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Eugene Terentjev

`emt10000@cam.ac.uk`

University of Cambridge <https://orcid.org/0000-0003-3517-6578>

Bao Jin

University of Cambridge

Lichang Lu

University of Cambridge

Yuheng Huang

Cambridge Smart Plastics Ltd

Kailing Lin

Cambridge Smart Plastics Ltd

Mohand Saed

University of Cambridge

Article

Keywords:

Posted Date: July 2nd, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10018087/v1>

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Additional Declarations: There is **NO** Competing Interest.

Topologically Engineered Nematic Networks with Ultrastrong Adhesion and Light-Debonding on Demand

Bao Jin^{1,2}, Lichang Lu¹, Yuheng Huang³, Kailing Lin³, Mohand O. Saed¹, Eugene M. Terentjev^{1*}

¹Cavendish Laboratory, University of Cambridge, Cambridge, CB3 0HE, UK

²Department of Mechanical Engineering, Tsinghua University, Beijing, 100084, China

³Cambridge Smart Plastics Ltd., 18 Hurrell Road, Cambridge, CB4 3RH, UK

*Corresponding author: emt1000@cam.ac.uk

Abstract: Adhesives that combine high bonding strength with rapid, controllable debonding remain fundamentally constrained by an intrinsic trade-off between adhesion and reversibility. Conventional pressure-sensitive adhesives (PSAs) rely on viscoelastic dissipation to achieve strong bonding, but their performance is fixed by formulation and cannot be actively switched. Nematic liquid crystal elastomers (LCEs) provide a distinct route in which adhesion is governed by bulk orientational order and can be modulated by the nematic–isotropic phase transition, although the achievable adhesion strength has so far remained limited.

Here we show that network topology engineering transforms nematic elastomers into ultrastrong and reversibly switchable adhesives that exceed industrial benchmarks while enabling remote release on demand. By reducing the fraction of multifunctional crosslinkers, we introduce a dense population of dangling oligomer chains within a percolated nematic network, producing adhesion strengths above 4500 N/m and substantially surpassing commercial duct tapes. Incorporation of MXene nanoplatelets further enhances adhesion while simultaneously enabling rapid photothermal switching through localized heating across the nematic–isotropic transition. Illumination collapses adhesion by more than an order of magnitude (>97%), while cooling restores the adhesive state.

The resulting adhesive behaviour differs fundamentally from that of conventional PSAs because the bonded state evolves through slow reorganization of orientational order under confinement. These results establish a general strategy for ultrastrong yet reversibly switchable adhesion in which network topology, orientational order and photothermal activation are coupled within a single soft material system.

Introduction

Liquid crystalline elastomers (LCEs) occupy a singular position among soft materials, combining the orientational order of liquid crystals with the elasticity of polymer networks.^{1–3} This coupling produces a range of unconventional mechanical responses, including soft elasticity and, separately, an anomalously high viscoelastic dissipation in the nematic phase—two effects that, while often conflated in the literature, arise from distinct physical mechanisms⁴ and need not be directly linked.

Adhesion in soft polymeric systems, particularly in pressure-sensitive adhesives (PSAs), is conventionally understood as a balance between interfacial interactions and bulk viscoelastic dissipation during debonding. In classical PSA formulations, lightly crosslinked elastomeric networks are augmented by mobile, non-networked components, so-called tackifiers, that enhance adhesion by promoting surface wetting and increasing energy dissipation at the crack tip.⁵⁻⁸ Despite their widespread use, such systems remain fundamentally constrained: their adhesive properties are fixed by composition and can only be tuned incrementally through external variables such as temperature or loading rate.^{5,6,9} Considerable efforts have therefore been devoted to stimuli-responsive adhesives capable of external control by temperature, light, electric fields or chemical triggers. However, existing approaches typically achieve switchability only at the expense of adhesive strength, or rely on complex surface architectures and reversible chemistries that are difficult to integrate into robust load-bearing systems.¹⁰⁻¹⁴ Among commercial PSAs, heavy-duty duct tapes represent an important practical benchmark because they combine high peel strength with mechanical robustness under load. Achieving comparable adhesion together with rapid and reversible debonding remains a major unresolved challenge in soft materials.

Recent work has revealed that nematic LCEs exhibit an anomalously strong pressure-sensitive adhesion that departs qualitatively from this classical framework.¹⁵⁻¹⁹ Unlike conventional PSAs, where adhesion is primarily governed by dissipative processes during detachment, LCE's adhesion is strongly dependent on the internal state of orientational order¹⁹ and can be reversibly modulated by the nematic-isotropic phase transition.¹⁵⁻¹⁷ The mechanism underlying this behaviour has been traced to the slow relaxation and reorganization of the polydomain nematic texture under confinement, leading to the formation of a deeply metastable bonded state with significant stored elastic energy.¹⁶ This introduces a fundamentally different paradigm in which adhesion is controlled by the history-dependent evolution of bulk structure during bonding, rather than by interfacial physics alone.

Here we advance this concept by introducing two independent and synergistic design strategies that transform LCE adhesion from an anomalous material property into a controllable functional platform. First, we engineer the topology of the elastomer network by deliberately reducing the fraction of multifunctional crosslinkers, thereby generating a substantial population of dangling oligomer chains within a percolated nematic network. While the use of mobile chains (tackifiers) within a network is a well-established strategy in conventional PSA design,^{5,20} in the present system these chains are embedded within a liquid crystalline matrix, where their dynamics are coupled to orientational order and its slow relaxation. Second, we incorporate 2D MXene nanoplatelets, which not only enable rapid photothermal switching under light illumination, but also further enhance adhesive performance. The resulting material combines two properties that are conventionally mutually exclusive: adhesion strengths significantly exceeding those of industrial duct tapes, and rapid, reversible debonding remotely activated by light. This behaviour extends beyond the conventional PSA framework and instead emerges from the interplay between network topology, nematic order and phase-transition-driven mechanics. These results establish a new design principle for soft adhesives, where strong bonding and controlled release are achieved within a single, structurally simple material system.

