1. Introduction

The conversion of solar energy into electricity is of great significance to the sustainable development of renewable energy and the economy. In the past decade, the emergence of a new type of third-generation solar cells, inorganic–organic hybrid perovskite solar cells (PSCs), has attracted tremendous attention due to its type-II energy band alignment and smaller charge recombination increment remains unclear. First-principles studies can analyze the interfacial interactions of TDMs with the perovskite at the atomic level to explore the underlying mechanism for the PCE increment remains unclear.

First-principles studies can analyze the interfacial interactions of TDMs with the perovskite at the atomic level to explore their capacity for application in PSCs and provide guidance for laboratory fabrication. Very recently, diverse TDMs such as graphene,SnS, black phosphorus, and 2D materials (TDMs) have demonstrated their great potential as functional materials in perovskite solar cells (PSCs) to boost conversion efficiency. As a versatile TDM, g-C3N4 has far less applications in PSCs than in medical science, energy storage, supercapacitors, and catalysis. In this work, the capacity of two kinds of triazine-based g-C3N4 as interfacial modifiers for CH3NH3PbI3 (MAPbI3) perovskite based on density functional theory is discussed. Due to the existence of two feasible atomically exposed surfaces in MAPbI3 perovskite, denoted as Pbl and MAI interfaces, four heterojunction structures are constructed. The interfacial carrier transport kinetics and charge recombination distribution of the heterojunctions are systematically investigated. The results show that the NPCN/MAI heterojunction facilitates the separation and transport of charges due to its type-II energy band alignment and smaller charge recombination region. Herein, a promising interface-modified 2D material for perovskite photovoltaic layer is introduced and an avenue to reduce the charge recombination loss in PSCs is provided.

PCE of PSCs was boosted to 25.7%, which is comparable with that of commercial silicon-based solar cells. Even though impressive progress was achieved, the PSCs still confront the issues of massive nonradiative recombination that hinders approaching the efficiency limits and the environmental stability that severely impedes the commercialization. These issues essentially arise from the intrinsic properties of the inorganic–organic hybrid formula, whereas overcoming the drawbacks by altering the composition would be challenging, as it typically deteriorates the photoelectric performance simultaneously. The alternative strategy is to induce functional materials inside the photovoltaic layer or the interface to passivate the trap states as well as improve the stability.

2D materials (TDMs) are promising functional materials to modify the perovskites due to their excellent thermal, mechanical, electrical, and optical properties, which are used as electrodes, carrier transporting layers, buffer layers, and perovskite additives. Among the numerous TDMs, graphitic carbon nitride (g-C3N4) is a versatile TDM with a similar structure to graphene, which has been widely used in applications of medical science, energy storage, supercapacitors, and catalysis. However, the investigation of g-C3N4 in the field of PSCs seems to be still in its infancy compared with other research hotspots. Pure g-C3N4 is rarely reported in the field of PSCs, except as additives for functional layers. As an example, Jiang and co-workers found that the PSC prepared by adding g-C3N4 to the perovskite precursor solution can reach PCE of 19.49%, while the efficiency of the device without g-C3N4 was only 16.22%. The authors attribute the improvement to the high perovskite crystal quality and electrical conductivity. Similarly, Liu et al. reported an all-inorganic CsPbBr3 PSC that uses the g-C3N4 additive to reduce charge recombination losses, resulting in a PCE of 8.0% with a fill factor of 80.25%. Though improvement was obtained by incorporating g-C3N4, the underlying mechanism for the PCE increment remains unclear.

First-principles studies can analyze the interfacial interactions of TDMs with the perovskite at the atomic level to explore their capacity for application in PSCs and provide guidance for laboratory fabrication. Very recently, diverse TDMs such as graphene, SnS, black phosphorus, and 2D
perovskites[31] have been proposed to construct heterojunctions with 3D perovskites to explain the mechanism or guide experimental synthesis. Taking the hot TDM, graphene, as an example, Cao et al. proposed that the contact of different surfaces of MAPbI$_3$ perovskite with graphene would lead to the n-type and p-type doping of graphene, respectively, and then envisaged a p–i–n (or n–i–p) architecture solar cell.[27] In addition, as a member of the perovskite family, the use of 2D perovskites to improve the stability and performance of 3D perovskites has been experimentally confirmed.[12–14] In terms of theoretical research, Liu and his colleagues studied the interfacial electronic properties and charge transfer characteristics of 2D BA$_2$PbI$_4$/3D MAPbI$_3$ heterojunctions, which further improved the performance of 2D/3D perovskite heterojunction-based solar cells.[31] Obviously, the study of TDM/perovskite heterojunctions is of great significance to promote the development of PSCs. To the best of our knowledge, 2D g-C$_3$N$_4$/perovskite heterojunctions have not been reported yet.

