Nano-Achiral Composite Films with Strong Circular Polarization

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Composites from two-dimensional (2D) nanomaterials display uniquely high electrical, thermal, and mechanical properties[1,2]. Pairing their robustness with polarization rotation is needed for hyperspectral optics in extreme conditions[3,4]. However, the rigid nanoplatelets have randomized achiral shapes, which scrambles the circular polarization of photons with comparable wavelengths. Here we show that multilayer nanocomposites from 2D nanomaterials with textured surfaces strongly and controllably rotate light polarization, despite being nano-achiral. The intense circular dichroism in nanocomposite films originates from the diagonal patterns of wrinkles, grooves or ridges leading to an angular offset between axes of linear birefringence and linear dichroism. Stratification of the layer-by-layer (LBL) nanocomposites affords precise engineering of the polarization-active materials from imprecise nanoplatelets with an optical asymmetry *g*-factor of 1.0, exceeding those of typical nanomaterials by ~500 times. High thermal resilience of the composite optics enables operating temperature as high as 250 ºC and imaging of hot emitters in the near-infrared part of the spectrum. Combining LBL nanocomposites with achiral dyes results in anisotropic factors for circularly polarized emission approaching the theoretical limit. The generality of the observed phenomena is demonstrated by nanocomposite polarizers from MoS_2, MXene, and graphene oxide and by two manufacturing methods. A large family of LBL optical nanocomponents can be computationally designed and additively engineered for ruggedized optics.

Photonics devices for robotic perception systems, holographic technologies, information encryption protocols, through-space communications, and power plant

monitoring require variable and intense polarization rotation as well as tolerance to extreme environments[3–7] (nuclear reactors, ocean floor, desserts, etc.). Working temperatures above 100 °C are not accessible for the vast majority of devices based on liquid crystals (LCs), small molecules or organic polymers, due to temperature-sensitive phase transitions, recrystallization processes, and oxidation reactions[8,9]. Nanocomposite films from zero- and one-dimensional building blocks, that is, nanoparticles[10,11] and nanofibers[12–14], display resilience to oxidation paired with high circular dichroism (CD) and optical asymmetry *g*-factors that are essential figures of merit for chiral optics. However, current chiral nanocomposites remain vulnerable to temperature-induced dehydration and phase transitions.

Nacre-like layered composites from 2D nanomaterials are known for their toughness[2], fire-resistance[15], and optical transparency[16]. These properties are assets for ruggedized polarization optics, but their use in chiral photonic components seems counterintuitive. Weak CD could be obtained due to molecular scale chiral features[17–19], but making nanoplatelets with strong nanoscale chirality, optical polarization rotation and high *g*-factor is fundamentally problematic due to their rigidity and high energy required for their twisting and strong un-polarized scattering. Additionally, the layered structure of 2D nanomaterials is much more difficult to reconfigure without structural damage compared to LCs.

Herein, we demonstrate nanocomposite optics combining strong polarization rotation, mechanical robustness, and temperatures of operation as high as 250 °C. They were achieved by the LBL deposition of 2D nanoscale components into stacked optical media with tunable linear dichroism (LD) and linear birefringence (LB) that can be additively engineered for specific wavelengths including the near-infrared (NIR) range. All the components and composites are achiral at the scales relevant to experimentally

observed polarization rotation, which makes the optical response conceptually different from deformation-induced[20], lithographically-defined[21,22], and assembly-facilitated[10,23] out-of-plane chirality investigated in the past. The generality of the observed phenomena was demonstrated by composite polarizers made from three chemically diverse 2D nanomaterials, such as titanium carbide ($Ti_3C_2T_x$) representing MXenes[1,24,25], molybdenum sulfide (MoS_2) representing transition metal chalcogenides, and graphene oxide (GO) representing nanocarbons. Using primarily $Ti_3C_2T_x$ as an example, two manufacturing methods were used to generate nanoscale surface textures with centimeter-scale dimensions showcasing their scalability.

Multiscale chirality of nanoplatelets

The chiroptical activity of chemical structures is dependent on the amplitude of electromagnetic resonances and their left/right asymmetry. When the frequency match for resonance is satisfied, chiroptical activity will be the highest when the wavelength of photons is comparable to the size of chiral objects[21,23]. Nanostructured materials display chirality at multiple scales[18] and their chiroptical activity is maximized when they display high left/right asymmetry at the scale comparable to the wavelength of the photon[26]. As such, chiral plasmonic particles and assemblies have strong chiroptical activity for visible wavelengths because their dimensions of about 100 nm are complemented by high mirror asymmetry. The latter can be quantified as Hausdorff[27] chirality measure (HCM) or Osipov–Pickup–Dunmur[28] chirality index (OPD) exceeding 0.2 and 5.0[29,30], respectively.

Nanoplatelets of 2D materials, exemplified by $Ti_3C_2T_x$, MoS_2[31,32] or GO[17], show only weak, if any, chiroptical activities (Supplementary Figs. 1–5). Their mirror asymmetry at atomic scale can be high (HCM ~0.3, OPD ~0.001) due to, for instance, amino acid surface ligands with well-defined chiral centers, strong affinities to

inorganic cores and collective ligand-to-ligand charge transfers (Supplementary Note 2), but the characteristic size of the asymmetric tetrahedrons around the α-carbon is small ~0.3 nm. Thus, photons with 200–300 nm wavelengths matching in energy with resonant electronic transitions, produce only weak CD peaks with an amplitude of 1–10 mdeg, which are smaller than those for many nanoscale (bio)molecules. Chirality measures for the same nanoplatelets at 100 nm scale are near zero (HCM = 7.9×10^{-2}, OPD = -6.4×10^{-5}) due to the rigidity of the 2D material and irregular shapes, which makes them nano-achiral (Supplementary Fig. 6 and Supplementary Table 1). Consequently, the chiroptical activity for MoS_2 and $Ti_3C_2T_x$ nanoplatelets with various amino acid ligands around the electronic oscillation band and band gaps is weak or near zero.

Chiroptical activity of composites

The presence of LB- and LD-active strata along the light path results in polarization rotation and strong CD[33–35], which is typically treated as an artifact complicating measurements of 'true' isotropic CD related to chirality. The LB component of the optical media splits the light beam propagating along the *z*-axis into 'slow' and 'fast' wave packets with orthogonal polarizations. Subsequent passage through LD-active media serving as an imperfect linear polarizer retains their phase lag, resulting in strong ellipticity for the combined transmitted beam (Supplementary Fig. 7).