Results

1. Network topology engineering drives adhesion beyond industrial benchmarks

We first establish the effect of network topology on the pressure-sensitive adhesion of nematic LCEs. In contrast to the traditional stoichiometric elastomer networks we deliberately reduce the fraction of multifunctional crosslinkers, creating a percolated network that contains a substantial population of dangling oligomer chains (Fig. 1a-c). The oligomer backbone length is held constant at approximately ten mesogenic units (Fig. 1b), allowing the role of network topology to be isolated directly. The combined effects of network topology engineering and MXene nanoplatelet incorporation are immediate and striking.

As shown in Fig. 1d, the 'optimal' composite network exhibits peel strength exceeding 4500 N/m after thermal annealing, representing more than an order-of-magnitude enhancement over the previously reported LCE systems and placing the material well beyond the performance envelope of conventional pressure-sensitive adhesives. Commercial duct tapes, widely regarded as a benchmark for high-performance PSA, typically exhibit peel strengths in the range of 400-800 N/m under comparable conditions (Supplementary Fig. S1). The present LCE adhesive therefore surpasses industrial standards by a substantial margin, exploiting the enhanced damping of the nematic phase. Importantly, the outstanding adhesion strength achieved by combining the elastomer network with MXene nanoplatelet can be released on demand under remote light control (Fig. 1e, switching adhesion strength ratio reaching 97%).

Notably, the enhancement produced by MXene nanoplatelets is substantially larger than would be expected from conventional filler reinforcement alone. The nanoplatelets are present at low loading fractions and produce only modest changes in bulk mechanical properties (Supplementary Fig. S2), yet generate a pronounced increase in peel adhesion. This suggests that the nanoplatelets participate directly in the dissipative and structural processes governing adhesive bonding in the nematic state, rather than acting merely as passive mechanical fillers. This enhancement is not a trivial consequence of softening the network. All materials retain the defining characteristics of nematic LCEs, including a clear nematic-isotropic transition and elastomeric mechanical integrity (Supplementary Fig. S2 and S3). Instead, the increase in adhesion reflects a fundamental change in how the material engages with the interface: the introduction of a mobile chain population within a liquid crystalline network enables a degree of interfacial adaptation and energy storage that is inaccessible to conventional elastomeric PSAs.

