Conductive-bridge interlayer contacts for two-dimensional optoelectronic devices

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Abstract

Photodiodes based on two-dimensional semiconductors are of potential use in the development of optoelectronic devices, but their photovoltaic efficiency is limited by strong Fermi level pinning at metal–semiconductor contacts. Typical metal–interlayer–semiconductor contacts can address this issue, but can also lead to an increase in series resistance. Here, we report a conductive-bridge interlayer contact that offers both Fermi level depinning and low resistance. We create an oxide interlayer that decouples the metal and semiconductor, while embedded gold nanoclusters in the interlayer act as conductive paths that facilitate efficient charge transport. Using these contacts, we fabricate a tungsten disulfide (WS_2) photodiode with a photoresponsivity of 0.29 A W^{-1}, linear dynamic range of 122 dB, and power conversion efficiency of 9.9%. Our approach also provides a platform for probing photocarrier dynamics, and we find that contact recombination substantially impacts photovoltaic performance. In addition, we illustrate the potential of using photodiodes with these conductive-bridge interlayer contacts as full-colour 2D and 3D imagers.

Main

Photodiodes are widely used for light detection, harvesting, and emitting applications[1]. By scaling the devices to nanometre scales, it is possible to improve their operational speed, power efficiency, and integration density[2]. Layered two-dimensional (2D) transition metal dichalcogenides are expected to be ideal candidates for the absorber materials in miniaturized photodiodes because of their atomically uniform thickness and strong light–matter interactions[3–5]. However, many photodiodes based on 2D transition metal dichalcogenides suffer from low photovoltaic efficiency (power conversion efficiencies are typically less than 5%)[6,7] and a limited operating range, primarily due to inefficient charge collection at the electrical contacts.

One of the fundamental parameters that determine charge-collection efficiency is the Schottky barrier height at metal–semiconductor contacts[1,8]. Ideally, by careful selection of the contact metal, the Schottky barrier for photogenerated carriers (electrons and holes) can be made sufficiently low to facilitate charge transport in and out of an optoelectronic device. However, in reality, an ideal Schottky barrier is not formed due to interface phenomena at metal–2D semiconductor junctions, such as metal-induced gap states (MIGS) and defect-induced gap states (DIGS) (Fig. 1a(i)), leading to strong Fermi level pinning (FLP)[8–11]. This pinning, regardless of the metal work function (WF), usually results in a large Schottky barrier at the contacts, which severely hampers charge collection and consequently the performance of 2D photovoltaic devices[12,13]. Although various approaches to mitigate FLP have been proposed, such as creating van der Waals (vdW) contacts via metal transfer[8,12] or using 2D vdW metal contacts[9], these techniques have a limited choice of transferrable metals and transfer-process-related issues that can increase the cost and complexity of device fabrication.

An alternative method to avoid this problem is to create a metal–interlayer–semiconductor

(MIS) contact[14–16] (Fig. 1a(ii)). Introducing an interlayer to decouple the interactions between the metal and semiconductor can potentially prevent FLP, thus enabling precise control over the Schottky barrier and thus the charge-collection efficiency of optoelectronic devices. But so far the MIS approach has yielded limited success, primarily owing to the extreme sensitivity of tunnelling to the interlayer thickness: even a sub-nanometre increase in the interlayer thickness (and in the tunnelling width) causes an undesirable high series resistance[17,18]. This high series resistance is problematic as it severely limits the dynamic sensing range and photovoltaic performance of the device[17], especially under intense illumination. In addition, depositing an extremely thin interlayer on 2D surfaces with high uniformity is technically challenging.

In this Article, we report a method, which we term conductive-bridge interlayer contact (CBIC) (Fig. 1a(iii)), to form low-resistance and pinning-free interlayer contacts. The method incorporates gold atoms into an oxide interlayer, creating conductive pathways within the interlayer. These pathways can substantially reduce the series resistance induced by the interlayer, thus providing low-resistance contacts without the trade-off problem typically encountered in conventional MIS contacts[17,18]. We use CBIC contacts to create a tungsten disulfide (WS_2) Schottky photodiode that exhibits good photovoltaic performance over a wide operating dynamic range. The CBIC is also a useful platform to explore photocarrier dynamics, unveiling that contact recombination associated with the Schottky barrier greatly impacts the performance of 2D photovoltaics. Finally, we demonstrate full-colour 2D and 3D imaging using prototype CBIC-integrated image sensors.

Architecture design of the CBIC

Figure 1b shows a schematic of pre-patterned CBIC fabrication via sequential sputtering. First, a low-WF metal (Al, Ti, Cr, Cu; ~20 nm thick) and Au (1 nm nominal thickness) were sequentially deposited on a SiO_2 substrate without breaking vacuum. Note that the addition of a trace amount of Au during the sputtering process is crucial in this design (Supplementary Figs. 1 and 2), as further discussed later. Afterwards, the as-deposited metal layer was exposed to ambient atmosphere to form an approximately 5 nm thick oxidization layer on the surface of the metal, as confirmed by X-ray photoelectron spectroscopy (XPS) measurements (Supplementary Fig. 3). This oxide layer serves as a physical spacer that decouples the metal–semiconductor interaction, thereby suppressing FLP[16]. Finally, the resulting CBIC electrode was used to fabricate a vertical Schottky diode structure, in which multilayer WS_2 (~50 nm) was used as the absorber, and the CBIC and chlorine-doped $SnSe_2$ (Cl–$SnSe_2$, ~10 nm) were used as bottom and top electrodes, respectively. The Cl–$SnSe_2$ employed here not only offers ideal band alignment with multilayer WS_2 but also serves as a transparent conductive electrode that enables top illumination operation. The material properties of Cl–$SnSe_2$ are provided in Supplementary Figs. 4–6. Figure 1c presents a schematic illustration and optical image of a complete device (see Supplementary Fig. 7 for AFM measurements of the sample thicknesses).