2D nanomaterials can display LD and LB[36,37], but the typical layered nanocomposites made from them are *x*-*y* isotropic because nanoplatelets are placed randomly on the surface. Two methods impart LB/LD activity and, thus, CD by adding surface textures along the *z*-axis.

Method 1 (M1): Polydimethylsiloxane (PDMS) is coated onto polyethylene terephthalate (PET) substrates with strong LB followed by imprinting submicron scale

grooves with a typical interval and depth of 740 ± 60 and 120 ± 20 nm, respectively. Nanoplatelets of 2D nanomaterial are conformally deposited on the PDMS grooves by the LBL deposition of negatively charged nanoplatelets and positively charged poly(diallyldimethylammonium chloride) (PDDA) with a total thickness determined by the number of bilayers. As such, 10 bilayers of $Ti_3C_2T_x$-PDDA produce a coating of 27 ± 3 nm (**Fig. 1**, Supplementary Figs. 8–10, and Supplementary Table 2). Importantly, the platelets of 2D materials coating the walls of the grooves are stacked, which results in a composite with high refractive index and an electrical dipole orthogonal to the *z*-axis. When the dihedral angles between the grooves and 'built-in' LB axis of PET are +45° (left-handed, LH, composites) and -45° angles (right-handed, RH, composites), strong monopolar CD with opposite signs are observed (**Fig. 1e,f**, Supplementary Figs. 11 and 12); the shape of the spectrum matches the theoretical ones based on LB/LD interactions[33] nearly perfectly.

Method 2 (M2). LBL films are deposited onto twisted PDMS sheets followed by their relaxation to the flat state (**Fig. 2a,b**). The compressed side produced cracked films with a typical thickness of 130 ± 20 nm, while the stretched side produced wrinkled films with a height of 680 ± 82 nm (**Fig. 2c** and Supplementary Fig. 13). LB in these materials emerges from PDMS[38] (Supplementary Fig. 14) and increases when the substrate is stretched (**Fig. 2d** and Supplementary Figs. 15–19). The LD activity originates from the wrinkled and cracked patterns of the LBL films, again, resulting in electrical polarization vectors oscillating orthogonally to the *z*-axis. Testing different conditions of stretching and twisting, we produced wrinkles at +45° (LH composites) and -45° angles (RH composites) with respect to the long axis of the substrates.

A wide range of 2D materials can be used for both M1 and M2 as demonstrated by $Ti_3C_2T_x$, MoS_2 and GO (Supplementary Figs. 20–22). In all cases, centimeter-scale

samples with uniform optical properties were obtained. In all cases, the 2D materials produce composite solids with stiffness, hardness, and substrate adhesion far exceeding the conventional chiroptical materials (Supplementary Fig. 23). The rigidity of nanoplatelets, however, does not allow them to twist[10,11,20,39] and they remain nano-achiral or nano-racemic in both M1- and M2-composites.

Structural tuning of circular dichroism

Attaining high isotropic or 'true' CD requires simultaneous optimization of the chirality measures, sizes, and resonance energies of chiral chemical objects, which are subject to a long list of chemical and physical constraints. The stratified optical media with LB and LD components de-couples these parameters and simplifies the materials design process. Following Mueller calculus for light-matter interactions, CD of a transmitted light beam should obey the dependence[33–35] (Methods):

$$\mathrm{CD} \propto \mathrm{LB} \cdot \mathrm{LD}' = \mathrm{LB} \cdot \mathrm{LD} \cdot \cos(2\,\alpha - 90^{\mathrm{o}}) \qquad \text{(Eq. 1)}$$

where LB characterizes the linear birefringence of PET (M1) or PDMS (M2) along the *y*-axis (**Fig. 1a** and Supplementary Fig. 11), while LD represents linear dichroism at 45° in agreement with the convention of Mueller calculus; the latter can be altered by the dihedral angle, α, between LD and LB. We tested this dependence for several experimental parameters. For example, we varied α by changing the direction of grooves (M1) (**Fig. 1g** and Supplementary Fig. 24) or the direction of cracks/wrinkles (M2) (Supplementary Fig. 25). We observed a nearly perfect agreement with Eq. 1 for all cases of composites of different handedness, shape, peak position, polarity and α. CD maxima were observed at α = 45° and 135° with preferred LH and RH circular polarization effect for the transmitted photons.

CD amplitude rises approximately proportionally to the number of LBL deposition cycles (Supplementary Figs. 26–28) for both M1- and M2-composites instead of

expected quadratic dependence, which is indicative of the compensatory effects related potentially to additional light-matter interactions.

Strain applied to the material can strongly change LB, LD, and α. Stretching the M2-composites up to 31% increases LB, and thus CD (Eq. 1; Supplementary Fig. 29). After removing the strain, the spectra returned to their original states, which can be repeated at least 1000 times without polarization loss (**Fig. 2f**). Mueller matrix polarimetry (MMP) mapping reveals that the chiroptical response remained homogenous even under tensile strains as high as 50% (**Fig. 2e**). In addition, with increasing strain amplitudes, the direction of cracks/wrinkles gradually aligned towards the strain direction (Supplementary Fig. 13), resulting in the decrease of α and LD' as well as the saturation and slow decrease of CD beyond 15% strain.

Tuning the optical activities in different strata is facilitated by the additive nature of LBL films. Additional LD activity can be imparted by depositing a layer of aligned rod-like plasmonic materials (Supplementary Fig. 30), such as Ag nanowires by a grazing incidence spray-LBL[13] (**Fig. 2g**). A single layer of Ag nanowires enhanced LD amplitude 5–10 times compared to the nanoplatelet wrinkles (Supplementary Figs. 26–31), enabling high *g*-factors without difficulties in chemical synthesis. The magnitude of polarization rotation reached as much as 3500 millidegrees with a maximum *g*-factor >1.0 (**Fig. 2h,i**), which exceeds the highest *g*-factors of 0.1–0.4 reported for nanoparticles in dispersion[23,40,41] and comparable to or exceeding *g*-factors of 0.5–2 reported for Bouligand structures[42,43].

