

CO₂-mediated conversion of physical pressure into chemical pressure to realize lattice strain

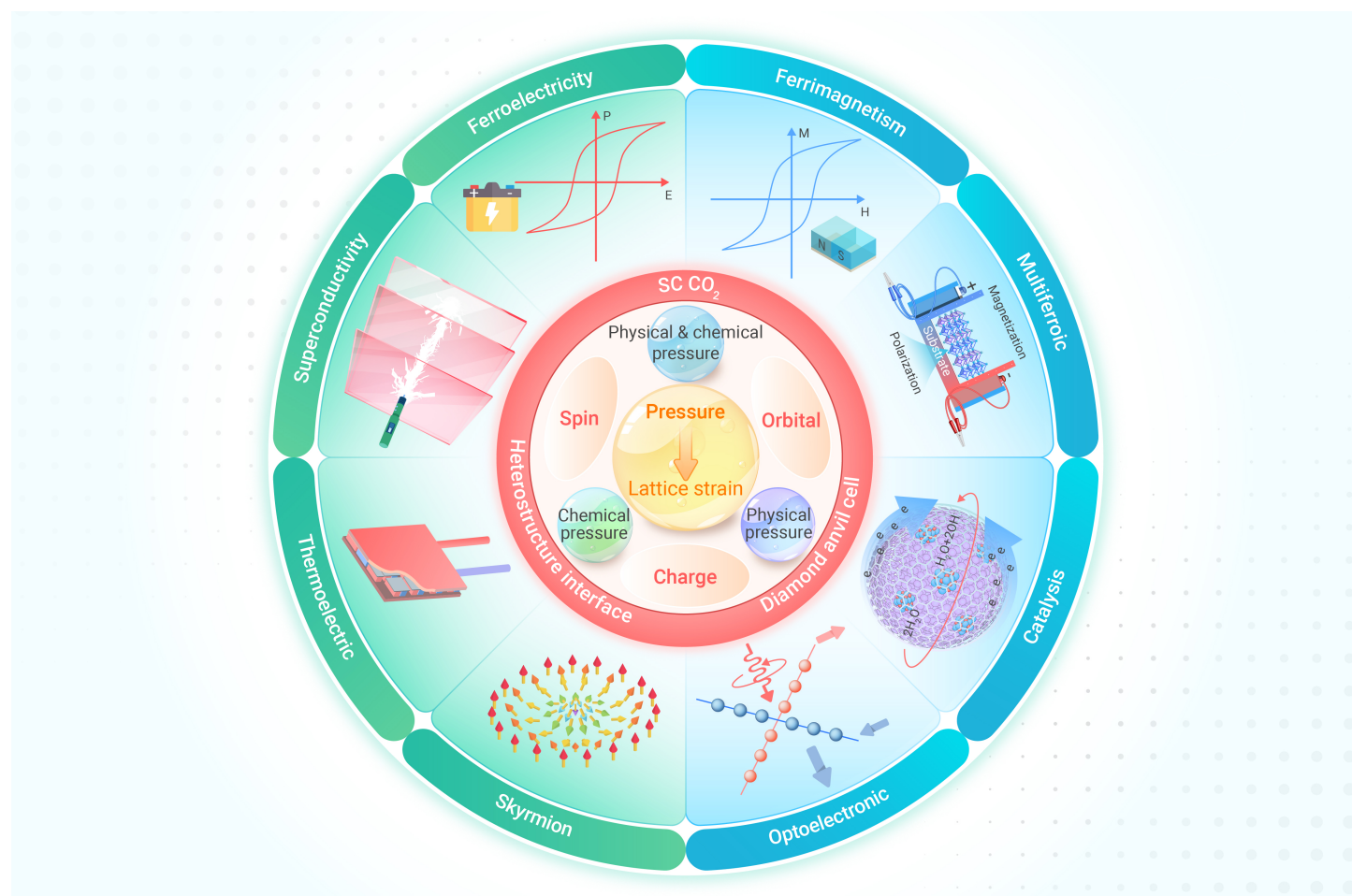
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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- Supercritical CO₂ mediates the conversion of physical pressure into chemical pressure induced lattice strain.
- Lattice strain effectively modulates the coupling between spins, charges and orbitals in materials.
- Lattice strain brings some novel physical phenomena.

CO₂-mediated conversion of physical pressure into chemical pressure to realize lattice strain

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Lattice strains often lead to novel phenomena, and they are used in a variety of applications, such as thermoelectricity, ferroelectricity, ferromagnetic, skyrmion and superconductivity. In this paper, we briefly summarize the novel phenomena brought about by lattice strain. An efficient method to realize lattice strain using supercritical CO₂ (SC CO₂) is proposed. When CO₂ molecules shuttle through the confined space of the crystal structure, it can generate huge energy subsequently. Specifically, CO₂ carries MPa-level external physical pressure inside the crystal structure, forming localized defects or phase transitions. This alters the original chemical coordination environment and generates internal chemical pressures up to the GPa level. The crystal is stripped from three-dimensional to two-dimensional under high pressure, and lattice strain is realized at the same time. This work will provide new insights into the realization of lattice strain and give valuable guidance to broaden its application.

INTRODUCTION

Throughout history, human civilization has been greatly influenced by the discovery and development of new materials. Historical epochs have been named after the most significant materials that were developed and widely used, such as the Bronze Age and Iron Age.^{1,2} However, the most essential effect on the material is the optimization of the microstructure and bonding states, in other words, the modulation of the lattice structure.³⁻⁵ The regulation of lattice structure is mainly manifested in the distortion and deformation, which often brings about changes in spin, charge, orbitals and lattice degrees of freedom.⁶⁻⁸ Their coupling opens the door to new materials and brings colorful new functions. For example, lattice strains can inhibit heat transport, which significantly improves the thermoelectric conversion efficiency.⁹ It can also improve ferroelectricity by enhancing polarization strength.^{10,11} In addition, lattice strains are often used in fields such as ferri-

magnetism,¹² optoelectronic,¹³ skyrmion,¹⁴ and superconductivity.¹⁵⁻¹⁷ The realization of lattice strain requires the action of pressure, which in the category of origins can be divided into internal chemical pressure and external physical pressure.

Physical pressure is generally achieved by the action of external mechanical pressure (e.g., diamond anvil cell) to achieve compression per unit cell size. Chemical pressures here are identified using chemical methods to generate power and effect, which can produce interfacial epitaxial strain, ion substitution and atomic vacancy, etc. For example, epitaxial strain has been successfully applied to stabilize the metastable phases in the form of hetero-junction films under ambient pressure, where the stress established by lattice mismatch between substrate and epitaxial heterojunction has been estimated to reach several GPa. Internal chemical pressures in a given accurate compound can be generated by chemical substitution of smaller/larger ions to induce cell contraction/expansion that are equivalent to positive/negative chemical pressure. It is a big challenge but of great value to pursue a path to achieve lattice strain by a combination of external physical pressure and internal chemical pressure. Accurately, by applying external physical pressure to the inside of the lattice structure, the physical pressure perturbation causes the atoms to move and as well as change the original chemical coordination environment to form local defects or phase transitions, thus realizing the transformation from external physical pressure to internal chemical pressure. Based on this idea, we propose a method to realize large-area fully homogeneous lattice strain using supercritical CO₂ (SC CO₂) (Figure 1B).

Supercritical fluids are substances at conditions above their critical temperature and pressure, which can display vapor- and liquid-like behaviors including near-zero surface tension, low viscosity, high diffusion coefficients, excellent surface wettability, and strong solvency¹⁸ (Figure 1A). Among them, SC CO₂, as a green and environmentally friendly solvent, has great application prospects in materials processing. Under the external physical pressure

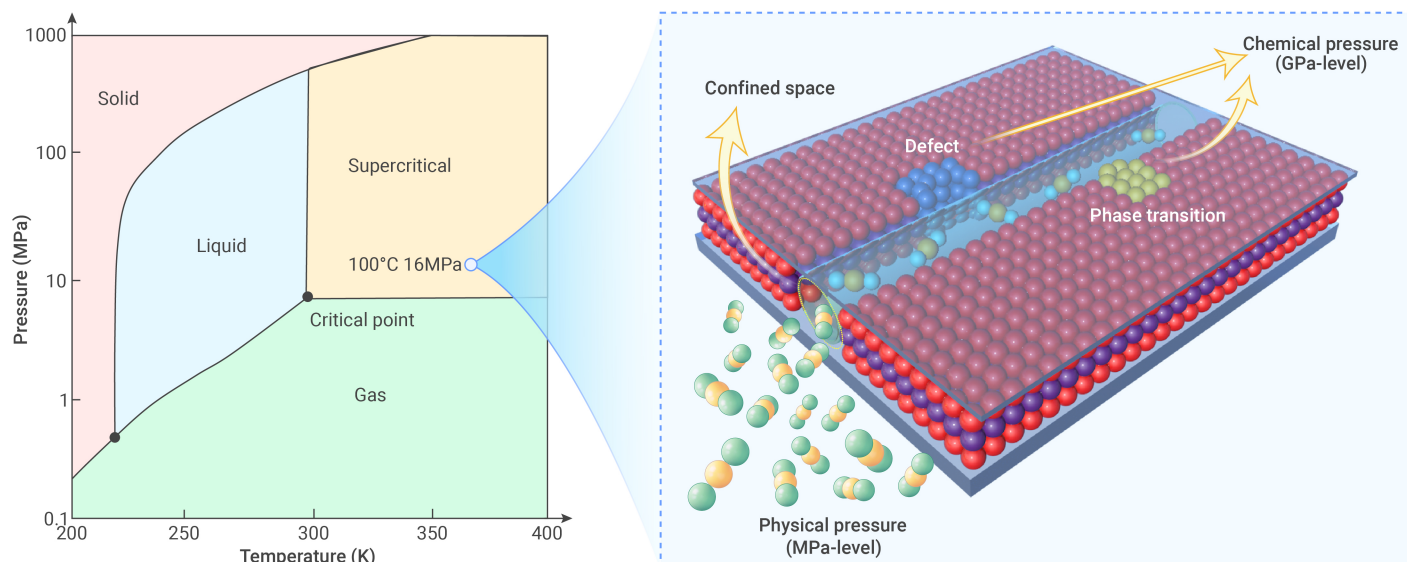


Figure 1. SCCO₂ action on materials Pressure-temperature phase diagram of CO₂ fluid (Left). Mechanisms for CO₂-mediated conversion of physical pressure to chemical pressure (Right).

at MPa level, the CO₂ molecules shuttling in the lattice selectively, break the covalent bonds in the lattice to change the internal coordination environment of the crystals, which in turn generates the internal chemical pressure at GPa level to realize the lattice strain. This CO₂-mediated conversion of external physical pressure into internal chemical pressure to realize lattice strain is expected to gain wider application.