We first performed electrical characterization of a CBIC made from Al, hereafter referred to as CBIC–Al, using conductive atomic force microscopy (CAFM). For simplicity, we mostly focus on CBIC–Al as an example in our discussions. Two different samples were prepared and measured for comparison, one with the addition of elemental Au and the other without, as illustrated in Fig. 1d,e. The currents recorded as the CAFM tip (applied constant bias of 1 V) scanned across the samples are presented in Fig. 1f,g. A sharp contrast was found, where the CBIC–Al with Au structure shows multiple conductive spots across the entire sample, with an abrupt current increase (Fig. 1f; see also Supplementary Fig. 8 for basic information such as size and density), whereas CBIC–Al without Au shows no noticeable current change (Fig. 1g).

This observed distinction clearly suggests the existence of electrical conduction paths within the oxide interlayer of CBIC–Al with Au. Cross-sectional scanning transmission electron microscopy (STEM) measurement followed to directly probe the conducting path in a complete device of CBIC–Al/WS_2/Cl–$SnSe_2$ (the same device as in Fig. 1c). A typical cross-sectional STEM image clearly revealed multiple Au nanoclusters (approximately 2 nm in diameter) uniformly embedded within the amorphous AlO_x interlayer (Fig. 1h,i), which is further supported by an energy-dispersive X-ray spectroscopy (EDS) mapping image (Supplementary Fig. 9). Most probably, electrical conduction across the interlayer primarily occurs via the Au-induced gap states, as evidenced by XPS measurement and density functional theory (DFT) calculations (Supplementary Fig. 10). The coupling between elemental Au atoms and the AlO_x layer can induce finite gap states within the bandgap. Consequently, unoccupied gap states near the Fermi level may contribute to efficient charge-carrier transport through the interlayer, which could be responsible for the observed enhanced conductance in CBIC–Al. Note that this operational mechanism differs markedly from that of a conventional MIS contact, which strongly relies on tunnelling transport (Supplementary Fig. 11). It is also observed that the WS_2/Cl–$SnSe_2$ interface features an atomically clean contact without apparent disorder (Supplementary Fig. 12). Taken together, the data in Fig. 1 demonstrate that the incorporation of a small amount of Au into the interlayer establishes robust electrical conduction pathways that improve the conductivity of the CBIC. As we show below, these conductive pathways effectively reduce contact resistance in the CBIC, which in turn facilitates charge extraction in 2D optoelectronic devices.

Characterizations of the CBIC

One intriguing feature of the CBIC method is its versatility with various metals. This allows for the tunability of work functions, which is crucial for the rational design and fabrication of electronics. To demonstrate that the WF of the CBIC can be tailored simply by changing the metal type, we characterized CBICs made from different metals with varying WFs (Ti, Cr, and Cu) following the identical process as with CBIC–Al. Kelvin probe force microscopy (KPFM) was used to evaluate the WF distributions of the CBICs; all KPFM images exhibit highly uniform features over a large area (Fig. 2a), indicating electrical homogeneity of the CBICs. The measured WF values of the CBICs were essentially similar to those of pristine metal films[8,19], as shown in Fig. 2b. The mean WFs were found to be 3.92 ± 0.04 eV for CBIC–Al, 4.12 ± 0.05 eV for CBIC–Ti, 4.36 ± 0.02 eV for CBIC–Cr, and 4.74 ± 0.04 eV for CBIC–Cu. This result suggests that the WF of the CBIC is largely dictated by the underlying deposited metal, highlighting the versatility of this method. Slight deviations from the intrinsic WFs are likely caused by possible interfacial alloying between metals[20] or interfacial dipoles at the metal–semiconductor junctions[21]. As shown in topographical AFM images, the surface roughness of the CBICs was negligibly different, with root mean square (RMS) values ranging from 0.56 to 0.69 nm (Supplementary Fig. 13). We additionally performed XPS and Raman spectroscopy to investigate the chemical states of the CBIC–Al/WS_2 interface. Here, we intentionally prepared the sample using thin WS_2 (~2.86 nm, four layers) to ensure a reliable evaluation of the interfacial chemistry (Supplementary Fig. 14). The XPS spectrum obtained from CBIC–Al/WS_2 (Fig. 2c) is almost identical to that of pristine WS_2. Also, non-stoichiometric W_xS_y peaks were not observed, suggesting that the WS_2 layer remained largely intact without any chemical perturbation. The Raman spectrum (Supplementary Fig. 14d) also shows no peaks associated with non-stoichiometric W_xS_y, further confirming that CBIC–Al in contact with WS_2 does not introduce any chemical reactions, defects, or strain at the interface or within the WS_2.

