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Recyclable luminescent solar concentrator from lead-free perovskite derivative

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Abstract

Luminescent solar concentrators (LSCs) offer a sustainable approach to power generation using fluorescent glasses, yet their green industrialization is impeded by the limited production scale and non-recyclability of embedded nanocrystals. Here, we introduce a lead-free perovskite derivative ETP₂SbCl₅ (ETP = (C₆H₅)₃PC₂H₅) with a reversible transition between powder and glass states. Through molecular dynamics and density functional theory, we elucidate the possible structural distortions of [SbCl₅] pyramids and their impact on luminescence. The fabricated LSCs, utilizing such fluorescent glasses with an efficient absorption for <420 nm, achieve the highest power conversion and optical efficiencies of ~5.56% and ~32.5%, respectively. In addition to self-healing by reheating at ~200 °C, impressively, it could be mass recycled to phosphor by ethanol or heating treatments, which still maintains nearly initial fluorescent performance and could be repurposed like freshly synthesized samples. This work presents a paradigm for the sustainable use of fluorescent materials and offers a reliable path toward low-carbon globalization.

Introduction

Luminescent solar concentrators (LSCs), which balance transparency with photovoltaic capabilities, harmoniously combining energy generation with architectural esthetics, are emerging as pivotal solutions in the quest for self-sufficient green energy¹. Using semitransparent fluorescent glasses, LSCs absorb a portion of sunlight (such as ultraviolet light) and undergo photoluminescence (PL), which is then conducted to the solar cells integrated on the edges for optical-electrical conversion². As a key component, in previous reports, the fluorescent glasses were fabricated by embedding nano-scale crystals, such as the carbon-³, silicon-⁴, and Se/S-based^{5,6} quantum dots (QDs), as well as Pb-based perovskite nanocrystals (NCs)^{7–10}, into various polymers. Despite the relatively

mature synthesis protocols for QDs/NCs at the laboratory stage, their protracted fabrication durations, massive solvent consumption, and constrained chemical yield result in elevated preparation costs, which present substantial challenges for future large-scale production of LSCs (Supplementary Table S1). Moreover, the embedded emitters are challenging to segregate and recycle, which leads to a disposable use characteristic, necessitating re-synthesis and re-embedding once the glasses are damaged. By comparison, one-step synthesis of fluorescent glasses by simply thermal treatment is more desirable for low-cost and mass-scale production, and exploring such new materials and corresponding preparation schemes are considered crucial to potential industrialization.

Ionic liquids, recognized for their environmental compatibility and versatile functionality, have been recently employed to modify interfaces and passivate NCs/QDs effectively^{11–13}. Aided by the self-solvent property and cationic components, they could combine with metal ions thus forming (semi-)transparent glasses with zero-dimensional structures at mild heating temperatures. For example, in 2002, Mitzi et al. originally reported on a perovskite film (2-FPEA)₂SnI₄ (2-FPEA = C₆H₄FC₂H₄NH₃),

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which possessed a relatively low melting temperature of $\sim 200^\circ\text{C}$, enabling its conversion into transparent thin film¹⁴. However, the immediate crystallization would occur once cooling to room temperature, hindering the investigation for amorphous state. Then, exploring the melting and quenching conditions for preparing stable hybrid glass became a popular research focus, of which the trend continued until that the S(1-1)NPB (S-(1-1)naphthylethylammonium lead bromide) was demonstrated to readily enable access to a glass state through quenching from the melt in 2020 (ref. 15,16). Recently, with the develop of lead-free perovskites, ns^2 and RE-based (RE = rare earth) hybrid luminescent glasses with shape-on-demand characteristics were gradually explored, such as (Bmmim)₂SbCl₅ (Bmmim = C₄H₆N₂C₄H₉, ref. 17), (HTP/ETP)₂MnBr₄ (HTP = (C₆H₅)₃PC₇H₁₅, ETP = (C₆H₅)₃PC₂H₅, ref. 18,19), ETP₂SbCl₅ (ref. 20,21), and RE(NO₃)₃(C₅H₂N₄)₂ (ref. 22), etc. Although the microscopic mechanisms of the powder-glass transition and specific structural distortion are still unclear, the hybrid glasses have indeed exhibited exceptional performance in various applications including memory and computing^{23,24}, energy storage^{25,26}, thermoelectric²⁷, photovoltaic²⁸ and X-ray detection^{29,30}, etc.

Such burgeoning perovskite hybrid glasses could greatly integrate the functions of fluorophores and polymers/glasses, which present an exemplary solution for the preparation strategy and chemical yields. By using their optical waveguide effect, this technique of fluorescent glass may offer a promising scheme for innovative LSCs. Additionally, in the age of low-carbon and sustainable globalization, heightened attention should be given to the recyclability of these novel fluorescent glasses once damaged or discarded, thereby eliminating disposable use, resource squandering or heavy metal pollution^{31,32}.

In this work, we synthesized the yellow emissive ETP₂SbCl₅ phosphor with a near-unity PLQY by a simple solution process at room temperature, of which the glass phase could be obtained by further thermal treatment. The as-fabricated ETP₂SbCl₅-based LSCs exhibited the highest power conversion and optical efficiencies of $\sim 5.56\%$ and $\sim 32.5\%$, respectively, on a $3 \times 3 \times 0.5\text{ cm}^3$ fluorescent glasses. Besides the self-healing property, reversible transitions between phosphor and glass phases have been detected. Even after undergoing 10 cycles of phosphor-glass transitions, the final recycled phosphors still maintained $\sim 95\%$ of their initial PL performance, enabling them to be further used in other fields, such as phosphor converted-LED or anti-counterfeiting, etc., as effective as freshly synthesized phosphors.

Results

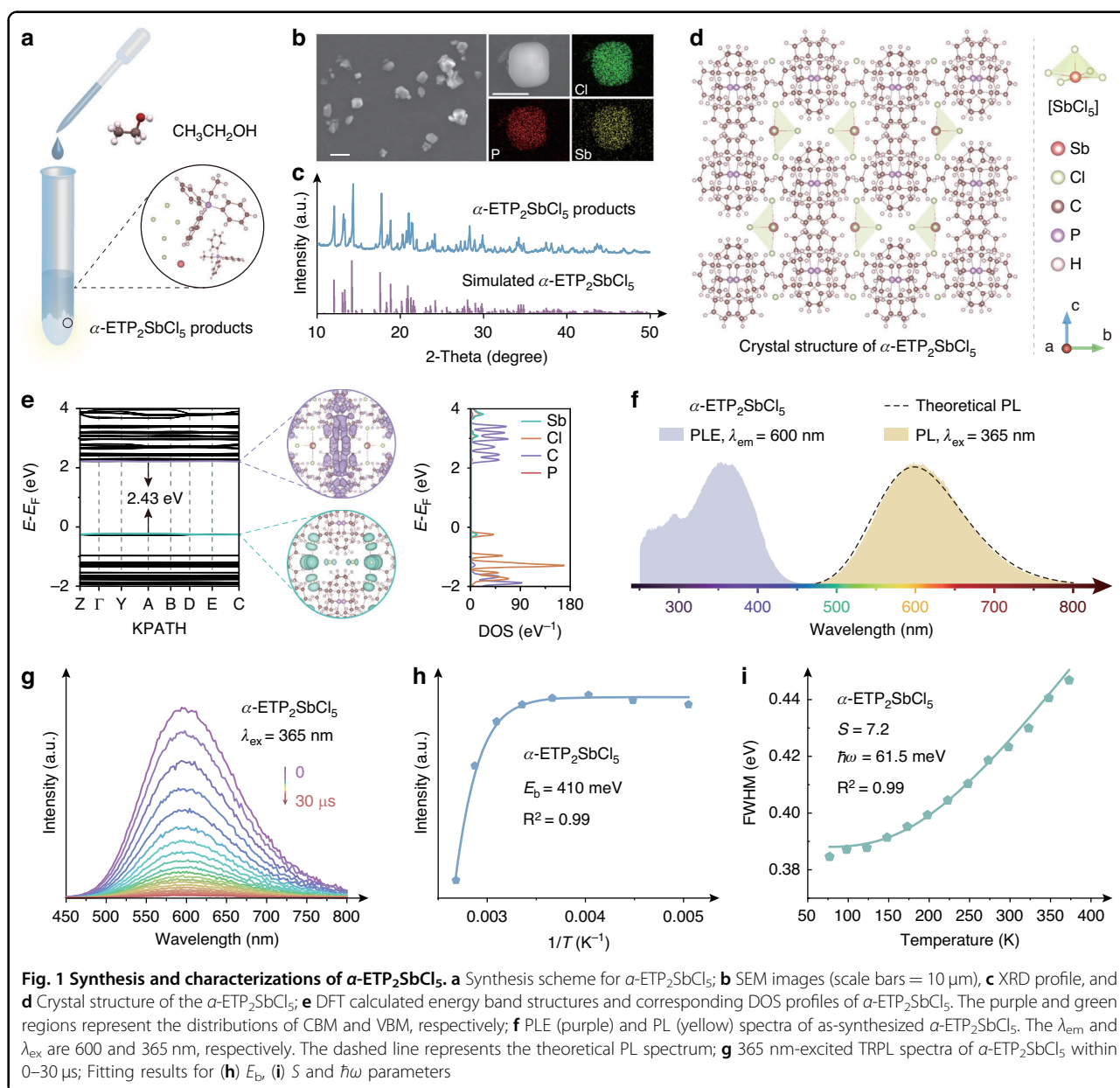
Preparation and fluorescence of α -ETP₂SbCl₅