Here, we discuss the potential of two kinds of triazine-based g-C$_3$N$_4$ as interfacial modifiers for MAPbI$_3$ perovskite based on density functional theory. Considering the existence of two possible atomically exposed surfaces for MAPbI$_3$ perovskite, denoted as PbI and MAI interfaces, four heterojunction structures were constructed. We investigate the variations in the electronic properties of their constituents before and after the formation of heterojunctions. The charge transport kinetics between heterojunction interfaces and the distribution of charge recombination regions are discussed in depth. The results show that the nonplanar g-C$_3$N$_4$/MAI heterojunction possesses a favorable type-II energy-level arrangement, that is, the energy levels of valence band maximum (VBM) and conduction band minimum (CBM) of the heterojunctions are determined by the different constituent unit, and there is no large charge recombination region inside the perovskite, which has promising potential for solar cells application. This work introduces a promising 2D carbon nitride material as an interface modifier for perovskites and provides an avenue to reduce the recombination losses of charge in PSCs.

2. Computational Methods

Vienna Ab initio Simulation Package (VASP) was used to perform the first-principles calculation, using the projector-augmented wave method based on density functional theory (DFT).[35] The generalized gradient approximation (GGA) of the Perdew–Burke–Ernzerhof (PBE) exchange-correlation functional was selected for calculation. Heyd–Scuseria–Ernzerhof functional (HSE06) has also been used. The correction of van der Waals interaction adopted the Grimme DFT-D3 method.[36] A planewave (PW) cutoff energy of 500 eV was set. The relaxation and self-consistent calculation were based on the $3 \times 3 \times 1$ k-point mesh generated by the Monkhorst–Park (M–K) scheme. In geometric optimization, the uppermost atoms in the perovskite were relaxed until the force was less than 0.01 eV Å$^{-1}$ and the energy threshold was less than $1 \times 10^{-5}$ eV, while the perovskite lattice parameters and the bottom two-layer atoms were fixed. A vacuum layer over 15 Å along the z-direction was provided to eliminate direct interaction between adjacent plates. Part of the VASP calculated data were postprocessed by the VASPKIT code.[37]

3. Results and Discussion

Graphitic C$_3$N$_4$ (g-C$_3$N$_4$), possesses a similar lamellar structure to graphene. Different possible phase structures of g-C$_3$N$_4$ have been proposed, for example, g-h-triazine, g-o-triazine, and g-h-heptazine.[38] The unit cells of the g-h-triazine and g-o-triazine phase are composed of triazine (C$_3$N$_3$) rings, which exhibit hexagonal and orthorhombic structures, respectively. Compared with g-o-triazine, the g-h-triazine phase structure has longer C–N bonds, which endow lower energy and better stability. Similarly, g-h-heptazine C$_3$N$_4$ that built on heptazine units (C$_7$N$_7$) is also fairly stable. In this article, we also consider a neglected triazine-based g-C$_3$N$_4$ phase structure distinct from g-h-triazine, derived from the Materials Project.[39] This structure is almost identical to the g-h-triazine structure, except that the C and N atoms are not coplanar. In the following discussion, we denote the two structures composed of C$_3$N$_3$ (CN) rings as planar CN (PCN) and nonplanar CN (NPCN) and the other one composed of C$_7$N$_7$ units as hexagonal CN (HCN). Only the monolayer structures of them are considered in our article.

