.13 eV is given in the inset of Fig. 1(b). This redistribution of free charges across the heterojunction has a significant impact on I-V characteristics.

3. PHOTOCURRENT CHARACTERISTICS FOR Al₂O₃ THICKNESS OF 3–4 nm

Upon exposure to light, the photocurrent through the MIS junction is observed for oxide thickness of 3 nm, as shown in Fig. 2. To measure the photocurrent, 532 nm pulsed laser light (Spark Antares laser, 80 MHz, pulse width 5–6 ps) is focused onto the sample by a 50x objective via an inverted Olympus microscope, as shown in Fig. 2(a). A wide field optical image of sample 1 with light focused on the CdSe MB-Al₂O₃-Ag heterojunction is displayed in Fig. 2(b). The generated photocurrent is also detected by a lock-in technique (see Appendix A for details). The calibrated peak currents as a function of bias voltage for sample 1 and sample 4 are presented in Figs. 2(c) and 2(d), respectively. In both cases, we observed non-zero photocurrent at zero bias voltage. However, the polarity of photocurrent at zero bias varies from one sample to another. Among all samples, 25% exhibit positive current, 58% have negative current, and 17% have zero current at zero bias. We attribute the change in the polarity of zero bias photocurrent to the variation in the conduction band edge position of the ultrathin Al₂O₃. As described in Figs. 2(e) and 2(f), the magnitude of photocurrent increases with the incident laser power even at zero bias.

The simulated photocurrent curves, by the COMSOL semiconductor module in combination with the wave optics module, with the Al₂O₃ conduction band/Au work function at 3.74 eV/5.13 eV and 3.77 eV/5.1 eV are given in Figs. 2(g) and 2(h), respectively, and yield good agreement with experimental observations (see Appendix B for details). The photocurrent at zero bias is sensitively related to the local conduction band edge of Al₂O₃, which can be related to the immobilized charges in the Al₂O₃ film [18,19] and the local work function of Au.

4. IMPACT OF CARRIER REDISTRIBUTION ON OPTICAL ABSORPTION IN Ag-Al₂O₃-CdSe MB

The simulation model we developed to describe the photocurrents can also be used to evaluate the optical absorption properties of CdSe in this heterojunction. As the net optical absorption coefficient α can be expressed as $\alpha = \alpha_0 (f_v - f_c)$ [20], where $\alpha_0$ is the maximum absorption coefficient at photon energy of $\hbar\omega$, and $f_v$ and $f_c$ are the electron occupancy factors in the valence band and conduction band at the given photon energy, respectively; $f_v$ and $f_c$ follow Fermi–Dirac distribution (see Appendix B for details) and can be obtained directly from the simulation model. The net optical absorption loss is therefore directly related to the difference in electron occupancy factors in the two bands. The band alignment across the heterojunction can directly influence the electron occupancy factors within the semiconductor. Figure 3(a) gives a diagram of the Ag-Al₂O₃-CdSe cross section used in the following simulations. The thicknesses of Al₂O₃ and CdSe are set as 3 nm and 200 nm, respectively. Figures 3(b) and 3(c) show energy diagrams at zero bias for the Ag-Al₂O₃-CdSe heterojunction and a model system, respectively. The model system assumes the metal work function positioned at the middle of the bandgap of CdSe as well as at that of the insulator. As discussed previously, the Ag-Al₂O₃-CdSe heterojunction allows the carriers to be redistributed to maintain the same effective Fermi level across the heterojunction. Figure 3(d) demonstrates the electron density distribution along the red line highlighted in Fig. 3(a) in the Ag-Al₂O₃-CdSe configuration [Fig. 3(b)] upon excitation of light at 532 nm as a function of position and incident power. Figure 3(f) gives the corresponding $f_v - f_c$ at the emission wavelength of 716 nm. As a comparison, we give the electron density distribution [Fig. 3(e)] and $f_v - f_c$ [Fig. 3(g)] of the model system [Fig. 3(c)]. The hole density, $f_v$ and $f_c$ distributions are given in Appendix E.