The α -ETP₂SbCl₅ (ETP = (C₆H₅)₃PC₂H₅) powder is synthesized by simply mixing ETPCl and SbCl₃ in

moderate ethanol (Fig. 1a), with a near-100% chemical yield. Similar to our previous report, this simple solution-process strategy greatly supports rapid and mass preparation for green industrialization³³. The as-synthesized α -ETP₂SbCl₅ exhibits a main particle size of $\sim 1\text{--}7\text{ }\mu\text{m}$ (Fig. 1b and Supplementary Fig. S1a–c), with uniformly distributed P, Sb, and Cl elements. X-ray diffraction (XRD) patterns of the α -ETP₂SbCl₅ products are consistent with that of the standard crystal (Fig. 1c, Supplementary Fig. S2a and Table S2)^{20,21}, suggesting a uniformly pure phase and the reliability of our strategy. The crystal structure of the α -ETP₂SbCl₅ is shown in Fig. 1d, where the Sb atoms bond with Cl atoms thus forming [SbCl₅] pyramids, and they are separated by the organic units (ETP) to form perovskite derivative with a zero-dimensional structure. Figure 1e shows the energy band structure and density of state (DOS) of α -ETP₂SbCl₅ by density functional theory (DFT), in which the valence band maximum (VBM) is mainly consistent with Sb and Cl atoms, that is the [SbCl₅] pyramids, suggesting the electrons in [SbCl₅] pyramids could absorb the photonic energy for excitation. The conduction band minimum (CBM) is mainly from C atoms of organic units, and the formed energy band gap with VBM is $\sim 2.43\text{ eV}$. The VBM and CBM are both flat, inducing large effective masses of both holes and electrons, which lays a foundation for self-trapped excitons (STE)-based emission^{34–36}.

As shown in Fig. 1f, under the photoluminescence excitation (PLE) of 365 nm, the α -ETP₂SbCl₅ products emit a bright yellow light covering from ~ 470 to $\sim 800\text{ nm}$ (central wavelength: $\sim 600\text{ nm}$; PLQY: $97.2 \pm 2.6\%$), which should be assigned to the triplet STEs of Sb³⁺ ions, specifically ³P₁ $\rightarrow$ ¹S₀ transition^{37,38}, consistent with the theoretical STE emission behavior. Furthermore, photoluminescence (PL) bands at ~ 330 and $\sim 450\text{ nm}$ could be detected under a 280 nm excitation, which could be assigned to the transitions of the band gap and the organic ETP, respectively (Supplementary Fig. S1d–f). The time-resolved PL (TRPL) profiles show consistent spectral shapes and central wavelengths (Fig. 1g and Supplementary Fig. S1g), further indicating the pure luminous center of [SbCl₅] pyramids under 365 nm excitation, without obvious defects. With the assistance of temperature-dependent PL profiles (Supplementary Fig. S1h, i), the exciton binding energy (E_b) could be fitted as $\sim 410\text{ meV}$ (Fig. 1h) by Formula 1 (T is the testing temperature. $I(T)$ and I_0 represent the integrated emission intensities at T and 0 K, respectively. A and k_b are the constant and Boltzmann constant, respectively.), which is much higher than 26 meV, suggesting the preference of free carriers to form excitons for luminescence at room temperature^{37,39,40}. Huang-Rhys factor (S) is commonly used for qualifying the electron-phonon coupling strength, which could be calculated by Formula 2 (ref. 41). The $\hbar\omega$





represents phonon energy, in connection with the excited state and self-trapping time, and the T is temperature. The S value of α -ETP₂SbCl₅ is estimated as ~ 7.2 through fitting temperature dependence of full width at half maximum (FWHM), shown in Fig. 1i, which denotes an effective STE emission (Supplementary Fig. S3), corresponding to its PL behaviors described above⁴².

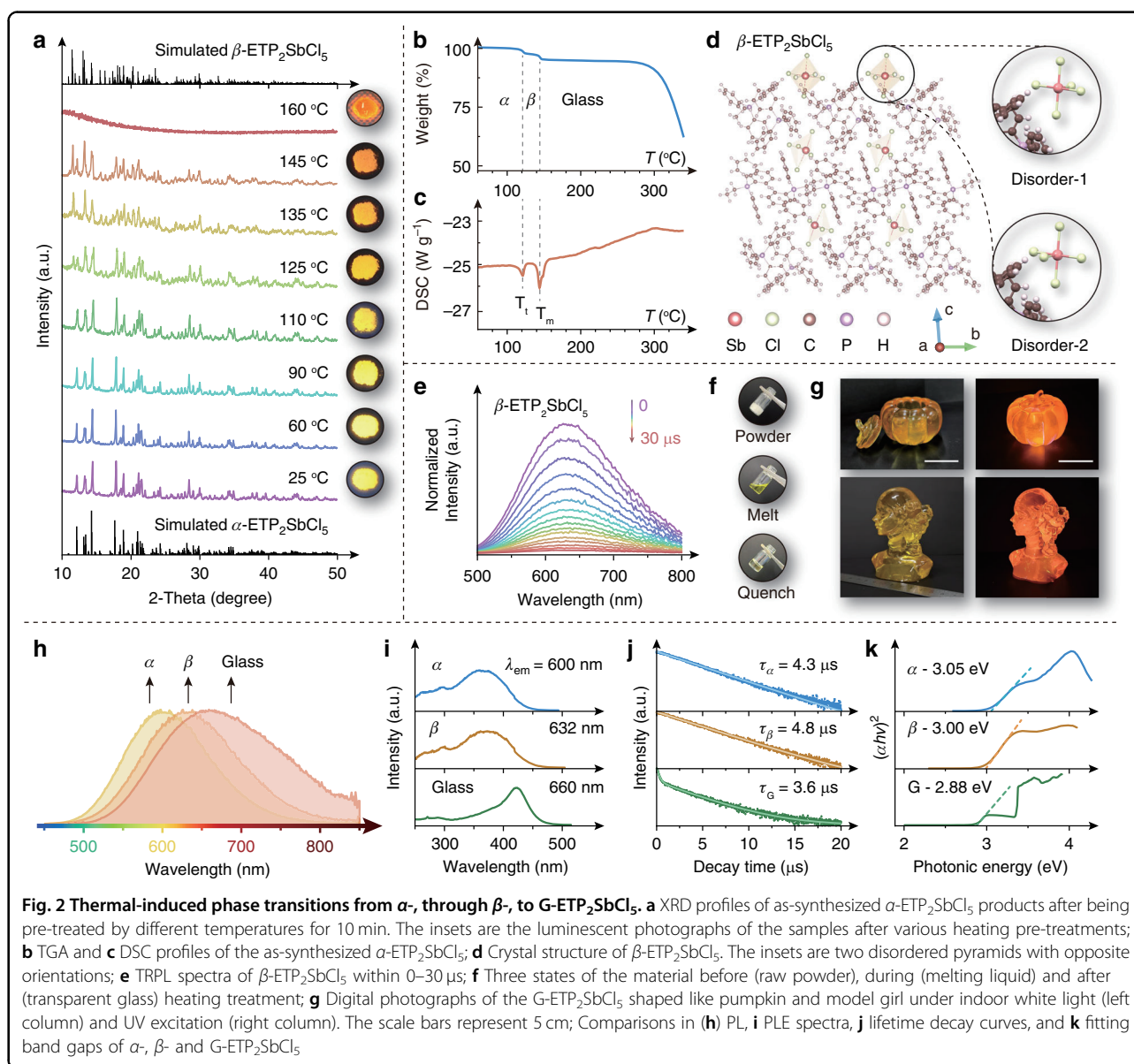
$$I(T) = \frac{I_0}{1 + Ae^{-E_b/k_bT}} \quad (1)$$

