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# Fluorescent coordination-polymer single crystals with tunable elastic–plastic transformations

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Achieving and controlling mechanical responses in single crystals with complex coordination architectures remains challenging. Here, we report the design and synthesis of a homologous series of one-dimensional cadmium(II) coordination polymers that exhibit controllable bending under an external load. Systematic modification of imidazole-based ligands modulates the periodic Cd⋯Cd separation along the chains, thereby tuning the macroscopic response from elastic to plastic. Based on experimental results and data analysis, we identified an inverse correlation between the metal–metal repeat distance and the attainable elastic strain, consistent with a molecular-spring-like Cd–Cl–Cd backbone. In parallel, the imidazole chromophores endow all materials with stable blue photoluminescence under 365 nm excitation. The convergence of mechanical adaptability and intrinsic emission identifies these crystals as promising candidates for flexible optoelectronic components and optical waveguides.

## Introduction

Mechanically responsive single crystals capable of pronounced shape transformations under external stimuli have recently emerged as a focal topic in crystal engineering.[1–4] Such materials hold significant promise for applications in flexible electronic devices,[5,6] optical waveguides,[7–12] and soft robotics.[13–16] The mechanical compliance of single crystals encompasses both elastic and plastic properties, and their corresponding deformation mechanisms have been extensively elucidated in organic single crystals.[17] Elastic bending arises from reversible compression of the inner arc and extension of the outer arc,[18,19] whereas plastic bending is governed by irreversible layer slippage that permanently disrupts the lattice.[20,21]

To date, research on flexible single crystals has predominantly focused on organic molecules.[22–26] These materials typically feature planar molecular conformations with criss-cross or parallel packing within the crystal and rely on weak intermolecular interactions to facilitate deformation.[27] By contrast, mechanically adaptive coordination polymers (CPs) remain comparatively unexplored. Their structural complexity—stemming from polynuclear nodes, variable coordination numbers, and diverse ligand environments[14–28]—poses significant challenges to achieving structural deformation through coordination bonds. Nonetheless, coordination compound crystals exhibit unique optical,[29–31] electrical,[32–35] magnetic,[36,37] and catalytic properties.[38,39] Progress in fabricating mechanically flexible CP single crystals will therefore provide a powerful platform for next-generation flexible optoelectronic materials.

An early study on mechanically responsive coordination polymer crystals was reported by the Đaković group,[40] who showed that a series of one-dimensional Cd(II) halide polymeric chains exhibit tunable elasticity by altering noncovalent interactions. Subsequently, Emmerling and co-workers discovered that the one-dimensional CP $[Zn(\mu\text{-}Cl)_2(3,5\text{-dichloropyridine})_2]_n$ displays pronounced plastic deformability.[41] Beyond elastic and plastic behavior, researchers have sought to introduce new functionalities into these mechanically responsive coordination polymer crystals. Gao *et al.*[42] found that one-dimensional lead halide elastic crystals exhibit strong electron–phonon coupling. Zhang *et al.*[43] introduced helicity (*P* or *M*) into the crystal $[CdCl_2(1\text{-methyl-2-pyridone})]_n$, synthesizing elastic CPs that display circular dichroism and second-harmonic generation by combining a spring-like framework with weak intermolecular interactions. Nevertheless, such reports remain rare, particularly for optical materials with tunable elastic–plastic responses.

Herein, we report three fluorescent CPs with finely tunable mechanical flexibility. Imidazole derivatives, valued for their inherent rigidity and distinctive bipolar architecture, are versatile building blocks for luminescent systems.[44–48] Concurrently, alkyl functionalization is routinely employed to modulate elastic–plastic responses.[49–52] We therefore envisage

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fluorescent assemblies in which imidazole-based ligands coordinate to chloride-bridged, one-dimensional $(CdCl_2)_n$ double chains, with strategic alkyl substitution enabling precise control over mechanical behaviour (Fig. 1).