APPLICATIONS OF LATTICE STRAIN

Thermoelectric technology

Thermoelectric technology can utilize waste heat to generate electricity, opening up a new way to improve energy utilization. The configurational entropy of a material can be manipulated by introducing different atomic species to modulate the phase composition and provide greater scope for property optimization. Jiang et al.⁹ investigated the formation of n-type PbSe-based high-entropy materials through entropy-driven structural stabilization. The highly disordered lattice in this high-entropy system causes anomalous shear strains and provides strong phonon scattering for lower lattice thermal conductivities (Figure 2A). The distorted lattice is able to suppress the heat transport while maintaining the electrical properties, providing a significant improvement in the thermal conversion efficiency of the material. The thermoelectric figure of merit (zT) at 900 K was enhanced to 1.8 in this material (Figure 2B). Optimized thermoelectric properties enable the manufacture of segmented thermoelectric modules with conversion efficiencies up to 12.3% at 507 K. (Figure 2C). This work provides a completely new paradigm for optimizing thermoelectric properties in high-entropy thermoelectric materials. Effective enhancement of thermoelectric properties using lattice strain has also been achieved in other similar materials such as (Cu/Ag)₈Ge(Se/Te)₆, (Cu/Ag)(In/Ga)Te₂ and (Sn/Ge/Pb/Mn)Te.^{19–22}

Ferroelectricity

Enormous strains can exist in thin films when one material is deposited on another,²³ resulting from differences in crystal lattice parameters and thermal expansion behavior between the film and the underlying substrate or arising from defects formed during film deposition.^{24,25} Biaxial compressive strain was used to significantly enhance the ferroelectric properties of BaTiO₃ thin films as early as 2004 by K. J. Choi et al.¹⁰ This strain, imposed by coherent epitaxy, can result in a ferroelectric transition temperature nearly 500°C higher and a remanent polarization at least 250% higher than bulk BaTiO₃ single crystals. The strong coupling of the lattice, spin, and orbit under pressure brings diverse properties into solid-state materials, such as superconductivity, multiferroics, ferromagnetism, etc.^{26–28} BaTiO₃:BaO composite films (c-BTO) and single-phase BaTiO₃ films (s-BTO) were grown by magnetron sputtering.²⁹ The large lattice of cubic BaO exerted considerable negative chemical pressure on the tetragonal BaTiO₃, which significantly enhanced the ferroelectric properties of BaTiO₃. The tetragonal BaTiO₃ (Figure 3A) (110) facets and the "tetragonal-like" BaO (001) facets (Figure 3C) are well-matched, and along these specific orientations, a coherent interface is formed between the BaTiO₃ and the "tetragonal-like" BaO (Figure 3B). The lattice constants obtained by synchrotron X-ray diffraction and reciprocal space mapping measurements confirm that c-BTO obtains a chemically negative pressure of 5.7 GPa. In addition, the atomic level of c-BTO was investigated by cross-sectional HAADF-STEM and found that the displacement of Ti atoms in the cells close to the interface was significantly increased compared to the cells far from the interface (Figure 3D). The ferroelectric polarization versus electric field hysteresis loop (P-E loop) confirms that c-BTO has the highest remanent polarization Pr of up to 100 μC/cm² (Figure 3E), while s-BTO and BaTiO₃ ceramics have Pr values of 30 μC/cm² and 15 μC/cm², respectively (Figure 3E).

Inner coherent interface, through size effect or hetero structural inter phase strain imposed by PbO, is introduced to the matrix to achieve remarkably strengthened negative chemical pressure compared to the mild chemical pressure in heterogeneously doped lattice, and negative pressure was demonstrated to be vital for the enhancement of ferroelectricity in PbTiO₃.^{30,31} However, through delicate manipulation, PbTiO₃ nanoparticles with sizes down to 12 nm could possess enhanced ferroelectricity due to the negative pressure introduced by the PbO-terminated surface.³¹ The interphase strain applied through the coherence of PbO and PbTiO₃ yields a giant out-of-plane

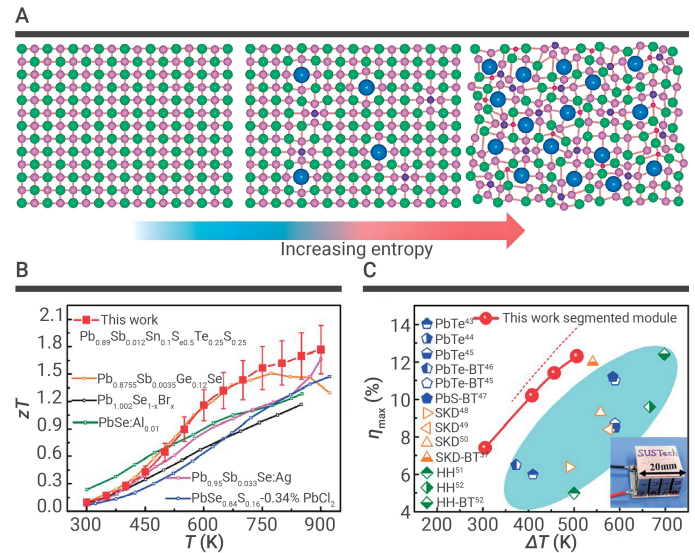


Figure 2. Improving the performance of thermoelectric materials and modules through entropy engineering (A) Diagram of lattice strain with increasing entropy. (B) zT values as a function of temperature for the high-entropy n-type PbSe-based materials in this work. (C) Maximum conversion efficiencies (η_{max}) as a function of temperature difference for the high-entropy segmented thermoelectric module in this work and some reported results from the literature. Reprinted with permission.⁹ Copyright 2021, American Association for the Advancement of Science.

negative pressure, leading to tremendous remanent polarization and high Curie temperature.³⁰ PbTiO₃ has a tetragonal chalcogenide structure with lattice parameters $a = 3.899$, $c = 4.154$, and $c/a = 1.065$ (Figure 3F).³² The precursor PbO used for the preparation of PbTiO₃ has a similar tetragonal structure, albeit derived from a fluorite structure, with a large c/a ratio ($a = 3.9729$, $c = 5.0217$, $c/a = 1.264$) (Figure 3G).³³ Figures 3I–J show that the substable PbO is randomly distributed in PbTiO₃. The HAADF-STEM image (Figure 3K) provides direct evidence of the heterogeneous epitaxial structure between PbTiO₃ and PbO. The intensity distribution along the red dashed line in Figure 3L reveals a transition from Ti in PbTiO₃ to Pb in PbO, suggesting a good lattice match at the PbTiO₃/PbO interface (Figure 3M). Similarly, the c -axis lattice constant, tetragonal, was greatly increased by tuning the distribution of O in BaTiO₃ thin films. The realization of polarization-enhanced interface improves the ferroelectricity of BaTiO₃.³⁴ Ti/O-deficient chalcogenide nanoregions exert large out-of-plane tensile strains and electron doping effects on the SrTiO₃ substrate to form a tetragonal structure that drives the eccentric motions of Ti and Sr atoms. Strong ferroelectricity was also obtained for SrTiO₃ by defect-induced lattice strain.³⁵

Ferrimagnetism

Currently, a range of Two-dimensional (2D) ferromagnets—including Cr₂Ge₂Te₆,³⁶ CrI₃,^{37,38} and Fe₃GeTe₂,^{39,40} have been discovered,⁴¹ which exhibit a range of interesting behaviors such as layer-dependent magnetism in CrI₃,³⁷ and room-temperature ferromagnetism with ionic gating in Fe₃GeTe₂.⁴⁰ Unlike most 2D ferromagnetic materials with only one phase, it is predicted that Cr₅Te₈ exists in a two-phase structure, where different lattice strains not only bring about different magnetic responses, but also structural strains are more likely to make it exhibit extraordinary transport properties, such as the topological Hall effect and the large anomalous Hall effect.⁴² The large anomalous Hall effect, which is generated by the magnetic ordering and spin-orbit coupling⁴³ are beneficial for building next-generation dissipation-free electronic and spintronic devices because the Joule heat associated with the longitudinal current can be effectively suppressed.