Circularly polarized emission

Additive engineering of LB/LD active composites can also be applied to the circularly polarized conversion emission (CPCE) of photoluminescence. Different from straightforward circularly polarized emission requires the asymmetry of fluorescent

chromophores, the indirect CPCE hinges on the engineering of LB and LD components inherent in the light emission process. This strategy streamlines molecular designs and offers diverse solutions for the conundrum of weak circularly polarized emission anisotropy (g_{em}) and luminescence quantum yield (Φ_{lum}). While 2D nanomaterials display strong luminescence, achieving 'true' circularly polarized emission with a high g_{em} is more difficult than 'true' CD[44]. In the case of chiral molecules, chemists' dilemma is that increasing optical asymmetry typically results in decreased Φ_{lum}, owing to the non-radiative dissipation of excited state energy[45].

Programmable LD and LB of nanocomposites can be harnessed to achieve a high g_{em} with a Φ_{lum} approaching unity, even for achiral fluorescent dyes (**Fig. 3a**). For example, rhodamine 6G (Φ_{lum} = 0.95) is an achiral fluorescent molecule without any circularly polarized emission (Supplementary Fig. 32); however, when it was incorporated with M2-composites of $Ti_3C_2T_x$, strong CPCE was observed.

Elaborating on Eq. 1 for emissive media, the structure-property relations for CPCE can be encapsulated in the relationship:

$$\mathrm{CPCE} = \mathrm{CD} \cdot \Phi_{\mathrm{lum}} \propto \mathrm{LB} \cdot \mathrm{LD}' \cdot \Phi_{\mathrm{lum}} = \mathrm{LB} \cdot \mathrm{LD} \cdot \cos(2\,\alpha - 90^{\mathrm{o}}) \cdot \Phi_{\mathrm{lum}} \quad \text{(Eq. 2)}$$

Experimental data firmly validates this equation. Similar to CD, LH and RH emission can be attained from opposite α via changing the direction of cracks/wrinkles (**Fig. 3b,c** and Supplementary Fig. 33), enabling us to obtain g_{em} as high as 0.11. The value is several orders of magnitude higher than the typical asymmetry factors for chiral fluorescence emitters with $g_{em} = g_{lum} = g_{CPL}$ that are generally less than 0.01[46] and comparable to the asymmetry factors of 0.1–0.5 reported for lanthanide complexes[47] and liquid crystal composites[48].

The strain of PDMS from 0–25% changes LB while maintaining LD constant. A linear correlation between CPCE and g_{em} responses with LB has been observed

(Supplementary Fig. 34), in agreement with Eq. 2. Stretching M2-composites also demonstrates the real-time modulation of CPCE's degree of ellipticity.

When aligned Ag nanowire films were used to enhance LD, the same α-dependence was observed (**Fig. 3d**). The ellipticity of emitted light was as high as 3.6 deg while g_{em} was as high as 0.19. Further optimization of LD' with a linear polarizer enabled bright CPCE intensity with an ellipticity of 17 deg and g_{em} of 1.6 (**Fig. 3e**), approaching the theoretical limit of 2.0, corresponding to perfect separation of LH and RH photons. Having a unique combination of high g_{em} and Φ_{lum} made possible visualization of CPCE with different helicities using a standard camera (**Fig. 3g**).

Optical stacks with tunable α, LB and LD were made from a wide variety of achiral dyes and a large family of composite materials with gradually variable strong CPCE in different parts of the spectrum that would be nearly impossible using molecular or quantum-confined chiral structures. Incorporation of tryptophan ($\Phi_{\mathrm{lum}} = 0.12$), cascade blue acetyl azide dye ($\Phi_{\mathrm{lum}} = 0.54$), and IR-783 dye ($\Phi_{\mathrm{lum}} = 0.11$) with M2-composites of $Ti_3C_2T_x$, both positive and negative circular polarizations emissions at 360, 450, and 780 nm were obtained (**Fig. 3f,g** and Supplementary Fig. 35).

Thermal-resilient polarization imaging

Elevated temperatures typically destroy circular polarization effects in current LCs[8,49], composites[50], or solutions[23,40]. Commercial circular polarizers lose their optical activity at temperatures as low as 50 °C (**Fig. 4c** and Supplementary Fig. 36d,f). Cholesteric liquid crystals lose their optical activity above 90 °C. In contrast, M2-composites of $Ti_3C_2T_x$ with and without added Ag nanowires exhibit intense circular polarization from 20 to 250 °C (**Fig. 4c** and Supplementary Fig. 36a,c).

Benefiting from its thermal stability, LBL $Ti_3C_2T_x$ composites may serve as an effective circular modulator for polarized imaging of hot emitters, particularly in the

NIR range (**Fig. 4a**). We demonstrated this by imaging flames from burning organic fuels with strong NIR radiation in the range of 1300–1500 nm and their circular polarization modulated by M2-composites from $Ti_3C_2T_x$ (**Fig. 4b**, Supplementary Figs. 37 and 38).

The polarization contrast can be optimized in a highly predictable manner by the sequence of LBL layers (**Fig. 4d**). This capability can be expanded to any part of the NIR spectrum including those currently inaccessible with LCs and needed for a variety of technologies[51,52].

In conclusion, the combination of chiroptical, mechanical, and thermal properties demonstrated for layered nanocomposites from 2D materials with LB/LD effects are conceptually difficult to achieve for organic materials and nanostructures with isotropic CD. The manufacturing simplicity, materials modularity, and computational predictability of the additively engineered composites lead to a large family of optically active materials with millions of potential permutations of nanoscale components, organizational patterns, and optical effects.