With minimized interface disorders on the 2D semiconductor, the CBIC can effectively suppress FLP originating from DIGS[22]. Additionally, decoupling the metal–2D semiconductor interaction by the interlayer can greatly suppress the MIGS that strongly pin the Fermi level[10]. To demonstrate this, we fabricated top-gated WS_2 transistors with CBICs as source and drain electrodes and investigated their contact properties using temperature-dependent charge transport measurements (see Methods for details about the device fabrication). Figure 2d shows a schematic (upper) and optical image (lower) of a top-gated WS_2 transistor fabricated with CBICs. As shown in Fig. 2e–h, all devices with CBICs exhibited similar n-type characteristics, regardless of the CBIC type (room temperature I–V characteristics of the same devices are shown in Supplementary Fig. 15). However, both CBIC–Cr and CBIC–Cu devices exhibited a much stronger temperature dependence of current compared to CBIC–Al and CBIC–Ti devices, indicating high Schottky barriers at the CBIC–Cr and CBIC–Cu contacts. In particular, the CBIC–Cu device with a relatively high WF exhibited an ambipolar behaviour, with nearly symmetric p- and n-branches (see Supplementary Fig. 15d for a log-scale plot). The reproducibility of this behaviour was also verified with another five CBIC–Cu devices all fabricated with similar device geometries (Supplementary Fig. 16). A statistical analysis of carrier polarity, threshold voltage (V_T), and electron field-effect mobility (μ_e) for different CBIC types of the aforementioned devices is shown in Fig. 2i. Here, the carrier polarity was characterized by the current ratio of I_{5V} (I_{DS} at V_{GS} = 5 V) to I_{-5V} (I_{DS} at V_{GS} = –5 V), which represents the majority carrier type. As the WF of the CBIC increased, a clear transition in the majority charge carrier was observed along with an increased V_T. The observed trend of WF dependence can be attributed to a highly tunable Schottky barrier height due to Fermi level depinning, which is consistent with previous studies[8]. Significant degradation in μ_e was observed in both CBIC–Cr and CBIC–Cu devices, due to their large electron barriers[8,23].

To gain insight into the observed distinctions resulting from different CBIC types, we further estimated the Schottky barrier height (SBH) using temperature-dependent I–V measurements[24]. A detailed description of SBH extraction can be found in Supplementary Fig. 17. Plots of effective barrier Φ_B versus V_{GS} for the different CBICs are presented in Fig. 2j along with the 2D Cl–$SnSe_2$ contact exploited in this work. The SBH (Φ_{SB}) for each contact was determined under the flat band condition ($V_{GS} = V_{FB}$) and plotted against the metal WF (Fig. 2k). For the CBICs, the SBH value was highly dependent on the CBIC work functions. The pinning factor ($S = |d\Phi_{SB}/dW|$) was calculated to be 1.17, largely following the Schottky–Mott rule ($S = 1$), which clearly indicates the Fermi level depinning effect. This contrasts with direct metallization samples, all fabricated by electron beam evaporation (Supplementary Fig. 17). The absence of pinning can be ascribed to an effective suppression of MIGS[25] and/or DIGS[26], provided by minimized metal–semiconductor interaction. We further confirmed that this depinning effect is also valid for ultrathin WS_2 (monolayer and bilayer) devices, as shown in Supplementary Fig. 18. Additionally, for the device with Cl–$SnSe_2$, we observed p-type behaviour, with a hole SBH of 317 meV, consistent with the Schottky−Mott rule and its field-effect transistor characteristics (Supplementary Fig. 19). The above results demonstrate the versatility and pinning-free characteristics of the CBIC, which are key prerequisites for efficient 2D optoelectronic devices. Below, by fabricating 2D Schottky photodiodes, we reveal unique advantages and the potential of the CBIC.

CBIC-based WS_2 diode as a photodetector

The CBIC can be employed to fabricate a 2D photodetector that exhibits a robust photodetection ability, including a large linear dynamic range (LDR), high responsivity, and fast photoresponse. Figure 3a illustrates a schematic of our designed vertical WS_2 Schottky diode (top) and the energy-level alignment of the materials (bottom) considered in this study. In principle, the WF of the electron-selective (hole-selective) contact should be aligned with the conduction (valence) band of the absorber layer to facilitate efficient extraction of photogenerated carriers. We first explored this picture experimentally by measuring a device fabricated with palladium metal (Pd) as hole-selective contacts. The device showed poor rectifying behaviour (Fig. 3b) due to severe FLP near the conduction band edge of WS_2. To solve this issue, we adopted 2D Cl–$SnSe_2$ metal as a pinning-free electrode, as previously reported[9]. As shown in Fig. 3c,d, the Cl–$SnSe_2$-contacted devices exhibited clear rectification behaviour, with high on/off current ratios exceeding 10^4. To ensure satisfactory performance, we applied Cl–$SnSe_2$ as hole-selective contacts throughout the remainder of this study unless otherwise specified.