$$FWHM = 2.36\sqrt{S\hbar\omega_{\text{phonon}}}\sqrt{\coth\frac{\hbar\omega_{\text{phonon}}}{2k_bT}} \quad (2)$$

Phase transition from α -, through β -, to G-ETP₂SbCl₅

The XRD patterns in Fig. 2a show that, the α -ETP₂SbCl₅ products are relatively stable below the pre-treated temperature of $\sim 110^\circ\text{C}$. However, additional diffraction patterns appear while further increasing the pre-treated temperature to $\sim 125^\circ\text{C}$, which should be assigned to the new phase of β -ETP₂SbCl₅, and it becomes the only remaining phase after pre-heated at 135°C for 10 min, denoting the complete phase transition from α - to β -ETP₂SbCl₅ (Supplementary Fig. S2b and Table S2). This phase transition could also be tracked by thermogravimetric analysis (TGA), shown as the first step in Fig. 2b (mass loss $\sim 2\%$), during which the sample absorbs thermal energy and exhibits a negative differential scanning





calorimetry (DSC) peak, shown as the signal at $T_{\text{transition}}$ (called T_t hereafter, $T_t \sim 121^\circ\text{C}$) in Fig. 2c. In the crystal structure of the β -ETP₂SbCl₅, Sb still maintains [SbCl₅] pyramid configuration, but with a disordered orientation. For instance, in Fig. 2d, the edge of the crystal appears two octahedrons, while each of them is actually the superposition of two possibilities, namely disorder-1 and -2 with opposite orientations, and the statistical probability that a Cl atom occupies a symmetric position is 1/2. Nevertheless, in TRPL spectra, β -ETP₂SbCl₅ still exhibits uniform orange-yellow emissions with central wavelengths of ~ 632 nm (Fig. 2e and Supplementary Fig. S4a, b), probably due to the similar configurations including the lengths and angles of Sb-Cl bonds between each [SbCl₅] pyramid. The β -ETP₂SbCl₅ exhibits a similar high

PLQY of $(96.5 \pm 3.7)\%$ and a slightly higher S value of ~ 9.7 (Supplementary Fig. S4c–f), suggesting a little stronger electron-phonon coupling strength^{39–41}, which could be one of the reasons for the red-shifted band from ~ 600 to ~ 632 nm, as well as other slight changes including red-shifted PLE spectra (from ~ 365 to ~ 368 nm), prolonged lifetime (from ~ 4.3 to ~ 4.8 μ s), and narrower band gap (experimentally from ~ 3.05 to ~ 3.00 eV, and theoretically from ~ 2.43 to ~ 2.42 eV), shown in Fig. 2i–k and Supplementary Fig. S4g, h.

Once further increasing the temperature up to $T_{\text{melting}} \sim 144^\circ\text{C}$ (called T_m hereafter), the sample would undergo the second thermal absorption, shown as the second step of TGA curve in Fig. 2b (mass losses are $\sim 5\%$ and $\sim 3\%$ compared to the α - and β -phase, respectively)



and the second negative DSC signal in Fig. 2c. The melting liquid shows no emission property, while it becomes a transparent solid (Fig. 2f) with a bright orange-red emission but without any XRD peaks after natural quenching to room temperature (Fig. 2a, 160 °C pre-heated condition), illustrating the formation of the amorphous luminescent ETP₂SbCl₅ glass (G-ETP₂SbCl₅). Thanks to the great fluidity of the melting liquid, it could be designed into any complex 3D shapes utilizing customized silicone molds, such as a pumpkin or a model girl, as shown in Fig. 2g, indicating a potential application in the field of 3D luminescent mold.

The G-ETP₂SbCl₅ shows further red-shift in PL spectra, with a central wavelength of ~660 nm (Fig. 2h and Supplementary Fig. S5a, b), a broader FWHM value (α -0.42 eV, β -0.43 eV, G-0.52 eV), and a decreased PLQY of (52.6 ± 3.3)%. Additionally, the PLE spectrum of G-ETP₂SbCl₅ has also been extended and shows obvious red-shift with the highest peak at ~413 nm, shown in Fig. 2i. The G-ETP₂SbCl₅ still possesses an E_b of 231 > 25 meV, with a further increased S value of ~24.2 (Supplementary Fig. S5c–f), indicating a further enhanced electron-phonon coupling strength^{39–41}. Furthermore, compared with the fluorescent lifetimes of α - and β -phase, the G-ETP₂SbCl₅ appears a double exponential decay behavior with a lifetime decreases to ~3.6 μ s (Fig. 2j). This may be attributed by defects that induced by disordered organic groups and [SbCl₅] octahedrons, which will be discussed in the following text), and its band gap further shortens to ~2.88 eV (Fig. 2k). These phenomena demonstrate the micro-structure of G-ETP₂SbCl₅ has been greatly transformed by thermal stimulation, and the specific [SbCl₅] pyramids (such as disorder condition and bond length) may differ from the α - and β -ETP₂SbCl₅ powders.

