

Enduring Mechanical Memory from the Constitutive Response of Elastically Recoverable Nanostructured Materials

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Mechanical memory and computing are gaining significant traction as potential means to complement traditional electronics for robust and energy efficient performance in extreme environments. However, progress has largely focused on bistable metamaterials, while traditional constitutive memory phenomena have been largely overlooked—primarily due to the absence of compelling experimental demonstrations in elastically recoverable materials. Here, we report constitutive return point memory (RPM) in elastically recoverable, vertically aligned carbon nanotube (VACNT) foams, analogous to magnetic hysteresis-based RPM utilized in hard drives. Unlike viscoelastic fading memory, VACNTs exhibit nonvolatile memory arising from rate-independent nanoscale friction. We find that the interplay between RPM and frictional dissipation enables independent tunability of the VACNTs' dynamic modulus, allowing for both on-demand softening and stiffening. We leverage this property to experimentally demonstrate tunable wave speed in a VACNT array with elastic interlayers, paving the way for novel shock limiters, elastodynamic lensing, and wave-based analog mechanical computing.

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I. INTRODUCTION

The ability to encode, retrieve, and erase information by leveraging a material's constitutive response, known as the material-memory effect, manifests in diverse forms across various material systems [1,2]. However, a large body of work is focused on geometric-instability-based “digital” mechanical memory, where bistable building blocks, like beams [3,4], shells [5], origami cells [6,7], shape memory, or magnetic actuators [8], snap between two stable configurations to encode states 0 and 1. Although these configurational states can be retained without power and operate reliably in harsh environments, the stored information is encoded in the geometric configuration of the structure [3,9–14]. Distinct from this configurational memory, the constitutive material-memory effect arises from the material's intrinsic constitutive mechanisms, reflecting the material's remembrance of the history of applied stimuli. In such cases, simply understanding and exploiting the constitutive response is sufficient to achieve

tunable functionalities, without the need for complex metamaterial design. This approach is analogous to examples of constitutive memory observed in current-voltage hysteresis in memory resistor (memristor) based devices [15], shape memory triggered by various physical stimuli [16,17], memory of previously applied maximum stress observed in the Mullins effect [18], and return point memory (RPM) in magnetic hysteresis for data storage in hard drives and magnetic tapes [19,20].

In ferromagnetic materials exhibiting magnetic hysteresis, RPM refers to the ability to retain memory of the magnetization (M) and field (H) state when switching the external magnetic field's direction, as the hysteresis curve returns to this point when the field's direction is switched back [19,21] [Fig. 1(g)]. This behavior fundamentally requires a rate-independent and fully reversible hysteretic response [21], enabling nonvolatile memory and access of constitutive states. Magnetic systems closely approximate this ideal RPM due to rapid, reversible spin reorientation and domain wall motion with negligible relaxation. By analogy, in mechanical systems, RPM is the memory of the stress (σ) and strain (ϵ) state in the constitutive response of the material at the point where the loading direction reverses [Fig. 1(f)]. While apparent RPM-like behavior has been observed across diverse mechanical systems [13,22–24], the nature of underlying mechanisms differs fundamentally from that of magnetic RPM. In granular and

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rock systems, hysteretic responses often exhibit RPM-like features alongside pronounced quadratic nonlinearity in wave propagation. However, these arise from dissipative processes such as dilatancy, frictional rearrangements, and microcracking, which are inherently irreversible and often time dependent [22,25,26]. In elastoplastic solids, classic studies show RPM-like features in the stress-strain response. After loading beyond yield and unloading, subsequent reloading retraces to the prior reversal point and rejoins the monotonic curve [2,24,27]. However, this mechanism intrinsically involves permanent plastic deformation that alters the material's internal structure, rendering the memory inherently irreproducible. Similarly, crumpled sheets exhibit RPM-like loops due to bistable geometric features (e.g., d-cones and ridge networks), yet the memory progressively degrades with cycling due to slow plastic flow and structural aging [13]. Amorphous granular and colloidal assemblies can encode "trained" amplitudes under oscillatory shear, with the ability to remember the most recently trained amplitude, analogous to RPM [28,29]. However, unlike the RPM seen in constitutive response, here the memory is encoded through training and it resides in the configuration, making this phenomena training protocol dependent and distinguishing it from the readily available RPM seen in constitutive response of materials.

Until now, RPM arising from intrinsically rate-independent and reversible mechanisms, analogous to those in magnetic systems, has not been reported in constitutive response of elastically recoverable material systems, leaving their potential for practical applications untapped. Here, we demonstrate such highly consistent and repeatable RPM in elastically recoverable vertically aligned carbon nanotube (VACNT) foams synthesized by a scalable chemical vapor deposition (CVD) process [Fig. 1(d) herein and Fig. S1 in Supplemental Material (SM) [30]]. We further demonstrate that VACNT foams exhibit a time-independent (or rate-independent) and reversible hysteretic response that enables nonvolatile retention of constitutive states, fundamentally distinguishing it from previously reported RPM-like behavior in mechanical systems. VACNT foams are renowned for their high specific energy absorption [31], fatigue resistance [32], and thermal stability [33,34]. While their mechanical and thermal properties have been extensively studied [35], little is known about their hysteretic stress-strain response under nonmonotonic loading, such as partial cyclic loading [21,36], which is crucial for understanding the constitutive memory effects identified in this paper. Moreover, their dynamic mechanical behavior at high frequencies and across various strain amplitudes remains elusive. In this study, we comprehensively characterize the mechanical behavior of VACNT foams as a function of strain rate and strain magnitude through stress-relaxation, quasistatic cyclic compression, and dynamic mechanical analysis (DMA) experiments. We found that unlike viscoelastic polymeric foams, VACNT foams do not

exhibit stress relaxation and demonstrate strain-rate-independent behavior. Contrary to previous speculations [32,33,37], we provide evidence for a nonviscoelastic mechanism of energy dissipation in VACNT foams, governed by frictional interactions between nanotubes. We developed a novel deformation-dependent stick-slip frictional (DDSSF) model, consisting of springs and frictional sliders [38], to explain the rate-independent behavior. Our model accurately captures the RPM in VACNT foams and provides insights into its origin, attributed to stick-slip frictional interactions among carbon nanotubes (CNTs) [27,39]. We show that RPM and deformation-dependent friction lead to dual tunability in the dynamic modulus of VACNT foams: strain-amplitude-dependent softening, known as the Payne effect [40], and static precompression-dependent stiffening. We experimentally demonstrate that these effects enable slowing of traveling stress pulses with increasing impulse amplitude, yet speed up under greater static precompression in a multilayer array of VACNT foams with elastic aluminum interlayers. This dual tunability facilitates amplitude-programmable wave propagation crucial for various wave lensing and diffusing applications [41]. Our work expands the scope of material memory to address long-standing engineering challenges and establishes RPM as a novel addition to the growing field of mechanical memory, alongside metamaterials.

II. FADING MEMORY AND RETURN POINT MEMORY

In this section, we establish why time-independent constitutive behavior is a prerequisite for true RPM and why materials with fading memory cannot realize it. The foremost requirement for any nonvolatile memory is the indefinite stability of the encoded information. For example, the polarity of ferromagnetic domains established during magnetic hysteresis remains permanently aligned even after the external magnetic field is removed. In contrast, the mechanical stress-strain hysteresis observed in soft dissipative materials, such as elastomers and polymeric foams, does not retain a lasting memory of the applied stress. Instead, they display fading memory due to their viscoelastic behavior. Fading memory implies that the magnitude of the effect of a recent cause is significantly higher than that of an earlier cause of the same magnitude [42,43]. To elaborate, consider a standard linear solid (SLS) viscoelastic material subjected to a strain that increases linearly with dimensionless time ($\bar{t}$) [Fig. 1(a)]. Let $d\epsilon$ represent an infinitesimal increase in strain from $\bar{t} = \bar{\tau}$ to $\bar{t} = \bar{\tau} + d\bar{\tau}$ [Fig. 1(a)]. The corresponding increase in dimensionless stress [Fig. 1(b)] can be expressed as follows (see details in SM [30]),

$$d\bar{\sigma} \downarrow = \mathcal{W}(\bar{t} - \bar{\tau}) \downarrow \times d\epsilon, \quad (1)$$

