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# Pressure treatment enables white-light emission in Zn-IPA MOF via asymmetrical metal-ligand chelate coordination

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Metal-organic frameworks that feature hybrid fluorescence and phosphorescence offer unique advantages in white-emitting communities based on their multiple emission centers and high exciton utilization. However, it poses a substantial challenge to realize superior white-light emission in singlecomponent metal-organic frameworks without encapsulating varying chromophores or integrating multiple phosphor subunits. Here, we achieve a highperformance white-light emission with photoluminescence quantum yield of 81.3% via boosting triplet excitons distribution through pressure treatment in single-component Zn-IPA metal-organic frameworks. A novel metal-ligand asymmetrical chelate coordination is successfully integrated into the Zn-IPA after a high-pressure treatment over ~20.0 GPa. This modification unexpectedly endows the targeted sample with a new emergent electronic state to narrow the singlet-triplet energy gap, which effectively accelerates the spinflipping process for boosted triplet excitons population. Time delay phosphorconverted light-emitting diodes are fabricated with long emission time up to ~7 s after switching off, providing significant advancements for white-light and time-delay lighting applications.

Phosphorescent metal-organic frameworks (MOFs), as one of the most developing functional materials, have witnessed their superiority in recent years based on their promising applications in information encryption<sup>1-4</sup>, chemical sensing<sup>5,6</sup>, optoelectronics<sup>7-9</sup>, drug delivery<sup>10</sup>, and bioimaging<sup>11</sup>. Recently, white-light emission MOFs based on encapsulating varying phosphorescent chromophores have been widely explored and well-studied<sup>12-14</sup>. However, this method has always been limited by challenges in achieving precise color control of various encapsulated phosphors and instability in emission colors<sup>15,16</sup>. Notably, single-component MOFs manifesting fluorescence and phosphorescence can be tailored to emit distinct white light as they could offer intrinsic multiple emitting centers and high exciton utilization, simultaneously<sup>17-19</sup>. However, the insufficient contribution of phosphorescence hinders it toward the balanced tone needed for white light.

Trapping into the lone-pair electrons of ligands, the heavy atom effect of metals, and the rigid frame configuration, single-component MOFs undertake significant promise in promoting spin-orbit coupling and suppressing nonradiative dissipation for a fast spin-flipping process and singlet/triplet-excitons radiative transition. Significantly, a wide range of emission types, including *n*-π<sup>\*</sup> transitions<sup>20</sup>, π-π<sup>\*</sup> transitions, ligand-to-metal charge transfer<sup>21-23</sup>, metal-to-ligand charge transfer<sup>24,25</sup>, ligand-to-ligand charge transfer<sup>26,27</sup>, etc. render the single-component MOFs with regulable singlet-triplet energy gap ( $\Delta E_{ST}$ ), further benefiting the manipulation of intersystem crossing (ISC) process<sup>28,29</sup>. Theoretically, any desired emission could be realized through a subtle design of structural subunits and their topological extended network. However, the immensely multifaceted building blocks and countless possible topologies have made such delicate

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engineer on their chemical configurations quite challenging. Notably, the photophysical processes are highly sensitive to structural variations<sup>30-32</sup>. Through directly manipulating crystal configurations, pressure has fabricated various superior optical materials by electron hybridization and strengthening intermolecular interactions<sup>33–39</sup>. Therefore, the single-component MOFs could further orchestrate high-pressure strategies to regulate the intrinsic ISC process and ultimately meet the balanced tone needed for white light.