References

1. VahidMohammadi, A., Rosen, J. & Gogotsi, Y. The world of two-dimensional carbides and nitrides (MXenes). *Science* **372**, eabf1581 (2021).
2. Nepal, D. et al. Hierarchically structured bioinspired nanocomposites. *Nat. Mater.* **22**, 18–35 (2023).
3. Toyoshima, M. et al. Polarization measurements through space-to-ground atmospheric propagation paths by using a highly polarized laser source in space. *Opt. Express* **17**, 22333–22340 (2009).
4. Ott, M. N. et al. Space flight requirements for fiber optic components: qualification testing and lessons learned. *Reliab. Opt. Fiber Components, Devices, Syst. Networks III* **6193**, 619309 (2006).
5. Dupeyroux, J., Serres, J. R. & Viollet, S. AntBot: a six-legged walking robot able to home like desert ants in outdoor environments. *Sci. Robot.* **4**, eaau0307 (2019).
6. Guo, X. et al. Full-color holographic display and encryption with full-polarization degree of freedom. *Adv. Mater.* **34**, 2103192 (2022).
7. Fang, X., Ren, H. & Gu, M. Orbital angular momentum holography for high-

security encryption. *Nat. Photonics* **14**, 102–108 (2020).
8. Khoo, I.-C. *Liquid crystals* (John Wiley & Sons, 2022).
9. Kawamoto, H. The history of liquid-crystal displays. *Proc. IEEE* **90**, 460–500 (2002).
10. Probst, P. T. et al. Mechano-tunable chiral metasurfaces via colloidal assembly. *Nat. Mater.* **20**, 1024–1028 (2021).
11. Han, H. et al. Multiscale hierarchical structures from a nanocluster mesophase. *Nat. Mater.* **21**, 518–525 (2022).
12. Giese, M., Blusch, L. K., Khan, M. K. & MacLachlan, M. J. Functional materials from cellulose-derived liquid-crystal templates. *Angew. Chem. Int. Ed.* **54**, 2888–2910 (2015).
13. Hu, H. et al. Nanoscale bouligand multilayers: giant circular dichroism of helical assemblies of plasmonic 1D nano-objects. *ACS Nano* **15**, 13653–13661 (2021).
14. Lv, J. et al. Gold nanowire chiral ultrathin films with ultrastrong and broadband optical activity. *Angew. Chem. Int. Ed.* **56**, 5055–5060 (2017).
15. Laufer, G., Kirkland, C., Cain, A. A. & Grunlan, J. C. Clay–chitosan nanobrick walls: completely renewable gas barrier and flame-retardant nanocoatings. *ACS Appl. Mater. Interfaces* **4**, 1643–1649 (2012).
16. Magrini, T. et al. Transparent and tough bulk composites inspired by nacre. *Nat. Commun.* **10**, 2794 (2019).
17. Suzuki, N. et al. Chiral graphene quantum dots. *ACS Nano* **10**, 1744–1755 (2016).
18. Jiang, W. et al. Emergence of complexity in hierarchically organized chiral particles. *Science* **368**, 642–648 (2020).
19. Kurtina, D. A. et al. Induction of chirality in atomically thin ZnSe and CdSe nanoplatelets: strengthening of circular dichroism via different coordination of cysteine-based ligands on an ultimate thin semiconductor core. *Materials* **16**, 1073 (2023).
20. Kim, Y. et al. Reconfigurable chiroptical nanocomposites with chirality transfer from the macro- to the nanoscale. *Nat. Mater.* **15**, 461–468 (2016).
21. Gansel, J. K. et al. Gold helix photonic metamaterial as broadband circular polarizer. *Science* **325**, 1513–1515 (2009).
22. Mark, A. G., Gibbs, J. G., Lee, T. C. & Fischer, P. Hybrid nanocolloids with programmed three-dimensional shape and material composition. *Nat. Mater.* **12**, 802–807 (2013).
23. Lu, J. et al. Enhanced optical asymmetry in supramolecular chiroplasmonic assemblies with long-range order. *Science* **371**, 1368–1374 (2021).
24. Zhang, D., Shah, D., Boltasseva, A. & Gogotsi, Y. MXenes for photonics. *ACS Photonics* **9**, 1108–1116 (2022).
25. Liu, R. & Li, W. High-thermal-stability and high-thermal-conductivity $Ti_3C_2T_x$ MXene/poly(vinyl alcohol) (PVA) composites. *ACS Omega* **3**, 2609–2617 (2018).
26. Kumar, P. et al. Photonically active bowtie nanoassemblies with chirality continuum. *Nature* **615**, 418–424 (2023).
27. Buda, A. B. & Mislow, K. A Hausdorff chirality measure. *J. Am. Chem. Soc.* **114**, 6006–6012 (1992).
28. Osipov, M. A., Pickup, B. T. & Dunmur, D. A. A new twist to molecular chirality: intrinsic chirality indices. *Molec. Phys.* **84**, 1193–1206 (1995).
29. Kim, J.-Y. et al. Assembly of gold nanoparticles into chiral superstructures driven by circularly polarized light. *J. Am. Chem. Soc.* **141**, 11739–11744 (2019).
30. Zhou, S. et al. Chiral assemblies of pinwheel superlattices on substrates. *Nature*