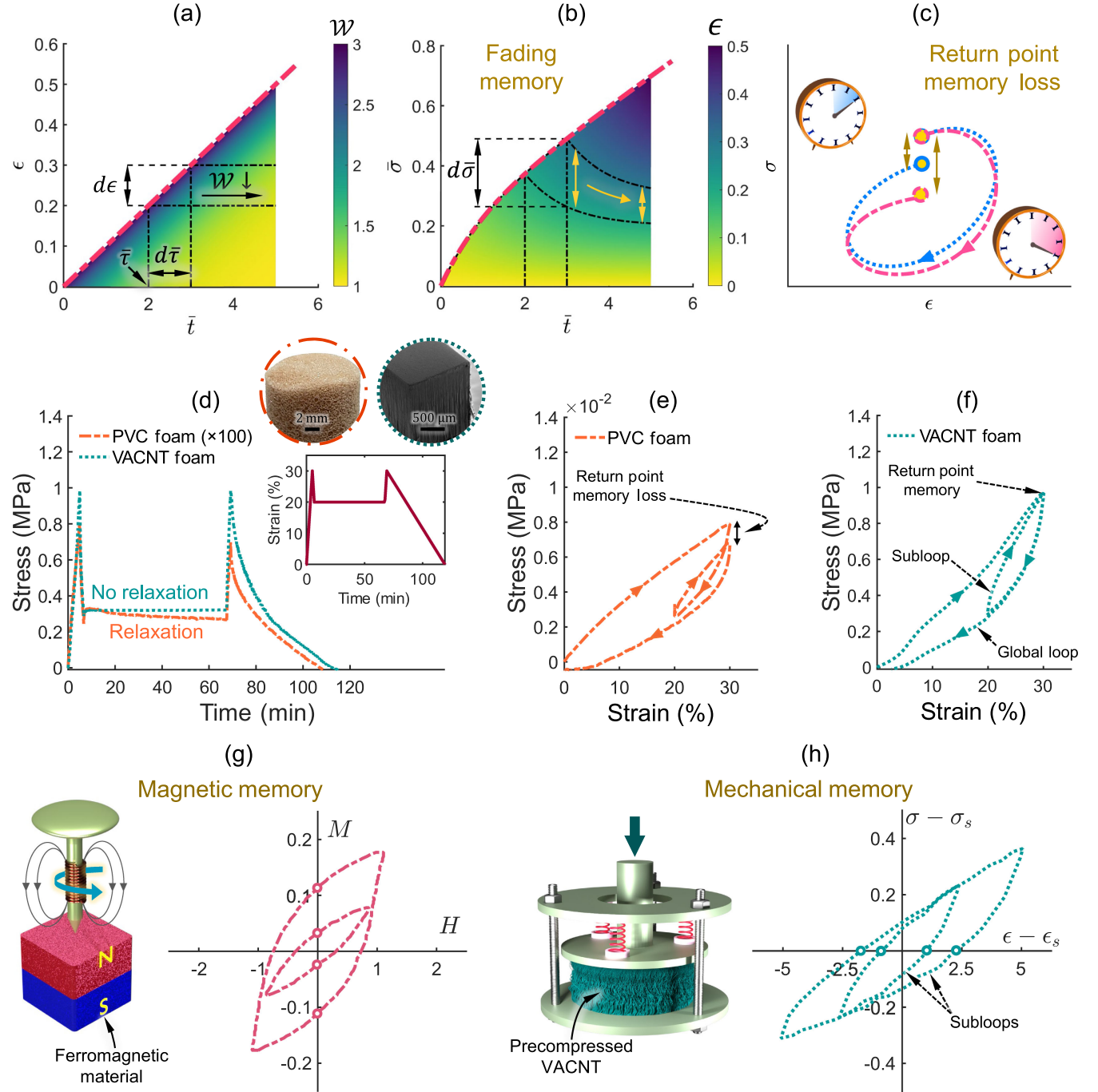


FIG. 1. Fading memory and return point memory. (a) Plot of strain applied on an SLS material as a function of dimensionless time. A small increment in strain $d\epsilon$ applied from $\bar{t}$ to $\bar{t} + d\bar{t}$ is shown. The weight of each strain increment as a function of time is represented using a color map (see details in SM [30]). (b) Resultant dimensionless stress as a function of dimensionless time with contribution of each strain increment to the stress is shown using color map. (c) An illustration of return point memory loss for different time intervals. (d) Comparison of stress as a function of time for VACNT foam and viscoelastic PVC foam, corresponding to the applied strain shown in the inset. PVC foam's stress is multiplied by 100 for better comparison. (e) Return point memory loss in viscoelastic PVC foam. (f) Return point memory in VACNT foam. (g) Illustration of magnetic memory achieved via ferromagnetic hysteresis [21]. (h) Illustration of mechanical memory through stress-strain hysteresis subloop in a precompressed VACNT sample.

where $\mathcal{W}$ represents the weight associated with $d\epsilon$, such that $\mathcal{W}(\bar{t} < \bar{\tau}) = 0$, and the down arrow ($\downarrow$) indicates temporal decay. The magnitude of $\mathcal{W}$ decreases with time due to relaxation [Fig. 1(a)], resulting in a fading memory

effect [Fig. 1(b)]. Alternatively, if the strain is increased in equal steps at equal time intervals, the stress increment $d\bar{\sigma}$ resulting from the most recent strain increment $d\epsilon$ will contribute more to the total stress ($\bar{\sigma}$), whereas the $d\bar{\sigma}$

associated with earlier de values will have faded more significantly [Fig. 1(b)], as they have had more time to relax. RPM loss is observed during the cyclic loading of viscoelastic materials because the memory of the switching point, where the loading direction is reversed, fades over time. The longer the duration, the greater the fading effect, as illustrated in Fig. 1(c).

Unlike viscoelastic polymeric foams, the VACNT foams do not undergo stress relaxation, enabling the memory of loading-direction reversal to persist indefinitely, which leads to RPM. In Fig. 1(d), we present the experimentally measured stress response of a VACNT foam sample alongside a viscoelastic polyvinyl chloride (PVC) foam subjected to a prescribed strain profile over time [see inset of Fig. 1(d)]. Both materials were first compressed (loaded) to a certain maximum strain (30%), partially unloaded (-10%), and then held at a constant strain for an extended period. As shown in Fig. 1(d), the PVC foam exhibits stress relaxation, whereas the stress in the VACNT foam remains constant, indicating the absence of relaxation. Upon reloading to the maximum strain (30%), the PVC foam fails to retrace the previous unloading point, resulting in RPM loss [Fig. 1(e)]. In contrast, the time-independent behavior of VACNT foam enables it to close the hysteresis subloop, exhibiting RPM [Fig. 1(f)]. This focused experiment provides direct, quantifiable evidence of enduring, true RPM in VACNT foams, in clear contrast to the fading memory in viscoelastic systems. Notably, we also examined sample-to-sample variation of RPM across different VACNT specimens and found that, although the stress levels vary with density, all samples exhibit RPM (Fig. S11 in SM [30]) despite the slight morphological variations introduced by the CVD synthesis process (Appendix A). To maintain consistency across the paper, we establish the following terminology: The hysteresis loop obtained from cyclic loading of an unstrained VACNT sample is referred to as the global loop, while the hysteresis loop resulting from any cyclic perturbation applied to a statically precompressed VACNT sample is termed the subloop [Fig. 1(f)].

Figure 1(f) presents the first demonstration of stress-strain RPM in a soft, elastically recoverable material. Notably, once the hysteresis subloop is fully enclosed by reloading, the subsequent unloading to zero strain follows the original global unloading path, as if the subloop had never occurred. In other words, when a hysteresis cycle is completed, whether it is a subloop or a global loop, the loading history responsible for that cycle is erased. This phenomenon is known as the wiping-out effect associated with RPM [19]. In Figs. 1(g) and 1(h), we draw analogy between ferromagnetic memory and VACNT-based mechanical memory. In ferromagnetic hysteresis [21], the remanent magnetization (M) after the removal of an external magnetic field ($H = 0$) can exist in two different states ($\pm$), representing bits. Similarly, the hysteretic subloop of a VACNT sample precompressed under a

constant static stress can retain two stable remnant strain states, representing mechanical bits analogous to magnetization in Fig. 1(g). These remnant strains depend on the loading history of the externally applied stress, analogous to a magnetic field. The absence of both stress relaxation and creep in VACNTs preserves the established state of stress and strain, ensuring stable mechanical bits for enduring material memory. Moreover, in magnetic hysteresis, the size of the hysteresis loop can be shrunk or expanded by varying the amplitude of the cyclic external magnetic field (H) [21], allowing the remnant magnetization (M) to continuously take any value along the y axis, as shown in Fig. 1(g). Similarly, in VACNTs, the size of the hysteretic subloop can be shrunk or enlarged depending on the extent of strain partially unloaded [Fig. 1(h)]. This allows remnant strains to take continuous values along the x axis in Fig. 1(h) at a constant static stress. In contrast, metamaterials with geometric bistable states are limited to two discrete states: on and off [6,10].

To understand the fundamental origin of RPM, its influence on the dynamic mechanical properties of VACNT foams, and the potential applications enabled by the resulting tunable behavior, we perform comprehensive experimental characterization across a broad range of strains and strain rates.

III. TIME-INDEPENDENT VACNT FOAMS

VACNT foams exhibit a porous morphology composed of entangled multiwalled CNTs that behave like slender elastic struts (Fig. S1 [30]), undergoing bending and buckling [44] under compression—similar to open-cell polymeric foams. Owing to their morphological similarity and hysteretic response, the energy dissipation in VACNT foams has been erroneously attributed to viscoelasticity in the literature [32,33,37]. While energy dissipation in polymeric foams primarily arises from viscoelasticity, dissipation in VACNT foams is thought to occur predominantly through interfacial friction between nanotubes [31,45]. Experimental observations of viscoelastic time-dependent behaviors such as stress relaxation and creep deformation in VACNTs likely result from either incomplete preconditioning of the sample [32] or misinterpretation of strain overshoot from the load frame as relaxation of the VACNT foam [37].