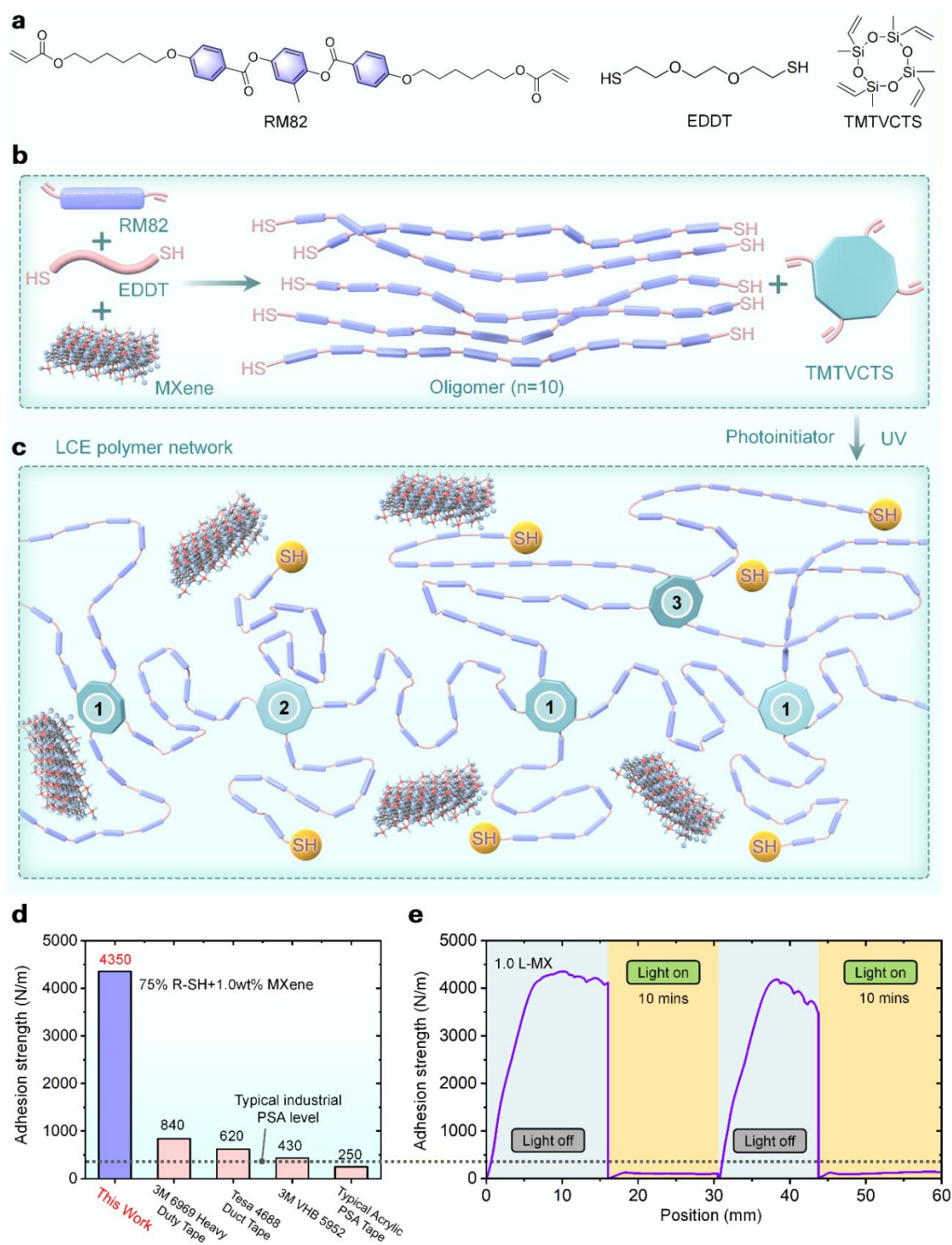


Figure 1 — Benchmark Performance and Switching

- (a) Molecules of mesogen (RM82), spacer (EDDT) and crosslinker (TMTVCTS).
- (b) Schematic of pre-polymer involved: thiol-terminated oligomer of chain length approximately 10 mesogenic units; the MXene nanoplates are employed as photothermal conversion units.
- (c) Scheme of the LCE network topology with free dangling ends, and MXene nanoplates uniformly embedded in the polymer network. The figure in each crosslink indicates the number of unconnected ends.

(d) The comparison of peel test adhesion strength results on our ‘best performer’ (the 25% crosslinked LCE - 75% R-SH dangling ends - with added 1 wt% MXene), compared with the benchmark of several commercial adhesive tapes.

(e) Illustration of switchable adhesion in the 25% crosslinked 1wt% MXene LCE composite, reversibly alternating between a very high adhesion strength in the nematic phase and low adhesion strength in the isotropic phase.

2. Dangling nematic chains amplify a fundamentally different adhesion mechanism

To resolve the origin of this enhancement, we examine the dependence of adhesion on the fraction of dangling oligomer chains at fixed oligomer length. The monotonic increase in the proportion of dangling oligomer chains in the topological network leads to a significant enhancement of extension strain (Fig. 2a), thereby exhibiting a much greater range of the soft elasticity plateau region: up to 500% strain for the 25% crosslinked (75% R-SH) LCE. Figure 2b shows a monotonic increase in peel strength as the network becomes increasingly under-constrained and soft, consistent with the growing population of dangling chains. Due to the extremely strong bonding strength, when the nematic 75% R-SH LCE is peeled off at 20°C, the bonding layer gets ripped off and a large amount of residue appears on the surface (Supplementary Fig. S4). When the peeling occurs at the isotropic phase at 100°C, the adhesion strength is low and the peeled surface remains clean, with no residue. As expected, the increase in the fraction of dangling chains leads to a significant enhancement of the second-order characteristic nematic peak of loss factor $\tan \delta$ (Supplementary Fig. S5). Remarkably, incorporation of MXene nanoplatelets produces a further substantial increase in adhesion strength (Fig. 2c), despite only modest changes in bulk mechanical properties (Supplementary Fig. S6), while still maintaining its excellent tensile strain (Supplementary Fig. S7-S9, and Supplementary Video 1-3).

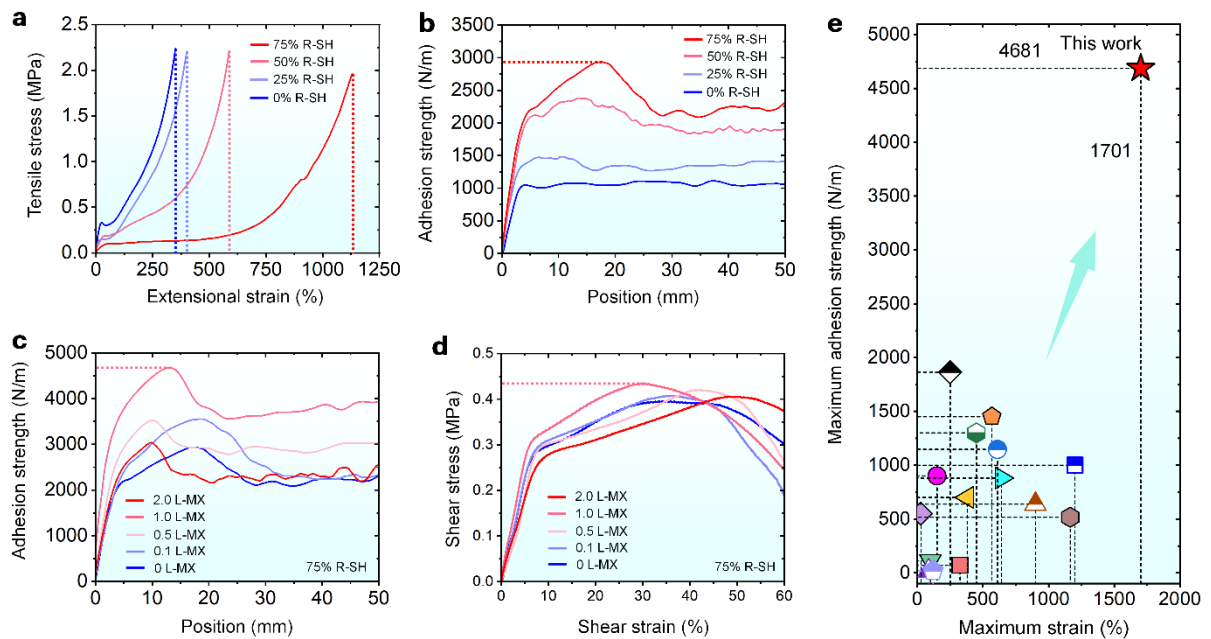


Figure 2 — Cooperative reinforcement mechanisms

(a) Tensile stress-strain curves for the increasing fraction of dangling chains.

(b) Peel strength for the increasing fraction of dangling chains.