The optimized structures of PCN, NPCN, and HCN are shown in Figure 1a–c, and their band structures are displayed in Figure 1d–f. The structural parameters and the bandgaps of them calculated by GGA–PBE and HSE06 functional are listed in Table 1. For consistency, the unit cell c-axes of these monolayer structures are all set to 20 Å. The lattice parameters of PCN model are $a = b = 4.806$ Å and those of HCN model are $a = 7.136$ Å and $b = 7.134$ Å. The PBE bandgap and HSE06 bandgap of PCN model are 1.62 and 3.23 eV, respectively, and that of HCN model are 1.2 and 2.77 eV, respectively. The calculated results of the unit cell parameters and bandgaps are almost consistent with the reported literature.[36,40] Moreover, the bandgaps calculated by HSE06 are in good agreement with the experimental value, 3.1 and 2.7 eV.[41,42] For NPCN model, the lattice constants and the PBE bandgap are 4.716 Å ($a = b$) and 2.42 eV, which are basically in good agreement with 2DMatPedia (2dm-4847).[43] The HSE06 bandgap value is 3.65 eV. Neither PBE bandgap and HSE06 bandgap of NPCN model are close to the experimental bandgap of g-C$_3$N$_4$. The graphitic C$_3$N$_4$ with NPCN phase structure requires further study. In addition, for large models, it is worth considering a PBE functional calculation, especially when the PBE functional calculation is sufficient to obtain qualitative conclusions.

The unit cell parameters of the optimized tetragonal I4/cm MAPbI$_3$ structure are also listed in Table 1, and the front and top views of the structure are displayed in Figure S1, Supporting Information. We calculated the energy band structure of the bulk MAPbI$_3$ perovskite based on the PBE functional (Figure S2, Supporting Information), and the bandgap value is 1.52 eV, which is in agreement with the experimental values.[44] Previous reports have shown that PBE functional can predict lead-based perovskite bandgaps fairly accurately, albeit thanks to the error neutralization due to ignoring the spin–orbit coupling (SOC) effect.[36,31]
We chose the two triazine-based graphitic \( \text{C}_3\text{N}_4 \) mentioned earlier, PCN and NPCN structures, to construct heterojunctions with \( \text{MAPbI}_3 \) perovskite. The (001) plane slab models obtained by the \( \text{MAPbI}_3 \) structure exist in two distinct atomic surfaces, namely PbI and MAI terminations, denoted in this article as PbI and MAI interfaces, respectively. Then, \( \frac{1}{2} \times 2 \times 1 \) monolayer CN (PCN and/or NPCN) structure and \( 1 \times 1 \) \( \text{MAPbI}_3 \) perovskite unit cell was used to construct the heterojunctions, and four heterojunctions with distinct structures were denoted as PCN/PbI, PCN/MAI, NPCN/PbI, and NPCN/MAI heterojunctions, respectively (Figure 2). Both the PbI and MAI interfaces used to construct the heterojunctions have a four-layer perovskite structure to ensure accurate conclusions. For the structure optimization of these heterojunctions, we fixed the unit cells’ shape and volume, completely optimized the coordinates of the atoms in the uppermost layer of the perovskite structure, and only optimized the in-plane (x–y plane) coordinates of the \( \text{g-C}_3\text{N}_4 \) structure (orange dotted box in Figure 2). The unit cell parameters, lattice mismatch ratios, interfacial distances, and binding energies of all optimized structures are listed in Table 2.

The lattice mismatch ratios are obtained by the formula \( \sigma = (a_0 - a)/a_0 \), where \( a_0 \) and \( a \) represent the unit cell axis lengths before and after unit cell transformation, respectively. The positive and negative values represent the amount of tension and compression, respectively. The interfacial distance, \( d \), is defined as the vertical distance between the lowest atom of the \( \text{g-C}_3\text{N}_4 \) structure and the highest Pb or I atom in the perovskite moiety. The stretched or compressed lattices of \( \text{CN} \) (PCN and NPCN) structures were utilized to match the lattice of \( \text{MAPbI}_3 \) perovskite structure. The interface distances are all in the range of 2.95–3.25 Å (Table 2), which exceeds the bond lengths of C–H (1.1 Å) and N–H (1.0 Å), indicating that the interfaces of \( \text{g-C}_3\text{N}_4 \) and \( \text{MAPbI}_3 \) are connected through van der Waals (vdW) force in the heterojunction structures. To evaluate the thermodynamic feasibility of these heterojunctions, the binding energy was calculated by the Equation (1).