Here, we experimentally demonstrate rate-independent mechanical behavior and a nonviscoelastic mechanism of energy dissipation that underpins the RPM effects in VACNT foams. We synthesize VACNT foams using a floating catalyst thermal chemical vapor deposition (tCVD) process (Appendix A) resulting in multi-walled carbon nanotubes that are nominally aligned vertically along the direction of growth [Fig. S1(c) [30]] [44]. When compressed, pristine VACNT samples exhibit temporary irreversible softening [Fig. 2(c)] as a function of compression cycles—a phenomenon known as the preconditioning

effect—which originates from the rearrangement of CNTs and permanent strain induced at the nanoscale [44]. The material progressively becomes compliant during the first few cycles before the softening eventually ceases, and the sample reaches a preconditioned state after 4–5 cycles [Fig. 2(c)]. During preconditioning, local buckling of nanotube bundles within the VACNT sample can cause fluctuations in the monotonicity of the loading-curve slope during the first compression cycle [44]. However, this effect diminishes as preconditioning progresses and the nanotubes become less vertically aligned. Figure S1(d) [30] shows an SEM micrograph of a preconditioned sample, indicating that the initial vertical alignment of CNTs has been altered. To be clear, preconditioning does not play a role in encoding a configurational memory into the material, rather it simply removes transient effects so the stress-strain response stabilizes. Once VACNT foams are preconditioned, their hysteretic stress-strain curves become reproducible and fully elastically recoverable over an indefinite number of cycles, provided the applied strain does not exceed the preconditioning strain [31,32,44]. To ensure a consistent mechanical response across various experiments, we first preconditioned our sample by applying cyclic compression up to preconditioning strain of 60%

at a strain rate of 0.01 s^{-1} for 5 cycles [Figs. 2(b) and 2(c)] (Appendix B). Notably, our study focuses exclusively on the compressive behavior of VACNTs. Tensile testing is impractical because gripping the thin samples is difficult, and tensile loading tends to pull the CNTs apart rather than reliably loading the whole foam [46], so it offers no practical utility. However, in a precompressed state, a VACNT sample can reliably experience local tension while the net strain remains compressive, allowing us to perform DMA and wave transport experiments.

A. Strain-rate-independent constitutive behavior

Figure 2(d) shows the cyclic quasistatic hysteretic stress-strain response of a preconditioned VACNT sample measured over strain rates spanning 3 orders of magnitude, with the maximum strain kept below the preconditioning strain (60%). The hysteretic global loop, comprising both loading and unloading curves, remains unaffected by varying strain rates, indicating rate-independent behavior [47]. In contrast to the strong rate dependence observed in viscoelastic polymeric foams and elastomers [43], this rate independence in VACNT foams suggests that energy dissipation arises from non-viscoelastic mechanisms. Additionally, the rate-independent

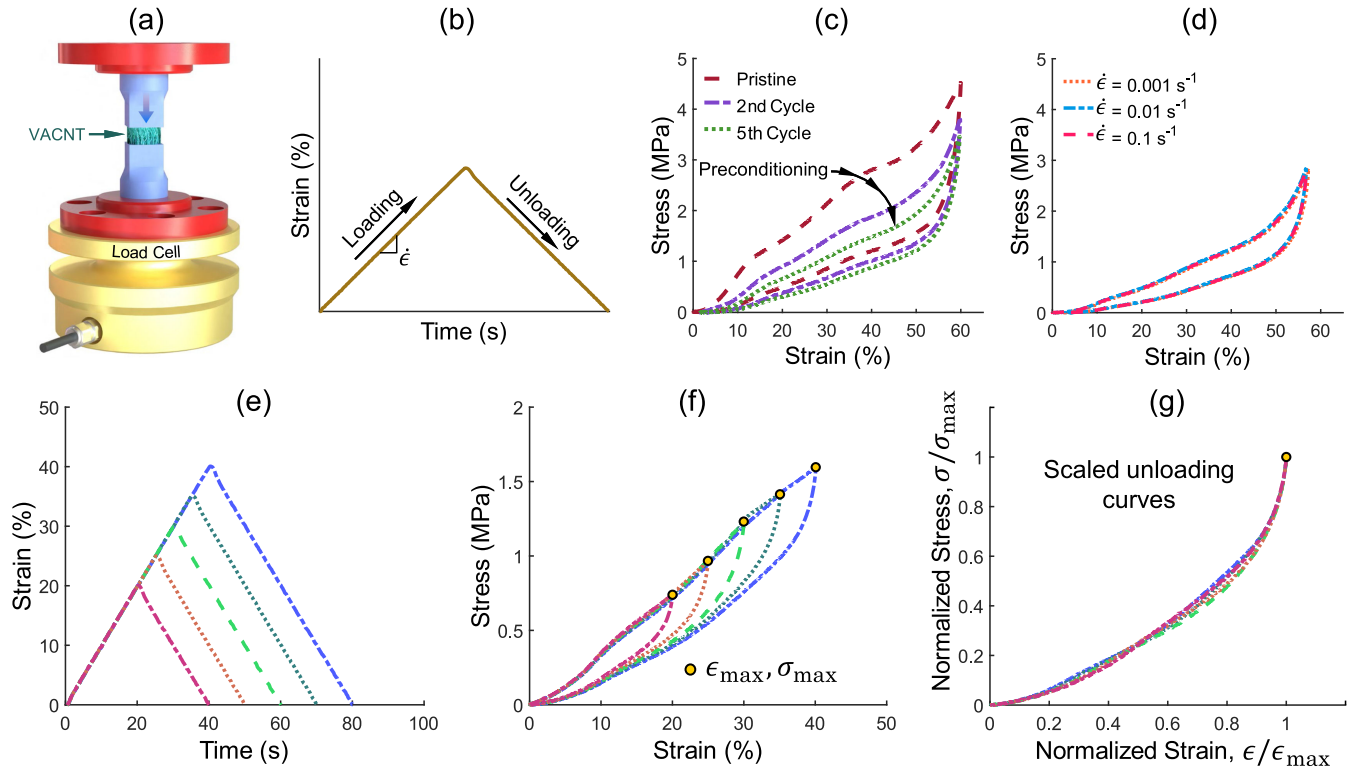


FIG. 2. Quasistatic compression response of VACNT foams. (a) Illustration of the Instron ElectroPulse E3000 load frame used for quasistatic compression and stress-relaxation experiments. (b) Applied ramp strain with a constant strain rate for quasistatic cyclic compression. (c) Quasistatic ramp compression of a pristine VACNT sample undergoing mechanical preconditioning. (d) Preconditioned stress-strain global-hysteresis loops, showing rate independency. (e) Strain applied as a ramp at a strain rate of 0.01 s^{-1} up to various maximum strain levels, followed by unloading. (f) Resulting cyclic stress-strain responses plotted as a function of maximum strain. (g) Normalized unloading curves illustrating the observed scaling behavior.

behavior is consistent with the absence of stress relaxation observed earlier [Fig. 1(d)]. Our detailed stress-relaxation experiments on VACNT foam further confirmed the absence of stress relaxation across all applied strain magnitudes (Appendix C herein and Fig. S6 in SM [30]).

Figures 2(e) and 2(f) show the cyclic quasistatic global-hysteresis loops obtained by ramping the strain to various maximum values ($\epsilon_{\max}$). Figure 2(g) compares the unloading portions of the hysteresis loops, scaled by normalizing stress and strain by their respective maximum values. The scaled unloading curves overlap, suggesting the unloading curve scales self-similarly as a function of the maximum strain (or maximum stress) while retaining its overall shape. Notably, this scaling no longer holds in regimes where the loading curve becomes nonlinear—for example, near the onset of densification ($\geq 49.64\%$) [Fig. 2(d)] or in the initial nonlinear region ($\leq 10\%$) caused by surface irregularities in the sample [Fig. 2(f)]. The scaling holds in the regime where the loading curve remains approximately linear, which informs our choice of the $\epsilon_{\max}$ range considered in Figs. 2(e)–2(g). In Sec. III C, we demonstrate that the self-similar scaling property of the unloading curve plays a crucial role in enabling tunable dynamic mechanical properties in VACNT foams.

B. Frequency-independent dynamic behavior

While the quasistatic ramp compression and stress-relaxation experiments unambiguously indicate time-independent mechanical behavior, these experimental methods probe only quasistatic strain rates. For structured and hierarchical materials, it is often argued that the bulk behavior becomes rate dependent when the frequency of the external load matches the characteristic resonances of the microscale or nanoscale constituent features that remain dormant at low strain rates.