Investigation of structural distortion

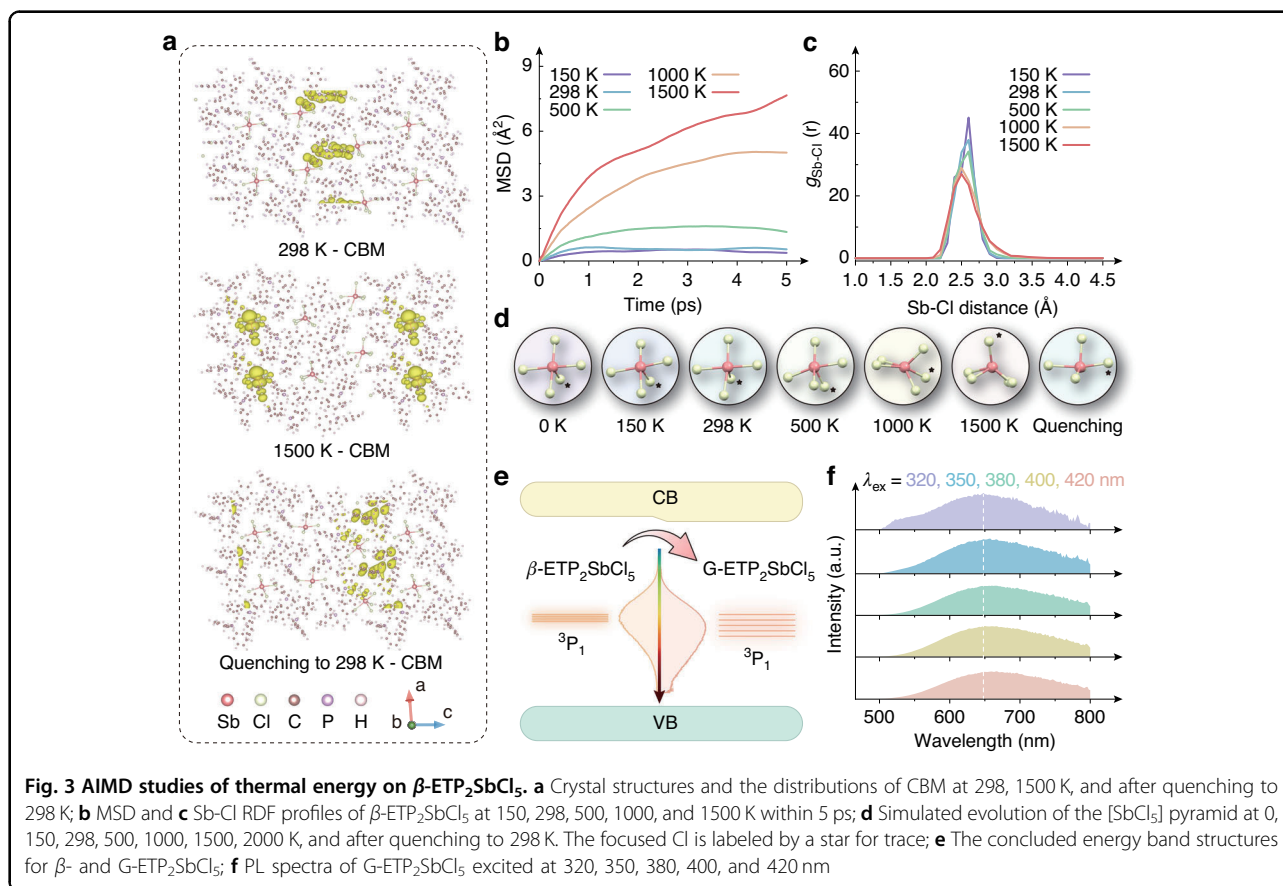
It is noted that, although G-ETP₂SbCl₅ could be obtained by directly heating the α -ETP₂SbCl₅ in a real operation, during which the phase transition through β -ETP₂SbCl₅ is an inevitable microscopic process. We therefore carried out the Ab initio molecular dynamics (AIMD) calculations on the β -ETP₂SbCl₅ lattice to simulate the crystal transition for deeply investigating the specific structure and luminescent mechanism of G-ETP₂SbCl₅. Since the simulation time (ps-scale) is far less than the actual condition (min-scale), the temperatures are set as 150–1500 K in our AIMD calculation for accelerating the molecular dynamics, thereby simulating long-term macroscopic phenomena^{43,44}. The mean squared displacement (MSD) is first simulated at 150–1500 K, as shown in Fig. 3b. The curves of 150–1000 K exhibit flat trends with time, suggesting that the lattice could reach a relatively balanced state, that is,

the sample may still maintain the powder state. While the MSD at 1500 K shows a monotonous increase trend in 5 ps, indicating the movement of particles may be free from the morphological constraints of powders, which could be considered as a near-molten state^{45,46}.

The simulated crystal structures of β -ETP₂SbCl₅ at different temperatures are shown in Fig. 3a and Supplementary Fig. S6, S7. In the range of 0–500 K, the organic molecular units and [SbCl₅] pyramids show a gradually distorted character with increasing temperature, along with a slightly decreased band gap (Supplementary Fig. S4g, Fig. S8, and Table S3, S4), still their overall arrangement and orientation do not change significantly (Videos S1–S3). As the temperature further increases to 1000–1500 K, the vibration of each atom is no longer limited to its original site, and the various components appear obvious distortions. For example, in one [SbCl₅] close-up in Fig. 3d, the focused Cl atom labeled by a star symbol breaks away from the initial position and relocates to a new site at 1000 K, accompanied by similar obvious displacements of 4 other Cl atoms, resulting in a rotation and disorder of the pentagonal pyramid, and this condition could be further aggravated at 1500 K, corresponding to a further disordered lattice (Videos S4, S5). Meanwhile, the band gap dramatically decreases below 2.00 eV, and the compositions of the VBM and CBM appear qualitative changes at 1500 K, both of which are mainly composed of Sb and Cl atoms, shown in Fig. 3a and Supplementary Fig. S9a, suggesting a radical change in the electronic transition method for the melting liquid.

Besides, the FWHMs of the radial distribution function (RDF) curves for Sb–Cl bonds gradually increase with the temperature, as shown in Fig. 3c. Similar broadening properties could also be observed from the RDF curves for Sb–P and Sb–Sb bonds in Supplementary Fig. S9b, c. These phenomena demonstrate that the crystal field strength of Sb³⁺ in the molten state has wider coverage than the powder state, which lays the foundation for the broadened PL spectrum of the quenched G-ETP₂SbCl₅. Note that the slight peak shift towards a short distance direction may be caused by the canonical ensemble (NVT) model with constant volume, which may restrict the lattice expansion thus increasing the pressure on the lattice and indirectly affecting the peak position. In the actual condition, the lattice is supposed to expand with the temperature, which could be deduced from the slight PL blue-shift with increasing temperature (Supplementary Fig. S1i, S4d, and S5d)^{42,47,48}. However, the isothermal-isobaric Ensemble (NPT) model, which maintains constant pressure, is extremely arduous on our servers. Our NVT-based AIMD aims to visualize the possible distortion of [SbCl₅] pyramids during $\beta \rightarrow G$ transition and analyze its subsequent impacts on luminescence.