In this work, the high-brightness white-light emission was successfully harvested from the original weak blue-light emitted Zn-IPA MOF via high-pressure treatment engineering over ~ 20.0 GPa. The targeted sample was unexpectedly decorated with new asymmetrical metal-ligand chelate coordination, which triggers the generation of a new type transition to fertilize the emission centers and narrow  $\Delta E_{ST}$ for an accelerated ISC process. This remarkably boosted the phosphorescent emission and manipulated the population of singlet and triplet excitons. We thereby acquired a high-performance broadband white-light emission that covered the entire visible range from 350 nm to 800 nm, where the photoluminescence quantum yield (PLQY) reached ~81.3% and Commission International de l'Eclairage (CIE) color coordinate was measured as (0.29, 0.37). Moreover, we further conducted a mild pressure treatment engineer on Zn-IPA MOF within ~11.0 GPa. A greater rigid configuration with strengthened C-H···π interactions was captured in the pressure-treated sample (Zn-IPA-11), which drives a remarkable enhancement of blue-light emission and phosphorescent lifetime prolongation. The PLQY was dramatically enhanced from 20.9% to 87.0% and the phosphorescent lifetime was prominently prolonged from 0.91 s to 1.01 s. We further fabricated the time delay phosphor-converted light-emitting diodes (pc-LEDs) based on these pressure-treated Zn-IPA MOFs with long emission times up to ~7 s after switching off, which preliminarily present a platform for the practical applications of this kind of phosphorescent materials.

## **Results and discussion**

Zn-IPA MOF crystallizes in the tetragonal P41212 space group and adopts a characteristic building unit with four crystallographic independent isophthalic acids (IPA) that coordinate tetrahedral Zn<sup>2+</sup> (Fig. 1a). Benzene rings of two adjacent IPA linkers assemble to form a herringbone geometry with a straight edge-to-face configuration. A prior report on Zn-IPA revealed that the intrinsic coordination interactions and C-H··· $\pi$  interactions can effectively enhance the rigidity of the molecular conformation, which was advantageous to suppress the nonradiative dissipation towards emission enhancement<sup>40</sup>. Theoretically, the Zn-IPA MOF decorated with fluorescent and phosphorescent emitting centers holds great potential in achieving white light. However, we found that the pristine Zn-IPA MOF only exhibited a blue light emission with PLQY of 20.9% irradiated by a 355 nm laser, accompanied by a week green phosphorescence emission at the band tail (~508 nm) with a lifetime of 0.91s (Fig. 1b, c and Supplementary Fig. 1a). The CIE color coordinate of the original sample was estimated to be (0.19, 0.18) (Supplementary Fig. 1b). Currently, the insufficient contribution hinders the fabrication of white light in Zn-IPA. In fact, alongside the advantageous rigidification, those intermolecular interactions still had huge potential for photophysical processes. We, therefore, employed pressure as an external trigger to alter these intermolecular interactions for the sake of boosting the contribution of phosphorescence in Zn-IPA.

A symmetric diamond anvil cell (DAC) was used to investigate the emission properties of Zn-IPA MOF from ambient pressure to 20.2 GPa. The crystal sample was loaded into the DAC together with liquid argon (Ar) as a pressure-transmitting medium (PTM). In situ high-pressure PL evolution was performed in Fig. 1d and Supplementary Fig. 2. The initially weak emission grew progressively toward higher intensities upon compression to 10.4 GPa, where the PL intensity at 10.4 GPa exhibited a remarkable 7-fold emission enhancement compared to that of the initial state (Fig. 1d). As the pressure further increased to 20.2 GPa, the emission intensity decreased progressively (Supplementary Fig. 2a). Subsequently, a drastically unique behavior was observed when the pressure was released from 20.2 GPa to 11.0 GPa. The PL spectra exhibited remarkable dual-emission centered at 467 nm and 538 nm, respectively (Supplementary Fig. 2b). This suggested that a new emerging state was generated up till that point. After pressure was completely released, we successfully observed a highperformance broadband white-light emission covering the entire visible range from 350 nm to 800 nm with CIE coordinate of (0.29, 0.37) in the targeted Zn-IPA (Fig. 1e-h). The PL intensity of the targeted sample exhibited a remarkable enhancement, which was 16 times stronger than the initial value (Fig. 1g). We estimated the pressuretreated PLQY values using the method developed by Lü et al.<sup>41-44</sup>. The PLQY of the high-pressure-treated sample (Zn-IPA-20) reached 81.3% (Supplementary Fig. 3 and Supplementary Table 1). In order to differentiate and analyze the origin of the white-light emission, we further performed PL evolution of Zn-IPA within 11.4 GPa (Supplementary Fig. 4). During this decompression process, there was no occurrence of new peaks. After pressure was completely released from 11.4 GPa, the resulting PL exhibited a blue-light emission and the intensity also displayed 16 times more intensely than the original Zn-IPA, with CIE color coordinate of (0.21, 0.21) (Fig. 1f-h). The PLQY of this mild-pressuretreated sample (Zn-IPA-11) reached 87.0% (Supplementary Fig. 5 and Supplementary Table 2). In this regard, the formation of white-light emission did require a higher pressure-processing range and should be of high relevance to the new emergent emission state. In addition, we also conducted contrast high-pressure PL experiments without and with PTM of silicon oil (Supplementary Fig. 6-9). The results were consistent with that using liquid Ar as the PTM, indicating that the present PTMs can not affect the generation of the targeted white light.