\[
\Delta E = E_{\text{H}} - E_{\text{CN}} - E_{\text{P}}
\]

Table 1. The lattice parameters and calculated bandgaps of relaxed PCN, NPCN, HCN, and bulk \( \text{MAPbI}_3 \) structure.

| Lattice parameters | Bandgap [eV] |
|--------------------|--------------|
| **Edge [Å]** | **Angle** | **PBE** | **HSE06** |
| PCN | \( a = 4.806 \) | \( \alpha = 90° \) | 1.62 | 3.23 |
| | \( b = 4.806 \) | \( \beta = 90° \) | | |
| | \( c = 20.00 \) | \( \gamma = 120° \) | | |
| NPCN | \( a = 4.716 \) | \( \alpha = 90° \) | 2.42 | 3.65 |
| | \( b = 4.716 \) | \( \beta = 90° \) | | |
| | \( c = 20.00 \) | \( \gamma = 120° \) | | |
| HCN | \( a = 7.136 \) | \( \alpha = 90° \) | 1.20 | 2.77 |
| | \( b = 7.134 \) | \( \beta = 90° \) | | |
| | \( c = 20.00 \) | \( \gamma = 120° \) | | |
| MAPbI | \( a = 8.849 \) | \( \alpha = 90° \) | 1.52 | – |
| | \( b = 8.849 \) | \( \beta = 90° \) | | |
| | \( c = 12.642 \) | \( \gamma = 90° \) | | |

Figure 1. The optimized structures of graphitic carbon nitride and their electronic band structures. a–c) Aerial views (top) and front views (bottom) of planar g-h-triazine \( \text{C}_3\text{N}_4 \) (PCN), nonplanar g-h-triazine \( \text{C}_3\text{N}_4 \) (NPCN), and g-h-heptazine \( \text{C}_3\text{N}_4 \) (HCN) structures. d–f) The electronic band structures of PCN, NPCN, and HCN structures. The blue and red lines represent the band structures calculated by PBE and HSE06 functionals, respectively.
where $E_{14}$ represents the total energy of the heterojunction and $E_{CN}$ and $E_{FP}$ represent the total energy of the CN (PCN or NPCN) and perovskite moieties in the heterojunction, respectively. The binding energies of all these heterojunctions are negative, indicating that both of them are energetically available.

To study the alterations in electronic properties after the heterojunction formation, the electronic band structures of the PCN, NPCN, PbI, and MAI interface structures were calculated based on PBE functional (Figure 3a–d). All of these band structures behave as direct bandgap.[28] Figure 3e–h shows the projected band structures of PCN/PbI, PCN/MAI, NPCN/PbI, and NPCN/MAI heterojunctions, respectively, where the dark cyan and yellow solid lines represent the electron contributions from the CN and perovskite moieties, respectively. Overall, the band structures of heterojunctions basically retain the band shapes of their constituents, but their energy levels exhibit varying degrees of shift from the Fermi level. In detail, the band energy levels of perovskite moiety in PCN/PbI and PCN/MAI heterojunctions are shifted downward relative to the Fermi level, while that of NPCN moiety in NPCN/PbI and NPCN/MAI heterojunctions display a similar variation.

To find the reason for the band energy-level shift, we analyzed and compared the work functions of the PCN, NPCN, PbI, and MAI interface structures. The work function was obtained by the formula $W = E_V - E_F$, where $E_V$ and $E_F$ represent the vacuum level and the Fermi level, respectively. Their work functions are 3.15, 5.33, 5.55, and 4.17 eV, respectively (Figure S3, Supporting Information). We further obtained their energy-level arrangement, referenced to the vacuum level (Figure S4, Supporting Information). We reason that the band energy-level shift in the band structure of the heterojunction is due to the electron flow driven by the work function, that is, electrons flow from the part with a smaller work function to the part with a larger work function, which in turn leads to n-type doping of the influx part and a subsequent downward offset relative to the Fermi level. It is found that the energy-level shift variations in the band structures of all our heterojunctions, except for the NPCN/PbI heterojunction, can be attributed to work function differences. We speculate that the reason for the anomaly of the NPCN/PbI heterojunction is that the small difference in work function is not enough to cause the same alteration as the others, while at the same time the dominant electronic state of the PbI interface moiety causes a slight downward shift of the band energy of NPCN moiety.