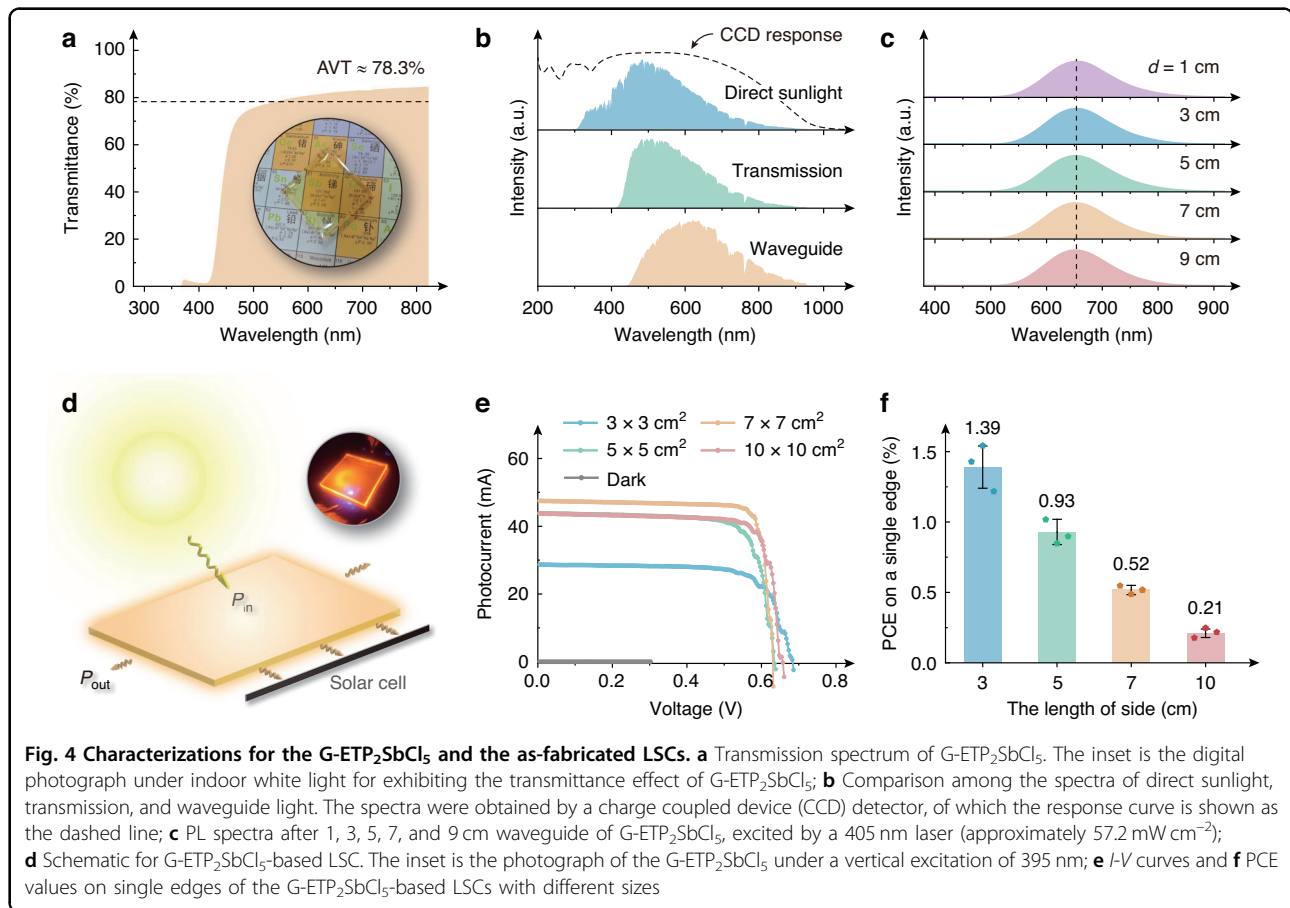


Next, the temperature is reduced to 298 K in a linear method (Supplementary Fig. S9d) for simulating the G-ETP₂SbCl₅. The kinetic energy of each atom gradually decreases, and the rotation of the [SbCl₅] pyramids slows down progressively until it comes to a halt below a certain temperature threshold (see Video S6). Due to the relatively fast natural quenching process, the material could maintain a disordered structure, thereby forming the transparent G-ETP₂SbCl₅ instead of a recrystallized scattering solid (Supplementary Fig. S10). And the aforementioned extended coverage of the crystal field strength could be maintained, thus further increasing the $^3P_1$ splitting degree of Sb³⁺ ions (Fig. 3e and Supplementary Fig. S11)³⁸, which, combined with the narrower band gap in both theoretical and experimental profiles (Fig. 2k and Supplementary Fig. S8b, f), results in the broadened and red-shifted bands in both PL and PLE spectra (Fig. 2h, i). Under 350–420 nm excitation, the G-ETP₂SbCl₅ exhibits consistent PL behaviors, as shown in Fig. 3f. While, in addition to the organic emissive band at ~520 nm, a slight blue shift of the orange-emissive band could also be observed under 320 nm excitation, which may be attributed to some local defects formed during the quenching process. These defects could also be one of the main reasons for the shortened lifetime and decreased PLQY of G-ETP₂SbCl₅.

Characteristics and performance of LSCs

The G-ETP₂SbCl₅ is then shaped into square glasses with 3, 5, 7, and 10 cm side lengths and ~0.5 cm thickness (Supplementary Fig. S12a, b) with the assistance of silicone molds. As shown in Fig. 4a, the G-ETP₂SbCl₅ exhibits great average visible transmittance (AVT, see Formula 3, ref. ⁴⁹). The $T(\lambda)$ is the transmittance of the glass. AM1.5 G(λ) represents the standard solar spectrum, and the $V(\lambda)$ is the vision function curve, shown in Supplementary Fig. S12c) of ~78.3%, which could greatly guarantee the characteristics of transparent glass. On the basis that the PL range meets the external quantum efficiency (EQE) response of Si cells, the G-ETP₂SbCl₅ with a broader PLE band shows remarkable absorption performance for the ultraviolet (UV) light (~300–420 nm, Fig. 4a and Supplementary Fig. S12d, e), thus near-zero UV light could be detected in the solar spectrum after transmission (Fig. 4b). Then the correlated color temperature of the sunlight could be decreased from ~7556 K to ~5616 K (Supplementary Fig. S12f), leading to a softer warm-white transmission light for indoors. This fascinating property endows it with a competitive ability to block UV radiation therefore adequately protecting the human eyes and skin in practical LSC applications. On the other hand, it could also suppress the irreversible





performance degradation of solar cells by UV radiation⁵⁰.

$$AVT = \frac{\int_{380nm}^{780nm} T(\lambda) \times AM1.5 G(\lambda) \times V(\lambda) d\lambda}{\int_{380nm}^{780nm} AM1.5 G(\lambda) \times V(\lambda) d\lambda} \quad (3)$$

Thanks to the effective waveguide behavior, the STE emission of Sb³⁺ could be conducted to the edges (Supplementary Fig. S13a–c), with the measured PLQY_{edge} values of 41.7% on 3 × 3 cm² (Table 1). Although it gradually decreases with the lateral size due to the inevitable photon escape (Supplementary Fig. S13d), the PLQY_{edge} of the 7 × 7 cm² glass could still maintain 30.4%, which is ~57.8% of total G-ETP₂SbCl₅, demonstrating a reliable edge output performance (The PLQY measurement for 10 × 10 cm² glass is not available because of the size limitation of the integrating sphere). Furthermore, due to the large Stokes shift of ~240 nm, nearly no overlap could be observed between PL and PLE spectra, therefore the G-ETP₂SbCl₅ glass exhibits consistent spectra under a 405 nm UV laser without obvious self-absorption even with 1–9 cm of the optical waveguide distance, shown as “d” in Fig. 4c and Supplementary Fig. S13d. Besides the PL, a part of the sunlight could also be conducted to the

edges. Hence, the resulting waveguide spectrum represents a combination of the emission from G-ETP₂SbCl₅ material that is excited by the UV portion of sunlight, along with the direct waveguide of the visible to near-infrared region of the solar spectrum (Fig. 4b, bottom).