- (c) Peel strength for 25% crosslinking (75% R-SH), and the increasing fraction of MXene nanoplatelets, showing the 1%MXene is the optimal loading.
- (d) Lap shear strength for 25% crosslinking, and the increasing fraction of MXene nanoplatelets, showing only a little difference in the lap shear strength.
- (e) Comparison of adhesion performance between materials in the literature and our optimal 25%xl-1%Mx LCE composite.

At first sight, this behaviour resembles the effect of tackifiers in classical PSA formulations,^{5,7,20} where mobile chain fractions promote wetting and dissipative processes. However, the magnitude and character of the adhesion enhancement observed here are incompatible with a purely classical interpretation. In particular, the ultrastrong adhesion develops in a material that simultaneously exhibits the slow, history-dependent relaxation of a polydomain nematic network.^{16,20} In conventional PSAs, adhesion originates predominantly from viscous dissipation generated during crack propagation and fibrillation in the debonding process.^{5,6,9,21,22} In the present system, however, the adhesive state evolves progressively through slow relaxation of the confined polydomain nematic texture prior to detachment, generating a bonded state that is both highly dissipative and mechanically metastable.

The dangling oligomer chains and nanoplatelet additives appear to amplify this process by facilitating local interfacial adaptation while preserving the slow cooperative dynamics of the surrounding nematic network. However, in the lap shear test, the enhancement of shear stress originated from MXene nanoplatelets was not significant (Fig. 2d). The subtle difference lies in the fact that the lap shear test generates an enormous magnitude of shear in the thin adhesive layer, where the adhesion strength is dominated by the binding strength and crack nucleation between the topmost distorted nematic domains in the topological network and the adjacent nematic domains.^{16,22-25} Therefore, embedded MXene nanoplatelets have no significant effect in this specific geometry. This also explains why the enormous adhesive strength recorded in the peel test is not transferred to the lap shear: the weaker LCE composite network breaks at a consistent tensile stress of 2 MPa, which translates into ca. 0.5 MPa shear stress for cohesive failure: almost exactly what we measure in Fig.2d and Supplementary Fig. S10.

Furthermore, we found that the probe-tack dynamic adhesion force of LCE is strongly affected by the pull-off velocity, temperature, and dwell time (Supplementary Fig. S11). These observations suggest a qualitatively distinct regime of adhesive behaviour. The dangling oligomer chains do not merely enhance dissipation during debonding, but actively participate in constructing the interfacial state during bonding. They facilitate rapid local rearrangement at the interface while the surrounding nematic network undergoes slow cooperative relaxation toward a lower-energy configuration. The resulting bonded state combines strong viscoelastic dissipation with substantial stored elastic energy associated with reconfigured nematic domains. Adhesion is therefore governed not only by interfacial mechanics, but also by the coupled evolution of network topology and orientational order. The LCE PSA platform designed through the network topology engineering strategy demonstrates significantly superior adhesion strength and extensible strain compared to the reported results in literature (Fig. 2e and Supplementary Table S1).

3. Photothermal activation collapses adhesion on demand

We next address the key practical limitation of high-performance adhesives: the lack of controlled debonding. To overcome this, we incorporate MXene (Ti_3C_2) photothermally active nanoplatelets into the LCE matrix (Fig. 3a). The MXene nanoplatelets play a dual role, simultaneously reinforcing adhesion and enabling photothermal switching by converting incident light into heat to trigger the nematic-isotropic transition. The temperature at the centre of the illuminated spot can reach almost 120 °C, with the temperature gradient distributed radially from the central spot (Fig. 3b).

Illumination produces a rapid temperature rise that drives the LCE material across the phase transition into the isotropic phase under modest light intensities (Fig. 3c, d). This phase transition has an immediate and dramatic effect on adhesion. Figure 1e shows that the peel strength collapses by almost two orders of magnitude under illumination, effectively cancelling the strong adhesive state. Crucially, this process is fully reversible; upon removal of illumination, the material cools back into the nematic phase and recovers its high adhesion strength after equilibration ("Light off" regions in Fig. 1e). The switching adhesion therefore directly reflects the intrinsic phase dependence of LCE adhesion,^{15,16,26} but is here actuated rapidly and locally through photothermal control.

The efficiency of photothermal switching depends strongly on MXene loading concentration because the nanoplatelets directly control the photothermal conversion efficiency.²⁷⁻²⁹ In the absence of MXene, the LCE adhesive remains largely insensitive to illumination even after prolonged exposure (Fig. 3e). However, when the loading concentration of MXene nanoplatelets reaches 1.0 wt%, the average adhesion strength of LCE PSA before and after illumination was 4283 N/m and 118 N/m respectively, and the switching ratio reaches 97.2% (Fig. 3f).