It is clear from the plots that for the Ag-Al₂O₃-CdSe heterojunction, the electron density of CdSe is significantly enhanced in the region close to Ag due to the accumulation of electrons, which leads to a significantly decreased $f_v - f_c$ value. At the pump power of 1 W, $f_v - f_c = 0.1$ at $z = 0$ in Fig. 3(f), meaning that the absorption coefficient $\alpha$ decreases to only 10% of the original value, while $f_v - f_c = 0.82$ at $z = 0$ in the model system without the transfer of carriers from the metal [Fig. 3(g)]. It is worth noting that the input power required to reach the onset of absorption loss reduction, for example, $f_v - f_c = 0.9$, occurs at input power of $1 \times 10^{-5}$ W for Ag-Al₂O₃-CdSe, which is four orders of magnitude smaller compared to that needed for the model system (occurring at input power of 0.2 W). Our analysis reveals the mechanisms for the pump intensity reduction observed in gain-assisted loss compensation in Ag-Al₂O₃-CdSe hybrid plasmonic waveguides compared to their photonic counterparts [5]. These results suggest that in gain-assisted hybrid plasmonic signal propagation, the presence of metal not only allows light to be confined into a deep subwavelength volume, but the redistribution of carriers across the heterojunction also allows significant modulation to the optical signal with only a fraction of pump energy as required to reach the same level of signal modulation in its photonic counterpart (more details can be found in Appendix F). This unique property can be very useful for all-optical signal modulation and computation [21,22].
5. ABRUPT PHOTOCURRENT INCREASE FOR Al₂O₃ THICKNESS LARGER THAN 4 nm

There is, however, a limitation on how thick the Al₂O₃ is allowed to be for the detection of photocurrent at zero bias. Once the thickness of Al₂O₃ increases to and above 4 nm, not only an energy gap is developed in the photocurrent $I-V$ characteristics, but also a step-like feature is observed in the photocurrent as illustrated in Fig. 4(a). It is more intuitive to convert the peak current into the unit of electron number per 5 ps, as indicated by the right vertical axis of Fig. 4(a). The three curves are obtained from two different experimental setups (see Appendix A). The bottom curve is obtained with the same setup as used for Fig. 2.

Fig. 2. Photocurrent across Ag-Al₂O₃ (3–4 nm)-CdSe MB-Au heterojunction. (a) Diagram of experimental setup. (b) Wide field optical image showing the laser beam incident on the Ag-Al₂O₃-CdSe MB heterojunction (sample 1). (c), (d) Peak photocurrent versus bias voltage on Au electrode of sample 1 and sample 4 at different incident laser powers. (e), (f) Zero bias photocurrent as a function of incident laser power for sample 1 and sample 4, respectively. The error bars indicate the fluctuation of reading from the lock-in amplifier. (g), (h) COMSOL simulation results of the photocurrent density versus bias voltage on Au electrode with Al₂O₃ conduction band at 3.74 eV (g) and 3.77 eV (h) and Au work function at 5.13 eV (g) and 5.1 eV (h) for incident laser power of $1 \times 10^{-4}$ W. The simulated zero bias photocurrents as a function of incident laser power are given as insets.
while the top two curves are obtained using a supercontinuum laser (repetition rate 40 MHz, \( \sim 80 \) ps pulse width), after passing through a 600 ± 20 nm bandpass filter. The curves for sample 2 and sample 6 are offset for clarity. The dashed lines indicate the zero current lines for each curve. From the curves shown in Fig. 4(a), we can see that only a few electrons transport through the heterojunction from each light pulse. This repeatable step-like negative current onset occurs between \(-0.7\) V and \(0.5\) V, too small to be related to the avalanche effect \[23\]. We speculate that the sharp increase in photocurrent is caused by the Coulomb blockade effect commonly observed in single electron tunneling events through a double-barrier heterojunction \[19,24–26\]. In the current case, the tunneling most likely happens through a localized state within the \(\text{Al}_2\text{O}_3\) film \[16\]. It is difficult to predict the Coulomb blockade energy, the energy required for the electron to overcome to tunnel into the localized state within the \(\text{Al}_2\text{O}_3\) layer, from the current simulation model. However, we can estimate the value to be less than half of the energy gap observed in Fig. 4(a) and larger than the thermal energy of \(k_B T\), therefore between \(0.03\) eV and \(0.75\) eV. Figure 4(b) shows the photocurrent as a function of bias voltage on Au for different incident laser powers for sample 6. As shown in the inset of Fig. 4(b), the generated photocurrent after the threshold voltage linearly depends on the incident laser power. The photocurrent also depends on the wavelength of excitation light and is substantially quenched upon exposure to light with a photon energy below the bandgap of CdSe, as shown in Appendix G.