612, 259–265 (2022).
31. Purcell-Milton, F. et al. Induction of chirality in two-dimensional nanomaterials: chiral 2D MoS_2 nanostructures. *ACS Nano* **12**, 954–964 (2018).
32. Zhang, H., He, H., Jiang, X., Xia, Z. & Wei, W. Preparation and characterization of chiral transition-metal dichalcogenide quantum dots and their enantioselective catalysis. *ACS Appl. Mater. Interfaces* **10**, 30680–30688 (2018).
33. Salij, A., Goldsmith, R. H. & Tempelaar, R. Theory of apparent circular dichroism reveals the origin of inverted and noninverted chiroptical response under sample flipping. *J. Am. Chem. Soc.* **143**, 21519–21531 (2021).
34. Albano, G., Pescitelli, G. & Di Bari, L. Chiroptical properties in thin films of π-conjugated systems. *Chem. Rev.* **120**, 10145–10243 (2020).
35. Yao, Y. et al. Extracting pure circular dichroism from hierarchically structured CdS magic cluster films. *ACS Nano* **16**, 20457–20469 (2022).
36. Ermolaev, G. A. et al. Giant optical anisotropy in transition metal dichalcogenides for next-generation photonics. *Nat. Commun.* **12**, 854 (2021).
37. Papadopoulos, N. et al. Large birefringence and linear dichroism in TiS_3 nanosheets. *Nanoscale* **10**, 12424–12429 (2018).
38. Tarjányi, N., Turek, I. & Martincek, I. Effect of mechanical stress on optical properties of polydimethylsiloxane II – birefringence. *Opt. Mater.* **37**, 798–803 (2014).
39. Aftenieva, O., Schnepf, M., Mehlhorn, B. & König, T. A. F. Tunable circular dichroism by photoluminescent moiré gratings. *Adv. Opt. Mater.* **9**, 2001280 (2021).
40. Xu, L. et al. Enantiomer-dependent immunological response to chiral nanoparticles. *Nature* **601**, 366–373 (2022).
41. González-Rubio, G. et al. Micelle-directed chiral seeded growth on anisotropic gold nanocrystals. *Science* **368**, 1472–1477 (2020).
42. Xiong, R. et al. Integration of optical surface structures with chiral nanocellulose for enhanced chiroptical properties. *Adv. Mater.* **32**, 1905600 (2020).
43. Wu, W., Battie, Y., Lemaire, V., Decher, G. & Pauly, M. Structure-dependent chiroptical properties of twisted multilayered silver nanowire assemblies. *Nano Lett.* **21**, 8298–8303 (2021).
44. Hubley, A. et al. Chiral perovskite nanoplatelets exhibiting circularly polarized luminescence through ligand optimization. *Adv. Opt. Mater.* **10**, 2200394 (2022).
45. Liu, J. et al. What makes efficient circularly polarised luminescence in the condensed phase: aggregation-induced circular dichroism and light emission. *Chem. Sci.* **3**, 2737–2747 (2012).
46. Jiang, S. & Kotov, N. A. Circular polarized light emission in chiral inorganic nanomaterials. *Adv. Mater.* **35**, 2108431 (2023).
47. Mukthar, N. F. M., Schley, N. D. & Ung, G. Strong circularly polarized luminescence at 1550 nm from enantiopure molecular erbium complexes. *J. Am. Chem. Soc.* **144**, 6148–6153 (2022).
48. Wu, Y., Li, M., Zheng, Z., Yu, Z.-Q. & Zhu, W.-H. Liquid crystal assembly for ultra-dissymmetric circularly polarized luminescence and beyond. *J. Am. Chem.Soc.* **145**, 12951–12966 (2023).
49. Dierking, I. Chiral liquid crystals: structures, phases, effects. *Symmetry* **6**, 444–472 (2014).
50. He, H. et al. Cholesteric-superhelix-enabled reconfigurable circularly polarized luminescence from uniaxially aligned upconversion nanorod films. *Laser Photonics Rev.* **16**, 2200115 (2022).

51. Neshev, D. N. & Miroshnichenko, A. E. Enabling smart vision with metasurfaces. *Nat. Photonics* **17**, 26–35 (2023).
52. Kim, W. et al. Thermally-curable nanocomposite printing for the scalable manufacturing of dielectric metasurfaces. *Microsystems Nanoeng.* **8**, 73 (2022).

Figure legends

Figure 1. Nanocomposites from 2D nanomaterials prepared by Method 1; uniform, tunable left- and right-handed polarization rotation. a, Schematics of Method 1. **b,** Photograph of LH and RH $Ti_3C_2T_x$ composites deposited on grooved PET. **c,d,** Three-dimensional (**c**) and high-magnification (**d**) atomic force microscope images showing that LBL deposition of $Ti_3C_2T_x$ nanoplatelets in the PDMS grooves results in nanoscale wrinkles. **e,f,** CD spectra (**e**) and CD mapping (**f**) of the composites at each maximum polarization rotation amplitude for LH (left) and RH (right) M1-composites of $Ti_3C_2T_x$ using MMP. **g,** Dependence of CD intensities on the dihedral angle between LB and LD, α, of the composites.

Figure 2. Nanocomposites from 2D nanomaterials prepared by Method 2; real-time reconfigurable polarization rotation. a, Schematics of Method 2. **b,** Photograph of $Ti_3C_2T_x$ composites LBL assembled on a twisted PDMS strip and the corresponding released state. **c,** Three-dimensional atomic force microscope images for the wrinkled (left) and cracked (right) sides of RH M2-composites of $Ti_3C_2T_x$. **d,** CD spectra for the composites under a 10% tensile strain. **e,** CD mapping at each maximum polarization rotation amplitude for the LH (left) and RH (right) composites under various strain amplitudes. **f,** Cycling test for the polarization rotation of M2-composites of $Ti_3C_2T_x$ under a periodic 10% tensile strain. **g,** Scanning electron microscopy image of Ag nanowire films with a controlled orientation. **h,** CD spectra for 25%-strained composites with Ag nanowire films under various α via rotating Ag nanowire films; **i,** corresponding dependence of *g*-factor with α.

Figure 3. Generation and modulation of strong CPCE. a, Schematics for the generation of CPCE with M2-composites that contain a LD layer of nanoplatelet wrinkles and a LB layer of strained PDMS. **b,** CPCE spectra for rhodamine 6G under the modulation of an unstrained M2-composite of $Ti_3C_2T_x$ with a 25%-strained PDMS, at various α via rotating the LD direction in the *x*-*y* plane. **c–e,** Dependence of g_{em} on α for the composites with LD from the unstrained $Ti_3C_2T_x$ composite (**c**), an aligned Ag nanowire film (**d**), and a linear polarizer (**e**). **f,** Normalized CPCE spectra (left) and corresponding emission photographs (right) for different fluorescent dyes modulated with $Ti_3C_2T_x$ composites at α of 30 (dash) and 330° (solid). **g,** Photographs to image the circular polarization of emission with LH and RH circular filters for the fluorescent dyes of cascade blue acetyl azide (blue) and rhodamine 6G (green) with polarization modulated with composites that had different LD originations.

Figure 4. Polarization imaging in the near-infrared range using thermally resilient composites. a, Schematics of circularly polarized optical imaging with LH and RH composites. **b,** Images and corresponding intensity profiles for the LH and RH polarized flame recorded by a NIR camera incorporated with M2-composites of $Ti_3C_2T_x$ and Ag nanowire layers for circular polarization filtering. **c,** Dependence of temperatures on CD for different composite films. **d,** Dependence of the polarization anisotropy for flame with LH NIR polarization. Data are mean ± s.d.

Methods

Fabrication of M1-composites

Both M1- and M2-composites were fabricated on meticulously designed substrates with the LBL assembly of nanoplatelets, which impart exceptional capabilities to the resulting films, including: (1) self-limited deposition enabling conformal coating on complex surfaces; (2) high-temperature resilience and mechanical robustness of the resulting composite films; (3) strong adhesion to the wide range of substrates; (4) predictability of the optical properties; and (5) suitability for all or nearly 2D nanomaterials and all optical substrates.