Previously, VACNT foams have been shown to exhibit frequency-independent dynamic modulus and loss factor at low frequencies up to 100 Hz [33]; however, their broadband behavior has not been well characterized. Since commercial load frames have limitations on operating frequency, we built a custom dynamic mechanical analyzer [Fig. 3(a)] consisting of a piezoelectric actuator and a dynamic force sensor capable of measuring the dynamic mechanical properties of soft materials up to 1000 Hz (Appendix D). To perform dynamic mechanical analysis, we first apply a static precompression strain on the sample (ϵ_s) and then subject it to a sinusoidal strain of a desired amplitude (ϵ_d) and frequency (f) using the piezoelectric actuator [Fig. 3(a)]. The total strain (ϵ_T) can be expressed as follows:

$$\epsilon_T(t) = \epsilon_s + \epsilon_d \sin(2\pi ft). \quad (2)$$

In response, the force sensor measures the oscillatory component of the force, which we convert to stress by normalizing it by the cross-sectional area of the sample

(Appendix D). The oscillatory components of stress and strain [Fig. 3(a)] form a hysteretic subloop centered at the origin as shown in Fig. 3(b). A frequency sweep from 1 to 1000 Hz at constant $\epsilon_d = 0.063\%$ and $\epsilon_s = 10\%$ reveals that the hysteretic subloops remain unchanged with frequency [Fig. 3(b)], indicating broadband frequency-independent behavior. From these loops, we extract two key dynamic mechanical properties of VACNT foams—dynamic modulus (E_d) and loss factor (ζ)—defined as follows (see details in SM [30]):

$$E_d = \frac{\sigma_d}{\epsilon_d}, \quad \zeta = \frac{E_l}{\sqrt{E_d^2 - E_l^2}}, \quad E_l = \frac{W_{\text{dis}}}{\pi \times \epsilon_d^2}. \quad (3)$$

Here, σ_d is the dynamic stress amplitude, E_l represents the effective loss modulus, $\sqrt{E_d^2 - E_l^2}$ denotes the effective storage modulus (see details in SM [30]), and W_{dis} is the energy dissipated per unit volume, calculated from the area inscribed in the hysteretic subloop. Both E_d and ζ are frequency independent [Figs. 3(c) and 3(d)], affirming rate-independent behavior and nonviscoelastic origin of energy dissipation. The apparent slight frequency dependence at low frequencies is attributed to charge leakage in the piezoelectric force sensor (see details in SM [30]). In contrast, the strain gauge-based load cell in the Instron ElectroPulse E3000 does not exhibit this limitation, allowing for more accurate measurements at low frequencies. Nevertheless, unlike the strongly frequency-dependent behavior of viscoelastic foams, the broadband, frequency-independent response observed here is uncommon among dissipative foams reported in the literature.

C. Dynamic softening and stiffening governed by the global-hysteresis loop's unloading curve

To examine the effects of ϵ_d and ϵ_s , we measured the dynamic mechanical properties as a function of ϵ_d , ranging from 0.0125% to 1.25%, for various values of $\epsilon_s \leq 30\%$, while keeping the total strain (ϵ_T) well below the onset of densification (49.64%). The dynamic modulus (E_d) decreases steadily with increasing ϵ_d for each applied ϵ_s [Fig. 3(e)], demonstrating strain-amplitude-dependent softening, commonly known as the Payne effect [39,40]. In contrast, the loss factor increases with amplitude [Fig. 3(f)] because E_l [Eq. (3)] exhibits only a weak dependence on amplitude [Fig. S2(a) [30]], while $\sqrt{E_d^2 - E_l^2}$ decreases sharply with amplitude [Fig. S2(b) [30]]. For a fixed ϵ_d , E_d increases with ϵ_s indicating stiffening [inset of Fig. 3(e)], whereas the loss factor remains almost unchanged [Fig. 3(f)].

To understand the contrasting effects of dynamic (ϵ_d) and static strain (ϵ_s) on the dynamic modulus (E_d), we examine the complete stress-strain trajectory from the DMA experiments, which involve ramping the strain to

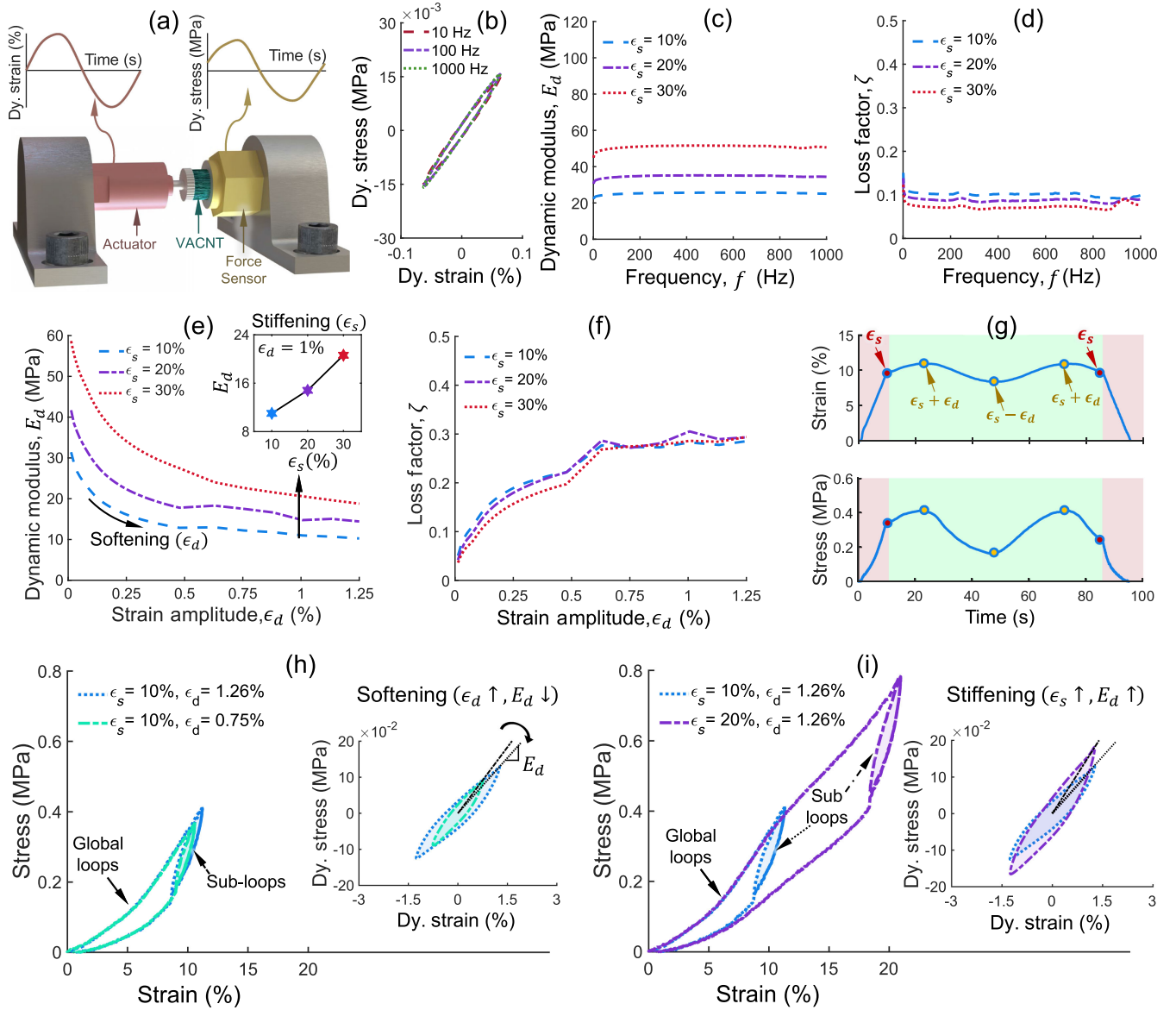


FIG. 3. Dynamic mechanical analysis response of VACNT foams. (a) Illustration of our custom-built experimental apparatus for performing dynamic mechanical analysis. (b) Stress-strain hysteresis subloops for applied sinusoidal strain at different frequencies. (c),(d) Dynamic modulus and loss factor as functions of frequency ($\epsilon_d = 0.063\%$). (e) Experimentally measured dynamic modulus and (f) loss factor as functions of dynamic strain amplitude (ϵ_d) and static precompression (ϵ_s). (g) Strain applied as a function of time (top) to measure the dynamic mechanical properties. Corresponding stress response as a function of time (bottom). (h) Comparison of complete hysteresis stress-strain responses from DMA testing for a fixed $\epsilon_s = 10\%$ and two different ϵ_d , with the corresponding hysteresis subloops enlarged and compared in the inset. (i) Comparison of complete hysteresis stress-strain responses for a fixed $\epsilon_d = 1.26\%$ and two different ϵ_s , with the corresponding hysteresis subloops shown in the inset.

ϵ_s , holding it constant, and then superimposing a dynamic sinusoidal strain [Fig. 3(g)]. Since the force sensor in our custom DMA setup records only time-varying forces and the actuator's piezostack has a limited stroke, we conducted this experiment using an Instron Electropulse E3000 system (Appendix D). Figure 3(g) shows the applied strain waveform and resulting stress response as functions of time. Note that while the dynamic component of the applied strain follows the intended sinusoidal form, the corresponding stress response is quasisinusoidal—it

maxima and minima align with those of the strain, but the curves diverge elsewhere. As a result, the hysteresis loop exhibits a biconvex shape with slope discontinuities, forming sharp corners when the direction of strain application is reversed [Fig. 3(b)], in contrast to the smooth, elliptical loops typically observed in viscoelastic foams [43]. This slope discontinuity upon strain reversal suggests that the underlying dissipative mechanism is direction dependent—a phenomenon we examine in detail in Sec. IV.

In Fig. 3(h), we compare the full stress-strain response for $\epsilon_d = 0.75\%$ and $\epsilon_d = 1.26\%$, both at the same pre-compression strain $\epsilon_s = 10\%$. For both ϵ_d , the initial loading in the global loop up to the maximum strain ($\epsilon_s + \epsilon_d$) follows the loading curve observed in the quasistatic ramp compression [Fig. S2(d) [30]]. The sinusoidal part of the applied strain [$\epsilon_d \sin(2\pi ft)$] [Fig. 3(g)] results in a small hysteretic subloop composed of an unloading half-cycle ($2\epsilon_d$ strain unloaded from $\epsilon_s + \epsilon_d$ to $\epsilon_s - \epsilon_d$) and a reloading half-cycle (loading the strain back from $\epsilon_s - \epsilon_d$ to $\epsilon_s + \epsilon_d$) [Fig. 3(h) herein and Figs. S2(c) and S2(d) in SM [30]]. Because of rate independence, the unloading half-cycle traces the corresponding portion of the global loop's unloading curve up to the extent of strain unloaded [Fig. S2(d) [30]]. In contrast, the reloading half-cycle closely matches an inverted version of the unloading half-cycle, indicating that the hysteresis loop exhibits Masing behavior [27] [Fig. S2(d) [30]]. The reloading half-cycle fully encloses the hysteretic subloop, achieving RPM.