The G-ETP₂SbCl₅-based LSC prototypes are then fabricated by coupling commercial Si solar cells on a single edge of each G-ETP₂SbCl₅ glass, shown in Fig. 4d. Under the irradiation of the standard solar simulator (AM1.5 G, 100 mW cm⁻²), the 3 × 3 cm² LSC reaches a short-circuit photocurrent (*I*_{LSC}) of ~28.2 mA, with the highest power conversion efficiency (PCE) of (1.39 ± 0.16)%. It is noted that the *I*-*V* curves, output powers, and PCEs in Fig. 4e–f and Supplementary Fig. S13e are depicted by solar cells on single edges of the fluorescent glasses. It is reasonable to consider that the other three edges could output similar powers, then the total PCEs could be calculated by four folds of a single cell⁵¹, and the specific parameters are shown in Table 1. Among them, the optical efficiency (*η*_{opt}) of the fluorescent glass, which is commonly defined as the ratio of the total number of emitted photons reaching the lightguide edge to the total number of solar photons incident onto the lightguide front surface (Formula 4, ref. 52–54), also achieved the highest value of



Table 1 The parameters of the as-fabricated LSCs

Size (cm ³)	PLQY _{edge} (%)	Single I_{LSC} (mA)	FF (%)	Single PCE (%)	Total PCE (%)	$\eta_{opt.}$ (%)
3 × 3 × 0.5	41.7	28.2	73.5	1.39 ± 0.16	5.56	32.5
5 × 5 × 0.5	35.9	43.8	77.0	0.93 ± 0.09	3.72	17.9
7 × 7 × 0.5	30.4	47.5	76.5	0.52 ± 0.03	2.08	9.9
10 × 10 × 0.5	—	43.7	77.4	0.21 ± 0.02	0.84	5.2

~32.5% on the 3 × 3 cm² LSC (see Supplementary Table S5 and Note for performance metrics and their derivation).

$$\eta_{opt.} = \frac{I_{LSC} \times A_{edge}}{I_{PV} \times A_{LSC}} \quad (4)$$

Where the I_{LSC} and I_{PV} represent the total short-circuit current of LSCs and Si solar cells, respectively, and the A_{LSC} and A_{edge} refer to the surfaces and total edge areas of LSCs, respectively. Similar to the previous reports^{1,7,10}, the PCE gradually decreases with the increase of lateral size, and it is not only affected by the decreased $\eta_{opt.}$ induced by photon escape, but also by the declined PCE_{PV} of Si solar cells in larger areas (Supplementary Fig. S13f, g and Table S4). Nevertheless, the LSC with a large scale of 10 × 10 × 0.5 cm³ still maintains PCE and $\eta_{opt.}$ of ~0.84% and ~5.2%, respectively, without any optical amplifiers or extra waveguide media, which exhibits comparable performance to the previously reported LSCs (Supplementary Table S6), suggesting the feasibility of LSCs by using such fluorescent glass-formed lead-free perovskite derivatives. Additionally, as shown in Supplementary Fig. S14, we have further prepared another green emissive glass ETP₂MnBr₄ with a higher PLQY of ~70%, and the as-fabricated 10 × 10 × 0.5 cm³ ETP₂MnBr₄-based LSC exhibits total PCE and $\eta_{opt.}$ of ~0.72% and ~3.9%, respectively. Such a slightly lower LSC performance than that based on G-ETP₂SbCl₅ is probably due to the obvious self-absorption induced by the overlap of PL and PLE spectra. These extended results further prove the importance of a large Stokes shift for LSC, and on the other hand, demonstrate the great extensibility of fluorescent glass-based LSCs.

Self-healing and recyclable repurposing

One attraction is that this kind of fluorescent glass could realize self-healing or re-shaping even if damaged, which possesses a much lower healing temperature (~150–250 °C) than conventional inorganic glasses (e.g. > 800 °C, ref. ⁵⁵), shown as processes 4 and 5 in Fig. 5a. The PL performance of G-ETP₂SbCl₅ exhibits nearly no attenuations during 10 damage-healing cycles (Fig. 5b–c). More intriguingly, the β - and G-ETP₂SbCl₅ could be

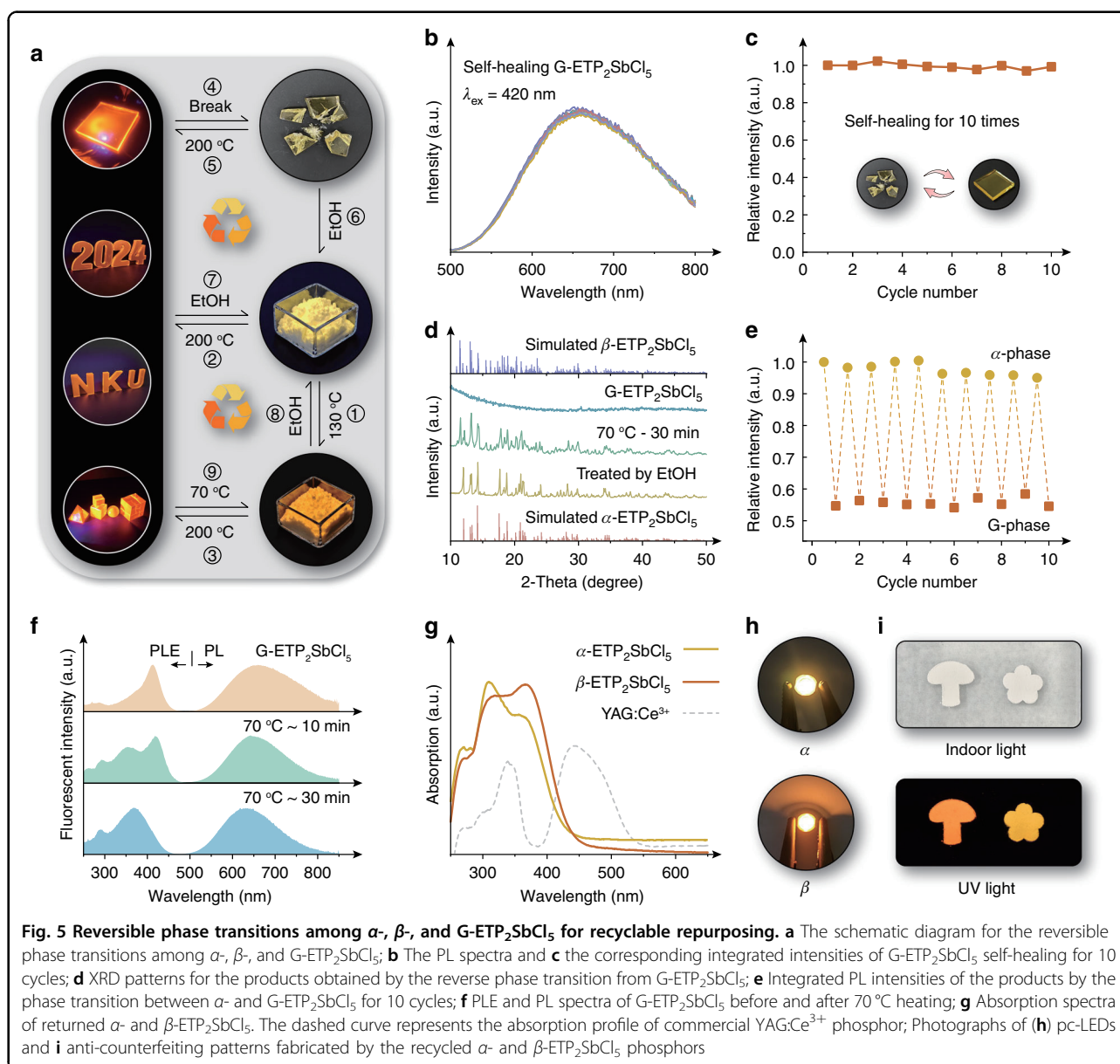
restored to the initial α -ETP₂SbCl₅ powders in mass production after adding moderate ethanol, with a transition yield of >82.5%. Such a remarkable recyclable ratio may be attributed to the ethanol-assisted dissolving and recrystallization processes, shown in Fig. 5a (processes 6–8), Fig. 5d, and Supplementary Fig. S15a (inset). This reversible phase transition exhibits great fatigue resistance, and the recycled α -ETP₂SbCl₅ could still maintain ~95% of the fresh PL intensity even after 10 consecutive α -G transitions (Fig. 5e and Supplementary Fig. S15a). Furthermore, β -ETP₂SbCl₅ could also be re-obtained by heating G-ETP₂SbCl₅ at 70 °C for ~30 min (Fig. 5d), during which the recrystallization of the amorphous phase is initiated and the uniform [SbCl₅] pyramids are re-produced (Fig. 5f), corresponding to the transition from a transparent glass to a non-transparent scattering solid.