To further study electronic properties of the heterojunctions, the projected density of states (PDOS) was calculated. Figure 4a–d shows the PDOS of PCN/PbI, PCN/MAI, NPCN/PbI, and NPCN/MAI heterojunctions, respectively. The gray, orange,
dark cyan fill represent the electronic state contribution of heterojunction, perovskite moiety (PbI or MAI interface), and CN (PCN or NPCN) moiety, respectively. It can be observed that the electronic states at the valence band maximum (VBM) and the conduction band minimum (CBM) for PCN/MAI heterojunction both belong to the PCN moiety. For the PDOS of PCN/Pbi heterojunction, the VBM and CBM electronic states belong to PCN and PbI interface moieties, respectively. The PDOS of NPCN/PbI and NPCN/MAI heterojunctions show different band-edge states. Typically, the electronic states at VBM and CBM of NPCN/Pbial heterojunction are both assigned to the Pbi interface moiety, while those of NPCN/MAI heterojunctions are assigned to the MAI interface and NPCN moieties, respectively.

The energy-level arrangement diagram assists to analyze the band arrangement of each part of the heterojunction. The energy-level arrangement of VBM and CBM of the heterojunction determined by the same constituent unit is a type-I arrangement; otherwise, it is a type-II arrangement. A type-II energy-level arrangement for heterojunction is more conducive to charge carrier transfer. Therefore, the energy-level arrangements of PCN/MAI and NPCN/PbI heterojunctions are type-I arrangements, while that of the others are type-II arrangements. Note that the heterojunctions exhibit different performances probably, even with the identical type-I or type-II arrangement. For the heterojunctions with type-I arrangement, that is, PCN/MAI and NPCN/PbI heterojunctions, when the electron–hole pairs were generated on the perovskite moiety by sunlight irradiation, the PCN structure in the former and NPCN structure in the latter would not effectively extract electrons and/or holes due to the presence of charge carrier recombination (both electrons and holes would flow from MAI interface moiety to PCN moiety in the PCN/MAI heterojunction) and energy barrier, respectively. For the heterojunctions with type-II arrangement, the

Figure 3. Electronic band structures. a–d) The electronic band structures of PCN, NPCN, PbI interface, and MAI interface structures in the heterojunction lattice. e–h) The projected band structures of PCN/PbI, PCN/MAI, NPCN/PbI, and NPCN/MAI heterojunction structures. The horizontal gray dashed lines represent their Fermi levels. The dark cyan and orange lines represent the electronic contribution of CN (PCN or NPCN) and perovskite moiety (PbI or MAI interface) in the projected band structures of heterojunctions.
PCN moiety in PCN/PbI heterojunction and NPCN moiety in NPCN/MAI heterojunction would validly extract photogenerated holes and electrons, respectively, under sunlight irradiation. Furthermore, we found that the bandgap value of PCN structure within the heterojunction lattice is significantly smaller than that of the PCN structure with the initial lattice (the PBE bandgap is altered from 1.62 to 0.35 eV), which is due to the electronic structure alteration led by lattice transformation (compress on the X-axis and stretch on the Y-axis). In contrast, the electronic structure variation of NPCN structure after lattice transformation is relatively tiny (the PBE bandgap is altered from 2.42 to 1.8 eV), and its electronic structure is better maintained, which may be due to the nonplanar geometry with better structural stability. Therefore, we deem that the NPCN/MAI heterojunction with type-II band alignment would have more potential for photovoltaic applications, such as solar cells, due to the better electron extraction capability of NPCN structure, while the PCN/PbI heterojunction with type-II band alignment requires prestress conditioning of PCN structure.