In samples with \(\text{Al}_2\text{O}_3\) of about 5 nm thickness, we have observed the onset voltage spanning from \(-1.5\) V to \(-4.2\) V (see Appendix H). These results suggest that once the \(\text{Al}_2\text{O}_3\) thickness goes beyond 5 nm, carrier redistribution across the MIS junction may be ignored at zero bias. As mentioned previously, the dark current of devices with \(\text{Al}_2\text{O}_3\)
Fig. 4. Photocurrent across Ag-Al2O3 (4–5 nm)-CdSe MB-Au heterojunction. (a) Peak photocurrent as a function of bias voltage on Au for three devices measured with two different laser systems. The black and blue curves are offset for clarity. The dashed lines indicate the zero current lines for each curve. The arrows indicate the step-like onsets of photocurrents. Samples 5 and 6 were excited with a supercontinuum laser (repetition rate 40 MHz, pulse width ~80 ps) after passing a bandpass filter (600 ± 20 nm). The inset shows a wide field optical image of the laser spot focused on the Ag-Al2O3-CdSe MB heterojunction of sample 6, with scale bar of 5 μm. Sample 2 was excited with the Spark Antares laser at 532 nm (80 MHz, pulse width 5–6 ps). The peak input intensities associated with the three curves are 18.3 kW/cm², 2585 kW/cm², and 1495 kW/cm² for samples 6, 5, and 2, respectively. (b) Sample 6 photocurrent as a function of bias voltage on Au for different incident laser powers. After the initial sharp increase, the current plateaus or follows a gradual linear increase. The inset shows the linear dependence of the plateaued current on the input power. The error bars indicate the range of current variation in the gradual linear increase regions.

In conclusion, we have studied the photocurrent generation and transportation through the Ag-Al2O3-CdSe heterojunction with Al2O3 thickness varying from 3 nm to 5 nm. The direct observation of nonzero photocurrents at zero bias voltage in the oxide thickness range of ~3–4 nm suggests a charge redistribution across the heterojunction when CdSe is optically excited, and this charge redistribution leads to a substantially reduced optical absorption coefficient upon excitation of the hybrid plasmonic mode supported by the heterojunction, making gain-assisted plasmonic signal modulation possible at low pump intensity. As the thickness of the oxide increases to ~4–5 nm, the Coulomb blockade effect is observed in the photocurrents, corresponding to a sharp onset in the I-V characteristics. The extremely low dark current and large photocurrent detected upon exposure to light at relatively low voltage make this configuration a promising candidate in photodetection applications where low dark current and low bias voltages are of paramount importance.

APPENDIX A: METHODS

1. Sample Fabrication

The CdSe MBs are grown by a chemical vapor deposition method. In short, an alumina boat containing CdSe powder is placed in the middle of a quartz tube furnace (single zone, Elite Thermal Systems). A Si substrate coated with a thin layer of Au (~5 nm) is then positioned at the downstream side of the tube. The furnace is first pumped down by a mechanical pump for 30 min. Ar gas is then introduced in the tube with a flow rate of 50 sccm (standard cubic centimeters per minute) while the pump is kept on. After the system is stabilized for 30 min, the furnace is heated up to 690°C and kept at this temperature for another 30 min for CdSe nanobelt growth. During growth, the Si sample is kept at a temperature of 500°C–600°C.