To map out the mechanism of the new states and white-light emission, we combined in situ high-pressure absorption explorations (Fig. 2a and Supplementary Fig. 10). At ambient conditions, the absorption edge was located at ~ 300 nm. When the pressure increased beyond 12.1 GPa, the absorption edge at ~ 380-700 nm exhibited a significant enhancement compared to that at 9.9 GPa, which might be related to the appearance of the new electronic state. Upon further compression to 20.2 GPa, the absorption edge moved distinctly in the direction of long wavelength. When the pressure was released to the ambient conditions, the identifiable new absorption bands at ~ 390 nm (band I) could be clearly identified in Fig. 2b. Given the existing two original components, fluorescence and phosphorescence, the new absorption band indicated that the targeted white-light emission was comprised of three components. Especially, the phosphorescence emission band of the pressure-treated sample (Zn-IPA-20) was measured at ~553 nm (Supplementary Fig. 11). According to the peakdifferentiation-imitating analysis, the new state emission might be located at ~ 518 nm (Fig. 2c). We then measured the time-resolved PL decay curves at 518 nm and 553 nm, revealing a lifetime of 1.62 ns and 0.78 s, respectively (Fig. 2d). Furthermore, based on these measured emission lifetimes and PLQYs, it can be estimated that the phosphorescence efficiency of Zn-IPA-20 increased from 7.7% to 44.8% (Supplementary Fig. 12). It is worth noting that the newly emerged state emission at ~518 nm would play a significant role in narrowing the energy gap between the singlet and triplet states ( $\Delta E_{ST}$ ). Therefore, we estimate the value of  $\Delta E_{ST}$  based on these measured absorption and phosphorescent spectra. Before pressure treatment, the value of  $\Delta E_{ST}$ was estimated as 1.63 eV (Supplementary Fig. 13). In the treated sample, the  $\Delta E_{\rm ST}$  was narrowed to 0.37 eV. Such decreased  $\Delta E_{\rm ST}$  could thereby accelerate the ISC process to generate a higher fraction of triplet excitons for phosphorescence enhancement based on the energy gap law<sup>45,46</sup>. According to the measured quantum yields and lifetimes, the ISC rates ( $K_{ISC}$ ) before and after pressure treatment were then estimated in Supplementary Table S3. Notably, the  $K_{ISC}$  was



