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Letter

2D Perovskite Nanosheets with Intrinsic Chirality

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ABSTRACT: Chiral perovskite materials have been intensively studied because of their unique properties and wide range of potential applications; however, the synthesis of perovskite nanocrystals with improved chirality has been scarcely investigated. In this Letter, two-dimensional perovskite nanosheets with intrinsic chirality are demonstrated. Inserting chiral amines into the perovskite framework leads to the chirality transfer from amine molecules to perovskite structure. The protecting agent, specifically, achiral octylamine, is found to influence the chiral optical signal or dissymmetric factor of nanosheets significantly. By controlling the amount of octylamine, we have synthesized



perovskite nanosheets with the highest *g*-factor ever reported. We expect our primary demonstration could attract more attention toward the synthesis of intrinsic chiral perovskite nanocrystals and the development of nanocrystal-based chiral-optical devices with improved functions.

alide perovskites, a type of semiconductor materials, are Lattractive for applications in fields ranging from photovoltaics¹⁻⁴ and optoelectronics^{5,6} to light emission.^{7,8} In the past decade, the certified power conversion efficiency (PCE) of halide perovskite solar cells has reached 25.5%, which is comparable to that of monocrystalline silicon photovoltaic devices.⁹ Perovskite red and green light-emitting diodes (LEDs) have displayed over 20% external quantum efficiency (EQE),¹⁰⁻¹² which is competitive to that of CdSe quantum dots (QDs) and organic semiconductor LEDs. On the other hand, with the introduction of chirality, the central inversion symmetry of the crystal structure is broken to form chiral perovskite.¹³ With the combination of excellent optoelectronic properties and unique chiral characteristics, chiral perovskites have expanded the functionality to areas such as circular polarized light detection.^{14,15} circular polarized luminescence,^{16–23} nonlinear optics,^{20,24,25} ferroelectrics,^{26–29} and spintronics.³⁰

At present, there are basically four methods developed to introduce chirality into perovskites: (i) inserting chiral molecules into perovskite bulk to form chiral crystalline structure, (ii) perovskite nanocrystal (NC) surface distortion induced by using chiral molecules, (iii) electronic coupling between the chiral molecules and achiral perovskite, and (iv) screw dislocation of perovskite nanocrystal assemblies. In method i, chiral amines are directly inserted into the perovskite framework where the chirality is transferred from the small molecules to the perovskite crystal structure. Usually, the dissymmetric factor of this type of chiral perovskites is relatively higher because it has chiral space groups and displays intrinsic chiral characteristics.^{31,32} For methods ii and iii, the

chirality of the perovskite is actually derived from the interaction of the chiral ligand "shell" and the achiral perovskite NCs "core"; in other words, their chirality is the induced chirality. In these chiral perovskites, the chiral components are mainly concentrated on the surface, and consequently, the chiral optical signal is relatively weak.^{19,20,33,34} In method iv, there are no chiral molecules and the chirality of perovskites can be realized by the screw dislocation as the assembling of nanocrystals, displaying a higher order of aggregation chirality.³⁵

Despite the intensive studies on chiral perovskite materials, the synthesis of chiral perovskite NCs has scarcely been reported. At the beginning, a post ligand exchange method, widely used for the synthesis of chiral quantum dots (*e.g.* CdSe), was employed to prepare chiral perovskite NCs (see method ii above). The g-factor of these perovskite NCs can reach 1.5×10^{-3} ; however, no apparent circular dichroism (CD) signals were presented at the absorption maximum.³³ Upon attaching chiral *R*- or *S*- α -methylbenzylamine (*R*-*/S*-MBA) to the surface of methylammonium lead bromide perovskite during the crystal growth, chiral perovskite nanosheets (NSs) with chiroptical response in excitonic absorption region were prepared (see method iii above).³⁴ Recently, the coassembly of perovskite NCs and chiral gels was