## Work description and characterization

Single-crystalline metal CPs $[Cd_2Cl_4(Im)_2]_n$ (**1**), $[Cd_2Cl_4(MeIm)_2]_n$ (**2**), and $[Cd_2Cl_4(EtIm)_2]_n$ (**3**), where Im, MeIm, and EtIm denote imidazole, 1-methylimidazole, and 1-ethylimidazole, respectively, were obtained by solvothermal synthesis and consistently adopt a needle-like morphology. The mechanical properties of individual single crystals were investigated using a three-point bending test. Crystals **1** and **2** demonstrated notable elasticity (Fig. 2a–f). Upon the application of an external force, the crystals could be substantially bent, and they would spontaneously revert to their original linear state following the removal of the load (Movies S1 and S2). This process could be repeated multiple times without inducing fracture. Beyond their elastic limits, however, the two compounds displayed divergent behaviors. Crystal **1** underwent brittle fracture upon reaching its maximum bending strain. In contrast, crystal **2** exhibited a distinct plastic deformation region beyond its elastic threshold; once deformed past this point, it retained a bent configuration, signifying an elastic-to-plastic transition (Movie S3). To quantitatively evaluate and compare their elastic capacities, the maximum bending strain ($\varepsilon$) for each crystal was calculated based on the Euler–Bernoulli beam theory. The calculated $\varepsilon$ values for crystals **1** and **2** were 4.98% and 4.43%, respectively (Fig. S3). Conversely, the mechanical response of crystal **3** was markedly different. It exhibited no discernible elastic deformation under mechanical stress, instead undergoing exclusively plastic deformation, thus classifying it as a purely plastic crystal (Fig. 2g–i and Movie S4). This work presents a homologous series of 1D CP single crystals that exhibit systematically tunable mechanical responses, spanning purely elastic (**1**), elastic–plastic (**2**), and purely plastic (**3**) behaviors.

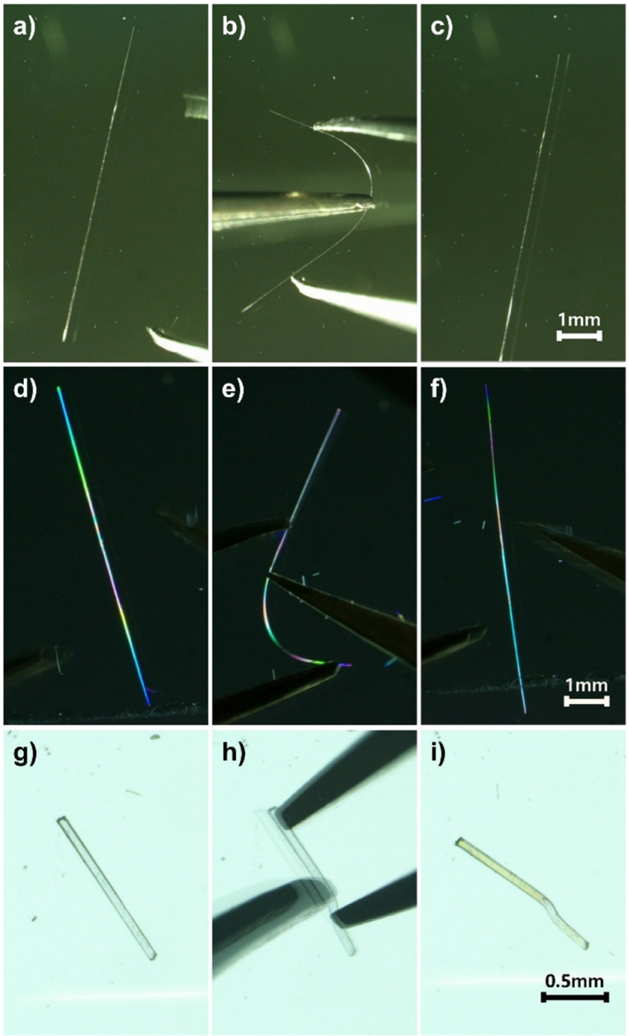

**Fig. 2** Mechanical response of crystals. (a–c) Elastic bending and recovery of crystal **1**. (d–f). Elastic bending and recovery of crystal **2**. (g–i) The plastic mechanical response process of crystal **3**.

## Single-crystal structural analysis

The single crystal structures of compounds **1–3** were determined through single-crystal X-ray diffraction (SC-XRD). These compounds crystallize in the space groups *Pnma*, $P2_1/c$, and $I2/m$, respectively (Table S4) and are all characterized as 1D CPs.