It is evident that the unloading and reloading segments of the hysteretic subloop are composed of a small portion of the global unloading curve near the maximum strain and its inverted counterpart, respectively. As ϵ_d increases, a longer portion of the curve is traversed [Fig. 3(h)]. Because the slope of the global unloading curve decreases from the maximum strain toward zero strain [Figs. 2(f) and 2(g)], the hysteretic subloop exhibits a lower average slope and appears tilted clockwise [inset of Fig. 3(h)], resulting in a lower dynamic modulus. In contrast, for a fixed ϵ_d , increasing ϵ_s causes the subloop to tilt counterclockwise [inset of Fig. 3(i)], indicating stiffening. Notably, this two-way dependence of the dynamic modulus arises solely from the global unloading curve, while the global loading curve is nearly linear and plays no significant role. This unique behavior is not seen in other material systems, where such properties predominantly originate from the loading response. Here, we theoretically demonstrate that these properties originate from the scaling behavior of the global unloading curve discussed earlier [Figs. 2(e)–2(g)].

As discussed earlier, the quasistatic unloading curve scales as a function of the maximum compressive strain ($\epsilon_{\max}$) [Figs. 2(e)–2(g)]. We assume for the scaled unloading curve [Fig. 2(g)], the normalized stress ($\sigma/\sigma_{\max}$) is a function of the normalized strain $f(\epsilon/\epsilon_{\max})$ as follows:

$$\sigma = \sigma_{\max} \times f\left(\frac{\epsilon}{\epsilon_{\max}}\right). \quad (4)$$

Using the above equation, applying RPM for hysteresis, we derive the following approximate equation for E_d as a function of ϵ_s and ϵ_d (see details in SM [30]):

$$E_d \approx 3.981 \times \left[f'(1) - \frac{\epsilon_d}{(\epsilon_s + \epsilon_d)} f''(1) \right]. \quad (5)$$

Here, $f'(1)$ and $f''(1)$ denote the first and second derivatives of $f(\epsilon/\epsilon_{\max})$, evaluated at $\epsilon/\epsilon_{\max} = 1$. Since the scaled unloading curve is monotonically increasing and concave up [Fig. 2(g)], both derivatives are positive. As a result, the analytical expression predicts that E_d decreases with increasing ϵ_d but increases with increasing ϵ_s . The general nature of the derivation underlying Eq. (5) makes this relationship applicable to any material that exhibits similar scaling of unloading curve. The comprehensive mechanical characterization of VACNT foams revealed their rate-independent behavior and tunable dynamic mechanical properties, but left the following questions unresolved. What physical mechanism gives VACNT foams their Masing behavior and RPM? And why does their unloading curve scale? We address these questions through our rate-independent, deformation-dependent stick-slip friction model.

IV. DEFORMATION-DEPENDENT STICK-SLIP FRICTIONAL MODEL

We propose a novel rate-independent frictional damping model that captures the experimentally observed mechanical behavior and provides insights into the origin of RPM and Masing behavior in VACNT foams. While viscoelastic models equipped with spring-dashpot elements have been widely used [48], their inherent rate dependence makes them unsuitable for VACNT foams. Moreover, the rate independence, Masing behavior, and slope discontinuities observed in VACNT foams suggest that a frictional damping model is appropriate, as these features are distinctive characteristics of such models. Instead of dashpots, frictional damping models use Coulomb sliders, typically illustrated as a pin sliding on a plate [Figs. 4(a) and 4(b) herein and Fig. S3 in SM [30]]. The slider remains locked (stick) until the external force exceeds a break-free friction force (F_{bf}), triggering sliding (slip) motion [27,39,49]. Frictional models, such as the Iwan model (Fig. S3 [30]), have been shown to effectively capture unloading-point memory and the Masing behavior observed in elastoplastic materials and bolted joints [27]. Notably, these models are fundamentally analogous to the hysteron-based Preisach model used to describe ferromagnetic memory [19,21]. The hysteron-based Preisach model leads to remanent magnetization in ferromagnetic materials, analogous to plastic deformation in the Iwan model. Thus, similar to the Iwan model, hysteron-based models such as triboelastic models with quadratic hysteresis [39] are not suitable for modeling the VACNT response.

We modify the elastoplastic frictional damping model to exhibit full elastic recovery by adopting Coulomb sliders with the break-free friction force (F_{bf}) proportional to the applied deformation (x)—a DDSSF model. When connected in parallel with a spring ($F_s = kx$), the friction force in such a Coulomb slider (F_{bf}) opposes the relative sliding motion between the pin and the plate. As a result, it can

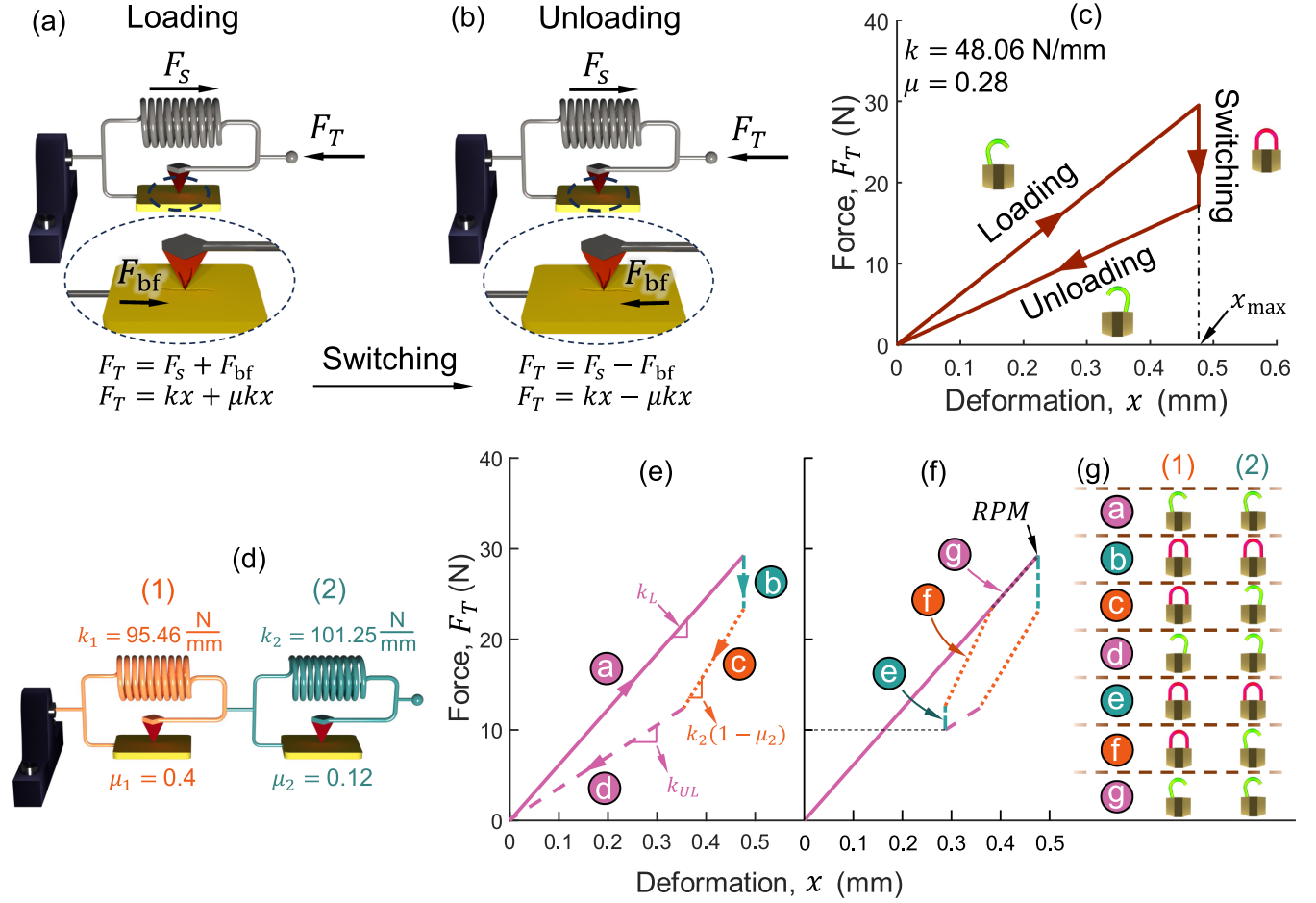


FIG. 4. Deformation-dependent stick-slip frictional (DDSSF) model. (a) A spring and a deformation-dependent Coulomb slider connected in parallel, undergoing compression (loading) and (b) unloading. (c) The hysteretic force-deformation cyclic response for a single spring-slider pair. (d) Two spring-slider pairs connected in series, each with different stiffness and friction coefficients. (e) The resulting cyclic force-deformation response. (f) Two spring-slider pairs exhibiting RPM. (g) Lock-unlock illustrations depicting the locking of Coulomb sliders during various phases of loading and unloading.