Materials with magnetic ordering are tightly coupled to crystal structures and lattice strains and usually have complex free energy profiles.^{37,40,44} For example, epitaxial strains are applied to ferromagnetic thin films through a predetermined substrate to change the Curie temperature,^{45–47} magnetization,^{48,49} and magnetic anisotropy.⁵⁰ To functionalize the materials, the strain can be electrically driven using a piezoelectric substrate, enabling

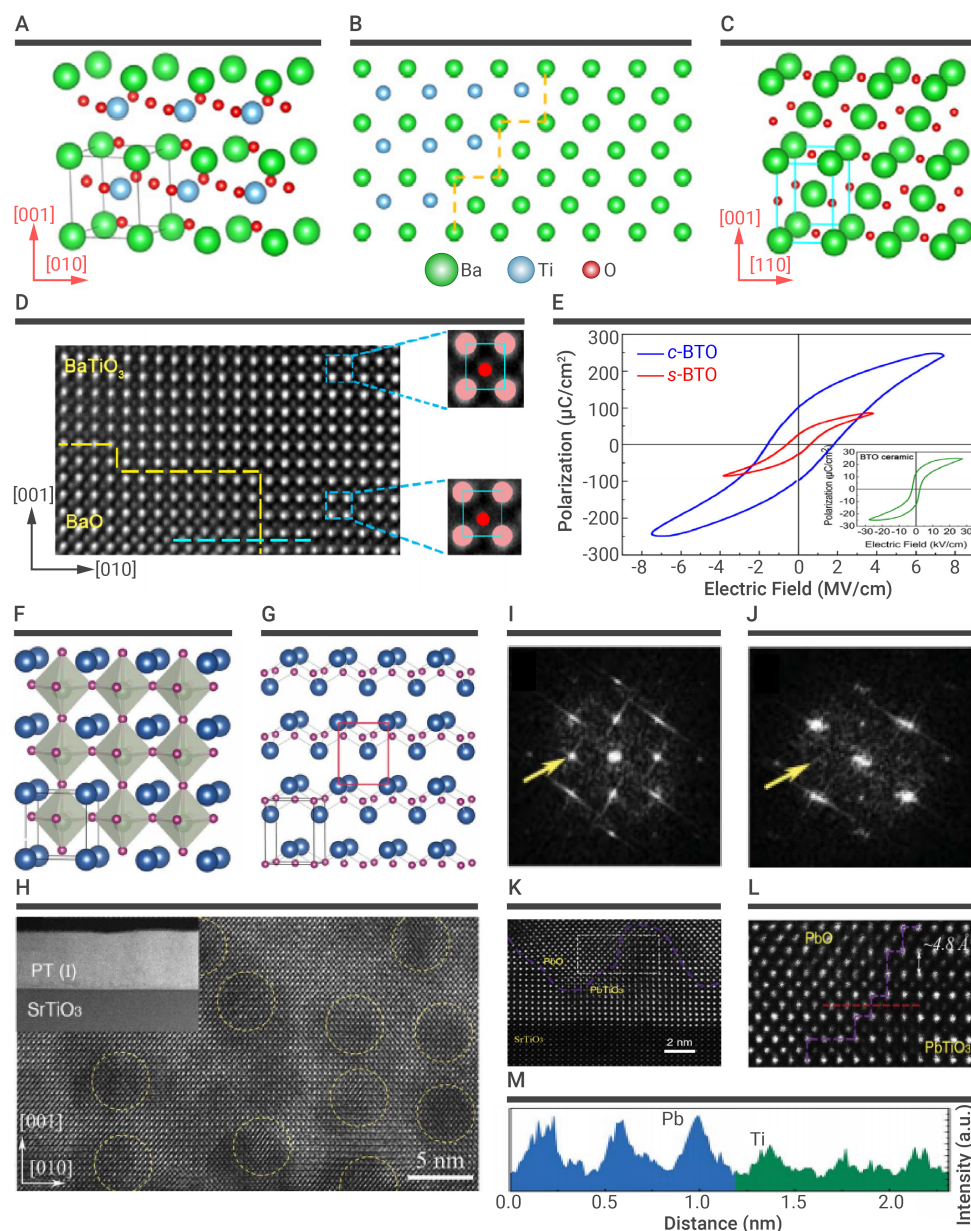


Figure 3. Strain-induced ferroelectricity in BaTiO₃ and PbTiO₃. Lattice structures of BaTiO₃ (A) and BaO (C), respectively. (B) Interface schematic. (C) Cross-sectional STEM image of c-BTO, with two BaTiO₃ unit cells magnified in right panels. (D) Ferroelectric polarization versus electric field hysteresis loops of c-BTO and s-BTO, with BaTiO₃ ceramics inserted. (E) Ferroelectric polarization versus electric field hysteresis loops. Reprinted with permission.²⁹ Copyright 2021, American Chemical Society. (F) Crystalline structure of PbTiO₃. (G) Crystalline structure of PbO. (H) High-resolution transmission electron microscopy image along the (a) axis of the PT (I) thin films. (I) The FFT pattern taken from the regions surrounding the yellow dashed circles of (H). (J) The FFT pattern taken from a typical example of the regions marked by yellow dashed circles of (H). (K) HAADF-STEM image of the heteroepitaxial interface between PbTiO₃ and PbO. (L) Enlarged view of the region enclosed by the white rectangle in (K). (M) Intensity profile along the red dashed line in (K). Reprinted with permission.³⁰ Copyright 2021, American Association for the Advancement of Science.

thin LaCoO₃ layers, surprisingly large magnetic moments (0.5 μ B/Co) (Figure 4E) and Curie temperatures (75 K) are observed (Figure 4F). These results demonstrate a strategy to create ultrathin ferromagnetic perovskite by utilizing atomic heterogeneous interface engineering, confinement-driven structural transitions, and spin-lattice coupling in strongly correlated materials. Using a similar approach, Ye et al. demonstrated that lattice strain favors the stabilization of the ferromagnetic state of LaCoO₃.⁵⁹ Li et al.⁶⁰ also found that the strong modification of Co 3d-O 2p hybridization associated with the increase of Co-O-Co bond angles and Co-O bond lengths weakened the crystal field splitting, promoted the ordered high spin state of Co ions, and induced the emergence of ferromagnetic insulating state.

METHOD OR STRATEGY FOR LATTICE STRAIN

Atomic hybridization (High entropy alloys)

Yeh et al.^{61,62} proposed four "core effects" of high-entropy alloys (HEAs), i.e., the high-

entropy effect, the slow diffusion effect, the lattice strain effect, and the chemical "cocktail" effect, which have been widely cited in the literature to rationalize the excellent properties of HEAs. Among these "core effects", lattice strain is considered to be the fundamental factor leading to the impressive properties of HEAs.⁶³⁻⁶⁵ Lattice strains generate highly fluctuating elastic stress/strain fields, which are crucial for dislocation behavior.⁶⁶ In addition, lattice strains help to stabilize thermodynamic nonequilibrium states, thereby lowering the energy barrier of the active species.^{67,68} As a result, HEAs with lattices strain exhibit more impressive multifunctional properties such as ultraelastic,⁶⁹ excellent strength and plasticity,^{9,70,71} high thermoelectric properties, unique irradiation resistance^{72,73} and promising catalytic efficiency^{74,75} compared to conventional alloys.

As shown in Figures 5A-B, the atomic size and elastic modulus are different between solute and solvent atoms, and the binding of solute atoms to the metal leads to local lattice strain. From a thermodynamic point of view, the local lattice strain is caused by the minimization of the Gibbs free energy of the alloy.⁷⁶ As shown in Figure 5C, atoms relocate away from the ideal lattice position, resulting in severe lattice strains and a highly fluctuating elastic stress field. Fundamentally, lattice strain is characterized by fluctuations or changes in lattice strain, which is also associated with changes in elastic energy^{77,78} and mismatch volume.⁷⁹⁻⁸¹ In order to quantify the atomic lattice strain, the deformed samples were analyzed by geometric phase analysis