**Fig. 1** | **The photoluminescence (PL) properties of Zn-IPA MOF upon compression and decompression. a** The crystal structure of Zn-IPA MOF. **b** PL spectra of Zn-IPA at 1 atm irradiated by 355 nm laser. **c** The time-resolved PL decay curves of the pristine Zn-IPA measured at 417 nm and 508 nm, respectively. **d** The PL evolution of Zn-IPA upon compression. **e** PL spectra of Zn-IPA after completely releasing

pressure from 20.2 GPa irradiated by 355 nm laser. **f** The CIE color coordinates of Zn-IPA-11 (0.21, 0.21) and Zn-IPA-20 (0.29, 0.37). **g** PL spectra of Zn-IPA at 1 atm, after completely releasing pressure from 11.4 GPa (Zn-IPA-11) and 20.2 GPa (Zn-IPA-20) irradiated by 355 nm laser. **h** Photographs of the pristine Zn-IPA (1 atm), Zn-IPA-11, and Zn-IPA-20. Source data are provided as a Source Data file.

effectively increased from 0.029 ns<sup>-1</sup> to 0.277 ns<sup>-1</sup> after pressure treatment. Therefore, the distribution of singlet and triplet excitons was regulated via an accelerated ISC process based on the narrowed  $\Delta E_{\rm ST}$ , leading to the phosphorescence enhancement and target broadband white-light emission.

In order to explicitly recognize and trace the generation of the new absorption bands, we performed a two-dimensional projection of the absorption intensity upon compression and decompression in Fig. 2e and f. A broadened bluish area was clearly observed from 12.1 GPa to 20.0 GPa. As the pressure was gradually released, the broadened bluish area survived down to the ambient conditions, which matches the position of the new absorption band at  $\sim$  330–450 nm. Thus, the generation of the new absorption band could be tracked by observing the expansion of the bluish area at  $\sim$  12.1 GPa in Fig. 2e.

To further verify this conclusion, we then estimated absorption dependence on pressure within 11.4 GPa. The shape of the absorption spectra did not undergo significant changes, and the absorption edge reverted to the started state after releasing pressure completely (Supplementary Fig. 14). Namely, the mild-pressure-treated sample (Zn-IPA-11) was still composed of the two original components. Therefore, we can conclude that the generation and the caption of the newly emergent state did require high-pressure treatment over ~ 11.4 GPa. Notably, although there is no significant change in the color of Zn-IPA-11, its PL intensity has been greatly improved. After turning off the laser excitation, the phosphorescent emission band of Zn-IPA-11 was measured at ~ 517 nm (Supplementary Fig. 15). We then performed the time-resolved PL decay curves at 433 nm and 517 nm, revealing a prolonged lifetime of 3.24 ns and 1.01 s, respectively (Supplementary Fig. 16). Based on these experimental results, it is estimated that the



**Fig. 2** | **The electronic structure transition of Zn-IPA MOF. a** UV-vis absorption spectra of Zn-IPA MOF upon compression to 20.0 GPa. **b** UV-vis absorption spectra of Zn-IPA at 1 atm and after pressure was released from 20.0 GPa. **c** PL spectra of Zn-IPA-20 irradiated by 355 nm laser. **d** The time-resolved PL decay

curves of the Zn-IPA-20 measured at 518 nm and 553 nm, respectively. Twodimensional projection of the absorption intensity upon (e) compression and (f) decompression. Source data are provided as a Source Data file.

fluorescence and phosphorescence efficiency of Zn-IPA-11 dramatically increased from 13.2% and 7.7% to 55.1% and 31.9% (Supplementary Fig. 17 and Supplementary Table 3).