Based on the aforementioned results, we have successfully demonstrated the reversible phase transitions among α -, β -, and G-ETP₂SbCl₅ (Fig. 5a), which significantly facilitates the resource recovery and repurposing even if the LSCs are damaged or decommissioned. The recycled phosphors still retain near-unity PLQYs and exhibit remarkable absorption intensities, surpassing that of the commercial YAG:Ce³⁺ (PLQY ~ 80–85%, Fig. 5g). These features position the recycled phosphors for further utilization in a variety of applications, such as phosphor converted LEDs (pc-LEDs), anti-counterfeiting, and luxury decoration, etc., on par with freshly synthesized phosphors (Fig. 5h, i and Supplementary Fig. S15b–d).

Discussion

The stability of the G-ETP₂SbCl₅ is also evaluated in this work, shown in Supplementary Fig. S16, S17. Under sunlight irradiation in air, the G-ETP₂SbCl₅ begins to show slight recrystallization by the 10th day, and it becomes more pronounced by the 20th day (Supplementary Fig. S16a), which could be attributed to the influence of the humidity. To improve its air-stability, we have further fabricated the laminated G-ETP₂SbCl₅ glass by quenching G-ETP₂SbCl₅ between two quartz slides, as shown in Supplementary Fig. S16b. The laminated glass shows consistent transmittance spectra after exposure to sunlight in air for 20 days, without any significant recrystallization. As shown in Supplementary Fig. S16c, d, the laminated G-ETP₂SbCl₅ exhibits





similar PL and PLE spectra after sunlight irradiation in air for 20 days, with its PLQY remaining virtually unchanged. On the other hand, this laminated approach could also prevent direct mechanical impact, thereby addressing the issue of fragile in practical applications, which is therefore considered as an effective engineering optimization solution. Furthermore, the G-ETP₂SbCl₅ also exhibits a reliable stability under long-term mild heating at 40–50 °C (Supplementary Fig. S17), which make it a candidate for future recyclable LSC application.

In summary, this study has presented the preparation of hybrid lead-free perovskite derivatives ETP₂SbCl₅, which exhibits reversible α -, β -, and G-phase, exhibiting remarkable PLQYs of (97.2 ± 2.6)%, (98.5 ± 3.2)% and

(52.6 ± 3.3)%, respectively. Through AIMD and DFT simulations, we have demonstrated the disordered [SbCl₅] pyramids with more complex Sb–Cl bonds and shortened band gap of G-ETP₂SbCl₅. These changes result in broadened and red-shifted PL and PLE bands, enabling it to effectively block the entire UV region (~300–420 nm) of sunlight for STE luminescence. The inaugural series of self-healing G-ETP₂SbCl₅-based LSCs have achieved the highest PCE and η_{opt} of ~5.56% and ~32.5%, respectively, on the 3 × 3 × 0.5 cm³ glass. Despite a decrease in PCE with increasing surface area, such novel fluorescent glass-based LSC prototypes, devoid of additional wave-guide medium, have demonstrated performance on par with prior reports. Moreover, the potential for



repurposing G-ETP₂SbCl₅ has been validated through the successful fabrication of pc-LEDs and anti-counterfeiting patterns by using recycled α - and β -ETP₂SbCl₅ phosphors from LSCs, underscoring its role in sustainable energy solutions for the low-carbon era.

Materials and methods

Raw materials

Ethyl-triphenylphosphonium chloride (ETPCL, 98%), Ethyl-triphenylphosphonium bromide (ETPBr, 98%), SbCl₃ (99.9%), and MnBr₂·H₂O (98%) were purchased from Macklin and were used directly for synthesis without further purification.

Preparation of α -ETP₂SbCl₅ powder

To synthesize 1 mmol of α -ETP₂SbCl₅ powder, 2 mmol of ETPCL (0.6536 g) and 1 mmol of SbCl₃ (0.2281 g) were weighed and transferred into a tube. Subsequently, 5 mL of ethanol was added to the mixture. The reaction mixture was vigorously stirred for approximately 5 minutes, with intermittent ultrasonic treatment (*e.g.* ultrasonication for 30 s after every 60 s of stirring) to facilitate the reaction process, resulting in the formation of a homogeneous target product.

Preparation of β -ETP₂SbCl₅ powder

Heating the as-synthesized α -ETP₂SbCl₅ powder at 130 °C for 10 min, results in the gradual color change from white to pale yellow, suggesting the formation of β -ETP₂SbCl₅ powder.

Preparation of G-ETP₂SbCl₅ glass

Heating the as-synthesized α - or β -ETP₂SbCl₅ powder to 200 °C for 30 min results in the complete melting of the powders liquid, which was poured into a silicone mold with a specific shape and quenched naturally to room temperature. The transparent G-ETP₂SbCl₅ could be obtained after completely quenching.

Reverse phase transition from β - to α -ETP₂SbCl₅

Mixing moderate ethanol with the β -ETP₂SbCl₅ sample (*e.g.* 5 mL for 1 g of sample, stirring for 1 min) drives the phase transition from β - to α -ETP₂SbCl₅.

Reverse phase transition from G- to α -ETP₂SbCl₅

Mixing moderate ethanol with the G-ETP₂SbCl₅ sample (*e.g.* 5 mL for 1 g of sample) and vigorously stirring the mixture for 30 min affords the recycled α -ETP₂SbCl₅ phosphors. It could be accompanied by grinding, for a faster reverse transition.

Reverse phase transition from G- to β -ETP₂SbCl₅

To observe the reverse phase transition process, we heated the G-ETP₂SbCl₅ at 70 °C, and collected PL

spectra over a period of 30 min. To achieve a rapid and complete reverse transition from G- to β -phase, the glass should be ground into a powder and then heated at 70 °C for 60 min or longer.

Preparation of ETP₂MnBr₄ powder and glass

The preparation of ETP₂MnBr₄ powder is similar to the α -ETP₂SbCl₅, while the ETP₂MnBr₄ glass is obtained by heating ETP₂MnBr₄ powder at 200 °C for 20 min and quenching to room temperature.

Fabrication of pc-LED devices

The pc-LED was fabricated by the recycled α -, β -ETP₂SbCl₅ phosphors and commercial 365 nm LED chip (3 W). The mass ratio of curing glue and phosphor is 5:1, and the dispensed pc-LED was irradiated by a 5 W, 395 nm light source for 30 s to complete curing.

Characterizations

The XRD patterns were acquired utilizing an Ultima X-ray diffractometer from Rigaku Corporation, Japan. A Cu K α X-ray source, with a wavelength of 1.5405 Å, was employed at an operational voltage and current of 40 kV and 40 mA, respectively. The diffractometer scanned samples at a rate of 10 degrees per minute. Morphological analysis and elemental mapping were conducted on the JSM-7800F (Japan), which was integrated with an EDS system for elemental distribution studies. The TGA and DSC curves were detected by a Simultaneous Thermal Analyzer from Mettler TGA/DSC3+ (Switzerland). The index of refraction was obtained by an ellipsometry from J.A. Woollam Co., Inc. RC-2.