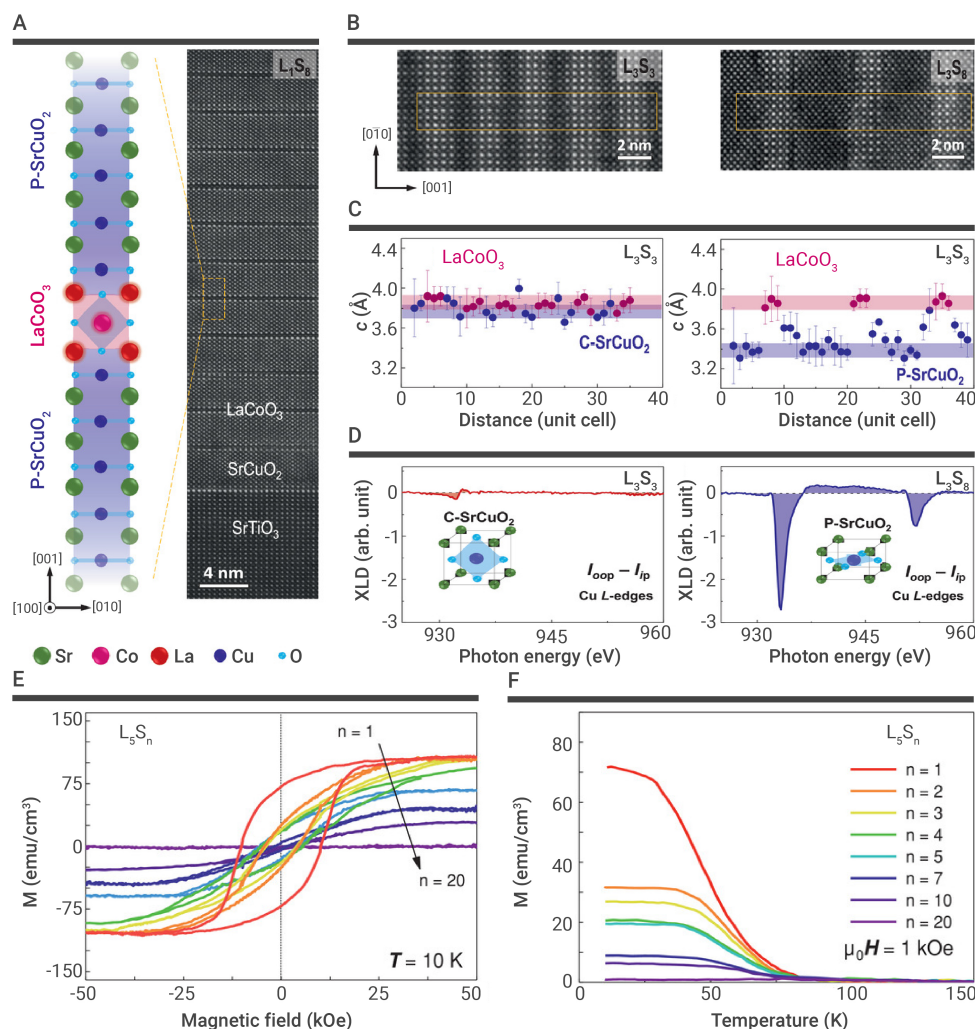


Figure 4. Structural and electronic state characterizations of the (L_mSn) superlattices (A) Schematic (left) and HAADF-STEM image (right) of a L₁S₈ superlattice. (B) HAADF-STEM images of L₃S₃ and L₃S₈ superlattices, respectively. (C) Atomic distances between the A-site cations along the [001] direction (D) X-ray linear dichroism (XLD) at the Cu L-edges for the L₃S₃ and L₃S₈ superlattices, respectively. (E) (F) M-H loops and M-T curves of L_mSn superlattice series for 1 ≤ *n* ≤ 20, respectively. M-H loops were recorded at 10 K. Reprinted with permission.⁵⁸ Copyright 2021, Wiley-VCH.

(Figure 5I) is a self-assembled nanocomposite in which one phase forms a 1D rod in a matrix of another phase (1-3 composite¹⁰⁹), both of which are epitaxially oriented with respect to the substrate. For this approach, the epitaxy occurs not only in the in-plane direction (on the substrate) but also in the out-of-plane direction between the nanocomposite components.

Diamond anvil cell

External physical pressure as a fundamental thermodynamic variable is one of the largest ranges of thermodynamic variables,¹¹⁰ which has been widely used not only to characterize the extreme conditions of matter, but also experimentally controlled to induce several important effects on interatomic interactions.¹¹¹ Diamond anvil cells as a common external physical pressure can likewise profoundly change the state of matter. It can alter chemical, structural, mechanical, electronic, magnetic, and phonon properties. This drives materials across the boundaries between soft and superhard, brittle and ductile, insulating and metallic, ferromagnetic and superconducting, amorphous and crystalline, ionic and covalent, and

vigorously reactive and inert. Through the interplay of atomic (molecular) and electronic interactions, pressure is a fertile dimension for new chemistry and new compounds. Many new types of bonds and material structures emerge under high pressure, and our traditional descriptions of the behavior of atoms and electrons no longer apply under pressure. The periodic table of elements under high pressure is very different from that under ambient pressure. New chemical "rules" emerge under extreme conditions.¹¹²⁻¹¹⁴ High pressures increase internal electron interactions, and atoms may become irregularly shaped and non-spherical, leading to very complex crystal structures and very large cells.¹¹⁵ Striking structural complexity has been found in many structures, including incommensurate structures that are widespread at high pressures.^{116,117} The valence of Eu atoms in EuO was measured experimentally at 90 GPa.¹¹⁸ The Eu atoms show a mixed valence consisting of discrete Eu²⁺ and Eu³⁺, with the average valence increasing with increasing pressure. Unexpectedly, at higher pressures, the measurements all clearly show a regressive shift of Eu to a lower valence state (close to Eu²⁺) with the NaCl → CsCl transition.

Heterogeneous epitaxy

Another common method of preparing strained lattices is heterogeneous epitaxy. Epitaxial growth provides the means to manipulate new states of matter that arise as a result of interactions between spins, charges, orbitals, and lattice degrees of freedom. Vacuum deposition methods incorporating epitaxial growth can be used to self-assemble materials with atomic layer precision and controllable properties (Figures 5G-I). Synthesis methods include strain engineering^{10,83,85-89} (Figure 5G), interface engineering^{84,90-97} (Figure 5H), epitaxial nanocomposites^{84,98-101} (Figure 5I). Strain engineering (Figure 5G) uses substrates with lattice constants different from those of the films grown on them. Thus, a uniform biaxial strain is applied to the film. Within the limits of strain and film thickness, it is made to produce strain relaxation defects¹⁰² (e.g., dislocations, cracks, or point defects¹⁰³⁻¹⁰⁵). While strain engineering has been used to improve the properties of materials in semiconductor devices,^{106,107} strain engineering also can perturb the energy from a substable polycrystalline (or hidden ground state) to a ground state. The resulting phase may have properties that are not present in the bulk form of the material. This may release hidden ground states^{85,87,89,108} with unrivaled properties. Interface engineering (Figure 5H) is the ability to create interfaces with atomic level precision. This capability is a prerequisite for validating the principles of electronic device formation. Reactive evaporation,⁹⁰ sputtering,⁹⁷ pulsed laser deposition^{92,94} and molecular-beam epitaxy^{91,93,95,96} demonstrate examples of precision interface engineering. The epitaxial nanocomposite

We summarize a brief evaluation of the above methods for achieving strain. For chemical pressure-induced lattice strain formed by atomic hybridization (high entropy), the location of lattice strain depends entirely on the location of atomic substitution, which leads to inhomogeneous strain. For the lattice strain formed by heterogeneous epitaxy, it is mainly concentrated at the interface of the heterostructure and may affect only 2-3 layers of the lattice, while there is no strain in the lattice away from the interface. In addition, this method generally requires laser deposition or vapor phase deposition techniques, which demand high on equipment. For external physical pressures it is usually necessary to apply a pressure of several hundred GPa using a diamond anvil cell to change the state of the material. This method is also characterized by non-uniform forces resulting in non-uniform lattice

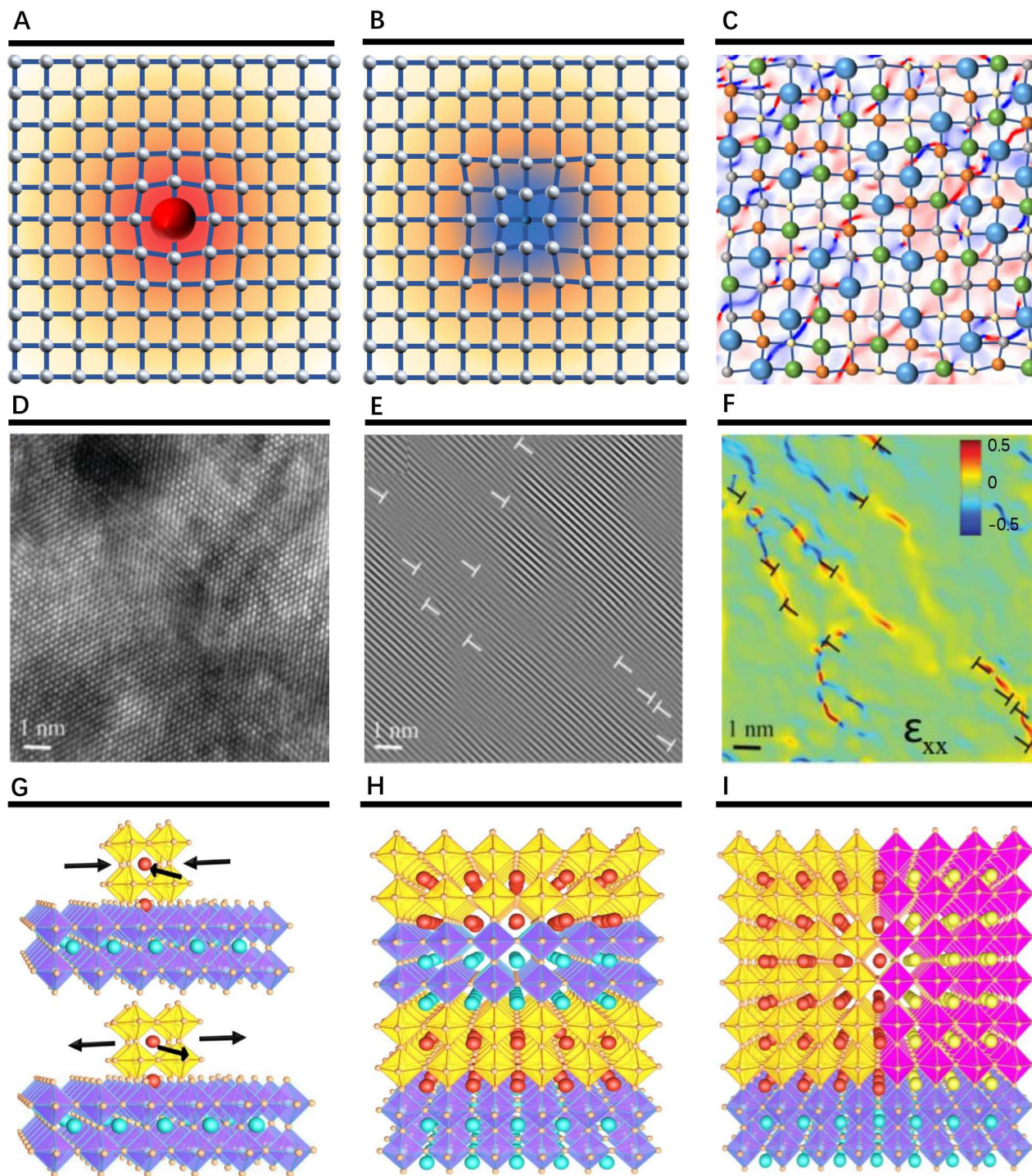


Figure 5. Generation of lattice strain Schematic plot of local lattice strain in (A) conventional solid solution alloys where the solute atom is larger than solvent atom, (B) conventional solid solution alloys where the solute atom is smaller than solvent atom, and (C) high entropy alloys. (D) The high-resolution TEM image of the deformed sample with the [011] zone axis. (E) The filtered inverse FFT image of (111) plane. (F) The corresponding atomic-scale strain map. Panel (D)-(F) reprinted with permission.⁵² Copyright 2022, Proceedings of the National Academy of Sciences. (G) Strain engineering through lattice mismatch. (H) Interface engineering to integrate different materials in an atomically abrupt manner and create interfacial phenomena. (I) An epitaxial nanocomposite created using the principles of epitaxy along the out-of-plane direction as well as the in-plane direction. Panel (G) reprinted with permission.⁵³ Copyright 2014, Cambridge university press. Panel (H)-(I) reprinted with permission.⁵⁴ Copyright 2008, Cambridge university press.