To gain further insights into the observed new electronic state and emission enhancement, in situ high-pressure structural characterization of Zn-IPA MOF was performed with angle-dispersive X-ray diffraction (ADXRD) experiments. As depicted in Fig. 3a, the d-spacing of the (004) lattice plane remained unchanged (8.08 Å) upon compression to 12.2 GPa, suggesting the zero-linear compression along the *c*-axis. A slight spacing narrow of (008) lattice plane from 4.04 Å to 4.02 Å occurred in the 1 atm ~ 12.2 GPa pressure range, which should be related to the distortion of the ligands (Fig. 3b). With a further increase of the loading pressure from 14.0 GPa to 21.3 GPa, the c-axis might be driven to expand evidenced by the countermovement of the (004) and (008) patterns. Meanwhile, the amorphization of the sample was tracked by observing the loss and widening of the remaining diffraction peaks after 14.0 GPa. Although we lost numerous long-range ordered information at 14.0-21.3 GPa, the residual peak of the (004) plane can exactly mirror the negative linear compression trend seen in experiments. After pressure was completely released, this amorphous phase survived down to the ambient conditions (Supplementary Fig. 18). We further explored the detailed structural parameters via Rietveld refinements of the ADXRD data. At the compression process, the c axis maintained zero-linear compression up to 12.2 GPa, which is consistent with the experimentally derived incompressibility of (004) and (008) planes. The *a/b*-axes decreased from 9.66 Å to 8.64 Å below 12.2 GPa (Supplementary Fig. 19), thus allowing the tighter staked herringbone geometry to enhance intermolecular C-H $\cdot$  $\pi$  interactions for emission enhancement (Fig. 3b). The unit cell volume of the sample kept decreasing with the increase in pressure (Supplementary Fig. 20). In addition, as the ligands were stacked closer and closer upon compression, we found that the carboxylate group began to chelate  $Zn^{2+}$ 

ion in a highly asymmetric mode instead of the original syn-anti bridging coordination at ~11.0 GPa (Fig. 3b). The difference between the two Zn–O distances is as much as 0.604-0.666 Å (Supplementary Fig. 21). As the generation of the new electronic state has been traced to ~ 12.1 GPa, this new chelate mode should be highly associated with the new state emission.

We further seek detailed structural characteristics using in situ high-pressure infrared (IR) absorption spectra (Fig. 3c). The signals at  $662 \text{ cm}^{-1}$ ,  $751 \text{ cm}^{-1}$ , and  $829 \text{ cm}^{-1}$  were assigned to  $\gamma$ (C-H) deformation vibrations of the IPA ligands<sup>47</sup>. It is worth noting that the mode at 751 cm<sup>-1</sup> remained substantially unchanged below 9.2 GPa, suggesting that the intermolecular vibrations were restricted on pressurization. Meanwhile, there is a new shoulder peak appearing at 744 cm<sup>-1</sup> when the pressure was increased to 5.4 GPa, accompanied by peak splitting of 662 cm<sup>-1</sup> and 829 cm<sup>-1</sup> modes. These changes in y(C-H) modes suggested a drastic reinforcement of the intermolecular C-H···π interactions upon compression, which favors the suppression of nonradiative dissipation and the enhancement of emission intensity before ~ 10.4 GPa. In addition, as the pressure further increased to 11.0 GPa in IR spectra, a new peak at the low-frequency side of OCO in-plane bending ( $\delta$ (OCO)) emerged. This mutation of  $\delta$ (OCO) mode well proved the formation of the new chelating coordination mode as mentioned before<sup>48</sup>. Confirmation of these behaviors was also found in the Hirshfeld surface of the system, where the red areas represented strong interactions (Fig. 3d). Upon compression, the red regions of C-H $\cdot\cdot\cdot\pi$  interactions were enlarged as expected, suggesting the promoting C-H···π interactions<sup>49</sup>. Moreover, the additional identified red areas at 12.1 GPa in Fig. 3d further confirm the formation of the new Zn-O coordination interactions.