either add to $(F_s + F_{bf})$ or subtract from $(F_s - F_{bf})$ the spring force, depending on the direction of sliding [Figs. 4(a) and 4(b)]. The force in the spring (F_s) represents the elasticity of the CNT network undergoing bending and buckling, whereas F_{bf} represents the internal friction between nanotubes. During compressive loading, the compaction of the VACNT foam enhances van der Waal frictional interactions [50], motivating our assumption that $F_{bf} \propto F_s$ [38,51]. Since $F_s = kx$, the magnitude of friction force F_{bf} becomes deformation dependent ($F_{bf} \propto kx$). As shown in Figs. 4(a) and 4(b), an external force F_T during loading (compression) and unloading (decompression) can be expressed as follows (see details in SM [30]):

$$F_{bf} = \pm \mu F_s, \quad F_s = kx, \quad (6)$$

$$\begin{aligned} F_T &= kx \times (1 + \mu) \quad (\text{loading}), \\ F_T &= kx \times (1 - \mu) \quad (\text{unloading}), \end{aligned} \quad (7)$$

where μ is the coefficient of friction ($0 \leq \mu < 1$), and k is the spring stiffness. Under cyclic compression, the hysteretic response of a single spring-slider pair [Figs. 4(a) and 4(b)] consists of three segments: a linear loading branch, a linear unloading branch, and a switching segment [Fig. 4(c)]. When compressed (loaded) to a deformation of x_{max} , the spring force is $F_s = kx_{max}$, and the friction force is $F_{bf} = \mu kx_{max}$. When the pair is unloaded, the direction of the friction force in the slider switches from $+\mu kx_{max}$ to $-\mu kx_{max}$ to allow slipping in the opposite direction [Figs. 4(a) and 4(b)]. During this switching, the total force drops by $2\mu kx_{max}$ while the slider remains locked, resulting in no change in deformation (x) [Fig. 4(c)]. Once the friction direction switches, the slider unlocks, and both the spring and the slider unload completely, following a linear force-deformation curve [Eq. (7)]. Unlike the Iwan model, which uses a constant F_{bf} (Fig. S3 [30]), our model results in no plastic deformation due to the deformation-dependent F_{bf} . Slope discontinuities occur at the transition points

between loading and switching, and between switching and unloading, resembling those observed experimentally in VACNT foams.

Connecting multiple spring-slider pairs with varying stiffnesses and friction coefficients in series produces a gradual unloading response [Figs. 4(d) and 4(e)], similar to that observed experimentally in VACNT foams. When an external compressive load is applied, all pairs compress simultaneously, following a linear loading curve [Fig. 4(e)] whose slope corresponds to the effective loading stiffness k_L (see details in SM [30]). After loading to a certain maximum force ($F_{T,\max}$), the sliders unlock sequentially in order of increasing μ during unloading. As shown in Figs. 4(e) and 4(g), during segment (b) of unloading, while the force decreases, both pairs stay locked, and the deformation remains unchanged. Once the force drops sufficiently to unlock pair (2), the next unloading segment (c) follows a slope of $k_2(1 - \mu_2)$, while pair (1) stays locked, with its slider's friction still in the process of switching direction. Once pair (1) also unlocks, both pairs unload simultaneously with a combined slope of k_{UL} (see details in SM [30]).

Using this same model of two spring-slider pairs, the fundamental origins of return point memory and Masing behavior can be understood. Figure 4(f) shows the hysteretic response when the model is partially unloaded to $F_T = 10$ N, and then reloaded back to the previously applied maximum force, forming a hysteretic subloop. When the loading direction is reversed at $F_T = 10$ N, the friction direction in the sliders, initially $-ve$, must switch back to $+ve$ to enable reloading, causing the sliders to unlock sequentially once again in the order of increasing μ . Once both pairs have unlocked, the reloading curve merges with the original global loading curve, ultimately exhibiting RPM. Figure 4(g) illustrates how the friction direction in the individual Coulomb sliders acts like mechanical bits, encoding the loading history. The evolution of friction direction during the subloop cycle reveals that RPM arises directly from both sliders resetting their friction direction to positive upon completing reloading. This reset allows the force (F_T) and deformation (x) to return to their previous magnitudes, thereby producing RPM. Furthermore, since the spring-slider pairs unlock in the same sequence each time the loading direction is reversed, an inverted symmetry emerges in the hysteretic subloop—characteristic of Masing behavior. However, for higher extents of partial unloading, this inverted symmetry gradually fades, and the Masing behavior ultimately ceases to exist, as detailed in SM [30].

Connecting many spring-slider pairs in series effectively captures the experimentally measured global-hysteretic stress-strain response of VACNT foams [Fig. 5(a) herein and Figs. S4(a) and S4(b) in SM [30]]. We developed a MATLAB [52] script (see also SM [30]) to compute the appropriate values of spring stiffnesses (k_i , $i = 1, 2, \dots, n$)

and friction coefficients (μ_i) for a given number of spring-slider pairs (n) to fit the experimental hysteretic response [Fig. S4(b) [30]]. Once the fitted values of k_i and μ_i are determined, the force-deformation response for any arbitrary force input can be calculated using another custom MATLAB script [see Ref. [52] and Fig. S4(b) in SM [30]]. Force and deformation are then normalized by the VACNT sample's cross-sectional area and thickness, respectively, to obtain stress and strain. Figure 5(a) shows the stress-strain response of a 25-pair DDSSF model accurately capturing the experimental hysteretic stress-strain response measured for various maximum strain levels. The inset of Fig. 5(a) shows the DDSSF unloading curves also exhibit the scaling property previously observed in experiments (Fig. 2). The DDSSF model provides insight into the origin of this scaling: The friction force in each slider at the onset of unloading is proportional to the maximum applied force as $\mu_i F_{T,\max} / (1 + \mu_i)$ (see details in SM [30]). Since the friction force in all sliders scales uniformly with increasing $F_{T,\max}$, the unloading curve also scales while preserving its shape.

Additionally, the model captures the strain-amplitude-dependent softening and static-precompression-dependent stiffening behavior observed in DMA experiments, accurately predicting the experimentally measured dynamic moduli [Figs. 5(b) and 3(e)]. We further demonstrate the accuracy of the DDSSF model by predicting the stress-strain response under an arbitrary strain input applied as a function of time [Fig. 5(c)]; the experimentally measured response [Fig. 5(e)] closely matches the earlier model prediction [Fig. 5(d)], validating the predictive capability of the DDSSF model. The hysteretic responses estimated using the DDSSF model, shown in Figs. S4(c) and S4(d) [30], also closely match the experimental results [Figs. 3(h) and 3(i)], further confirming the model's accuracy. The compelling experimental evidence and theoretical verification we have presented demonstrate that our DDSSF model is the most accurately devised constitutive model so far for VACNT foam materials compared to existing literature [34,53–55]. Moreover, our model overcomes the plastic-deformation limitation inherent to Jenkins-element-based Iwan models, thereby extending its applicability to material systems that exhibit elastic recovery. The DDSSF model can be further equipped with nonlinear springs to capture initial nonlinearity in the loading curve for $F_T < 10$ N and nonlinearity in the loading curve for compressive strains beyond the onset of densification [Fig. 2(d)], but this is beyond the scope of the present work.

The striking agreement between the DDSSF model and the experimentally measured constitutive response supports a stick-slip friction-motivated effective constitutive description of VACNT foams at the bulk scale, rather than providing direct microscopic evidence of individual nanotube stick-slip events. However, this stick-slip friction-motivated basis for modeling bulk VACNT foams is consistent with