strains. For lattice strain realized by SC CO₂, uniform lattice strain can be produced since CO₂ is in a fluid state that produces a uniform higher hydrostatic pressure. Moreover, the intensity of lattice strain can be effectively regulated by different CO₂ pressures. So, this method is simple, easy to operate, and can be realized to produce GPa-level strains using lower pressures.

LATTICE STRAIN UNDER THE ACTION OF SC CO₂

SC CO₂ is uniquely attractive and irreplaceable in chemical reactions and

materials preparation, including natural products, polymers, pharmaceuticals, metallic nanomaterials, carbon materials, metal oxides, metal-organic frameworks (MOFs), and covalent-organic frameworks (COFs).¹¹⁹⁻¹²² For material preparation, shear stress and high pressure are important processing methods that can bring about changes in nanomaterials through local coordination, phase transitions, crystallization, defects, and amorphization.

The thermal heave of CO₂ in crystal tunneling leads to its entropy loss, resulting in controlled internal shear stresses (Figure 6A).¹²³ The shear stress

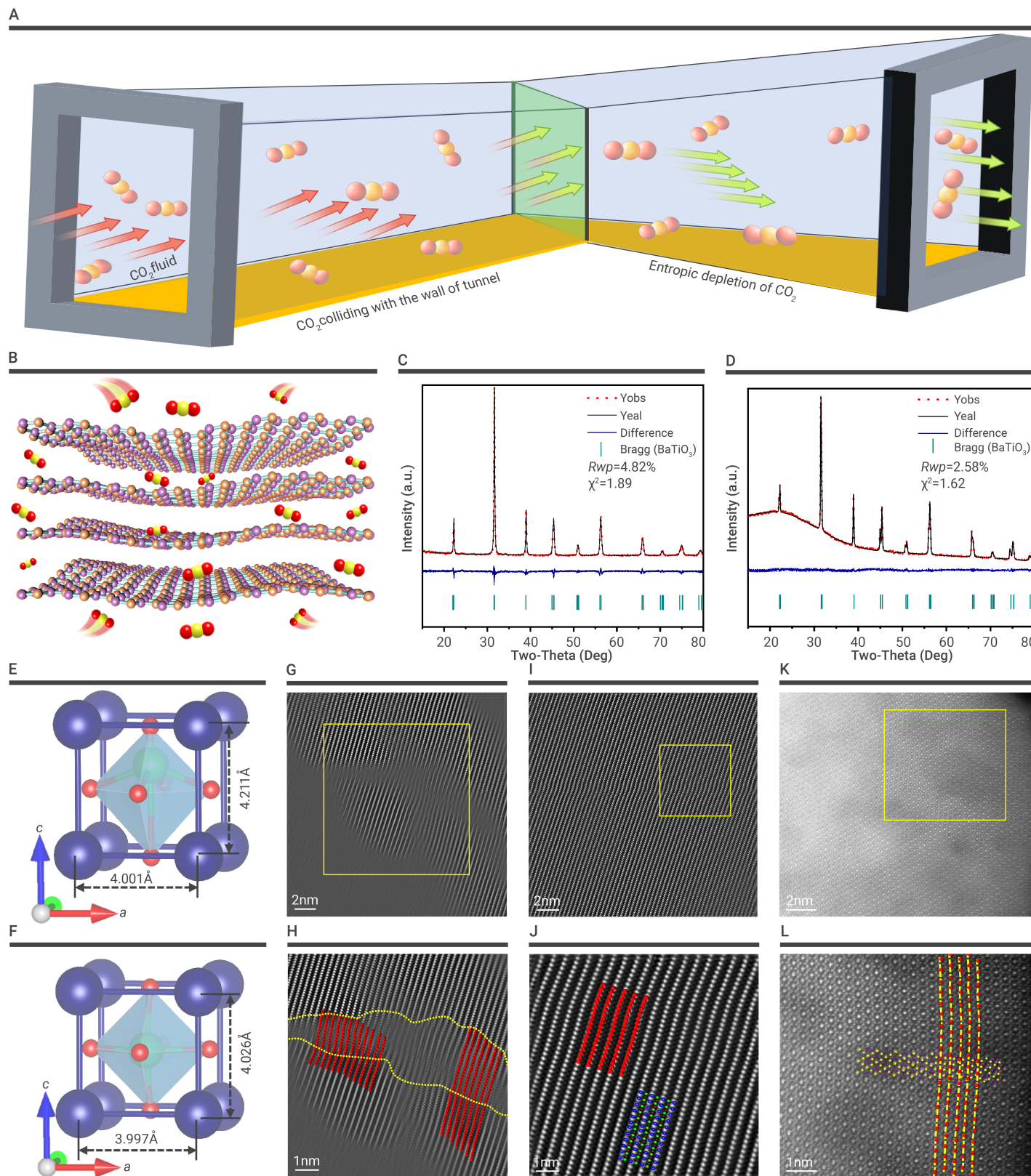


Figure 6. Lattice strain under the action of SC CO₂ (A) Schematic diagram of stripping preparation under SC CO₂. (B) Formation of a complete strained lattice structure under SC CO₂. (C) XRD refinement results of BaTiO₃ samples without SC CO₂. (D) XRD refinement results of BaTiO₃ samples treated with SC CO₂ at 16 MPa pressure. Schematic representation of the changes in lattice structure before (E) and after (F) SC CO₂ treatment, the data in the figure represent the a and c axis lattice constants. (G)-(J). STEM images of BaTiO₃ after SC CO₂ treatment. (H), (J). Localized enlarged views of the yellow boxes in Panel (G) and (I). The red balls represent atomic positions. (K)-(L). STEM images of CoFe₂O₄ after SC CO₂ treatment. (L) Localized enlarged views of the yellow boxes in Panel (K). The red spheres represent trends in atomic arrangement, where the Ba, Ti, O, Co and Fe atomic columns are outlined by blue, green, red, purple and yellow spheres, respectively.

field can cause plastic deformation of the material, resulting in a complete twisted structure (Figure 6B). In the process of preparing materials by SC CO₂,

SC CO₂ can be considered as a hydrostatic pressure (physical pressure). Under a certain hydrostatic pressure, CO₂ molecules in supercritical state can

Table 1. Changes in volume and the resulting chemical pressure of different samples after treatment with SC CO₂ at 16 MPa

	$V_0(\text{\AA}^3)$	$V(\text{\AA}^3)$	dV/V_0	$K(\text{GPa})$	$dP(\text{GPa})$
BaTiO ₃	67.41	64.32	0.0458	172.6	7.91
SrTiO ₃	59.55	60.14	0.0100	172.8	1.73
PbTiO ₃	63.09	63.25	0.0025	229.0	0.58
CaTiO ₃	223.77	223.85	0.0004	203.5	0.08
BiFeO ₃	375.18	374.44	0.0020	225.0	0.45

enter into the tunnels of the lattice structure, and the entry of large amounts of CO₂ can lead to expansion and deformation of the lattice structure. This can selectively break of chemical bonds, which enable material stripping and lead to deformation in the lattice structure to achieve a certain chemical pressure. Alternatively, it can force the atoms in the tunnel walls to change their original positions, forming local defects or phase transitions, changing the original chemical coordination environment, leading to local lattice distortion to generate a certain chemical pressure.

By XRD refinement we investigated the crystal structure of BaTiO₃ untreated (Figure 6C) and treated by SC CO₂ at 16 MPa (Figure 6D). The lattice parameters of untreated BaTiO₃ are $a = 4.001 \text{ \AA}$ and $c = 4.211 \text{ \AA}$, respectively, which indicate a clear tetragonal structure (Figure 6E). After being treated with SC CO₂, the value of c significantly decreases to $4.026 \text{ \AA}$ (Figure 6F), which is close to the value of a , indicating that BaTiO₃ has produced some lattice contraction in the c -axis direction. This indicates that a transition from tetragonal to cubic phase was produced under SC CO₂, while a reduction of the lattice volume from $67.41 \text{ \AA}^3$ to $64.32 \text{ \AA}^3$ can be confirmed. We quantitatively determine the chemical pressure from the equations relating lattice deformation and lattice elasticity:²⁹

$$dP = -K * dV/V_0$$

where dP is the chemical pressure, K is the bulk modulus, which is 172.6 GPa for BaTiO₃,^{124,125} and dV/V_0 is the variation of the lattice volume. Under the action of SC CO₂, BaTiO₃ is subjected to negative pressure of 7.91 GPa, which is much larger than the traditional heterogeneous substitution. In order to

confirm the universality of chemical pressure generated by SC CO₂, we calculated the change of lattice volume and chemical pressure before and after treatment by SC CO₂ at 16 MPa in more compounds, and the results are displayed in Table 1. Most of the samples produced GPa-level chemical pressures in the presence of SC CO₂ at 16 MPa. It can be shown that SC CO₂ at MPa level can generate internal pressures at GPa level for distortion of materials for different applications.