Compared with MIS devices, CBIC devices generally exhibited better rectification, largely due to an increased on-current (Fig. 3e). This observation suggests that electrical conduction through the conductive paths of the CBIC is the primary factor responsible for the improved charge injection and the resulting higher on-current, as compared to MIS devices. No significant difference in rectification was observed between CBIC–Ti and MIS–Ti devices, probably owing to a sufficiently low interfacial barrier of the TiO_x interlayer (Supplementary Fig. 20). Under visible light illumination (532 nm), the Schottky diode with CBIC–Al demonstrated a clear photoresponse, in which the photocurrent (I_{ph}) linearly increased with light intensity, exhibiting light-sensing capability (Fig. 3f). A very broad photocurrent plateau was also observed at a bias ranging from 0 V to –1.5 V, which is crucial for stable photodetector operation[27]. The spectral responsivity of the same device was also measured, revealing that the photoresponse follows the absorption spectrum of the WS_2/Cl–$SnSe_2$ heterostructure (Supplementary Fig. 21).

Besides light-sensing capability, a large dynamic range with excellent linearity is of great importance for practical applications and successful commercialization of 2D photodetectors. The majority of previous investigations have mainly focused on reporting the responsivity of devices at a specific illumination condition—note that a high photoconductive gain of a device under specific light intensities does not necessarily indicate superior light detection capability due to photogating effects[28,29]—while relatively few studies have attempted to evaluate the device LDR, defined as the range of linear responsivity under different light intensities[30,31]. Moreover, even less attention has been paid to device performance under intense illumination above 100 mW cm^{-2}, which indeed resembles the operating conditions of real applications. Figure 3g plots the photocurrent at $V = 0$ V (photovoltaic mode) as a function of incident light power. Our device exhibited an excellent linearity from the lowest detectable limit, 4.3 μW cm^{-2}, to 5.4 W cm^{-2}, corresponding to an LDR of 122 dB, which outperforms previously reported 2D photodetectors (Supplementary Table 1) at a similar wavelength and achieves comparable performance to commercial photodetectors (Si or InGaAs)[32,33]. Most importantly, evidence for the effectiveness of the CBIC in reducing the series resistance of the contacts is shown in Fig. 3h. Compared with MIS devices, the CBIC devices generally featured a higher LDR (100–126 dB, Supplementary Figs. 22 and 23). Owing to the substantial reduction in series resistance, photodetection at higher light levels becomes possible, thereby extending the photodetection range and leading to an enhanced LDR (further discussion is provided in Supplementary Fig. 24). We also observed a nearly consistent responsivity (R) of ~0.29 A W^{-1} and a specific detectivity (D^*) of ~2.11×10^{10} cm $Hz^{1/2}$ W^{-1} over a wide photodetection range spanning six orders of magnitude (Fig. 3i, and see Supplementary Fig. 25 for the frequency-dependent noise power spectral density). Frequency response measurement (Fig. 3j) revealed a fast response of the device with a 3 dB bandwidth of > 100 kHz, which is beyond the bandwidth limit of our instrument set-up (Supplementary Fig. 26). Simultaneously, we obtained a fast response time of less than 5 μs (Fig. 3j, inset), agreeing well with the measured –3 dB cutoff frequency. We believe that the response speed can be further improved by reducing the carrier transit time and minimizing device resistance and capacitance, as reported previously[2,34]. Photoswitching behaviours at modulated frequencies (100 Hz and 1 kHz) are shown in Supplementary Fig. 27. Additionally, the CBIC approach can also be extended to other 2D semiconductors, including ReS_2, MoS_2, and WSe_2 (Supplementary Fig. 28). Similar to the WS_2 devices, all these devices exhibited a clear photoresponse, demonstrating the universality of the CBIC method.

CBIC-based WS_2 diode as a photovoltaic cell

In addition to photodetection, CBIC-based WS_2 Schottky diodes can be operated as photovoltaic cells, as illustrated in Fig. 4a. Figure 4b shows an optical image of a CBIC–Al device along with corresponding spatial-resolution photocurrent mapping (at V = 0 V), measured with a 532 nm laser spot scanned over the device. The strongest photocurrent was observed to occur only in the heterojunction, suggesting that the photoresponse of our device originates from the photovoltaic effect. Figure 4c compares representative J–V characteristics between CBIC–Al and control MIS–Al devices, measured under 532 nm laser with a power of 44 mW cm^{-2}. Compared to the MIS–Al device, the CBIC–Al device exhibited much improved performance with a fill factor (FF) of 54% and an open-circuit voltage (V_{OC}) of ~0.66 V, yielding a high power conversion efficiency (PCE) of 9.91%; this value is notably comparable to or higher than previous 2D semiconductor Schottky or p–n photodiodes (Supplementary Table 2). We further note that devices with other CBICs also demonstrated similarly enhanced performance compared to the control MIS device (see Supplementary Figs. 29 and 30 for more details on the CBIC and MIS devices, respectively). The noticeable enhancement in PCE primarily arises from a substantially increased FF, as follows: the relatively high conductivity of the CBIC can substantially reduce electron extraction resistance to enhance the FF. Long-term stability was also assessed, as shown in Supplementary Fig. 31. After one month of storage in ambient environments, the CBIC–Al device did not exhibit such a high PCE (less than 1%) due to interfacial oxidation. Nevertheless, by employing subsequent encapsulation or other measure, high photovoltaic performance with air stability can potentially be achieved in the future.