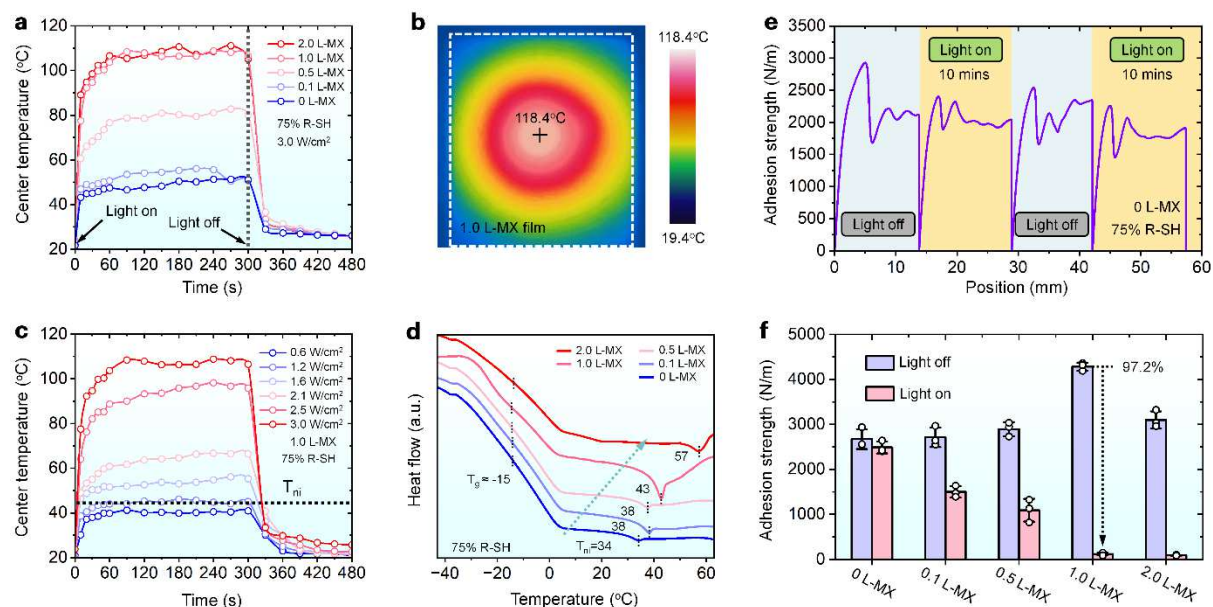


Figure 3 — Photothermal Switching

(a) The temperature increase in the sample on illumination for different MXene loading. The nematic–isotropic transition occurs near 60 °C; only minimal nanoplatelet concentrations required to achieve photothermal switching were therefore used.

(b) Illustration of the temperature gradient around the center of illuminated spot.

(c) The temperature increase in the 1%Mx sample at different light intensities.

- (d) DSC data on phase transitions in 75% R-SH (25%xl) samples for different MXene loading concentration, showing the small increase of the nematic-isotropic transition temperature.
- (e) The lack of thermal switching in the 0%Mx sample – compare with Figure 1(e) for the maximum range between 4300 N/m and 120 N/m in the isotropic phase.
- (f) Comparison of adhesion strength in the nematic and isotropic phase induced by illumination of 25%xl samples for different MXene loading.

The ability to trigger the isotropic transition locally through photothermal activation fundamentally changes the practical accessibility of LCE adhesion switching. In previous studies, switching required global thermal treatment of the sample, typically involving ovens or heated stages^{15,16,23,24,26,30}. By contrast, MXene-enabled photothermal control allows rapid and spatially selective release using light as a remote trigger, greatly expanding the potential utility of these materials in adaptive adhesion systems. This capability sharply contrasts with conventional PSA systems, where adhesion is fixed by formulation and cannot be dynamically switched without irreversible changes.

4. Dynamic covalent chemistry and network reprocessing

The topology-engineered LCE network remains dynamically adaptable through thermally activated siloxane exchange reactions within the crosslinked structure. Stress-relaxation experiments reveal a pronounced acceleration of relaxation dynamics with increasing temperature (Fig. 4a and Supplementary Fig. S12a), consistent with activated bond-exchange processes within the siloxane-containing network.³¹⁻³³ Importantly, incorporation of MXene nanoplatelets produces negligible changes in the relaxation kinetics, indicating that nanoplatelet addition does not significantly perturb the underlying dynamic exchange mechanism (Supplementary Fig. S12a).

The characteristic relaxation time $\tau(T)$, defined by the condition $\sigma/\sigma_0 = 1/e$, decreases systematically with temperature and follows classical Arrhenius behaviour: $\text{Log}[\tau] = \Delta E/k_B T + \text{const}$, where ΔE represents the activation energy associated with siloxane exchange.³² Arrhenius analysis yields activation energies of approximately 48 kJ/mol for both MXene-free and MXene-containing systems (Fig. 4b), confirming that the nanoplatelets do not measurably alter the exchange energetics. These activation energies are substantially lower than those reported for conventional siloxane vitrimer networks,³⁴⁻³⁶ indicating a comparatively facile dynamic exchange process within the present topological architecture.

The low activation barrier enables efficient hot-press reprocessing and self-healing of the adhesive films (Fig. 4c and Supplementary Fig. S12b). Cut films readily reform continuous structures after thermal pressing, while preserving the characteristic nematic elastomer behaviour and reversible adhesion switching. Unlike conventional permanently crosslinked elastomers, the present system therefore combines mechanical integrity with dynamic network adaptability and self-healing.

More importantly, dynamic covalent exchange appears to contribute directly to the anomalous adhesive behaviour of the nematic state. The adaptive exchange process provides additional relaxation pathways within the confined interfacial region, facilitating local rearrangement of dangling oligomer chains and distorted nematic domains during bonding. The adhesive interface is therefore not statically defined by network architecture alone, but evolves

continuously through the coupled effects of orientational order, chain mobility and dynamic bond exchange.

These observations further distinguish the present system from conventional PSAs, where adhesion is governed predominantly by dissipative processes during debonding. Here, dynamic covalent adaptability participates in the gradual construction of a metastable bonded state prior to detachment, coupling nonequilibrium nematic relaxation with adaptive network topology within a single adhesive material.