In order to study the effect of lattice strain under SC CO₂, the morphology of the nanostructures was investigated by HAADF-STEM. Figures 6G–J show the BaTiO₃ structure under the effect of SC CO₂ at 16 MPa. In Figure 6G, it shows an obvious clear lattice structure accompanied with lattice strain. A local zoomed-in view (Figure 6H), gives a clearer detail of the crystal structure where a lattice mismatch occurs at the yellow dashed line. Figure 6I presents another region of the BaTiO₃, in which can be observed obvious wrinkles, and the local zoom-in clearly shows the strained lattice (Figure 6J). The similar obvious lattice strain can be observed when SC CO₂ acts on CoFe₂O₄ at 16 MPa (Figures 6K–L).

APPLICATION OF SC CO₂ PRESSURE INDUCED LATTICE STRAIN

The interaction of multiple ordered factors such as spin, charge, orbital and lattice of materials under strain is complex. It is very sensitive to external factors such as magnetic field, electric field, light field, strain and temperature, thus presenting many novel physical phenomena and providing a fertile playground for revealing new paradigms and innovative applications in physics. Materials treated with SC CO₂ have been widely used.^{126–130} When SC CO₂ acts on oxide perovskite and transition metal dichalcogenides, it can lead lattice strain, and subsequently the ferromagnetism, magnetoelectric coupling, luminescence, and even efficient catalysis effect can be obtained.

Ferromagnetism of perovskite oxides

SC CO₂ was utilized to treat SrTiO₃ into a tetragonal phase with room temperature ferromagnetism.¹³¹ It was found that lattice strains were generated under the effect of SC CO₂, leading to the stretching of the cubic phase SrTiO₃ lattice into the tetragonal phase. GPA was performed on the HRTEM images, which showed that the SrTiO₃ nanosheets have a significant strain effect (Figure 7A). The crystal surface is stretched along the vertical direction and the highest occupied molecular orbital (HOMO) of Ti atoms on the (111)

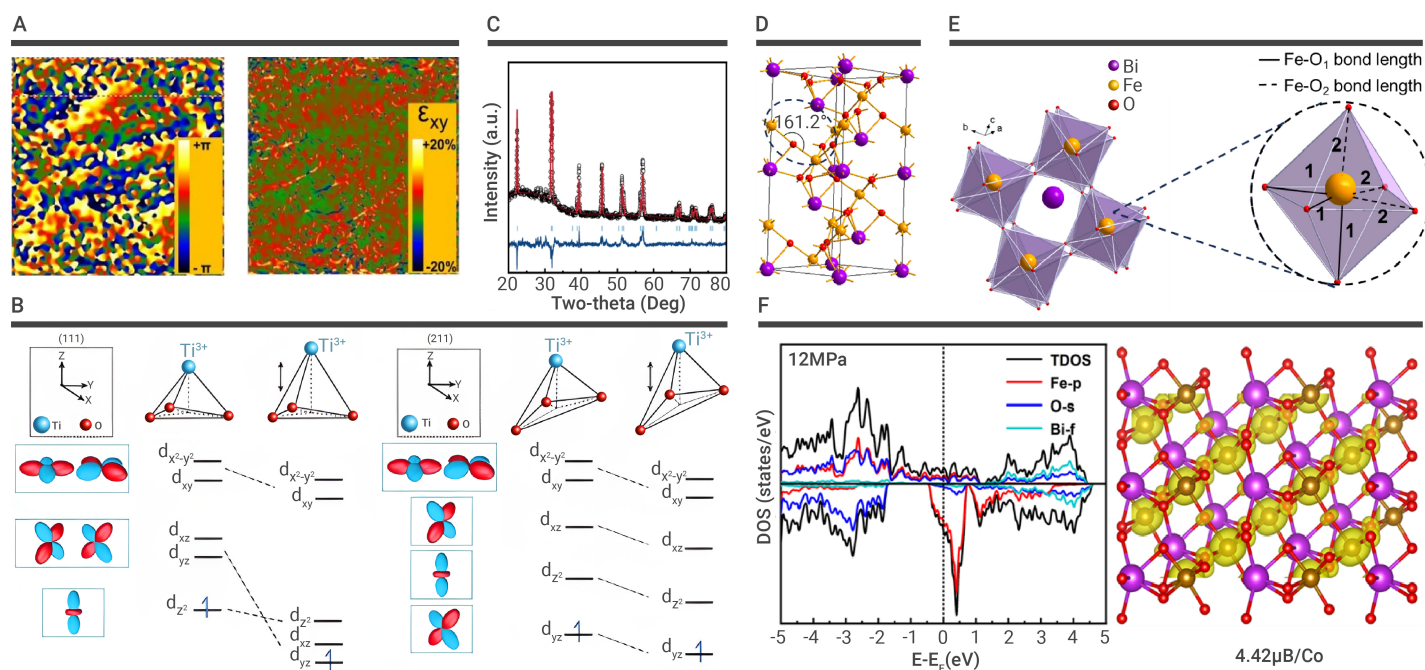


Figure 7. Strain-induced ferromagnetism of SrTiO₃ and BiFeO₃ (A) The phase map performed on the HRTEM image by geometric phase analysis (GPA). (B) The energy state of d orbital of Ti on (111) and (211) plane. (C)–(D) The Rietveld-refined XRD patterns and Lattice structure fitting diagram of BiFeO₃ under 12 MPa. (E) The FeO₆ octahedral structure indicates Fe–O bond. (F) The density of states and spin charge density distribution of BiFeO₃ treated with SC CO₂. Panel (A)–(B) reprinted with permission.¹³¹ Copyright 2023, Wiley-VCH. Panel (C)–(F) reprinted with permission.¹³⁵ Copyright 2023, Wiley-VCH.

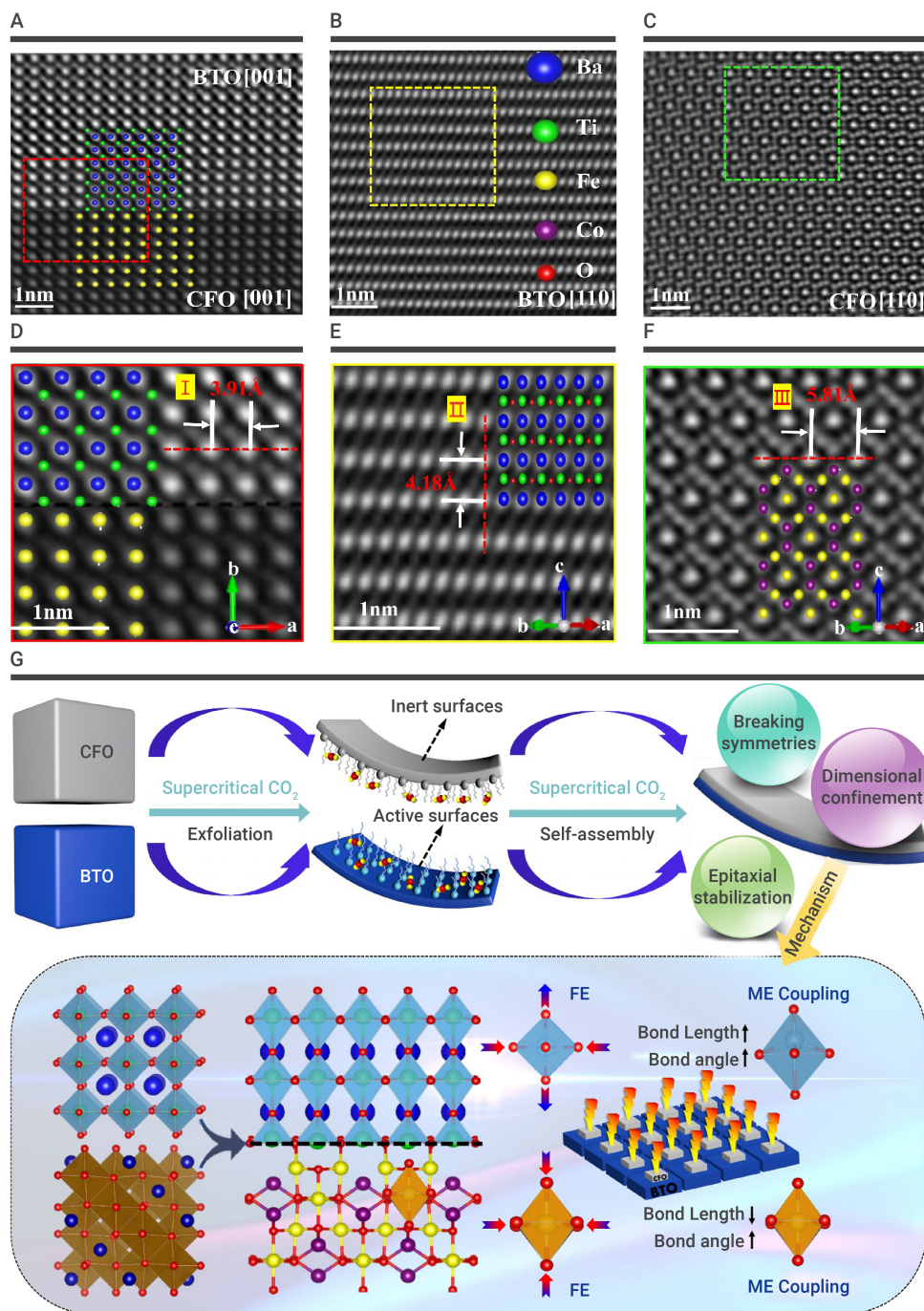


Figure 8. Microstructural characterization of nanosheets (A) Nanocomposite structure under high-angle annular dark-field scanning transmission electron microscope. The interface between the BaTiO₃ and CoFe₂O₄ (001) plane. (B) BaTiO₃ lattice structure along the [110] direction. (C) CoFe₂O₄ lattice structure along the [110] direction. (D) Locally enlarged view of the nanocomposite interface. (E) Local enlargement of the BaTiO₃ lattice structure along the [110] direction and the corresponding atomic structure. (F) Local enlargement of the CoFe₂O₄ lattice structure along the [110] direction and the corresponding atomic structure. (G) The enhancement mechanism of BaTiO₃ ferroelectricity and CoFe₂O₄ ferromagnetism. Reprinted with permission.¹²⁷ Copyright 2023, Wiley-VCH.