For M1-composites, their fabrication was via the LBL assembly of nanoplatelets on biaxially oriented PET with grooves patterned by wrinkled stamps in a soft-lithography technique. The process includes three steps:

(I) Preparation of wrinkled stamps. The wrinkled stamps were prepared by oxygen plasma etching to strained PDMS strips. PDMS strips were cut into 20 × 35 mm from PDMS films that were previously made by 20 mg Sylgard 184 silicone elastomer and 2.0 mg cross-linker base in a 12 × 12 cm petri dish. The obtained PDMS strip had a thickness of 1 mm and was 40%-strained along the length, fixed by clamps under the strain, and etched by oxygen plasma (Evactron® Plasma De-Contaminator™, 20 W for 20 min). Then 5 alternating PSS/PDDA bilayer units were LBL deposited on the strained PDMS to act as a spacer between the PDMS stamp and patterned grooves. Releasing the PDMS strip from the clamps obtained wrinkled stamps with a groove interval and height/depth of 840 ± 60 and 230 ± 60 nm (Supplementary Table 2). Similarly, the wrinkled stamps were made with 10%-, 20%-strained PDMS and commercial digital versatile discs that got wrinkles with groove intervals of 1220 ± 60, 1030 ± 60, and 760 ± 60 nm and heights of 90 ± 20, 220 ± 50, and 150 ± 10 nm, respectively.

(II) Soft-lithography to pattern grooves on PET. The PET film (150 μm in thickness) was cut into 75 × 50 mm, fixed on a glass slide, and treated with plasma clearing (5 W for 5 min) before dropping 300 μL mixture solution of silicone elastomer and cross-linker base (1:10). After the addition of spacer (150 μm) and spreading out of dropped solution, the wrinkled stamps prepared in step (I) were covered onto the top of the solution with an angle of 135° and 45° to the uniaxially stretched axis of PET film for the fabrication of LH and RH composites (**Fig. 1a**). The obtained PET film was placed in a desiccator to remove bubbles and then cured at 60 °C overnight. Afterward, the PDMS stamp was peeled off to obtain PET film with pattered PDMS grooves. The PDMS layer has a thickness of 150 μm determined by the spacer with the same groove intervals as the wrinkled stamps.

(III) LBL assembly on the patterned PET. The PET film with patterned grooves underwent a cleaning and activation process to impart hydrophilic properties under an ultraviolet ozone surface cleaner (5 W for 1 min). The activated strip was dipped into a 0.5 wt% PDDA solution for 5 min, then transferred to DI water to wash out unbound polymers, and dried with compressed air. Next, the positively charged PDMS strip was immersed into negatively charged Lys-capped $Ti_3C_2T_x$ nanoplatelets solution for 10 min to assemble one layer of $Ti_3C_2T_x$, then transferred to DI water to wash out unbound $Ti_3C_2T_x$ nanoplatelets, and dried with compressed air. The obtained strip was dipped into a PDDA solution to begin the next cycle of LBL assembly until 10 bilayers of $Ti_3C_2T_x$-PDDA deposition in the grooves. The formed wrinkled patterns of nanoplatelet films were well-aligned with a tilt angle of 45° and 135° with respect to the long axis of PET. A similar LBL assembly procedure was used to fabricate MoS_2 and GO composites with 5 bilayers of nanoplatelet-PDDA deposition considering the dominant

multilayer nanostructures.

Fabrication of M2-composites

M2-composites were fabricated by the LBL assembly of nanoplatelets on the twisted substrate of PDMS strips, followed by a releasing process to flat the twisted assembles to obtain composites with cracked and wrinkled patterns on the compressed and stretched sides. This process was divided into two steps:

(I) Fabrication of twisted substrate. PDMS strips were cut into a size of 10 × 45 mm, twisted 360° in either a clockwise or anticlockwise direction to get the LH and RH PDMS substrate, and fixed on the glass slide with clamps for further usage.

(II) LBL assembly of nanoplatelets on the twisted PDMS. The twisted PDMS strip was cleaned and activated with hydrophilic properties under plasma cleaner (20 W for 20 min). Then the activated strip was covered by a layer of PDDA and rinsed by negatively charged Lys-capped $Ti_3C_2T_x$ nanoplatelets solution for 5 min to fully deposit one layer of $Ti_3C_2T_x$. Repeating the LBL cycles until a composite film with 6 bilayers of $Ti_3C_2T_x$-PDDA formed on each side of the twisted PDMS. Afterward, releasing the twisted strip from clamps obtained M2-composites of $Ti_3C_2T_x$ for characterization. The same LBL assembly procedure was used to fabricate M2-composites of MoS_2 via the electrostatic interaction of positively charged PDDA with negatively charged MoS_2 nanoplatelets covered with polyoxometalates.

Fabrication of Ag nanowire films

A glass slide was cleaned with ethanol under sonication and then plasma-cleaned to make the slide hydrophilic. A layer of polyethylenimine (PEI) was deposited on the hydrophilic substrate by LBL assembly under the spraying of PEI and rinsed with water to wash out unbound PEI. Afterward, the suspension of Ag nanowires was sprayed on the PEI-coated substrate along airflow with a spraying nozzle (B1/4J, Spraying Systems) to align the Ag nanowires on the substrate via a grazing incidence spraying method[13,53]. Under the airflow, the Ag nanowires suspension was atomized into small droplets and aligned on the substrate under the shear force along the direction of airflow. The liquid flow rate was set to 1 mL/min using a liquid pump (M50, Valco Instruments Co.), while the airflow rate was set to 30 L/min with a flow meter (Red-Y, Vögtlin Instruments GmbH). The incidence angle of the shear force spray was 10°. The distance from the nozzle to the substrate was 1.0 cm. The deposition took 200 seconds to get a dense film with one layer of Ag nanowires. The obtained glass with well-aligned Ag nanowires was rinsed with water and dried with compressed air.