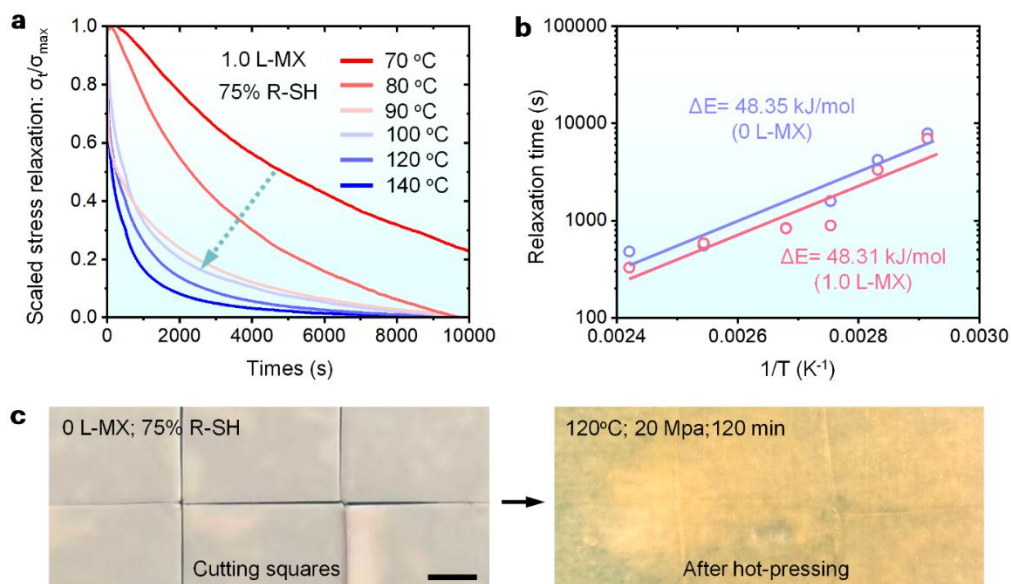


Figure 4 — Dynamic network adaptability and reprocessability

- (a) Temperature-dependent stress relaxation of the topological LCE network.
- (b) Arrhenius analysis of relaxation time for MXene-free and MXene-containing systems, showing nearly identical activation energies.
- (c) Hot-press reprocessing and self-healing of cut adhesive films. The scale bar is 3 mm.

5. Macroscopic manifestations of switchable ultrastrong adhesion

To illustrate the macroscopic consequences of switchable ultrastrong adhesion, we performed a series of representative demonstrations using the optimized MXene-containing LCE PSA platform (Fig. 5). The material firmly captures and transports a 25 mm steel sphere (64 g), followed by rapid remote release under illumination (Fig. 5a and Supplementary Video 4). Similar behaviour is observed for smaller objects and different loading conditions (Supplementary Fig. S13, S14 and Supplementary Videos 5 and 6).

More strikingly, a small LCE adhesive strip (5 mm × 10 mm) is capable of repeatedly transporting sequential 500 g loads while retaining reversible photothermal debonding behaviour (Fig. 5b and Supplementary Video 7). The same material, used as a double-sided adhesive interface, supports loads up to 1.77 kg while still undergoing rapid and clean release upon illumination (Fig. 5c and Supplementary Video 8). This macroscopic switching behaviour is accompanied by collapse of lap shear strength from 0.5 MPa to 0.03 MPa after photothermal activation, a reduction of over 94% (Fig. 5d). More impressively, a narrow LCE PSA film (25 mm) can stably lift up a 10 kg weight and quickly debond as needed under illumination condition (Fig. 5e and Supplementary Video 9).

Localized optical activation further enables spatially selective debonding. During 90° peel experiments, only illuminated regions of the adhesive interface detach, while adjacent non-illuminated regions remain strongly bonded (Supplementary Fig. S15, S16 and Supplementary Video 10). The ability to combine ultrastrong adhesion with rapid, localized and reversible release highlights the fundamentally distinct nature of adhesion in these topologically engineered nematic elastomers.