When decompressed to atmosphere pressure, the split and newly emerged peaks of  $\gamma$ (C-H) modes all survived down to the ambient conditions, suggesting the enhanced C-H… $\pi$  interactions were



Fig. 3 | The structure evolution of Zn-IPA MOF. a ADXRD spectra of Zn-IPA MOF upon compression to 21.3 GPa. b Schematic diagram of the structure evolution of Zn-IPA MOF. c Selected IR spectra of Zn-IPA MOF upon compression. The blue shaded areas correspond to the  $\gamma$ (C-H). The red-shaded areas correspond to the  $\delta$ (OCO). d Hirshfeld surfaces for the Zn-IPA MOF at 1 atm, 10.3 GPa, and 12.1 GPa

mapped with a  $d_{norm}$  distance. The enlargement of the red regions is displayed by the black dotted circle and orange arrows. **e** Selected IR spectra of Zn-IPA MOF at 1 atm and after releasing pressure from 11.0 GPa (Zn-IPA-11) and 20.8 GPa (Zn-IPA-20), respectively. Source data are provided as a Source Data file.

captured to the targeted structure, benefiting the increased emission intensity (Fig. 3e and Supplementary Fig. 22). Furthermore, the v(CC)<sub>ring</sub> modes dramatically shifted to lower wavenumbers from 1467 cm<sup>-1</sup> to 1443 cm<sup>-1</sup> and 1487 cm<sup>-1</sup> to 1483 cm<sup>-1</sup> after pressure treatment, further proving the harvested strong C-H···π interactions in the targeted structure<sup>50–52</sup>. Meanwhile, the new peak of  $\delta$ (OCO) was also reserved down to the ambient conditions and was located at 719 cm<sup>-1</sup>, demonstrating that the new chelating mode was well present in the treated structure. In addition, the v<sub>s</sub>(COO – ) band of the pressure-treated sample was red-shifted from 1408 cm<sup>-1</sup> to 1397 cm<sup>-1</sup>, while the v<sub>as</sub>(COO – ) was blue-shifted from 1604 cm<sup>-1</sup> to 1610 cm<sup>-1 53</sup>. Their relatively pressure-treated large  $\Delta$ (v<sub>s</sub>-v<sub>as</sub>) value of 213 cm<sup>-1</sup> was also assigned to the highly asymmetrical chelating mode of the carboxylate group, further confirming the new coordination configuration in the pressure-treated sample (Zn-IPA-20)<sup>54–56</sup>.

In the following, in situ high-pressure ADXRD and IR experiments within a compression cycle of 1 atm to 11.0 GPa were further conducted to identify the mechanism of emission intensity and color. We denoted that the structure underwent a reversible ordered arrangement when the pressure was completely released from 11.0 GPa, which was evidenced by the legible ADXRD patterns in Supplementary Fig. 23. The IR spectra of the targeted sample (Zn-IPA-11) contain only splitting  $\gamma$ (C-H) deformation vibrations, while the rest of the vibration patterns recover to the initial state (Fig. 3e and Supplementary Fig. 24). In addition, the enlarged red regions marked by the black dotted circle in Supplementary Fig. 25 suggested the promoting C-H $\cdot\cdot\cdot\pi$  interactions in Zn-IPA-11. In this regard, the mild pressure treatment merely captured enhanced intermolecular C-H··· $\pi$  interactions, thus suppressing the nonradiative dissipation and benefiting the blue-light emission enhancement and phosphorescence lifetime prolongation. Therefore, the vital qualification to generate the new emergent electronic state

for the rewarding white-light emission was the new metal-ligand chelate coordination produced under high pressure.

To give theoretical insight into the electronic structure changes of Zn-IPA, we performed the calculated UV-vis absorption spectra, the associated oscillator strength, and the corresponding natural transition orbitals (NTOs) of the relevant excited states (Supplementary Figs. 26 and 27). As the generation of both new electronic states and chelate coordination occurred after ~ 12.0 GPa and then survived down to the ambient conditions, the calculations were carried out using the refined crystal structure at 1 atm and 12.1 GPa. At ambient conditions, the corresponding excitation located at ~290 nm belonged to the charge transfer transition (Supplementary Fig. 27). As the pressure increased to 12.1 GPa, the expected new transition matching the experiment appeared at ~499 nm with an associated oscillator strength of 0.0437 (Supplementary Fig. 26). Notably, their NTOs exhibited obviously boosted electronic delocalization between adjacent ligands, and the hexagonal Zn<sup>2+</sup> also attributed to the rewarding NTOs (Supplementary Fig. 27). These suggested that the chelate coordination promotes intermolecular electronic delocalization, thus rendering the new type of locally excited (LE) state to appear at a lower energy level. This remarkably narrowed the  $\Delta E_{ST}$  of Zn-IPA-20, which effectively accelerated the ISC process and altered the population of singlet and triplet excitons (Fig. 4a).