With the capability to control the WF, our CBIC approach offers a versatile platform for both fundamental photophysics studies and high-performance photovoltaic devices. Although significant progress has been made in other photovoltaic technologies[16] (such as silicon, perovskite, and organic photovoltaics) through modulation of the WF in electrodes, this aspect has not yet been investigated in 2D electronics due to the lack of a suitable contact architecture that simultaneously offers versatile WF control and a pinning-free nature. To obtain a statistical variation of device performance, we measured 28 devices all fabricated with similar geometries; the photovoltaic parameters of the corresponding devices are summarized in Fig. 4d. A clear and distinguishable trend shows that both V_{OC} and FF strongly decreased with increasing WF of the CBIC. For example, when the WF increased from ~3.9 eV to ~4.7 eV, the average V_{OC} (FF) decreased from 0.7 V (60%) to 0.4 V (30%). This WF dependency was more pronounced at high illumination intensities, which can be confirmed by light-intensity-dependent measurements. In Fig. 4e, for two contrasting contacts (namely, CBIC–Al and –Cu) we plot V_{OC} as a function of light intensity. For the CBIC–Cu device with a relatively high WF (~4.7 eV), V_{OC} showed saturation behaviour with increasing light intensity. In contrast, for the CBIC–Al device with a low WF (~3.9 eV), V_{OC} increased continuously with light intensity, with a constant ideality factor (n) of ~1.27, indicative of Shockley–Read–Hall recombination[35]. In addition, the FF degradation trends were identical but to different extents, where the trend correlated with the magnitude of the WF of the CBIC electrode (Fig. 4f).

Since both V_{OC} and FF are closely associated with nonradiative recombination[17,36], the observed trend can be explained by the effect of contact recombination resulting from the Schottky barrier at contacts[36,37], as illustrated by the simulated band diagrams in Fig. 4g,h. For the CBIC–Cu device under light illumination (Fig. 4g), the high SBH ($\Phi_{SB} \approx 840$ meV) could block some of the photogenerated electrons, increasing the probability of non-collected carriers undergoing carrier recombination near the contacts. This reduces the quasi-Fermi-level splitting and charge-collection efficiency, thereby limiting both V_{OC} and FF. In contrast, for the CBIC–Al device under light illumination (Fig. 4h), photogenerated electrons can readily overcome the contact barrier ($\Phi_{SB} \approx 20$ meV) and reach the collecting electrode before carrier recombination occurs, which leads to enhancement in both V_{OC} and FF (further details are given in Supplementary Fig. 32). To corroborate this interpretation, we performed numerical simulations using a drift-diffusion model (details are provided in Supplementary Table 3). For a reliable comparison, we used identical material parameters except for the WF of the electrode (3.92 eV versus 4.74 eV). As shown in Fig. 4e, our simulated data quantitatively matched the experimental data for V_{OC}. This result is also in accordance with the calculated contact recombination current (Supplementary Fig. 33), where contact recombination predominantly contributed to the total recombination current in the CBIC–Cu device with a high WF. Taken together, these results highlight the importance of contact recombination reduction in enhancing the efficiency of 2D photovoltaics.

Full-colour 3D imaging demonstration

Fast response speed and high detectivity to visible light make our CBIC-integrated photodiodes promising for full-colour imaging applications. To demonstrate this, we employed a CBIC–Al-integrated WS_2 photodiode as a full-colour (RGB, red/green/blue) image sensor to build a 3D imaging system as an alternative to commercially available imagers. 3D integral imaging, a promising technique for 3D visualization, captures depth information by using an array of microlenses that allows for the recording of multiple perspectives of a scene to create a realistic 3D image[38,39]. This approach eliminates the need for a specialized viewing device for depth perception, which is advantageous compared to other 3D imaging methods. Figure 5a illustrates the principle of the 3D integral imaging system, involving two major processes: a pickup process and a reconstruction process. In the pickup process, an image sensor captures light reflected off an object or objects from different perspectives, resulting in sub-images referred to as elemental images. Then in the reconstruction process, these elemental images are reconstructed into a 3D image using a lens array.

Our full-colour 3D integral imaging set-up is schematically illustrated in Fig. 5b. While previous imaging demonstrations have mainly relied on transmissive-type systems[30,39–41], often constrained by limited photoresponsivity in low-light conditions, we employ a reflective-type imaging system where a CBIC-integrated image sensor captures reflected light from objects illuminated by a light source, which is crucial for practical applications. For this study, the object was illuminated with white light at an intensity of ~1,000 lux, corresponding to office illumination conditions[42]. The light reflected from the object passes through the camera lens and is irradiated onto the CBIC–Al image sensor. The photocurrent of the device is then recorded with an external source meter. To obtain full-colour images, a series of scanning operations were conducted at various wavelengths (650 nm for red, 550 nm for green, and 450 nm for blue) using an optical bandpass filter. Because the proposed CBIC-integrated 2D devices have a broadband photovoltaic response and excellent linearity of photoresponsivity in low-light conditions, fast, full-colour, and high-quality image processing becomes feasible with the reflective-type imaging system. A photograph of the imaging system is shown in Supplementary Fig. 34, and further information on the experiment is provided in Supplementary Table 4, and Methods.