Mueller calculus analysis for M1- and M2-composites

The Stokes vector (*S*) describes the light-matter interactions of a beam of light passing through optically active media using four measurable parameters[54,55], *I, Q, U, V*:

$$S = \begin{pmatrix} I \\ Q \\ U \\ V \end{pmatrix} \quad \text{(Eq. 3)}$$

where I is the total light intensity, Q is the averaged intensity difference between the linear polarized components associated with the x-and y-axes (i.e. 0° and 90°), U is the averaged intensity difference between the linear polarized components along 45° and 135° axes, and V is the difference between the averaged intensities between LH and RH circular polarized components.

The polarization properties of the medium can be represented by a Mueller matrix,

M, that is related to vectors S_{in} and S_{out} for the incoming and outcoming beams as:

$$S_{out} = M \cdot S_{in}$$

$$\begin{pmatrix} I_{out} \\ Q_{out} \\ U_{out} \\ V_{out} \end{pmatrix} = \begin{pmatrix} M_{00} & M_{01} & M_{02} & M_{03} \\ M_{10} & M_{11} & M_{12} & M_{13} \\ M_{20} & M_{21} & M_{22} & M_{23} \\ M_{30} & M_{31} & M_{32} & M_{33} \end{pmatrix} \begin{pmatrix} I_{in} \\ Q_{in} \\ U_{in} \\ V_{in} \end{pmatrix} \quad \text{(Eq. 4)}$$

Considering the light-matter interactions characteristic for each element M_{ij}, the Mueller matrix M can be written as measurable polarization components:

$$M = \exp\begin{pmatrix} \mathrm{T} & -\mathrm{LD} & -\mathrm{LD}' & \mathrm{CD} \\ -\mathrm{LD} & \mathrm{T} & \mathrm{CB} & \mathrm{LB}' \\ -\mathrm{LD}' & -\mathrm{CB} & \mathrm{T} & -\mathrm{LB} \\ \mathrm{CD} & -\mathrm{LB}' & -\mathrm{LB} & \mathrm{T} \end{pmatrix} \quad \text{(Eq. 5)}$$

where T represents light transmittance, LD represents horizontal linear dichroism associated with the *x*- and *y*-axes, LB represents horizontal linear birefringence, LD' represents 45° linear dichroism, LB' represents 45° linear birefringence, CD represents circular dichroism, CB represents circular birefringence of the media, and '–' represents the opposite intensities of corresponding each signal of LB, LD and CB in the matrix.

The CD of anisotropic optical active media can be represented by the M_{03} and M_{30} elements in the Mueller matrix. They contain both the 'true' and 'apparent' CD[33–35,56]:

$$M_{03} = z\mathrm{CD} - \frac{z^2}{2}(-\mathrm{LD} \cdot \mathrm{LB}' + \mathrm{LB} \cdot \mathrm{LD}') \quad \text{(Eq. 6)}$$

$$M_{30} = z\mathrm{CD} + \frac{z^2}{2}(-\mathrm{LD} \cdot \mathrm{LB}' + \mathrm{LB} \cdot \mathrm{LD}')$$

where z is the light path in a medium, zCD is the 'true' isotropic CD, and the term of $\frac{z^2}{2}(-\mathrm{LD} \cdot \mathrm{LB}' + \mathrm{LB} \cdot \mathrm{LD}')$ is the 'apparent' CD from the LB+LD interaction that occurs due to the inhomogeneity of the media and the non-zero angle between the optical axes of LD and LB. The 'apparent' CD scales as z^2 due to the involvement of both LD and LB, which is different from the isotropic CD with a linear scale to the light path of z. Note that, in real testing, the z has been included in the final spectra of CD, LB and LD that actually correspond to zCD, zLB and zLD in Eq. 6. For simplification reasons, the z term in the front of each optical polarization component has been ignored for the data analysis of MMP.

The 'apparent' CD is not an artifact, as it is sometimes referred in the literature since it does change the circular polarization state of circularly polarized light when the light passes through an anisotropic medium. It is a different optical process in comparison to 'true' CD (Supplementary Fig. 7). The sum of all CD components is detected by spectropolarimeters.

The actual artifact signals, $\mathrm{CD}_{\mathrm{artifact}}$, may come from the residual static birefringence of photoelastic modulators, the anisotropy of photomultipliers, and other optical components.[34,35] Thus, the complete equation for $\mathrm{CD}_{\mathrm{detected}}$ becomes:

$$\mathrm{CD}_{\mathrm{detected}} = z\mathrm{CD} + \frac{z^2}{2}(-\mathrm{LD} \cdot \mathrm{LB}' + \mathrm{LB} \cdot \mathrm{LD}') + \mathrm{CD}_{\mathrm{artifact}} \quad \text{(Eq. 7)}$$

In commercial instruments, $\mathrm{CD}_{\mathrm{artifact}}$ is negligible and will be ignored in the following discussion, note, however, that $\mathrm{CD}_{\mathrm{artifact}}$ cannot be ignored in the custom-designed instruments and should be quantitively assessed.

To give a more tangible understanding of how the structure of composites affects the light-matter interactions let us consider M1-composites from $Ti_3C_2T_x$. The term of zCD is approaching zero since the $Ti_3C_2T_x$ nanoplatelets LBL-assembled on the grooves are achiral (Supplementary Fig. 19). The linear dichroism is along the groove direction of 45° and 135° with respect to the *x*-axis (0°) that should result in large LD'

and zero LD. The negligible amplitude of the -LD component across the spectrum of interest can be also seen in Supplementary Fig. 11c for the M_{01} element in the Mueller matrix. In addition, the 'built-in' linear birefringence axis of PET is along the *y*-axis, resulting in large LB (associated with the *x*- and *y*-axes) and small even negligible LB' (associated with 45° and 135°) as shown in the M_{23} and M_{13} elements in the Mueller matrix (Supplementary Fig. 11e,f). Therefore, Eq. 7 can be simplified as:

$$\mathrm{CD}_{\mathrm{detected}} \propto \mathrm{LB} \cdot \mathrm{LD}' \quad \text{(Eq. 8)}$$

Eq. 8 is also true for M2-composites since the LB' of the composite tends to be zero. This is because the linear birefringence under strain is aligned to the stretching direction (*y*-axis) of PDMS strips, bringing large -LB; whereas, the vector along 45° and 135° is equal, corresponding to the measurement of LB'. The negligible amplitude of LB' across the spectrum of interest can be seen in the M_{13} elements in the Mueller matrix (Supplementary Fig. 29g).