Inspired by this, the blue and white time delay pc-LEDs were fabricated using 365 nm near-UV LED chips and the pressure-treated Zn-IPA MOFs (Fig. 4b–e). A low voltage (3 V) with various operating currents was used to test the color stability of the pc-LEDs. With the increase of the current from 10 to 90 mA, the emission intensity of the blue and white pc-LEDs steadily increased, exhibiting reasonable chromaticity stability (Fig. 4c and e). Furthermore, after turning off the power, the phosphorescence emission of these pc-LEDs could be



**Fig. 4** | **The emission mechanism and applications of the pressure-treated Zn-IPA MOFs. a** The Jablonski diagram of Zn-IPA at 1 atm and after pressure was released from 11.0 GPa (mild pressure treatment) and 20.8 GPa (high-pressure treatment), respectively. S<sub>1</sub> and S<sub>n</sub> represent the excited singlet states, and T<sub>1</sub> represents the first excited triplet state;  $\Delta E_{ST}$  represents the single-triplet energy gap; ISC represents intersystem crossing; Fluo. represents fluorescence; Phos. represents phosphorescence; N.R. represents non-radiative dissipation. **b** PL emission spectra of the fabricated blue pc-LED utilizing Zn-IPA-11. The insets exhibit

the photographs of the blue pc-LED devices in the bright and dark states. **c** PL spectra of the blue pc-LED at different operating currents. **d** PL emission spectra of the fabricated white pc-LEDs utilizing Zn-IPA-20. The insets exhibit the photographs of the white pc-LED devices in the bright and dark states. **e** PL spectra of the white pc-LED at different operating currents. **f** Phosphorescence photographs of pc-LED devices fabricated by Zn-IPA, Zn-IPA-11, and Zn-IPA-20, respectively. Source data are provided as a Source Data file.

captured by the naked eye for a few seconds. Especially, the afterglow of the blue pc-LED fabricated by Zn-IPA-11 could even last for nearly 7 s (Fig. 4f and Supplementary Movie). The time delay pc-LEDs with long emission time after switching off the power offer a buffer for eyes to reduce transient blindness arising from the abrupt darkness.

In summary, we reported a new type of coordination-responsive luminescence transition in a single component Zn-IPA MOF via different degrees of pressure treatment engineering. The harvested pressure-induced strengthened intermolecular C-H···π interactions offer greater rigidity against non-radiative dissipation, resulting in emission enhancement in target MOFs. Metal-ligand chelate coordination occurred over ~11.0 GPa promotes the generation of the new electronic state. Such an additional emission state offered a major impact on narrowing  $\Delta E_{ST}$  for accelerated ISC process, leading to the phosphorescent emission enhancement and rewarding highperformance white-light emission. The discovered ability to purposefully construct blue- and white-light emissions with long emission time through the elaborate molecular design by pressure-treatment engineering would provide a promising platform for the exploration of single-component phosphorescent MOFs.

## Methods

#### Synthesis of Zn-IPA before pressure treatment

A mixture consisting of IPA (88 mg),  $Zn(NO_3)_2$ ·6H<sub>2</sub>O (150 mg), and 2-methylimidazole (82 mg) was dissolved in 8 mL of water and transferred into a 25 mL Teflon-lined stainless steel autoclave. The sealed vessel was heated at 150 °C for 24 h. Upon gradual cooling to room temperature, colorless block-shaped crystals of Zn-IPA were obtained. This synthesis method was carried out following a previously reported procedure<sup>40</sup>.