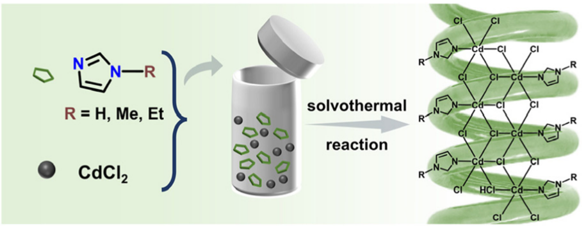

**Fig. 1** Synthesis and assembly strategies for compounds **1–3**.

In these structures, each Cd center is six-coordinate, adopting a distorted octahedral geometry defined by two $\mu_2$-Cl and three $\mu_3$-Cl ligands. This Cd–Cl framework extends into a 1D double-chain that propagates along the crystallographic *a*-axis—the long axis of the needle-shaped crystals. The nitrogen atoms of the imidazole ligands are positioned on the exterior of this framework (Fig. 3a). This 1D chain architecture accommodates external stress through variations in the coordination structure, potentially leading to significant mechanical deformation, analogous to “molecular springs”.[43,53–57] Adjacent 1D chains are interlinked by weak intermolecular C–H⋯Cl hydrogen bonds, the strength of which is a key determinant of the material’s elastic or plastic response. Although the individual chains possess an identical constitution, they adopt distinct stacking arrangements: in compounds **1** and **2**, the chains are arranged in an antiparallel, cross-stacked fashion, whereas compound **3** features a parallel stacking motif (Fig. 3b).

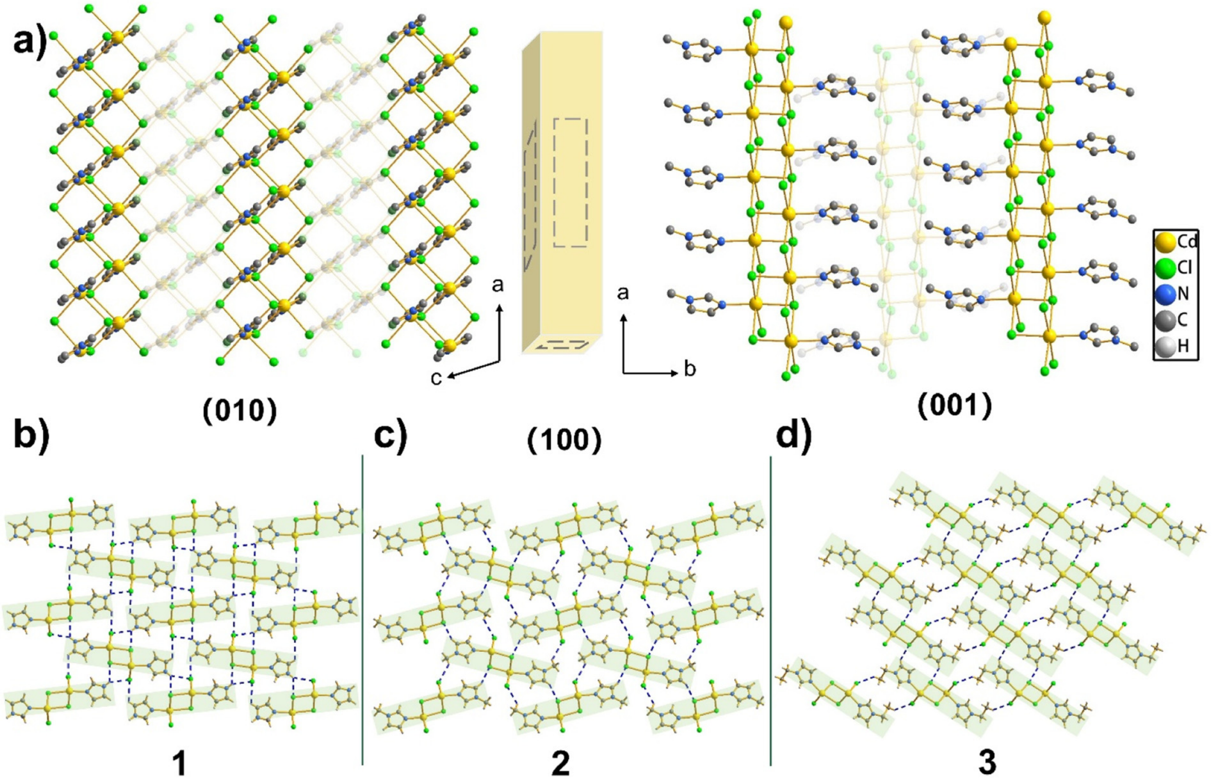

**Fig. 3** Single crystal structures. (a) and (c) Stacking diagrams of the different crystal faces of crystal **2**. (b–d) Differences in the stacking patterns between adjacent 1D chains: cross stacking in **1** and **2**, and parallel stacking in **3**.