Figure 5c displays the resulting spatial photocurrent images at different wavelengths and their corresponding full-colour images, taken with both conventional MIS–Al and CBIC–Al devices. These full-colour images were obtained by compiling normalized photocurrents from each of the RGB colour channels, as elaborated in Supplementary Fig. 35. In the case of the MIS–Al device, the weak photoresponse, regardless of wavelength, makes it difficult to recognize the object and its characteristics. In contrast, the CBIC–Al device allows for clear recognition of the target object, featuring the letters *P*, *I*, and *N* with enhanced contrast. This improved image quality can be attributed to the lower series resistance of the CBIC, which results in increased photoresponse and an extended light detection limit. The current distribution of the CBIC–Al device (Fig. 5d) also reveals a clear distinction between the photocurrent (I_{photo}) and the dark current (I_{dark}), in contrast to the MIS–Al device.

Figure 5e presents 3 × 3 elemental images and an optically reconstructed 3D image acquired with the CBIC–Al device. The successful execution of 3D image sensing through 3D reconstruction was confirmed using a lens array (see Supplementary Fig. 36 and Supplementary Video 1 for more details). Clearly, the resultant 3D image features noticeable depth and a realistic 3D effect when viewed from various angles, demonstrating the practical applicability of CBIC photodiodes in imaging applications. Parallax, defined as the variation in the perceived position of an object when observed from distinct lines of sight, is closely related to the number of sub-lenses used in the image capture and reconstruction phases; here, further improvements to the 3D imaging quality can be achieved by increasing the number of elemental lenses in the lens array. Additionally, a similar imaging system was employed to demonstrate the full-colour 2D imaging capabilities of the CBIC device (Supplementary Fig. 37). For practical industrial applications, reliable performance and scalability of CBIC devices should be further optimized; first, the scalable synthesis of high-quality 2D materials and their heterostructures needs to be addressed. We believe our CBIC approach can benefit from recent advancements in large-scale 2D material synthesis[43] to construct large-scale photosensor arrays for future full-colour imaging.

Conclusion

We have reported a conductive-bridge interlayer method for creating low-resistance and pinning-free contacts. The contacts consist of an oxide interlayer embedded with gold nanoclusters, which creates conductive pathways that enable efficient charge transport across the interlayer. WS_2 Schottky photodiodes with CBIC contacts showed a good photovoltaic performance over a wide dynamic operating range. We also apply this technique to investigate the photocarrier dynamics at the contacts, revealing how the Schottky barrier at the contacts determines the photovoltaic performance. Finally, we demonstrated full-colour 2D and 3D imaging using CBIC-integrated photodiodes. Our study achieves low-resistance and pinning-free contacts for efficient optoelectronic devices, and establishes an approach for making metal–interlayer–semiconductor contacts that could be generally applied to various metals with high work functions.

Methods

CBIC fabrication. Electron-beam lithography (EBL) was used to pattern the CBIC electrodes. After developing the resist, low-WF metal (20 nm; Al, Ti, Cr, Cu) and Au (nominal 1 nm) were sequentially deposited using sputtering without breaking vacuum. Subsequently, to obtain a dense native oxide interlayer, the samples were naturally oxidized in ambient atmosphere for more than 3 h. Finally, the samples were lifted off with acetone and rinsed in isopropanol.

Synthesis of Cl–$SnSe_2$. Cl–$SnSe_2$ ($SnSe_{1.98}Cl_{0.02}$) was synthesized via chemical vapour transport, following a procedure described elsewhere[44]. In brief, powders of Sn, Se, and $SnCl_2$ were mixed in stoichiometric proportions and loaded into a furnace. The reaction [$(2-x)Sn + xSnCl_2 + 2(2-x)Se \rightarrow 2SnSe_{2-x}Cl_x$] was performed via a two-step process: the mixture was heated to 250 °C for 24 h and then annealed at 400 °C for 48 h. Finally, the sample was melted at 750 °C for 24 h and slowly cooled to room temperature.

Device fabrication and measurements. For photodiode fabrication, we first loaded a pre-patterned CBIC onto a SiO_2 substrate. Using poly(dimethylsiloxane) (PDMS) films, WS_2 (40–50 nm) and Cl–$SnSe_2$ (5–10 nm) were sequentially transferred onto the pre-patterned CBIC using a micromanipulator. Subsequently, Ti/Au layers (1 nm/30 nm) were patterned and deposited using sputtering to serve as contact pads for electrical measurements. Photocurrent was measured under ambient conditions using an Agilent 4156C parameter analyser both in the dark and under illumination by diode lasers (Thorlabs). The light intensity was adjusted using neutral density filters in the beam path and calibrated with a commercial Si photodetector. The LDR was calculated as follows: $\mathrm{LDR} = 20\ \log(P_{\mathrm{high}}/P_{\mathrm{low}})$, where P_{high} and P_{low} represent the highest and lowest light power at which the measured photocurrent of the device exhibits a linear relation with the light power. Transient response and photoelectrical bandwidth measurements were made with a high-speed oscilloscope (Tektronix TDS2024) coupled with a function generator (Tektronix AFG31000).