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proven to be an effective approach to introduce chirality into perovskite NCs (belongs to method ii, see above). Interestingly, this coassembly endowed achiral perovskite NCs with circularly polarized emission functions.¹⁹ In addition, perovskite NCs prepared by using chiral α -octylamine as surface ligands have shown upconverted circularly polarized luminescence properties; however, the chiral intensity of these NCs is relatively weak (g-factor of $\sim 2.4 \times 10^{-4}$).²⁰ Without introducing chiral molecules, based on screw dislocation of CsPbBr₃ NC aggregates, the CD absorption of NC assemblies was demonstated, but it is approximately only one-tenth of that of hybrid perovskite with chiral crystal structure.35 To summarize the reported chiral perovskite NCs, we find that most NCs were mainly based on chirality induced by chiral ligand molecules, and apparently, this induction method produced NCs with relatively weak chiral optical response.

Herein, we report the synthesis of two-dimensional (2D) intrinsic chiral perovskite NSs in which the chiral amine *R*- or *S*- β -methylphenethylamine (*R*- or *S*-MPEA) is the internal structural component (see method i above). To the best of our knowledge, there have been no reports of perovskite NCs displaying intrinsic chirality. The *g*-factor of the perovskite NSs reaches 6.5×10^{-3} , which is the highest value of perovskite NCs at present. In addition, the influence of the protecting agent octylamine (OA) on the chiral optical signal and structure of perovskite NCs is also investigated.

To prepare intrinsic perovskite NCs, the structure of chiral amine is important because it is directly inserted into the framework. On the other hand, the steric hindrance of reactive molecules usually influences both the kinetics and dynamics of a chemical reaction. The substitutes adjacent to the reaction center occupying certain spatial position might greatly hinder the exposure of the reaction center, thereby reducing the reactivity of the molecule. Herein, we adopt a ligand-assisted reprecipitation method (LARP) to prepare the chiral 2D perovskite NSs in which the crystallization is completed very shortly, and therefore, the reactivity of chiral amines needs to be considered.

Four types of chiral amines, specifically, R-BA, R-HA, R-MBA, and R-MPEA, are selected (Figure S1a). In R-BA, R-HA, and R-MBA, the amino groups are directly connected to the chiral carbon. Apparently, the other bulk groups (e.g., methyl, alkyl, and phenyl) attached on the chiral center would greatly hinder the exposure of the amino group. However, in R-MPEA, an additional carbon separates the amino group from the bulk phenethyl substitute, and consequently, higher reactivity is expected. The absorption and chiral optical properties of perovskite NCs prepared by using four chiral amines are compared in Figure S1b,c. Significantly intense absorption peak is found for R-MPEA perovskite NCs at approximately 400 nm. However, very weak absorption is observed for the other three NCs (Figure S1b). The distinct absorbance indicates that the reactivity of R-MPEA and the yield of perovskite NCs are much higher in comparison to the other three amines. At the same time, the chiral optical property of these NCs is characterized by the CD spectrum. R-MPEA perovskite NCs exhibit a strong CD signal, whereas no apparent CD responses are found for R-BA, R-HA, and R-MBA NCs (Figure S1c). Both absorption and CD spectra demonstrate that R-MPEA is inserted into the perovskite framework where the chirality of R-MPEA has transferred to the NCs. In contrast, chiral amines with higher steric hindrance are hard to insert into the perovskite crystal.