The mechanical flexibility of crystals is typically governed by factors such as crystal packing and intermolecular forces. The antiparallel, cross-stacked arrangement in crystals **1** and **2**, connected through C–H⋯Cl interactions, furnishes an isotropic three-dimensional network that disperses local stress and suppresses crack propagation, thereby affording exceptional elasticity. By contrast, crystal **3** shows parallel packing on the (100) plane with discrete slip planes between chains (Fig. 3d), indicating a greater propensity for interchain slip that leads to plastic deformation.

Remarkably, the elastic bending strains of compounds **1** and **2** substantially surpass those of previously reported 1D CPs (Fig. 4 and Table S5). These values are not only approximately four to five times greater than those of analogous CPs constructed from single $(CdCl_2)_n$ coordination chains but are also comparable to those of π-stacked organic crystals and the notable benchmark of $Cu(acac)_2$ (4.4%).[58] Such exceptionally high bending strains underscore the significant promise of these materials for applications in flexible and wearable devices.

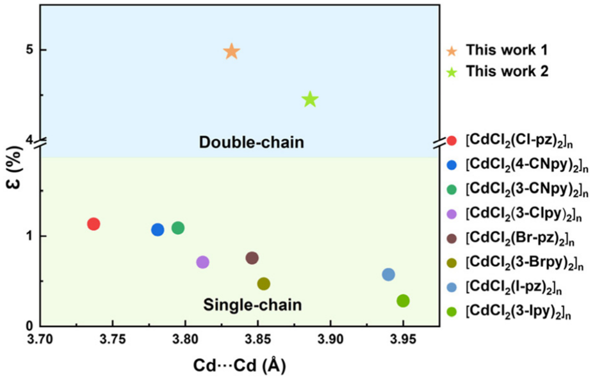

**Fig. 4** Comparison of bending strain $\varepsilon$ values for the reported 1D $(CdCl_2)_n$ chain crystals and the present compounds at different Cd⋯Cd distances.

According to the literature, the origin of this elasticity is attributed to the concomitant elongation of the crystal lattice on the outer arc and compression on the inner arc during bending.[57] In these 1D CPs, the crystallographic unit cell along the polymer chain is defined by the Cd⋯Cd separation. Consequently, the Cd–Cl–Cd coordination backbone is hypothesized to function as a molecular spring, wherein the Cd⋯Cd distance can be reversibly modulated by the applied mechanical force, thereby accounting for the macroscopic elastic deformation of the single crystals (Fig. 5a and b). The exceptional elastic performance observed in **1** and **2** suggests that their double-chain coordination framework plays a pivotal role in reinforcing the crystal lattice, making it capable of accommodating the large elastic deformations.

The deformability of a helical spring is largely governed by its pitch; analogously, we hypothesize that the elastic response of these 1D CPs correlates with the Cd⋯Cd distance (the pitch of a molecular spring). A survey of the literature on elastically deformable 1D $(CdCl_2)_n$ single crystals reveals an inverse correlation between the Cd⋯Cd distance and the attainable elastic strain: shorter separations tend to promote higher elasticity, whereas longer separations diminish it. As shown in Fig. 4 and Table S5, among known 1D Cd compounds adopting a $(CdCl_2)_n$ single-chain architecture, compound $[CdCl_2(Cl\text{-}pz)_2]_n$[40] exhibits the shortest Cd⋯Cd distance (3.7369 Å) and the highest elastic strain ($\varepsilon$ = 1.134%), whereas compound $[CdCl_2(3\text{-}Ipy)_2]_n$,[55] which has the longest Cd⋯Cd separation (3.95 Å), displays a substantially lower elastic bending strain of 0.28%. This principle is consistent with the behavior of our materials. The increasing steric hindrance of the alkyl substituents on the imidazole ligands (H < methyl < ethyl) incrementally widens the Cd–Cl–Cd coordination angle, thereby extending the Cd⋯Cd distance from 3.8319 Å in **1** to 3.8859 Å in **2** and 3.9649 Å in **3** (Fig. 5c). This structural trend directly corresponds to the observed rank order of their mechanical performance (**1** > **2** > **3**). From a coordination-structural perspective, a shorter Cd⋯Cd distance facilitates greater extension of the outer arc of a crystal under bending. We therefore propose that the intrinsic stretchability of the Cd–Cl–Cd links along the coordination backbone makes a decisive contribution to the elastic behaviour of these 1D CPs, complementing the roles of crystal packing and interchain interactions. Furthermore, the double-chain structures usually exhibit superior elasticity compared with single-chain structures at similar Cd⋯Cd distances. We propose that the additional Cd–Cl–Cd coordination linkages in the double-chain architecture stabilize the polymer backbone while enabling cooperative, hinge-like rotations. This arrangement distributes strain more evenly, reduces stress concentrations, and delays crack initiation and propagation, thereby enhancing the elastic response of single crystals.