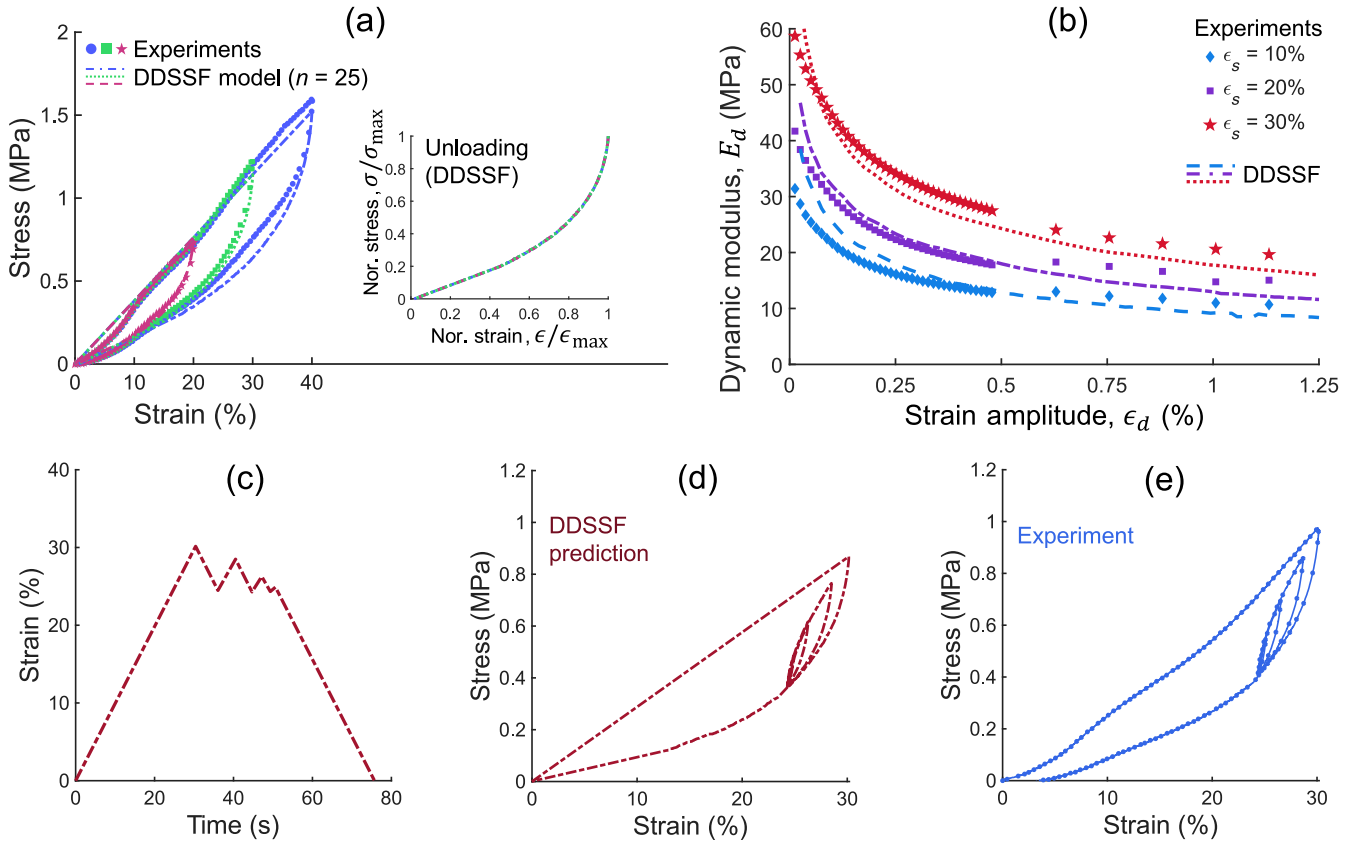


FIG. 5. DDSSF model validation. (a) DDSSF model capturing experimental global hysteresis and scaling property of the unloading curve. (b) Experimentally measured dynamic modulus as a function of strain amplitude and precompression strain, compared with DDSSF model predictions. (c) Strain applied as a function of time. (d) Stress-strain response predicted by the DDSSF model and (e) experimental verification.

previously reported nanoscale stick-slip phenomena in CNT systems. At the nanoscale, direct measurements using atomic force microscopy (AFM) show that friction on graphene and carbon nanotube surfaces exhibits stick-slip behavior as the AFM tip scratches the surface [56–58]. The AFM tip slips when the applied force exceeds the breakaway friction and exhibits sawtoothlike fluctuations about a steady value due to stick-slip behavior [58,59]. The magnitude of friction scales with the true contact area [60,61], consistent with the dry Coulomb friction law. AFM-based measurements are limited to the outer surface and exhibit frictional behavior resembling the Jenkins element used in the Iwan model [27]. At larger scales, such as in CNT yarns, mats, and VACNT foams, frictional behavior emerges from the statistically averaged, stochastic interactions among a large number of nanotubes within the material volume [33,34,62,63]. During loading, SEM imaging and coarse-grained molecular dynamics simulations show that neighboring nanotubes within the van der Waals interaction radius undergo rapid attachment and detachment at ultrafast time-scales (see SM [30]) [33,34,63]. This process, often termed zipping-unzipping, leads to energy dissipation [34,64,65]. Upon unloading and reloading, these same contacts follow a

reversible sequence of attachment and detachment, giving rise to RPM. While attachment occurs spontaneously, detachment dissipates energy as heat [33,34,63].

At the macroscale in VACNTs, as the bulk compressive strain increases, the average gap between nanotubes decreases [66,67], which strengthens van der Waals interactions and increases the frequency of attachment-detachment events. We show that the number of frictional interactions per unit volume is proportional to the bulk strain: For every 1% increase in bulk strain, CNT-CNT interactions increase by approximately 2% (see SM [30]) [66]. The combined effect of these interactions between polydispersed nanotubes with varying diameters, chirality, and orientation manifests as friction that scales with bulk deformation. This behavior is captured in the DDSSF model using Coulomb sliders spanning a range of friction coefficients, paired with springs of varying stiffness to represent nanotubes with different diameters and bending stiffnesses [Figs. S4(a) and S4(b) [30]]. In addition to capturing full stress-strain response and strain recovery [Fig. 5(a)], the DDSSF model reproduces the self-similar scaling of the unloading curve [inset of Fig. 5(a)], which emerges from deformation-dependent

friction. Other hysteron-based models fail to capture these features, further supporting that nanoscale friction in VACNTs gives rise to bulk-scale deformation-dependent friction. Although this rapid, reversible attachment-detachment motion has been observed in coarse-grained molecular dynamics simulations [34] and SEM studies [33], further strengthening the connection between atomic-scale friction and bulk behavior would require incorporating detailed information on the deformation-driven, dynamically evolving nanoscale morphology of VACNT foams. This would involve techniques such as synchrotron x-ray scattering with femtosecond pulses, tightly focused x-ray nanobeams, and *in situ* compression with micrometer-scale precision. However, this combination exceeds current synchrotron beamline capabilities and is beyond the scope of this work. In summary, our DDSSF model proved to be a successful effective constitutive model supporting the origin of RPM and how its interplay with deformation-dependent friction gives rise to a rich landscape of tunable dynamic mechanical properties, poised for novel engineering applications explored in Sec. V.

V. WAVE-SPEED TUNABILITY IN VACNT'S RPM-ENABLED WAVEGUIDES

Building on the DMA results, we next demonstrate how the constitutive memory of VACNT foams can be leveraged to program mechanical wave transport. As shown earlier, an imposed static precompression (ϵ_s) establishes a stable, nonvolatile constitutive state of the material, effectively serving as a stored mechanical memory. Whereas the dynamic modulus (E_d) acts as a functional readout of that state through its dependence on both the precompression (ϵ_s) and the dynamic strain amplitude (ϵ_d) [Eq. (5) herein and Fig. S12 in SM [30]]. Because the dynamic modulus (E_d) governs the medium's dispersion relation [68,69] and thereby its effective wave speed, this memory-dependent tunability in VACNT foams provides effective means to tailor wave propagation in VACNT-enabled waveguides (see details in SM [30]). Motivated by this, we leverage this precompression and the amplitude-dependent tunability of dynamic modulus, to realize programmable control of wave speed (group velocity) in VACNT-enabled periodic waveguides, directly linking the constitutive memory to adaptive stress-wave transport.

We design a periodic waveguide composed of VACNT foams alternately arranged with rigid aluminum interlayers to experimentally demonstrate tailored wave speed by leveraging the independent dual tunability of the VACNT's dynamic modulus as a function of dynamic strain amplitude (ϵ_d) and static strain (ϵ_s). The propagation speed of elastic waves in periodically layered structures is governed by the dispersion relation, which depends on the constitutive and geometric properties of the layers [70]. Nonlinearity in the constitutive response of VACNT layers can modify the dispersion relation, causing the group velocity to change with

amplitude [71,72]. To isolate this effect, we employ planar, repeatable impact inputs with consistent pulse width across amplitudes thereby minimizing waveform-dependent variations (see SM [30]). Such periodic waveguides featuring amplitude-dependent wave speeds can enable phenomena including wave delaying, focusing, defocusing, selective amplitude blocking in acoustic lensing [41,73], and amplitude-selective filtering [74,75].

Figure 6(a) illustrates the experimental setup, featuring the VACNT-aluminum waveguide housed inside a bespoke PTFE (polytetrafluoroethylene) sleeve. A screw-nut mechanism is mounted at one end to apply static precompression, while the other end features an impact anvil used to excite wave pulses. Using a gas gun setup [Fig. S5(a) [30]], we accelerated an aluminum striker to impact the anvil at various velocities (v_i), generating traveling stress pulses with different amplitudes but similar frequency content (Appendix E). We measured the evolution of the traveling pulse using two strain gauges mounted on aluminum cylinders separated by a known distance ΔL (Appendix E). The effective pulse speed (v_s), calculated by dividing ΔL by the time interval Δt measured from the strain gauge signals [Fig. S5(b) [30]], is plotted in Fig. 6(b) as a function of v_i for various levels of the static precompressive strain applied to the layered system (ϵ_s). Increasing ϵ_s stiffens the VACNT samples, raising the baseline wave speed. Because the incident pulses remained within the elastic regime of the aluminum interlayers, increasing v_i led to a proportional increase in the incident force amplitude to the system. Accordingly, increasing v_i subjects the VACNTs to higher dynamic strain (ϵ_d), causing softening and thereby decreasing the pulse speed. The dependence of pulse speed on both dynamic strain and static precompression follows trends similar to those observed in the dynamic modulus measurements [Fig. 3(e)]. To further validate these observations, we used the DDSSF model to estimate the group velocity, whose magnitude and trends closely match the experimental results [see details in SM and Fig. S9(d) [30]].