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The procedure to prepare chiral perovskite 2D sheets follows a modified antisolvent method.³⁶ During the NC growth, OA is added to stabilize the *R-/S*-MPEA perovskite NSs. At the beginning, the molar ratio of *R-/S*-MPEABr to PbBr₂ in the growth solution is studied. With the increase of *R-/S*-MPEABr, the CD signal of perovskite NCs increases and reaches the maximum at approximately 5:2 (Figure S2). Further increase leads to a slight decrease in CD intensity. To compare the chiral optical properties of NSs, the absorptions of both *R*-MPEA and *S*-MPEA perovskite NSs are normalized. These chiral NSs prepared with 1.25 μ L of OA exhibit mirror-symmetric CD responses in which the strongest peaks appear at ~399 nm and reach –58 and 60 mdeg for *R*-MPEA and *S*-MPEA NSs, respectively (Figure 1a). The strong chiroptical



Figure 1. (a) CD spectra and (b) *g*-factors of chiral MPEA perovskite nanosheets (n = 1). Absorption (solid curve) and emission (dashed curve) spectra of (c) *R*-MPEA and (d) *S*-MPEA perovskite nanosheets. During the synthesis of both NS, 1.25 μ L of OA is added.

signal is further confirmed by the high g-factors (defined as $\Delta \varepsilon / \varepsilon = \theta$ [mdeg]/(33 000 × *A*), where $\Delta \varepsilon$ and ε are the molar circular dichroism and molar extinction, respectively; *A* is the intensity in the absorption spectrum³⁷). As shown in Figure 1b, the g-factors of *R*- and S-MPEA NSs at ~399 nm reach -3.6×10^{-3} and 3.7×10^{-3} , respectively. This g-factor is on the same order of magnitude as that of chiral bulk perovskite³¹ while apparently larger than that of chiral NCs whose chirality originates from the interaction of the chiral ligands "shell" and the perovskite "core".²⁰

Upon comparison of the CD signal and absorption of R-/S-MPEA NSs, the strong CD signals between 375 and 425 nm correspond to the excitonic absorbance bands (Figure 1c,d, solid curves). At the excitonic absorption maxima, the CD value of R-MPEA NSs crosses zero and changes sign from negative to positive, while that of S-MPEA NSs changes sign from positive to negative. In other words, R- and S-MPEA NSs exhibit an opposite Cotton effect. Meanwhile, the CD spectra of R- and S-MPEA precursors indicate that there are no active chiroptical signals beyond 275 nm (Figure S3). Consequently, we surmise that the chirality is successfully transferred from the chiral molecules to perovskite NSs. In addition, the emission spectra are also recorded. No apparent differences are found for R- and S-MPEA NSs in which a sharp fluorescence peak

 $(\sim 402 \text{ nm})$ with a narrow full width at half-maximum (fwhm, $\sim 14 \text{ nm})$ is observed (Figure 1c,d, dashed curves).

The structure of chiral perovskite NSs is subsequently characterized by using TEM, AFM, and XRD. TEM and AFM images indicate that these chiral NSs are rectangular shape with the length of approximately $1-2 \mu m$, the width of 200–500 nm, and the thickness of around 2.0 nm (Figure 2a, TEM;



Figure 2. (a) TEM image and (b) AFM morphology and height profile of perovskite NSs prepared by using *R*-MPEA. (c) XRD patterns of *R*- and *S*-MPEA perovskite NSs. In the synthesis of NSs, 1.25 μ L of OA is added. (d) Schemes of perovskite nanosheet structure with *R*- and *S*-MPEA amines.

Figure 2b, AFM). This shape is in contrast to the quasi-square shape of achiral perovskite NSs.^{31,38,39} In chiral perovskite NSs, there is a polar axis which defines the preferential growth direction. It is proposed that the difference in growth rate between the polar axis direction and vertical direction is responsible for the formation of stripe shape. The XRD patterns of *R*- and *S*-MPEA NSs are almost identical, where both NSs exhibit a very sharp periodic peak around 4.88°, corresponding to the (002) plane with a stacking spacing of 1.8 nm (Figure 2c). This indicates perovskite NSs have only one atomic layer of [PbBr₄]⁻² (n = 1).