Characterization protocols

CD and extinction spectra were acquired using a JASCO J-1700 CD spectrophotometer with the *g*-factors to record the anisotropy of circular polarization: $g-\mathrm{factor} = \frac{\mathrm{CD}}{\mathrm{Extinction}} \times \frac{1}{32980}$, where the units of mdeg and a.u. were used to scale CD and extinction intensities. The switch from photomultiplier tube (190–800 nm) to InGaAs (800–1300 nm) detector results in the discontinuity at 800 nm for the intensities of CD, extinction and *g*-factor spectra obtained by JASCO J-1700, while not significantly influencing the spectral shape. MMP CD, other multiple optical polarization elements in the Mueller matrix, and polarization mapping were obtained with a Mueller matrix polarimeter by Hinds Instrument. Besides 'true' isotropic CD, both the mentioned instruments measure CD originating from the light interacting with LD- and LB-active strata. This optical activity can be referred to as 'apparent' CD; this term only underscores the difference with what other publications referred to as 'true' or isotropic CD originating from the mirror asymmetry of (nano)structures freely dispersed in the media and CD originating from other polarization components.

CPCE was measured with a JASCO CPL-300 spectrophotometer with g_{em} to record the anisotropy of circular polarization in the light emission: $g_{\mathrm{em}} = \frac{\mathrm{CPCE}}{\mathrm{DC}} \times \frac{1}{32980} \times \ln(10)$, where CPCE adopts a unit of mdeg, and direct current, DC, is the fluorescence intensity recorded by photomultiplier tubes with a unit of V. Near-infrared CPCE spectra were recorded by OLIS NIR CPL Solo spectrometer with Hamamatsu thermoelectric cooled near-infrared photomultiplier tubes from the range of 900 to1700 nm.

The mechanical properties of $Ti_3C_2T_x$ composite film were measured with nanoindentation by Hysitron TI 950 Nanoindenter (Bruker, USA) with a Berkovich probe on a silicon wafer. For the measurement of mechanical ruggedization of $Ti_3C_2T_x$ composites on soft PDMS strips with a height of 1 cm, a spherical probe with a diameter of 50 μm was used for the indentation test. After every 5 or 10 bilayers of $Ti_3C_2T_x$-PDDA deposition, nanoindentation was performed until 80 bilayers. All measurements were conducted in a displacement control mode with a peak displacement of 10, 30, and 50 nm for the composites with a total LBL thickness of 340 nm on a silicon wafer and 1 μm for the composites on PDMS substrates. 6 random points in an area of 5 × 5 mm were indented during the measurement. Reduced modulus (E_r) and hardness were fitted from the force-displacement curves, while Young's modulus (E) was calculated with[57]: $\frac{1}{E_r} = \frac{(1-\nu^2)}{E} + \frac{(1-\nu_i^2)}{E_i}$, where E_r is the fitted reduced modulus, ν is the Poisson's

ratios of PDMS (0.5) and $Ti_3C_2T_x$ (0.227), ν_i (0.2) is the Poisson's ratio of diamond indenter with corresponding Young's modulus (E_i) of 1220 GPa, and E is Young's modulus of the sample.

The refraction index of $Ti_3C_2T_x$ was measured with an ellipsometer of J.A. Woollam M-2000 VASE at three angles of 50°, 60°, and 70° to one layer of $Ti_3C_2T_x$ nanoplatelets LBL assembled on a Si wafer. The final refraction index used for simulation was averaged over the three angles. Scanning electron microscopy (SEM) images were taken by FEI Nova 200 Nanolab Dual Beam SEM and FEI Helios NanoLab 650 dual-beam SEM with an acceleration voltage of 5 kV and a current of 0.4 nA. Atomic force microscope (AFM) images were taken with a Veeco Dimension Icon AFM system with Bruker probes of RTESPA-150 and analyzed with NanoScope Analysis 2.0. X-ray Photoelectron Spectroscopy (XPS) spectra were obtained with the Kratos Axis Ultra spectrometer. The zeta-potential and hydrodynamic diameter of nanoplatelets were measured by a Zetasizer Nano ZSP (Malvern Instruments Ltd., GB).

Data availability

The authors declare that the data supporting the findings of this study are available within the article and its supplementary information files. Source data are provided with this paper.

Code availability

The codes used for chirality index calculations are available from https://github.com/aslozada/kanon and https://github.com/colombarifm/OPD_chirality_index.

53. Blell, R. et al. Generating in-plane orientational order in multilayer films prepared by spray-assisted layer-by-layer assembly. *ACS Nano* **11**, 84–94 (2017).
54. He, H. et al. Mueller matrix polarimetry–an emerging new tool for characterizing the microstructural feature of complex biological specimen. *J. Light. Technol.* **37**, 2534–2548 (2019).
55. Arteaga, O. & Kahr, B. Mueller matrix polarimetry of bianisotropic materials. *J. Opt. Soc. Am. B* **36**, F72–F83 (2019).
56. Arteaga Barriel, O. *Mueller matrix polarimetry of anisotropic chiral media* (University of Barcelona, 2010).
57. Oliver, W. C. & Pharr, G. M. An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments. *J. Mater. Res.* **7**, 1564–1583 (1992).

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Authors' Contributions: J.Lu and N.A.K designed the experiments and analyzed the data. J.Lu fabricated the composites. W.W. prepared the Ag film, ran the mechanical testing, and contributed to property discussions. J.Lu, K.W., and W.W. ran the AFM imaging and studied the structure of composites. A.J. and R.V. prepared MoS_2 nanoplatelets. B.S. and D.N. synthesized pristine $Ti_3C_2T_x$ nanoplatelets. F.C. and A.F.M. performed the DFT and MD simulations and chirality index calculations. X.Z. and J. Lahann performed the measurement and data analysis for the refractive index of $Ti_3C_2T_x$ composites, which was used by J.Lu on FDTD simulations. W.C. assisted J. Lu with MMP measurement and contributed to property discussions. J.Lu, W.W., F.C., A.F.M., D.N., R.V., and N.A.K. contributed to the writing of the paper with feedback from all authors. N.A.K. conceived and supervised the project.

Competing interests
The authors declare no competing interests.