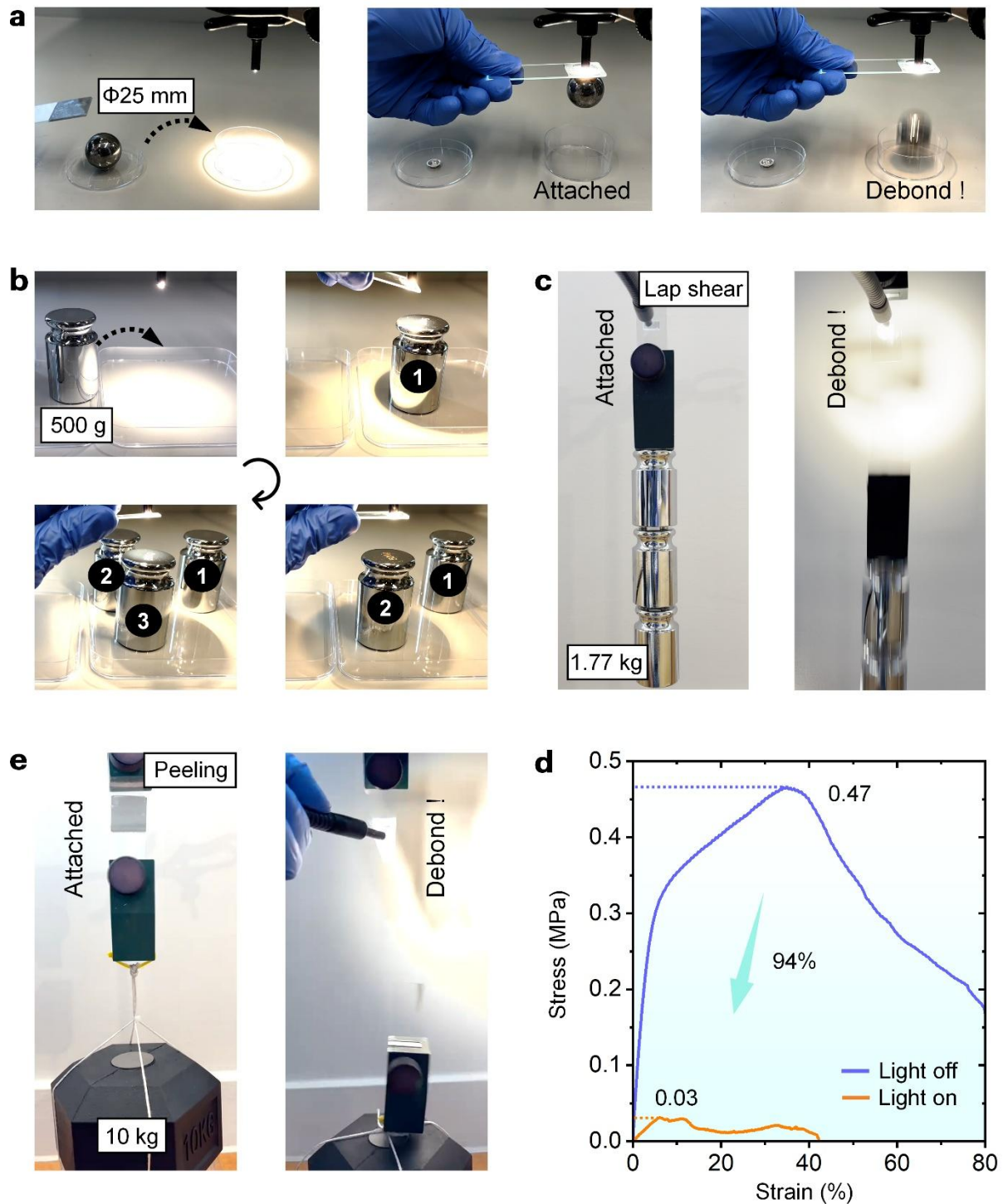


Figure 5 —Macroscopic manifestations of switchable ultrastrong adhesion

- (a) Remote capture, transport and photothermal release of a 25 mm steel sphere (64 g).
- (b) Reversible transport of sequential 500 g loads using a 5 × 10 mm LCE adhesive strip.

- (c) Double-sided LCE adhesive supporting a 1.77 kg load together with photothermally induced collapse of lap shear strength.
 - (d) Lap shear strength of the LCE film before and after illumination was 0.47 MPa and 0.03 MPa (decreased by 94%) respectively.
 - (e) LCE PSA with a width of 25 mm holding a 10 kg weight in the 180° peel geometry, followed by with the illumination induced debonding.
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Conclusions and Outlook

Conventional PSAs achieve strong bonding through viscoelastic dissipation within lightly crosslinked polymer networks, often enhanced by mobile tackifier components that promote interfacial wetting and fibrillation during peeling.^{5-9,37,38} In the present system, these dissipative processes are coupled to the slow cooperative relaxation of a confined nematic network, producing a deeply metastable bonded state whose adhesive strength substantially exceeds conventional PSA benchmarks.

We have shown that network topology engineering combined with nanoplatelet incorporation transforms nematic liquid crystal elastomers into a new class of ultrastrong and reversibly switchable PSAs. By introducing dangling oligomer chains into a percolated nematic network, adhesion strength substantially exceeding those of best commercial adhesive tapes is achieved while preserving reversible phase-dependent switching. MXene nanoplatelets further enhance adhesion and simultaneously provide a photothermal route for rapid, localized and reversible debonding through the nematic–isotropic transition.

The resulting adhesive behaviour differs fundamentally from conventional PSAs. In classical systems, adhesion is governed primarily by viscoelastic dissipation during debonding. The present materials evolve toward a deeply metastable bonded state through the slow relaxation of orientational order under confinement. The adhesive interface is therefore not static, but dynamically constructed through cooperative interactions between mobile chains, nanoplatelet fillers and the surrounding nematic network. This coupling of interfacial adaptation, bulk reorganization and phase transition introduces a new framework for adhesive design.

More broadly, our results suggest that liquid crystalline soft solids may provide a general route toward adaptive adhesive systems in which bonding strength, reversibility and activation pathway can all be independently controlled. The ability to combine ultrastrong adhesion with remote, localized release may be particularly attractive for robotic manipulation, reconfigurable assembly, transfer printing and biomedical fixation technologies, where conventional adhesives remain limited by the inability to reconcile strong attachment with facile detachment.

Future developments may extend this approach toward electrically, magnetically or chemically activated systems, or exploit anisotropic nanoparticle assemblies and aligned nematic textures to introduce directional or programmable adhesion. More generally, these findings highlight the broader potential of coupling network topology with nonequilibrium orientational dynamics in soft matter systems, opening opportunities for responsive interfaces whose mechanical behaviour is governed not only by composition, but by the evolving internal state of the material itself.

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Methods

Materials

The liquid crystal (LC) monomer 1,4-bis-[4-(6-acryloyloxyhexyloxy)benzoyloxy]-2-methylbenzene (RM82) was purchased from Chemfish Tokyo Co., Ltd. Chain extender, 2,2'-(ethylenedioxy)-diethanethiol (EDDT), and siloxane crosslinking agent, 2,4,6,8-tetramethyl-2,4,6,8-tetravinyl cyclotetrasiloxane (TMTVCTS) were purchased from Sigma-Aldrich. Catalyst Dipropylamine (DPA) was purchased from Sigma-Aldrich. The photoinitiator, Irgacure I-651, and photothermal conversion agent, Titanium carbide MXene (Ti_3C_2), and solvent toluene were obtained from Sigma-Aldrich. All chemicals were used as received.