To fabricate the device for photocurrent measurements, an Au pad along with alignment marks is first defined on a glass coverslip by photolithography and deposited by thermal evaporation. CdSe MBs are then transferred onto the coverslip. An Au stripe (thickness 50 nm) connecting the Au pad and one side of the CdSe MB is defined by E-beam lithography and deposited by thermal evaporation. An ultrathin layer of Al2O3 is then deposited on the sample by atomic layer deposition. The thickness of the Al2O3 is measured by an ellipsometer or surface profilometer. Last, an Ag electrode (thickness 60 nm) is defined on the other side of the CdSe MB by E-beam lithography and deposited by thermal evaporation.

2. I-V Measurement without Light

The applied voltage is supplied by a square wave function generator, which is applied to the Au electrode of the sample. The current is then fed into the current port of a lock-in amplifier.
Photocurrent Detection
To measure the photocurrent, pulsed lasers (Spark Antares laser at 532 nm, 80 MHz, pulse width 5–6 ps or NKT SC-400 Supercontinuum laser, 40 MHz, pulse width 76–90 ps, 450 nm to 2.5 μm) are used. To extract the small photo-induced current, a lock-in technique is used. The reference frequency of the lock-in amplifier is taken from an optical chopper, which is used to modulate the on-and-off of the light focused on the sample at a low frequency (~367 Hz). The modulated light is directed into an inverted microscope (Olympus) and then focused onto the sample by a long working distance 50× objective (Olympus, NA 0.55). A homemade LabVIEW program is used to control the voltage output/input from a National Instruments multifunction data acquisition (DAQ) device (NI USB-6212). The output voltage is applied to the Au electrode of the sample. The photocurrent is directly fed into the current port of a lock-in amplifier (Signal Recovery 7265) from the Ag electrode. The output voltage from the lock-in amplifier is then read back by the same LabVIEW program via the DAQ device.

4. COMSOL Simulation
The simulation on the I–V characteristic of the devices without and with exposure to light is carried out by the COMSOL Multiphysics simulation package 5.5 using the semiconductor module coupled with the wave optics module. Stationary study is used to calculate the I–V behavior without light and stationary-frequency study is used to calculate the photocurrent upon exposure to light (see Appendix B for more details).

APPENDIX B: COMSOL SIMULATIONS ON DARK CURRENT, PHOTOCURRENT, AND LOSS COMPENSATION
The semiconductor module in COMSOL 5.5 solves for the drift-diffusion equations of current density \(J_n(p)\) in the defined region:

\[
\begin{align*}
J_n(r, t) &= qnμ_n∇E_v + μkB T \left(\frac{n}{N_v}\right)∇n + qnD_{n,ph}∇\ln(T), \\
J_p(r, t) &= qpμ_p∇E_v + μkB T \left(\frac{p}{N_v}\right)∇p + qpD_{p,ph}∇\ln(T),
\end{align*}
\]

where \(n\) is the electron density, \(p\) the hole density, \(q\) the electron charge, \(E_v\) the conduction (valence) band energy level, \(\mu_n(p)\) the electron (hole) mobility, \(\kappa_B\) the Boltzmann constant, \(T\) the lattice temperature, and \(D_{n,ph}\) and \(D_{p,ph}\) the thermal diffusion coefficients for electrons and holes, respectively. \(G\) is the 1/2 order inverse Fermi–Dirac integral, and \(N_v (N_p)\) is the effective density of states for electrons (holes) in the conduction (valence) band.

The solution also satisfies current continuity equations:

\[
\begin{align*}
\frac{∂n}{∂t} &= \frac{1}{q} (\nabla \cdot J_n) - U_n, \\
\frac{∂p}{∂t} &= -\frac{1}{q} (\nabla \cdot J_p) - U_p,
\end{align*}
\]

where \(U_n(p)\) is the net electron (hole) recombination rate from all generation \((G_{n(p)}(t))\) and recombination mechanisms \((R_{n(p)}(t))\).