The structural scheme of *R*-MPEA NSs are shown in Figure 2d. These NSs are 2D structures in which *R*- or *S*-MPEA are the internal building components. Consequently, the chirality of amines has transferred to the NSs. Recent studies have shown that perovskites with chiral amine components have chiral space group (*e.g.* the chiral space group for *R*-/*S*-MBA perovskites is $(P2_12_12_1)$) where their chirality is regarded as intrinsic chirality.^{14,24,27,31} Hence, *R*-/*S*-MPEA perovskites are expected to have an intrinsic chiral crystal structure. Because of the chirality transfer, the lead bromide octahedral layers of the two chiral enantiomers possess mirror-symmetric distortion (see Figure 2d).

A previous report shows that the addition of surfactants such as OA molecules could not only regulate the morphology of perovskite NCs but also protect the NCs from aggregating.⁴⁰ In the synthesis of chiral *R*- and *S*-MPEA perovskite NSs, OA is also employed. As a type of low steric hindrance amine, OA can also enter the framework of the perovskite, and consequently, NSs prepared with a mixture of amines are the hybrid perovskites. Apparently, the amount of OA or the pubs.acs.org/JPCL

relative ratio of achiral and chiral amines would influence the structure and chiral optical property of NSs. To determine the effect of OA, a different amount of OA (from 0.5 to a maximum of 5 μ L) is added to synthesize *R-/S-MPEA* perovskite NSs. The CD spectra of these hybrid chiral perovskite NSs are recorded. With the increase of OA volume from 0 to 1 μ L, the CD signal was enhanced, whereas it gradually decreased upon further addition of OA (Figure 3a).



Figure 3. (a) CD spectra of *R*- and *S*-MPEA NSs prepared with different volume of OA. (b) Dependence of *g*-factor maximum in the excitonic region of *R*- and *S*-MPEA NSs on the amount of OA. Note: 0.5, 1, 1.25, 2.5, 3.75, and 5 μ L of OA correspond to the achiral (OA)/chiral (*R*-/*S*-MPEA) amines ratio of 33, 16.5, 13.2, 6.6, 4.4, and 3.3, respectively.

In addition, the dependence of the *g*-factor maxima in the excitonic absorption region on the amount of OA is plotted. Similar to CD intensity, the highest *g*-factor ($\sim 6 \times 10^{-3}$) appears when 1 μ L of OA is added (Figure 3b).

Apparently, to prepare perovskite NSs with the strongest chiral optical property, there is a balance between the OA's role as a stabilizer and component. In the absence of OA, there is the highest ratio of chiral amines; however, a relatively weak CD response is observed because these NCs are not stable without the protection of OA. Actually, without the OA molecule, the stability of perovskite NCs is poor, and we obtained only perovskite "aggregates" rather than monodispersed NSs. Introducing a small amount of OA (*e.g.*, 0.5 or 1 μ L), the *g*-factors of *R*-MPEA (*S*-MPEA) perovskite NSs reach the maximum of -4.6×10^{-3} (4.7×10^{-3} , $0.5 \,\mu$ L) and -6.5×10^{-3} (6.3×10^{-3} , $1 \,\mu$ L OA). We surmise that the enhancement of chiral optical response is due to the stabilizing functions and morphology regulation of OAs in the growth of NSs. Adding a small amount of OA led to the formation of NSs, stabilized these particles, and consequently enhanced the chiral signals. Upon further increase of OA from 1 to 5 μ L, the *g*-factors of *R*-MPEA perovskite NSs have decreased from -6.5×10^{-3} to -3.2×10^{-4} . A similar decrease of *g*-factor is found for *S*-MPEA perovskite NSs (from 6.3×10^{-3} to 2.7×10^{-4}). In this process, more OA molecules enter the perovskite framework, which consequently leads to the decrease of chiral active *R*-/*S*-MPEA perovskite components and final chiral optical response of NSs.