Fabrication of the top-gated CBIC-contacted transistors was similar to that of the photodiode devices. WS_2 (40–50 nm), h-BN (15–20 nm), and graphite (10–20 nm) were exfoliated onto PDMS films and subsequently transferred onto a pre-patterned CBIC, which served as both source and drain electrodes. Finally, gate metallization was performed similarly as for the photodiode devices. For the evaporated contact devices, a Ti (1 nm)/Au (10 nm) local bottom gate was first prepared on a SiO_2 substrate as the back-gate electrode. Afterward, h-BN (15–20 nm) and multilayer WS_2 flakes (40–50 nm) were transferred on top of the pre-deposited gate electrodes. Finally, the metal electrodes (20-nm-thick metal capped with 50-nm-thick Au to prevent reaction between metal and ambient oxygen and water) were defined using electron beam lithography followed by electron beam evaporation. All electrical characterizations of the transistors were performed using a parameter analyser (4200-SCS, Keithley) in vacuum ($\approx 10^{-3}$ Torr) at 170–350 K. No annealing was performed to avoid chemical interactions between the materials and surface oxidation.

Material characterization. The spatial distribution of the conductive paths in the CBIC was characterized via CAFM (NX-100, Park Systems). Current data were collected under a constant bias of 1 V applied to the Pt-coated tip while keeping the CBIC grounded. AFM and KPFM measurements were simultaneously performed with an NX-100 (Park Systems) in ambient environment. To obtain the work function of the electrodes, the KPFM tip (Cr/Au-coated Si tip) was calibrated using highly oriented pyrolytic graphite (HOPG), and differences in the contact potential values between the tip and the electrode were measured. Cross-sectional STEM images and EDS data were obtained using an aberration-corrected TEM (JEM-ARM200F, JEOL) operated at 200 kV. Chemical identification of the samples was performed via XPS (PHI, VersaProbe) with a micro-focused and monochromatic Al Kα (1,486.6 eV) excitation source. Raman spectra were obtained using a micro-Raman spectrometer (LabRAM HR Evolution, Horiba Jovin-Yvon) with a laser excitation wavelength of 532 nm.

Drift-diffusion simulations. Simulations of the photovoltaic devices were conducted using SCAPS-1D software[45]. SCAPS is an open-source software and can be obtained from https://users.elis.ugent.be/ELISgroups/solar/projects/scaps. The simulation parameters used in this study are presented in Supplementary Table 3.

3D integral imaging. The size of the object used for the 3D imaging application was approximately 15 mm × 15 mm. The scanning system consisted of an x-y planar linear stage (VT-800, PI), a camera lens, a white light-emitting diode, and measurement units (Keysight Technologies, B2901A). The camera lens was used to capture and focus the reflective image on the photodiode. The source measurement unit measured the photocurrents of the photodiode. All elemental images were acquired in reflection mode by illuminating the object with a white-light source emitting approximately 1,000 lux. Finally, the full-colour 3D integral image was obtained through the reconstruction process. During this process, the lens array had a focal length of 22 mm and a sub-lens size of 19 mm. The display panel was a Lenovo Yoga 920 (3840(H) × 2160(V), 13.9” display with a pixel density of 317 PPI).

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

Code availability

Any codes used in this study are available from the corresponding authors upon request.

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Author contributions

J.J., J.P.H., and S.-J.K. contributed equally. D.K.H. and J.J. conceived the idea and designed the experiments. J.J. and J.P.H. fabricated and characterized the devices. J.J. performed the first-principles and numerical drift-diffusion calculations. S.-J.K., J.-H.K., and M.-C.P. designed and set up the imaging system. J.H. and Y.J.S. performed the CAFM and KPFM measurements. K.L. and K.K. performed the HR-STEM and EDS measurements. W.K. and S.P. performed the XPS analysis. K.L. supplied Cl–$SnSe_2$ single crystals. T.T. and K.W. provided the h-BN crystals. J.A., B.-S.Y., S.J., H.W.K, S.K.Y., H.B., S.M., H.J.K., S.A.L., J.W.S., G.W., and J.-H.K. provided feedback throughout the experiments and data analysis. J.J., J.P.H., and

S.-J.K. cowrote the manuscript with input from all authors. D.K.H. and M.-C.P. supervised the project. All the authors discussed the results and approved the manuscript.

Competing interest

The authors declare no competing interests.

Figure Legends/Captions

Fig. 1 | Concept of the conductive-bridge interlayer contact. a, Comparison of the proposed and traditional contact architectures: (i) metal–semiconductor (MS) contact; (ii) metal–interlayer–semiconductor (MIS) contact; (iii) conductive-bridge interlayer contact (CBIC). Efficient carrier extraction can occur through conductive pathways in the CBIC, a feature absent in conventional MIS contacts. **b,** Schematic of the CBIC fabrication process via sequential sputter deposition. **c,** Schematic (upper) and optical image (lower) of a complete device, where a WS_2 absorber is sandwiched between CBIC–Al and Cl–$SnSe_2$ electrodes. **d,e,** Schematics of conductive atomic force microscopy (CAFM) measurements on CBIC–Al with Au (**d**) and without Au (**e**). **f,g,** Corresponding CAFM current maps (upper) and current line profiles (lower) of **d** and **e**, respectively. Conductive pathways are denoted by white arrows. The current line profile data are taken along the dashed red lines. **h,** Cross-sectional STEM image of the contact between WS_2 and CBIC–Al. Note that the bright spots near the bottom WS_2 layer might be residual interlayer particles after focused ion beam milling. **i,** Enlarged STEM image of the selected area in **h**, clearly revealing a uniform distribution of Au nanoclusters within the AlO_x interlayer.