The Wentzel–Kramers–Brillouin (WKB) tunneling model is used to allow tunneling current across the Ag-Al2O3–CdSe heterojunction to be calculated. Following Ref. [30], the electron normal thermionic current density across the heterojunction is multiplied by a factor of \(1 + \delta_{n,n}\). The extra current factor \(\delta_{n,n}\) is given by double integration along the electrical field line \((d\ell)\) and along the energy axis \((dV_e)\):
and reduced mass of CdSe, respectively. From Eq. (B4), we can see that \( \alpha_0 \) is closely related to the surrounding of CdSe through \( H_{12}^{\alpha} \) and \( g_{\text{red}} \). Appendix F gives two examples of \( \alpha_0 \).

The geometry we used to simulate the Ag-Al\(_2\)O\(_3\) (3 nm)-CdSe (200 nm)-Au heterojunction is given in Fig. 5. A 2D model is used. The top-left boundary is set as the Ag electrode and the top-right boundary is set as the Au electrode. For the simulation of photocurrent, the input and output ports are highlighted by orange lines and red arrows. The input power is distributed on the input port assuming an area with a length indicated by the orange line (800 nm) and out of plane depth of 1 \( \mu \)m. Figure 6 shows how the dark current across the heterojunction depends on the separation of Ag and Au electrodes and the thickness of CdSe MB.

**APPENDIX C: DARK CURRENT-VOLTAGE CHARACTERISTICS OF MORE SAMPLES**

Figure 7 shows the dark \( I-V \) characteristics of three more samples with Al\(_2\)O\(_3\) thickness \( \sim 3-4 \) nm. Sample 5 was characterized with an old setup with larger noise.

**APPENDIX D: HOLE DENSITY DISTRIBUTION AT ZERO BIAS WITHOUT EXPOSURE TO LIGHT**

The hole density distribution at zero bias across the Ag-Al\(_2\)O\(_3\) (3 nm)-CdSe (500 nm)-Au heterostructure is given in Fig. 8. The simulation parameters are the same as those used for the inset of Fig. 1(b).

**APPENDIX E: HOLE DENSITY, \( f_{\nu} \), AND \( f_{\text{c}} \) DISTRIBUTIONS RELATED TO FIG. 3**

The distributions of hole density, \( f_{\nu} \), and \( f_{\text{c}} \) at zero bias as a function of input power are shown in Fig. 9. Figures 9(a), 9(c), and 9(e) are simulated using the same parameters as those used for Fig. 3(d), and Figs. 9(b), 9(d), and 9(f) are simulated using the same parameters as those used for Fig. 3(e).
APPENDIX F: IMPACT OF CHARGE REDISTRIBUTION ACROSS Ag-Al₂O₃-CdSe ON GAIN-ASSISTED HYBRID PLASMONIC MODE PROPAGATION

As discussed in previous work [5], the propagation loss of the hybrid plasmonic mode supported by the Ag-Al₂O₃-CdSe heterostructure [inset of Fig. 10(a)] is significantly reduced at very low pump intensities. The threshold pump intensity, at which the loss compensation is observed, is 36 to 107 times smaller for a hybrid plasmonic waveguide than its photonic counterpart, in which the CdSe nanobelt is released on glass substrate [inset of Fig. 10(b)]. Figure 10 summarizes the results obtained from Ref. [5]. In these plots, the gain $g$ is defined as

$$g \text{ (dB)} = 10 \log \frac{I_{\text{on}} - I_{\text{sp}}}{I_{\text{off}}} = 10 \log \left( \frac{g_{\text{on}}}{g_{\text{sp}}} \right),$$

where $I_{\text{off}}$ indicates the intensity of the output signal without pump light, $I_{\text{sp}}$ the spontaneous emission intensity, and $I_{\text{on}}$ the intensity of the output signal with pump light.