method, and it was found that SC CO₂ pressure changed the cell parameters and crystal structure of BiFeO₃ (Figure 7C). The Fe-O-Fe bond angle of the SC CO₂ treated sample is 161.2°, which is larger than that of the untreated BiFeO₃ (Figure 7D), and thus the SC CO₂ treated BiFeO₃ exhibits stronger super-exchange interactions. Typically, there are two types of Fe-O bond lengths in FeO₆ octahedra (Figure 7E).¹³⁸ The enhancement of the magnetic properties was further confirmed by DFT calculations (Figure 7F).

Ferromagnetism of transition metal dichalcogenides

As proposed by Landau, the breaking of time-reversal symmetry leads to ferromagnetic ordering.¹³⁹ This theory is suggested to be realized through a combination of experimental approaches combining dimensionality reduction, phase transitions, and crystal surface exposure. In particular, tailoring the intrinsic structure of semiconductors allows for the simultaneous control of spin and charge, which then endows them with intrinsic ferromagnetism.¹⁴⁰ CO₂ permeate and swell the material at the molecular scale, and then "plasticize" the VO₂ solid at the architectural scale in order to form a 2D network of defects that "locks" the metastable phases into a new topology. Experimental results and theoretical calculations confirm that CO₂ molecules entering the interior of the VO₂ structure generate a strong shear stress to selectively break V-O bonds. The bulk VO₂ is then stripped off the high-energy surface

and (211) surfaces are stabilized, which promotes the spin density in the d_{yz} orbital and thus significantly enhances the magnetic moments (Figure 7B). Magnetic characterization indicates that the as-prepared samples have higher room-temperature ferromagnetism, which is about 7-15 times higher than that of other previously reported preparation methods.¹³²⁻¹³⁴

BiFeO₃ is a twisted rhombic perovskite structure at room temperature, and neutron diffraction experiments showed it is a G-type antiferromagnet.¹³⁶ The oxygen octahedra surrounding the transition metal cations are rotated around this axis, and the direction of the magnetic moments in the bulk rotates along a long-range cycloidal spin structure of 62–64 nm. It was found that if this cycloidal spin structure is suppressed, the system will display weak ferromagnetism.¹³⁷ It is found that SC CO₂ can destroy the cycloidal spin structure of BiFeO₃, leading to an increase in the Fe-O-Fe bond angle, which generates stronger super-exchange interactions. Ultimately, the saturation magnetization strength of BiFeO₃ is increased by nearly 23 times.¹³⁵ The XRD diffraction data were refined using the Rietveld structure refinement

and partially amorphized. At a pressure of 16 MPa, the diffusion of CO₂ leads to a transition from a three-dimensional bulk to a 2D network of defects, and formation of high-index (4306-), (5-)-3(-)-10 planes. The new topology obtained in this case exhibits a significantly enhanced ferromagnetic response.

Magnetoelectric coupling

In addition to the above single-phase structures, 2D nanocomposites were obtained by alternating in-plane and out-of-plane self-assembly growth of BaTiO₃ and CoFe₂O₄ realized by SC CO₂.¹²⁷ The coupling between BaTiO₃ and CoFe₂O₄ under the action of SC CO₂ leads to a certain amount of biaxial strain of each constituent, which enhances the ferroelectricity and ferromagnetism of BaTiO₃ and CoFe₂O₄. As a result, the coupling between BaTiO₃ and CoFe₂O₄ exhibits an unprecedentedly magnetoelectric (ME) coupling coefficient (348.5 mV cm⁻¹ Oe⁻¹) due to the lattice strain. The morphology of the nanostructures was investigated by HAADF-STEM, and a clear lattice structure and an

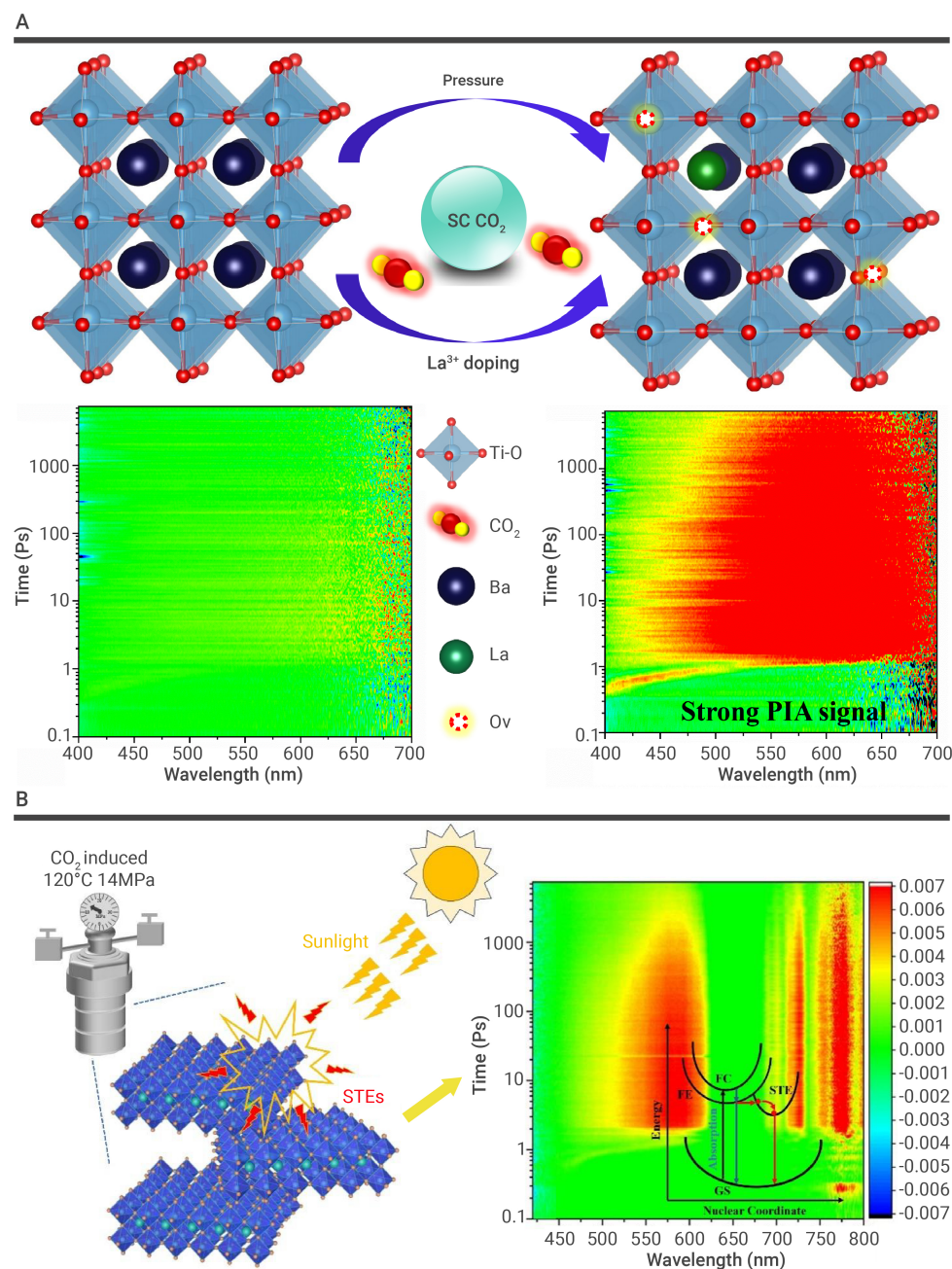


Figure 9. Luminescence Properties of BaTiO₃ and SrTiO₃ (A) Changes in transient absorption spectra under the combined effect of SC CO₂ pressure and La ion doping. (B) SC CO₂-induced lattice strain in SrTiO₃ realizes the emergence of STEs under photoexcitation. Panel (A) reprinted with permission.¹⁴¹ Copyright 2023, Wiley-VCH.

Luminescence properties

Lattice strain under SC CO₂ pressure has also been applied to improve the luminescence properties of BaTiO₃.¹⁴¹ Under the combined effect of SC CO₂ pressure and La ion doping, BaTiO₃ undergoes obvious lattice strain. With the increase of doping concentration, the degree of lattice strain increases, effectively promoting the generation of self-trapped excitons (STEs) (Figure 9A). At the same time, the increase in the degree of strain is accompanied by the enhancement of the photogenic positive absorption signal in the transient absorption measurements. This finding suggests that the lattice strain modulates the photoluminescence of BaTiO₃ nanocrystals. In addition, SC CO₂ modulated lattice strain was successfully generated STEs in SrTiO₃ in the absence of doping (Figure 9B). There is a phase transition from cubic to tetragonal phase in the lattice structure of SrTiO₃ under the effect of SC CO₂ pressure. This effectively contributes to the generation of STEs.