The effect of OA volume on the absorption and fluorescence spectra of perovskite NSs was subsequently characterized. No apparent difference between *R*- and *S*-MPEA perovskite NSs is observed. With the increase of OA, the absorption peak of both *R*- and *S*-MPEA perovskite NSs slightly red-shifts approximately 7 nm (Figures 4a and S4a), indicating the changes of



Figure 4. (a) Absorption and (b) PL spectra of *R*-MPEA perovskite NSs prepared by using different amounts of OA. (c) fwhm of PL spectra of both *R*- and *S*-MPEA NSs as a function of the OA volume. (d) XRD pattern of *R*-MPEA perovskite NSs with different amounts of OA.

exciton band. This band variation is due to the insertion of OA molecules which modifies the perovskite structure. In addition, this absorption peak shift is consistent with the CD spectra in which the zero CD intensity wavelength has also red-shifted approximately 4 nm (Figure 3a). Apparently, the shift of absorption peak is accompanied by the shift of fluorescence. Interestingly, severe tailing is found for the PL spectra of both R- and S-MPEA perovskite NSs with 0.5 and 1 μ L OA. The fwhm of these NSs is approximately 18 nm (Figures 4b and S4b). Further increase of OA has improved this tailing phenomenon in which the fwhm's of PL spectra significantly decrease and reach the steady value of around 10 nm (Figure 4c). In short, the luminescence quality of chiral perovskite NSs has improved; however, the chiral strength is the balance between the quality of crystalline structure and chiral amine ratios.

Next, the structure of chiral R-/S-MPEA perovskite NSs as a function of OA volume is characterized by using XRD and TEM. All of the R-/S-MPEA NSs have shown very similar XRD patterns in which a very sharp periodic peak, corresponding to the (002) plane, appears around 5° (Figures 4d and S4c). In the NSs with 0.5, 1, and 1.25 μ L of OA, the regular intervals of peaks are approximately 4.88°, which corresponds to the stacking distance of 1.8 nm. The increase of OA has little effect upon the stacking spacing of NSs, which demonstrates that very little OA has entered the perovskite framework. Upon further increasing the amount of OA to 5 μ L, the regular intervals of peaks reduce from 4.88° to ~4.68°, corresponding to the change of stacking distance from 1.8 to 1.9 nm. In this case, OA is inserted into the framework of chiral perovskite NSs, consequently leading to the decrease of chiral amine ratio and chiral optical signal of NSs. In addition, the TEM images of chiral perovskite NSs with excess of OA are recorded. As shown in Figure S5, these NSs synthesized with 2.5, 3.75, and 5 μ L OA have shown rounded square shape with the side length of several hundreds of nanometers. This morphological change of R-/S-MPEA perovskite NCs indicates the disappearance of the polar axis.

Finally, we must note that although these chiral perovskite NSs have shown strong chiral optical response, unfortunately, no circularly polarized luminescence (CPL) is recorded, at least at room temperature. This might relate to the temperature dependence of CPL demonstrated for bulk 2D chiral perovskite.¹⁷ Measurement performed at considerably lower temperature might be promising to detect CPL in these chiral NSs. In addition, the chiral intensity of chiral perovskite NSs is expected to further improve by using chiral molecules as protecting agents. The chiral stabilizer coated on the surface of NSs can induce the additional surface chirality. Consequently, the superposition of induced surface chirality and intrinsic chirality may enhance the chiral intensity of perovskite NSs.

In summary, two-dimensional chiral perovskite NSs with intrinsic chirality are prepared by inserting chiral amines into the perovskite framework. Upon controlling the volume of achiral OA, the chiral optical response and the structure of perovskite NSs can be regulated. A small amount of OA can improve the chirality of NSs, whereas excess OA leads to the insertion of achiral OA into the perovskite framework and thereby reduces the chiral strength of NSs. Notably, the *g*-factor of NSs reaches 6.5×10^{-3} , a value not achieved previously. Looking forward, we envision these intrinsic NCs could find appealing applications in chiral-optical devices, for example, CPL detection and spintronics.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jpclett.1c00315.

Experimental synthesis and characterization of perovskite nanosheets and additional results (PDF)

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Notes

The authors declare no competing financial interest.

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