As discussed in the main text, we can assume the absorption coefficient $\alpha$ of CdSe as

$$\alpha = a_0 (f_v - f_c),$$

where $a_0$ is the absorption coefficient without the pump light. As shown in Fig. 3 and Fig. 9, $\alpha$ decreases with the pump power due to the increased $f_v$ and decreased $f_c$ values [in the case of Fig. 9(c), the band bending yields $f_v$ value close to one at low pump power]. Figure 11(a) displays the electric energy density distribution along Ag-Al₂O₃-CdSe (200 nm) when the fundamental hybrid plasmonic mode at 716 nm is excited. The distribution shows that the electric energy density decreases rapidly away from the Ag-Al₂O₃-CdSe interface in the CdSe region, in a trend similar to Fig. 3(d). If we define an average occupancy difference as

$$\Delta f = f_v - f_c = \frac{\int W(r)(f_v - f_c)dr}{\int W(r)dr},$$

where $W(r)$ is the electric energy density along the dashed line highlighted in Fig. 11(a) and the position-dependent $f_v - f_c$ values are taken from Fig. 3(f), we can plot $\Delta f$ as a function of input power [Fig. 11(b)]. As a comparison, the $f_v - f_c$ values from the model system are also given in the same plot, which represents an approximation to the difference in electron occupancy factors for CdSe released on glass.

We can approximate the output intensity of light through a waveguide as $I = I_0 \exp(-\alpha L)$, where $I_0$ is the initial intensity of light at the start of the waveguide and $L$ the length of the
waveguide. Without pump light, $\alpha = \alpha_0$. With pump light, $\alpha = \alpha_0(f_v - f_c)$. So, $I_{\text{off}} = I_0 \exp(-\alpha_0 L)$, and $I_{\text{on}} = I_0 \exp[-\alpha_0(f_v - f_c)d - \alpha_0(L - d)] = I_{\text{off}} \exp[\alpha_0 d(1 - f_v + f_c)]$. Here $d$ is the distance on the waveguide that the pump light is on. If $I_{\text{sp}}$ can be ignored when the intensity of the stimulated emission is much larger than that of the spontaneous emission, $g(\text{dB})$ can be further expressed as

$$g(\text{dB}) = 4.3 \, \alpha_0 d(1 - f_v + f_c). \quad (F1)$$

For a hybrid plasmonic waveguide, $\alpha_0$ is usually much larger than that of the photonic waveguides. For the two waveguides shown in Fig. 10, the measured $\alpha_0$ are 6230 cm$^{-1}$ and 1260 cm$^{-1}$. From Eq. (F1), it is clear that the measured $g$ value is closely related to $\alpha_0$ and $f_v - f_c$ for a given $d$ (2.5 $\mu$m in Ref. [5]). For the photonic waveguide, for $g = 0.5$, $f_v - f_c = 0.63$. For the hybrid plasmonic waveguide of $g = 0.5$, $f_v - f_c = 0.92$. This implies that loss compensation can be observed at much lower input power for a hybrid plasmonic waveguide, as shown in Fig. 11(b). The dashed lines guide the eyes to the threshold input power [$g(\text{dB}) = 0$], at which the dashed lines intercept with the top $x$ axis. These results are consistent with what were observed in Fig. 10.

**APPENDIX G: SPECTRAL DEPENDENCE OF THE PHOTOCURRENT**

The photocurrent of sample 6 was measured at three different wavelengths, as shown in Fig. 12. The sample was excited with a supercontinuum laser (repetition rate 40 MHz, pulse width $\sim 80$ ps) after passing through bandpass filters ($500 \pm 20$ nm, $600 \pm 20$ nm, and $800 \pm 5$ nm). It is shown clearly that the photocurrent with 800 nm excitation is substantially smaller than those with 500 nm and 600 nm excitations.

**APPENDIX H: PHOTOCURRENT I-V CHARACTERISTICS OF Ag-Al$_2$O$_3$ (5 nm)-CdSe MB-Au**

Figure 13 shows the experimental results of peak photocurrent as a function of bias voltage for four samples with Al$_2$O$_3$ thickness $\sim 5$ nm. The samples were excited with a Spark Antares laser at 532 nm, with a repetition rate 80 MHz and pulse width
5–6 ps. The bias voltage for the negative photocurrent onset is in the range of −1.5 V to −4.2 V.

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**Data Availability.** Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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