Next, we investigate the NPCN-based heterojunctions, because the NPCN moiety in the heterojunctions preserves the intrinsic electronic properties well. To illustrate the interfacial charge transfer behavior, we calculated the 3D charge density difference (CDD) of NPCN/PbI and NPCN/MAI heterojunctions, which was obtained by Equation (2).

$$\Delta \rho = \rho_H - \rho_{CN} - \rho_P$$

where $\rho_H$ represents the charge density of PCN/PbI or NPCN/MAI heterojunction, $\rho_{CN}$ represents the charge density of NPCN moiety, and $\rho_P$ represents the charge density of PbI or MAI interface moiety in the heterojunction lattices. Figure 5a,b shows the CDD at the interfaces of NPCN/PbI and NPCN/MAI heterojunctions, respectively. The yellow electronic clouds represent electron accumulation, and the bright blue electronic clouds represent electron consumption. The value of the isosurfaces is $2.0 \times 10^{-4}$ e bohr$^{-3}$. The volume of the electron cloud reflects the density of accumulated or consumed electrons. For the NPCN/PbI heterojunction interface, the yellow electron clouds were observed on the surface and inside of the perovskite moiety, and the bright blue electron clouds were observed on the surface of NPCN structure. This result indicates that electrons mainly flow from NPCN/PbI heterojunction to PbI interface moiety during the formation of heterojunction.

Figure 5c,d shows the average differential charge density of the $x-y$ plane along the $z$-direction in these two heterojunctions. Figure 5c shows that the charge consumption mainly occurs on the surface of NPCN structure, and the charge accumulation mainly occurs on the surface of PbI interface moiety. At the same time, there is both charge consumption and accumulation inside the PbI interface moiety. Figure 5d shows that the charge consumption mainly occurs on the surface of MAI interface structure, and the charge accumulation mainly occurs on the surface of NPCN structure. Therefore, in the process of forming the heterojunction, electrons in NPCN/PbI heterojunction...
mainly flow from NPCN moiety to PbI interface moiety, while electrons in NPCN/MAI heterojunction flow from MAI interface moiety to NPCN moiety.

The charge carrier recombination of PSC devices was regarded as the primary concern that leads to voltage loss and thus limits the improvement of conversion efficiency.\cite{31} In the scenario of the heterojunction interfaces, the areas where charge accumulation and consumption occurred simultaneously are considered as the recombination center during the formation of the heterojunctions. In the NPCN/PbI heterojunction with type-I arrangement, the photogenerated carriers (electrons and holes) cannot be transferred to NPCN moiety because of the energy barrier. Therefore, a large density of photogenerated carriers will be recombined on the side of PbI interface moiety. The schematic diagram of the process is illustrated in Figure 5e, where the pink dotted rectangle represents the reorganization center. For the NPCN/MAI heterojunction, the charge recombination center is mainly located on NPCN moiety, and only a small part is located on the surface of the MAI interface structure. As a result, when the device based on NPCN/MAI heterojunction is exposed to sunlight, a small number of photogenerated carriers will recombine on the side of MAI interface moiety, and the majority of photogenerated carriers will be separated and transferred. The schematic diagram of the process is illustrated in Figure 5f. According to our theoretical calculation results, the NPCN/MAI heterojunction is more suitable for utilizing in PSC architectures.

4. Conclusion

In summary, the density functional first-principles study was used to analyze the interfacial interactions of two classes of triazine-based g-C\(_3\)N\(_4\)/MAPbI\(_3\) heterojunctions. As MAPbI\(_3\) perovskite has two different exposed atomic surfaces, which we denoted PbI and MAI interfaces, four heterojunction structures were constructed. The alterations in the electronic properties of their constituents before and after the formation of these heterojunctions were investigated. We reason that the band energy-level shift in the band structure of the heterojunction is due to the electron flow driven by the work function, which in turn leads to the n-type doping of the influx part, and a subsequent downward offset relative to the Fermi level. Furthermore, we calculated the PDOS of these heterojunctions and analyzed the energy-level assignments of them. It is found that the NPCN moiety of NPCN/MAI heterojunctions with type-II band alignment possesses a favorable electron extraction performance. Finally, the scenarios of electron transfer and charge carrier recombination under illumination in NPCN/PbI and NPCN/MAI heterojunctions are analyzed and
compared. The results show that the NPCN/MAI heterojunction is more favorable for charge separation and transfer due to the absence of large charge recombination regions. This work introduces a promising interface-modified 2D material for the perovskite photoactive layer and provides an idea to reduce the charge recombination loss in PSCs.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
2D materials, density functional theory, g-C3N4, heterojunctions, perovskites

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