Fig. 2 | Characterization of the CBIC. a, Kelvin probe force microscopy images of various CBICs. Scale bars, 1 μm. **b,** Work function (WF) distributions of the CBIC acquired from the indicated areas in **a**. The shaded stripes indicate the WFs of pristine metals taken from literature[8,19]. **c,** X-ray photoelectron spectroscopy spectra of WS_2 before and after CBIC–Al integration. **d,** Schematic (upper) and optical image (lower) of a top-gated WS_2 transistors fabricated with CBICs. **e–h,** Temperature-dependent transfer curves of WS_2 devices with CBIC–Al (**e**), Ti (**f**), Cr (**g**), and Cu (**h**) at $V_{DS} = 1$ V. **i,** Carrier polarity, threshold voltage (V_T), and electron field-effect mobility (μ_e) for different CBICs. All the error bars represent the mean values ± standard deviation (s.d.) (at least six devices are used for each case). **j,** Effective barrier height of each contact as a function of gate voltage. The Schottky barrier height is estimated under the flat band voltage condition ($V_{GS} = V_{FB}$), which corresponds to the onset of deviation from the linear trend. **k,** Estimated Schottky barrier heights for different contacts. The S value of the CBIC was extracted from linear fitting, denoted by the red dashed line.

Fig. 3 | Characteristics of the CBIC-integrated 2D photodetector. a, Schematic of the CBIC-integrated 2D WS_2 photodetector (upper) and energy level diagram (lower) of the materials used in this study. **b–d,** I–V characteristics of diodes for different contact configurations. **e,** Current rectification ratio between $I_{1.5V}$ (I_{DS} at $V = 1.5$ V) and $I_{-1.5V}$ (I_{DS} at $V = -1.5$ V) for different contacts. The central line in the error bar represents the median value, while the upper and lower bounds indicate the range between the 25th and 75th percentiles (at least seven devices were used for each case in the statistics). **f,** I–V curves of the representative CBIC–Al device measured under 532 nm laser illumination with power densities varying from 4.3 μW cm^{-2} to 5.4 W cm^{-2}. **g,** Short-circuit ($V = 0$ V) photocurrent as a function of light power density. The estimated linear dynamic range (LDR) of the device is over 120 dB. **h,** Comparison of the LDR of devices with different contacts. The CBIC devices generally exhibit a higher LDR compared to MIS devices. **i,** Responsivity and specific detectivity as a function of light power density. **j,** Normalized photoresponse as a function of modulated frequency, showing a −3 dB cutoff frequency exceeding 100 kHz. Insets: Plots of time-resolved photoresponse showing estimated rise and fall times of 4.21 μs and 3.95 μs, respectively.

Fig. 4 | Characteristics of the CBIC-integrated 2D photovoltaic cell. a, Schematic of the CBIC-integrated 2D WS_2 photovoltaic cell. **b,** Optical image (left) and corresponding photocurrent map (at $V = 0$ V) of the device. CBIC–Al and Cl–$SnSe_2$ electrodes are indicated by dashed and solid lines, respectively. **c,** *J*–*V* curves of the CBIC–Al and MIS–Al devices, measured under light illumination (532 nm, 44 mW cm^{-2}). **d,** Statistical distribution of photovoltaic parameters for devices with different CBICs (at least seven devices are used for each case). The distribution is represented in box-and-whisker plots (line within box: median, box limit: standard deviation, whiskers: 1.5 outliers). **e,** Plot of V_{OC} as a function of light intensity. Use of CBIC–Al can mitigate contact recombination and facilitate electron extraction, achieving a high V_{OC}. **f,** Plot of FF as a function of light intensity. **g,h,** Simulated band diagrams of the devices with CBIC–Cu (**g**) and CBIC–Al (**h**) under dark and light illumination conditions. The red dashed arrow in **g** indicates the contact recombination of electrons and holes. Inset: Enlarged view of the black dashed rectangle showing a negligible electron barrier of 20 meV.

Fig. 5 | Full-colour 3D imaging using CBIC-integrated image sensors. a, Principle of the 3D integral imaging method. **b,** Schematic and photographs of the 3D imaging system using a CBIC–Al device as a single-pixel imager. **c,** Spatial photocurrent images for various wavelengths (650 nm, 550 nm, and 450 nm, left) and corresponding full-colour images (right), acquired from MIS–Al (top) and CBIC–Al (bottom) devices. **d,** Photocurrent distributions of the MIS–Al and CBIC–Al devices for different wavelengths: 650 nm (left), 550 nm (middle), and 450 nm (right). Gaussian fittings are indicated by the solid lines. **e,** Full-colour elemental image (top) and the optically reconstructed 3D image (bottom), clearly depicting the 3D object with white letters *P*, *I*, and *N*. Details on the reconstruction process are provided in Supplementary Video 1.

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