Catalysis

The discovery and control of new phases of matter is significant in materials research. It is found that 2D layered MoS₂ exists in two phases, 2H and 1T, the 2H phase can exist stably in nature and the 1T phase is difficult to be found in nature. How to effectively convert the 2H phase into the 1T phase and reveal the related mechanism is an important issue. Through a SC CO₂-induced phase transition preparation system, the 2D atomic lateral heterostructures with 1T- and 2H-MoS₂ phases (1T@2H-MoS₂ nanosheets) were realized in the SC CO₂/ethanol/water system. Theoretical calculations demonstrated the

obvious two-phase interface can be seen in Figure 8A. According to Figure 8D, the atomic spacing of Ba atoms along the a-axis direction on the (001) plane is 3.91 Å, which is 2.13% smaller than the lattice constant (a) (3.995 Å), which suggests that there is a clear compression of BaTiO₃ along the equatorial direction of the octahedron. The lattice structure of CoFe₂O₄ along the [110] direction is given in Figures 8C & F, where the Fe atom spacing decreases from 5.93 Å to 5.81 Å, which is a typical value for the crystallographic space of the CoFe₂O₄ (220) plane. This decrease in atomic spacing indicates that CoFe₂O₄ is compressed along the axial direction. The thickness and lateral dimensions of the BaTiO₃-CoFe₂O₄ nanocomposites decrease with the increase of CoFe₂O₄ content during SC CO₂ treatment, which favors the interaction between the components. This interaction leads to a certain biaxial strain on each other, which enhances the ferroelectricity of BaTiO₃ and the ferromagnetism of CoFe₂O₄ (Figure 8G). Ultimately, the ME coupling properties of BaTiO₃-CoFe₂O₄ nanocomposites were effectively enhanced. This work demonstrates a new idea of preparing multiferroic nanocomposites by utilizing the biaxial strain generated by the interaction of the two phases in the presence of SC CO₂ to modulate the ME properties.

straining effect in single-layer MoS₂ nanosheets generated from the different adsorption strengths of CO₂ on 2H (0.18 eV) and 1T (0.62 eV) phases.¹⁴² Overall, the effects of local straining field can change the form of coordination number and symmetry of chemical bonding, and result in the phase transition. The obtained 1T phase exhibit excellent performance in photocatalytic and electrocatalytic hydrogen evolution reaction.

In our previous work, we successfully prepared PdCu alloys on C substrates using SC CO₂.¹⁴³ The process simultaneously realized the transformation of PdCu face-centered cubic phase (fcc) to body-centered cubic phase (bcc) and the structural distortion of the C substrate (Figures 10A-C). Their synergistic effects contributed to the ultra-high mass activity (3624.3 mA/mg_{Pd}) of the electrocatalytic formic acid oxidation reaction. In addition, an intralayer [Bi₂O₂]²⁺ structure was successfully constructed in ternary layered BiOCl using SC CO₂, and the structural distortion of intralayer [Bi₂O₂]²⁺ was successfully realized (Figures 10D-I). Combined with the experimental results of electrocatalytic CO₂ reduction reaction (CO₂RR) and DFT theoretical calculations, it was shown that the structure could significantly improve the reactivity and selectivity of HCOOH, exhibiting a Faraday efficiency of 90% and a formic acid current density of 32 mA cm⁻² mg⁻¹ within a wide voltage window

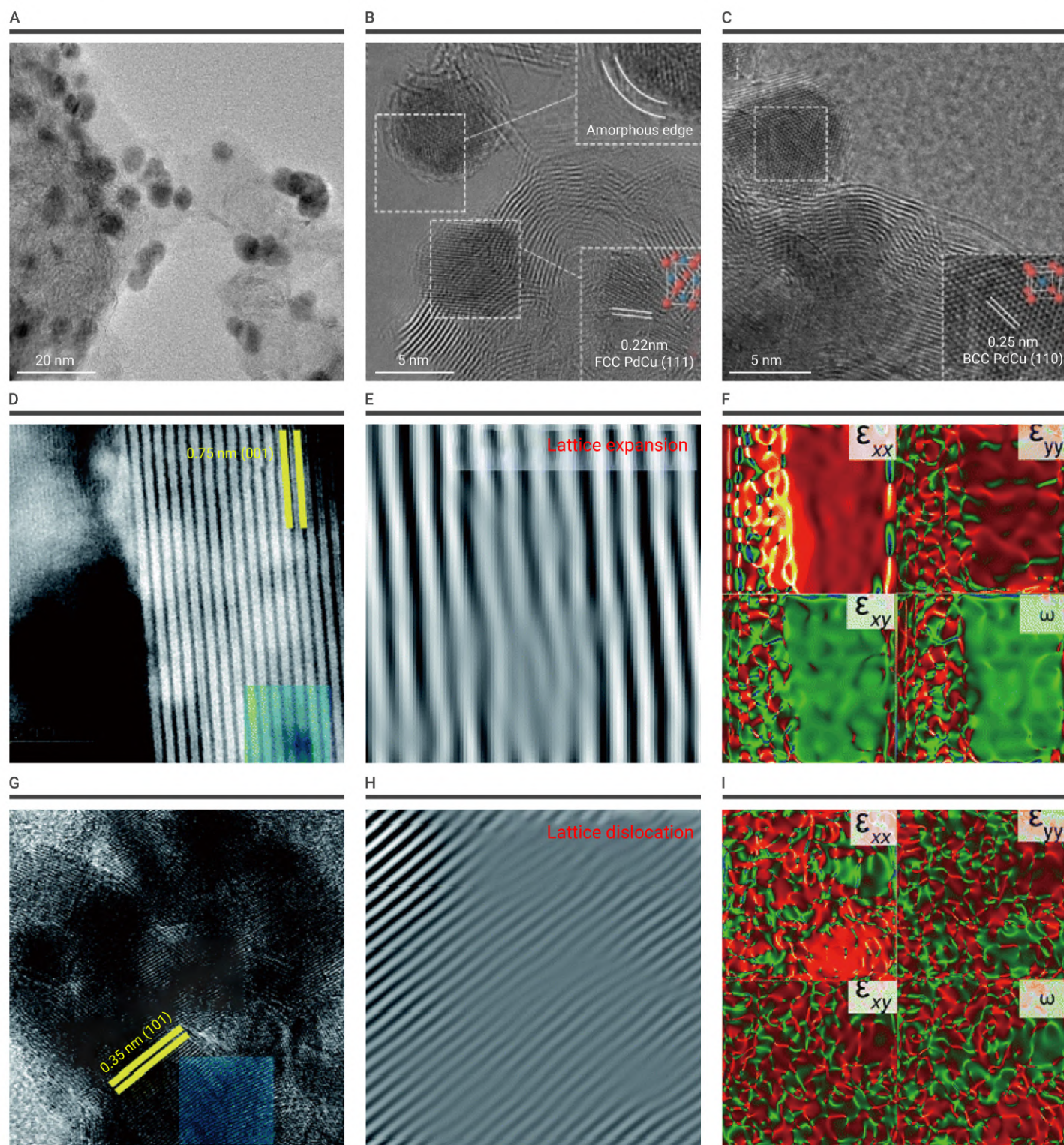


Figure 10. Strain-induced catalytic properties (A)–(C) TEM and HRTEM of the PdCu nanoflakes prepared with 20 MPa SC CO₂. (D)–(I) Microscopic characterization of distorted BiOCl nanoplates. Panel (A)–(C) reprinted with permission.¹⁴³ Copyright 2023, Wiley-VCH. Panel (D)–(I) reprinted with permission.¹⁴⁴ Copyright 2020, Royal Society of Chemistry.

from -0.6 to -0.9 V. The structure was also used in the reduction of HCOOH in a wide voltage window. The competitive side reactions of CO and H₂ were effectively suppressed in CO₂RR reduction engineering.¹⁴⁴

SUMMARY AND OUTLOOK

In summary, lattice strains realized by external physical or internal chemical stresses can modulate the spin, orbital, and charge of a material to induce emergent phenomena such as skyrmion, thermoelectricity, superconductivity,

ferroelectricity, ferrimagnetism, multiferroic, catalysis and electro-optical. CO₂ carries MPa-level external physical pressure into the crystal structure, destroying the original chemical coordination environment to form localized defects or phase transitions, generating internal chemical pressure up to the GPa level. This CO₂-mediated conversion of external physical pressure into internal chemical pressure for lattice strain has been demonstrated in the fields of ferrimagnetism, multiferroic, catalysis and optoelectronic. It is believed that SC CO₂ can be applied to more fields in the future. And, some

current new research results have confirmed that the ferroelectric properties can be excited by SC CO₂. This method of preparing materials with strained structures is very exciting and it will be a very interesting and promising in the field of materials fabrication. With the help of SC CO₂, complex strained structured materials with even greater impact are still to be developed, and some new physical phenomena are about to be revealed in more complex strained structures. We will learn how to harness this new lattice-strained material preparation method to find more wonders.

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AUTHOR CONTRIBUTIONS

B. Gao designed and performed the experiments, collected, and analyzed the data, wrote the manuscript and carried out the theoretical calculations. Q. Xu conceived the project, analyzed the data, and improved the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

There is no new data generated.

LEAD CONTACT WEBSITE

Xu group information is detailed at <http://www5.zzu.edu.cn/xuqun/>.