## Fluorescence

Beyond mechanical response, the chosen ligands endow all three crystals with luminescence (Fig. 6a). Under 365 nm irradiation, each emits stable blue fluorescence. Across the series **1** to **3**, the emission maxima undergo a progressive redshift and the full width at half-maximum broadens (Fig. 6b). Although free imidazole is only weakly emissive, coordination to $Cd^{2+}$ in an octahedral environment rigidifies the heteroaromatic ring and markedly enhances fluorescence.[59,60] Owing to the $d^{10}$ electronic configuration of $Cd^{2+}$, which precludes d–d transitions,[46,61,62] the observed emission is probably attributed to ligand-centred π–π* transitions within the imidazole framework.

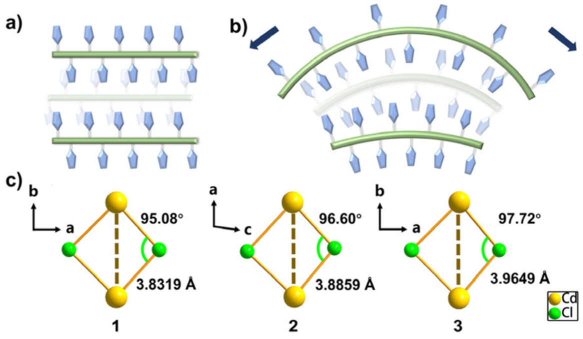

**Fig. 5** (a) and (b) Chain geometry before and after bending in this work. (c) Changes in the metal center Cd⋯Cd distance and frame angle in the direction of elongation of the 1D chain.

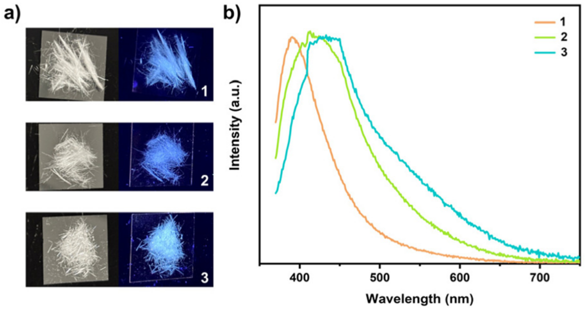

**Fig. 6** (a) Fluorescence phenomena of crystals **1–3** under 365 nm UV light; (b) normalized photoluminescence spectra of crystals **1–3** at their optimal excitation wavelengths.

## Conclusions

In this study, we prepared a series of mechanically flexible, fluorescent coordination-polymer single crystals by integrating imidazole ligands into $(CdCl_2)_n$ double-chain frameworks. A systematic variation of the alkyl substituents on the ligands tunes the mechanical response from elastic to predominantly plastic deformation. Owing to the robust double-chain scaffold and short Cd⋯Cd separations, compounds **1** and **2** achieve exceptional elastic strains of 4.98% and 4.43%, respectively, surpassing prior records for one-dimensional coordination polymers. Coordination to $Cd^{2+}$ imposes an octahedral constraint on the imidazole ring, enhancing emission; accordingly, all materials exhibit distinct blue fluorescence under 365 nm irradiation. Future work will incorporate more strongly emissive ligands to investigate the influence of elastic and plastic deformation on the luminescence properties of the materials. In addition, the introduction of chirality is expected to enable the flexible circularly polarized luminescence (CPL) functionality. These studies may ultimately contribute to the development of high-performance, flexible optical materials suitable for wearable technologies.

## Author contributions

Z.-S. Y. conceived the idea and designed the study. J.-H. Z. synthesized the materials and performed the experimental measurements. Z.-S. Y. and J. T. supervised the research. All authors discussed the results and commented on the manuscript.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

The data that support the findings of this study are available in the supplementary information (SI). Supplementary information: Movie S1 showing elastic deformation of crystal **1** (MP4); Movie S2 showing elastic deformation of crystal **2** (MP4); Movie S3 showing the elastic–plastic transition of crystal **2** (MP4); Movie S4 showing plastic deformation of crystal **3** (MP4). See DOI: https://doi.org/10.1039/d5dt02338f.

CCDC 2486224 (for **1**, 298 K), 2486225 (for **3**, 298 K), and 2486226 (for **2**, 298 K) contain the supplementary crystallographic data for this paper.[63a–c]

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