#### Characterization and high-pressure generation

All in situ high-pressure experiments were performed using a symmetric diamond anvil cell (DAC) apparatus at room temperature. The sample and a small ruby ball were loaded into the 150  $\mu$ m-diameter DAC chamber consisting of a T301 steel gasket pre-indented to a

thickness of 45  $\mu$ m. The pressure calibration was determined utilizing the standard ruby fluorescent technique<sup>57</sup>. In high-pressure experiments, liquid Ar was utilized as the pressure transmitting medium (PTM) for optical absorption, PL, and ADXRD experiments, while the KBr was employed as PTM for IR measurements. These PTMs did not have any detectable effect on the behavior of Zn-IPA MOF under pressure. All of the measurements were performed at room temperature.

#### In situ high-pressure experiments

A 355 nm line of a UV DPSS laser with a power of 4.8 mW was used for the PL measurements. The in situ high-pressure steady-state PL spectra of Zn-IPA MOF were collected by a modified spectrophotometer (Ocean Optics, QE65000) at various pressures. PL micrographs of the samples were obtained using a camera (Canon Eos 5D mark II) equipped with a microscope (Eclipse TI-U, Nikon). The camera can record the photographs under the same conditions including exposure time and intensity. The chromaticity coordinates (x, y) were calculated from the fluorescence data (355-800 nm) using the CIE1931xy.V.1.6.0.2a software package. The color of the fluorescent emission was identified by the CIE colorimetry system. Any colors could be described by the chromaticity (x, y) coordinates on the CIE diagram. The absorption spectra were performed with a Deuterium-Halogen light source and recorded with an optical fiber spectrometer (Ocean Optics, QE65000). IR spectra measurements of Zn-IPA MOF were carried out by Nicolet iN10 microscope spectrometer (Thermo Fisher Scientific, USA) using liquid-nitrogen cooled CCD. The PLQY was measured with Hamamatsu multichannel analyzer c10027. In situ high-pressure angle-dispersive X-ray diffraction (ADXRD) patterns were obtained with a wavelength of 0.6199 Å at 4W2 beamline, Beijing Synchrotron Radiation Facility (BSRF) the BL15U1 beamline, Shanghai Synchrotron Radiation Facility (SSRF). CeO<sub>2</sub> was applied to the standard sample to do the calibration. The FIT2D program was used to integrate the two-dimensional images, and the onedimensional diffraction angle 2-theta diagram was obtained. All the high-pressure experiments were conducted at room temperature.

## **First-principle calculations**

The energy and oscillator strength of the excited state of Zn-IPA MOF were calculated using time-dependent density functional theory, and the corresponding natural transition orbitals were analyzed. All calculations were performed using PBE0<sup>58</sup> hybrid functionals, with 6–31 G<sup>\*59,60</sup> basis sets used for non-metallic elements such as C, H, O, and SDD<sup>61</sup> pseudopotential basis sets used for Zn elements. All computing tasks are completed using the Gaussian 16 package.

## Data availability

The authors declare that the main data supporting our findings of this study are contained within the paper and Supplementary Information. Source data are provided in this paper.

## **Code availability**

Gaussian 16 code is available for download on the developer page: https://gaussian.com/.

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## **Author contributions**

X.Y. and B.Z. designed the project and supervised the work. Q.Y. performed the in situ high-pressure PL, UV-vis Absorption, and IR spectra measurements. Q.Y., W.W., and Y.Y. conducted the ADXRD, time-resolved PL decay curves, PLQY experiments, and pc-LED applications. P.L. and F.B. performed the calculations. Q.Y., X.Y., and B.Z. wrote the manuscript. All authors discussed the results and commented on the manuscript.

## **Competing interests**

The authors declare no competing interests.

## Additional information

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