The PLE, PL, PLQY, TRPL spectra and luminescence lifetime decay curves were recorded using a FLS-1000 spectrofluorometer (equipped with a 300 W Xenon lamp) from Edinburgh Instruments, England. For validation, a duplicate set of measurements was performed on an FS-5 spectrofluorometer (equipped with a 300 W Xenon lamp) from the same manufacturer. The measurement systems were calibrated against a commercially available YAG:Ce³⁺ phosphor, which serves as a standard with a known PLQY of ~80%. The specific PLQY values could be calculated by Formula 5, where the numerator and denominator represent the integrated emissive photons and the absorption photons of the sample, respectively. Specifically, the I_{PL} is the wavelength dependence of emission intensity, while the I_{sample} and I_{BG} are the excitation light intensity with and without sample, respectively⁵⁶. For fluorescent glasses, the PLQY_{edge} values are calculated by Formula 6, where the PLQY_{surface} and PLQY_{total} represent the PLQY values of fluorescent glasses with and without black taped edges. During the measurements, the excitation light beam was controlled to be incident on the center point of the fluorescent glass



surface.

$$PLQY = \frac{\int I_{PL} d\lambda}{\int I_{BG} - I_{sample} d\lambda} \quad (5)$$

$$PLQY_{edge} = PLQY_{total} - PLQY_{surface} \quad (6)$$

Temperature-dependent PL spectra were generated with the Aurora 4000, from GE-UV-NIR, Changchun New Industries Optoelectronics Tech. Co., Ltd, paired with a temperature control module, HCS421VXY, provided by Instec, Shanghai Hengshang Precision Instrument Co., LTD. The absorption spectra for all samples were determined using a UV-2600 spectrophotometer from Shimadzu Corporation, Japan.

The *I-V* characteristics and the corresponding PCEs of the fabricated LSCs were evaluated under a dual-light source solar simulator, conforming to the Wacom Class AAA standards, which simulates AM1.5 G conditions at 25 °C with an irradiance of 100 mW cm⁻². The corresponding *I-V* curves were monitored using a ADCMT 6246 source meter.

For the pc-LED devices measurements, a glovebox environment was employed to prevent environmental interference. The emission spectra of the pc-LED devices were measured with an integrating sphere (FOIS-1) from Ocean Optics, connected to a spectrophotometer (QE65 Pro). The electrical current-voltage characteristics were monitored using a Keithley 2400 source meter, with measurements taken at increments of 25 mA cm⁻². The pc-LED devices were directly characterized without any radiator.

Digital images of the samples and luminescence in this paper were all performed using a smartphone camera (vivo X200 pro mini, default mode).

DFT calculation

The Vienna Ab-initio Simulation Package (VASP) software was used to accomplish the DFT and AIMD calculations^{57–59}. The projector augmented wave (PAW) method⁶⁰ with the Perdew-Burke-Ernzerhof (PBE) functional⁶¹ was selected and the plane-wave cutoff energy was set to 500 eV. Geometric structures were fully relaxed until the energy and total forces were converged to 10⁻⁵ eV and 0.01 eV Å⁻¹, respectively. The Brillouin zone was sampled by the Γ -centered Monkhorst-Pack method with a *k*-spacing of 0.04 π Å⁻¹ (ref. ⁶²). Twenty points were inserted between every two high symmetry points for the band structure.

AIMD simulation

AIMD calculations were implemented at 150, 298, 500, 1000, and 1500 K based on the canonical ensemble

(NVT) and Nosé-Hoover thermostat algorithm^{63–65}. The AIMD simulation lasted 5 ps, with a time step of 1 fs. The quenching process was based on the same step and total time setting, and the temperature was set by Formula 7.

$$T = T_{begin} + (T_{end} - T_{begin}) \times \frac{N_{step}}{N_{total}} \quad (7)$$

Where the N_{STEP} is the current step, the N_{total} is the total step, and the T_{begin} and T_{end} represent the starting (1500 K) and ending temperature (298 K), respectively. The electronic structures of models subjected to molecular dynamics relaxation at various temperatures were all calculated. VASPKIT code was used for all post-processing analysis⁶⁶. The nonlinear MSD curves were caused by restricted behavior different from Brownian diffusion⁶⁷.

Calculation of STE spectra

The delta self-consistent field (Δ SCF) method was used to calculate energies of excited states^{68,69}, and ionic relaxation was further performed with fixed electron occupancy based on the ground state configuration, while spin polarization was applied. It can be described by replacing Formula 8 with Formula 9 of one spin state, where $n(\mathbf{r})$ is the density, N is the number of total electrons, and $\psi(\mathbf{r})$ is the KS orbital of the i -th electron and the orbital where CBM calculated under the ground-state was located⁷⁰. To evaluate the influence of electron-phonon coupling, finite differences with symmetry method was applied. A one-shot sampling (ZG configuration⁷¹) based on the structure after relaxation with Δ SCF was employed to obtain a single distorted structure under 300 K for each phase of materials. The frequency dependent dielectric matrix was finally calculated based on the got ZG configuration, as the real part of the dielectric tensor is obtained by the usual Kramers-Kronig transformation (Formula 10), and the imaginary part is determined by Formula 11, where the c refers to conduction band states, and v refers to valence band states, u_{ck} is the cell periodic part of the orbitals at the k -point $\mathbf{k}$, P is the principle value⁷².

$$n(\mathbf{r}) = \sum_{i=1}^N \psi_i^*(\mathbf{r}) \psi_i(\mathbf{r}) \quad (8)$$

$$n(\mathbf{r}) = \sum_{i=1}^{N-1} \psi_i^*(\mathbf{r}) \psi_i(\mathbf{r}) + \psi_{CBM}^*(\mathbf{r}) \psi_{CBM}(\mathbf{r}) \quad (9)$$

$$\epsilon_{\alpha\beta}^{(1)}(\omega) = 1 + \frac{2}{\pi} P \int_0^\infty \frac{\epsilon_{\alpha\beta}^{(2)}(\omega') \omega'}{\omega'^2 - \omega^2 + i\eta} d\omega' \quad (10)$$



$$\varepsilon_{\alpha\beta}^{(2)}(\omega) = \frac{4\pi^2 e^2}{\Omega} \lim_{q \rightarrow 0} \frac{1}{q^2} \sum_{c,v,k} 2w_k \delta(\varepsilon_{ck} - \varepsilon_{vk} - \omega) \times \langle u_{ck+e_{\alpha}q} | u_{vk} \rangle \langle u_{ck+e_{\beta}q} | u_{vk} \rangle^* \quad (11)$$

Waveguide simulation

The two-dimensional simulation on the waves propagation in the luminescent layer was performed by COMSOL Multiphysics, in which the wave optics module was adopted. The impedance and matching boundary condition, which were generally used for the calculation of total internal reflection in a waveguide, were set in four directions. To ensure the total reflection at the interface between the luminescent layer and the air layer, the incident angle should be larger than 42°. For the luminescence in all the directions, more than half of the light rays satisfy the total reflection condition, hence could be restricted and propagate in the above discussed waveguide. A fine rectangular mesh with an element size of one-fifteenth wavelength ($\lambda/15$) were used for the whole computational domain to guarantee the accuracy of numerical results.

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Author contributions

H.Y. and X.L. carried out the project and wrote the paper. X.L. supervised this project. H.Y. performed the experiments and analyzed the data. H.L. and G.L. performed the DFT, AIMD, and theoretical PL calculations. H.Y., X.W., and X.Z. characterized the performance of LSCs. W.S. and M.Y. measured the performance of pc-LED devices. Y.Y. carried out the waveguide simulation. H.Y., W.X., and J.X. performed the temperature-dependent PL spectra. All of the authors discussed the results and approved the final version of the manuscript.

Data availability

The data that support these findings are available from the corresponding author upon request.

Conflict of interest

The authors declare no competing interests.

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