Preparation of LCE

We first prepared the LC oligomers by the thiol-acrylate Michael addition reaction between RM82 monomer and EDDT extender. And the Michael addition was catalyzed via DPA. The mixture of RM82, EDDT and TMTVCTS was dissolved in toluene at 80°C. The molar ratio of RM82

to EDDT in the mixture was 1:1.1, corresponding to a thiol-terminated oligomer of chain length approximately 10 mesogenic units. The molar ratio of RM82 to TMTVCTS were 1:0.05 (0% R-SH), 1:0.0375 (25% R-SH), 1:0.025 (50% R-SH), and 1:0.0125 (75% R-SH) respectively. Subsequently, I-651 (2 wt%) and MXene nanoplates (0.1 wt%, 0.5 wt%, 1.0 wt%, and 2.0 wt%) were added to the mixture. Next, 1 wt% of the DPA catalyst was dropped into the mixture. The calculation basis for all the above-mentioned mass concentrations was the mass of RM82 monomer. Then, immediately perform a vigorous vortex mixing (Hauschild SpeedMixer) of the mixture in a dark environment for 60 minutes to fully oligomerized via Michael addition reaction. After vigorous rotation, the oligomers were coated onto the surface of the silicone paper or polyethylene terephthalate (PET) backing. Finally, the thiol-terminated oligomer was photopolymerized with TMTVCTS via 365 nm UV light for 20 min at 50°C. After the polymerization is completed, the samples were transferred to an oven of 80°C for 24h to remove the solvents.

180-Degrees Peel Test

The peel test followed the standard procedure outlined in ASTM D3330. We coated LCE PSA on the standard 25 mm-wide PET backing surface and evenly adhered it to the upper part of the clean glass surface. The free end of the PET film and the lower half of the glass were respectively clamped by vertically arranged fixtures (Tinius-Olsen ST1 tensiometer). The tape was pulled up with a rate of 100 mm/min, and the adhesion force was also recorded simultaneously. Ultimately, the standard peel test adhesion strength (N/m) could be obtained via dividing the adhesion force by the width of the PET backing (25 mm). Each group of samples was tested three times in duplicate.

Lap Shear Test

The standard lap shear test was conducted in accordance with the guidelines of ASTM D3163. A double-sided LCE PSA film with dimensions of 25 mm (L) X 10 mm (W) X 0.3 mm (T) was adhered to the overlapping area of two clean glass plates. The free ends of the two glass plates were respectively fixed on two clamps arranged vertically on the tensile machine (Tinius-Olsen ST1 tensiometer). The lifting speed of the upper clamp was set to a constant 1.25 mm/min. Each group of samples was tested three times in duplicate.

Probe Tack Test

A steel spherical probe with a diameter of 10 mm was mounted on the upper clamp of vertical dynamometer frame (Tinius-Olsen ST1 tensiometer). The LCE PSA film was adhered to the rigid surface of the heating platform, and the temperature of the LCE PSA film can be controlled by adjusting the temperature of the heating platform. The surface of the steel spherical probe was thoroughly cleaned with alcohol before each experiment. The steel spherical probe slowly descended and pressed into the surface of the LCE film until it reached the preload of -0.5 N. After that, the predetermined dwell time (300 seconds) was allowed, after which the probe was pulled up at a fixed rate of 10 mm/min. The adhesion force is defined as the peak value at which the LCE film detaches from the probe. Each group of samples was tested three times in duplicate.

Mechanical Analysis

Stress-strain curves of LCE PSA were measured on the Tinius-Olsen ST1 tensiometer using a rectangular film sample with dimensions of 50 mm (L) X 10 mm (W) X 0.4 mm (T) at ambient

temperature conditions. The rate of uniaxial tensile test is 5 mm/min. The uniaxial tensile test of each formulation of LCE film were conducted three times.

Dynamic Mechanical Analysis (DMA)

Temperature ramp tests were conducted using a TA Instruments DMA850 in tension film mode with LCE film dimensions of approximately 15 mm (L) X 5 mm (W) X 0.4 mm (T). The oscillatory tension frequency of the temperature ramp was 1 Hz, and the strain amplitude was 0.02%. The storage modulus (E'), loss modulus (E''), and loss factor ($\tan \delta = E''/E'$) were recorded as function of temperature from -80°C to 150°C at the rate of $2^{\circ}\text{C}/\text{min}$.

The same TA Instruments DMA850 was applied to measure the stress relaxation behavior of the LCE films with same geometric dimensions. After applying a constant 3% uniaxial tensile strain to each sample, it was allowed to relax at the specified temperature (70°C , 80°C , 90°C , 100°C , 120°C , 140°C) until equilibrium was reached.

Iso-stress experiments of LCE films were also conducted using a TA Instruments DMA850 in tensile mode. The sample dimensions were approximately 15 mm (L) X 5 mm (W) X 0.4 mm (T). A constant stress (5 kPa, 10 kPa, 15 kPa, 20 kPa, 25 kPa, 30 kPa) was applied to the sample, and the strain data of the sample from 20°C to 150°C with a rate of $2^{\circ}\text{C}/\text{min}$ were recorded.

Dynamic Scanning Calorimetry (DSC)

We employed DSC4000 PerkinElmer to obtained the transition temperatures of LCE. Approximately 15 mg of the samples was loaded into a standard aluminum DSC pans. The samples were heated from 30°C to 150°C with rate of $10^{\circ}\text{C}/\text{min}$, and held isothermally for 5 minutes. Subsequently, the samples were cooled at a rate of $10^{\circ}\text{C}/\text{min}$ to -50°C and then held isothermally for another 5 minutes. Finally, the samples were heated again to 150°C at a rate of $10^{\circ}\text{C}/\text{min}$.

Funding Statements

This work was supported by the ERC Synergy grant 'ALCEMIST' (101167171), the Royal Society through University Research Fellowships (G113670), the National Natural Science Foundation of China (52505198), the China Postdoctoral Science Foundation (BX20250429 and 2025M781310), and the Open Research Fund of the Beijing National Laboratory for Condensed Matter Physics (2024BNLCMPKF021).

Data Availability Statement

All experimental data obtained in this work is available on request from the corresponding author.

Supplementary Files

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- [SupportingInformation.pdf](#)