Notably, such wave-speed tunability is not achievable in typical viscoelastic materials due to their characteristic hyperelastic nonlinearity, which tends to increase wave speed at higher strain amplitudes [76]. Even nominally softening viscoelastic systems can exhibit apparent strain-rate-dependent stiffening with increasing impact velocity, which can counteract any softening effects that may be present. Additionally, their inherent frequency dependence introduces additional dispersion and fading memory, making robust wave-speed regulation extremely challenging. Similarly, pure torsional waves in granular chains and nonlinear phononic systems that exhibit amplitude- and precompression-dependent wave speeds, arising from nonlinear mesoscale contact mechanics, involve time-dependent effects and irreversible hysteresis, especially in strongly nonlinear large amplitude regimes [41,77].

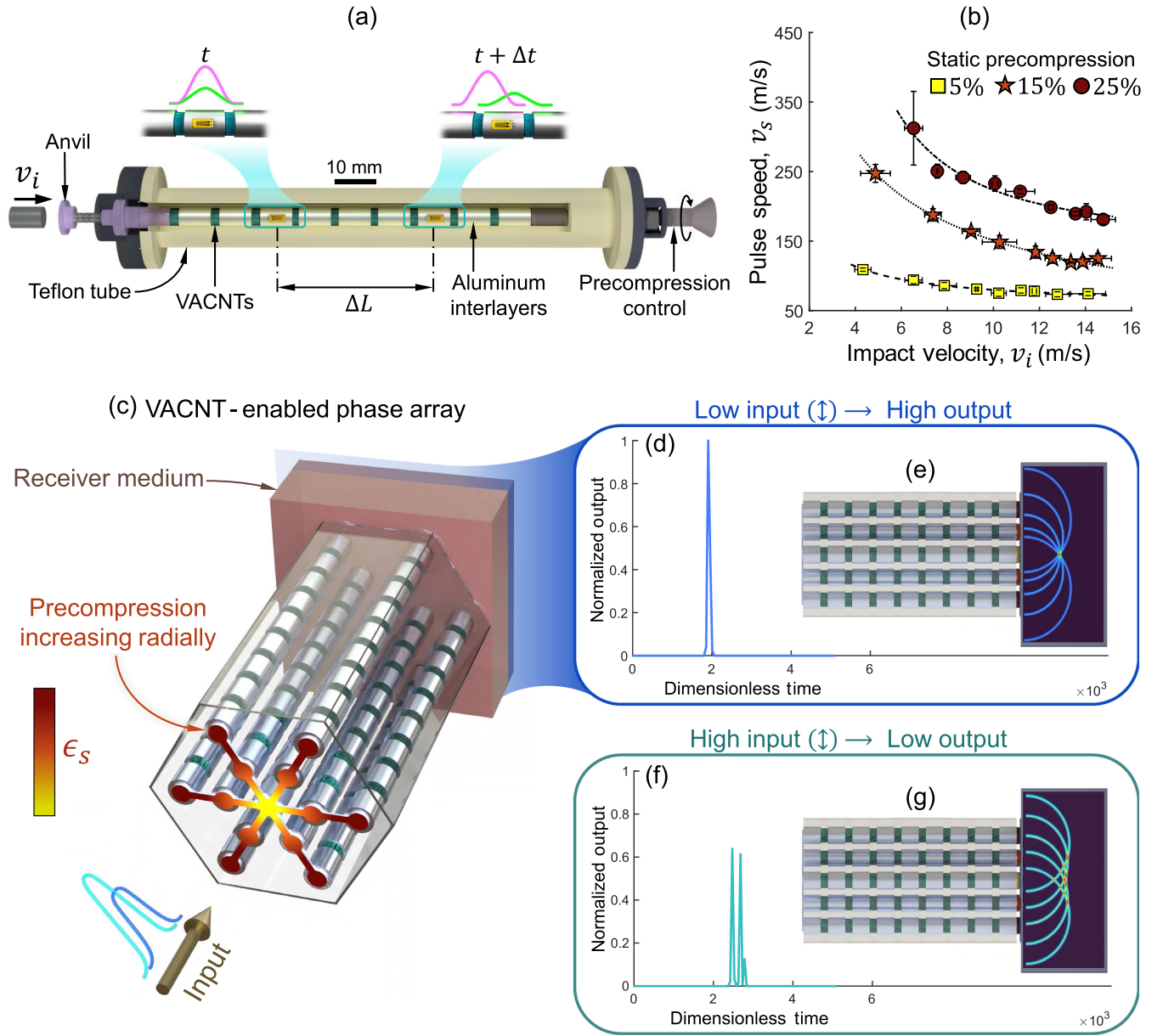


FIG. 6. Wave modulation using RPM-enabled waveguides. (a) Illustration of the experimental setup showing a VACNT-aluminum layered waveguide impacted by a projectile to generate traveling stress pulses of varying amplitude. (b) Experimentally measured speed of the traveling stress pulse plotted as a function of impact velocity (v_i) for various static precompression levels. The error bars represent the standard deviation from three experimental repetitions. The dotted lines represent an approximate analytical fit [see Eq. (61) in SM [30]], which is used for phased array calculations. (c) Conceptual illustration of a hexagonal phased array constructed with VACNT-aluminum metastructure with radially increasing precompression. (d),(e) Wave coalescing occurring within the linear elastic receiver medium at low input design amplitudes. (f),(g) The coalescing effect diminishes as the amplitude increases. In (d) and (f), the x axis is normalized by 0.98 ms and the y axis is normalized by 18.22 MPa—the peak output amplitude for the low input case (see details in SM [30]).

Moreover, the torsional wave tunability in granular systems depends on coupled Hertzian contact mechanisms—precompression-dependent stiffening from normal contact nonlinearity and amplitude-dependent softening from quadratic moment-angle behavior—introducing biaxial coupling that complicates modeling and control [77]. In contrast, our system derives both tunabilities from a single normal compression constitutive response, enabling a unified and

physically elegant framework for wave control. Beyond this, VACNT foams uniquely enable memory-controlled wave speeds, where identical amplitudes at fixed precompression can access distinct tangent moduli through loading-unloading path encoding, an effect inaccessible in viscoelastic or plastic systems due to relaxation and irreversibility. Importantly, VACNT foams' absence of stress relaxation and reproducible return to precompressed states ensure

precise and repeatable wave-speed tunability under repeated excitations. These robust functionalities potentially unlock a unique class of metamaterials based on VACNT-enabled elastic waveguides and modulators, suited for various vibration control, shock mitigation, and analog computing applications [78].

VI. DEVICE CONCEPT FOR PROGRAMMABLE AMPLITUDE-SELECTIVE WAVE MODULATION

The predictive control of elastic wave speed enables wave front shaping by fine-tuning phase gradients in acoustic devices and other engineered wave systems [79]. With embedded amplitude-sensitive layers, these gradients can be made to self-regulate based on the amplitude of the incoming pulse. This capability facilitates the design of wave limiters for protective systems that neutralize high-amplitude loads, as well as analog matched filters that identify target waveforms without the need for costly digital computation. To illustrate this concept, using the experimentally measured wave propagation characteristics, we present an analytically computed phased array model [Figs. 6(c)–6(g)] composed of individual VACNT-aluminum layered systems arranged in a radially symmetric pattern with uniform spacing [Fig. 6(c)]. Here, static precompression is applied in a radially symmetric manner, increasing with distance from the center, consistent with the symmetry of the phased array. As a result, an excitation pulse of a given amplitude incident uniformly across the entire phase array travels slowest at the center array and progressively faster in the outer arrays, thereby establishing a radial phase gradient across the array [Fig. 6(c)].

In Fig. 6(b), the individual curves corresponding to each precompression level are more widely spaced at low v_i and converge as v_i increases, indicating that the phase gradient is sensitive to incident wave amplitude. To demonstrate this, we applied a uniform incident pulse to each array from one end [Fig. 6(c)], allowing the pulses to propagate through the array and transmit elastic waves into a receiver medium placed at the opposite end [80]. Because of the higher precompression in the outermost arrays, those pulses arrive first, while the center array delivers its pulse last. A low-amplitude incidence preserves a larger speed contrast between the arrays, causing the waves generated in the receiver medium to coalesce and form a sharp pressure peak at the focal point $\mathcal{F}$ [Figs. 6(d) and 6(e)]. In contrast, a high-amplitude incidence reduces the speed contrast, disrupting the wave focusing and resulting in an attenuated pressure output at the same location [Figs. 6(f) and 6(g)]. This system functions as an amplitude-dependent wave limiter: Only a preselected low incident amplitude results in constructive coalescence, while higher amplitudes lose synchrony, spread out over time, and emerge significantly attenuated. By tuning the chains to coalesce at this low amplitude (see details in SM [30]), the system can suppress

a broad range of higher-amplitude incident waves—enabling smart, protective shock-absorbing liners that redistribute hazardous impact energy both spatially and temporally above a specified threshold. This novel engineered material design exploiting VACNT's RPM should be understood in contrast to conventional wave limiters that operate in linear regime and the nonlinear systems that undergo plasticity and permanent damage under higher intensity loads [74,75]. The robust tunability of the proposed periodic waveguide and phased array system arises from the constitutive memory of VACNT foams: the RPM generated by deformation-dependent stick-slip friction, which resets each layer to its original stress state after every pulse—unlike viscoelastic polymers whose stresses gradually fade (memory loss).

These systems also have the potential for wave-based mechanical analog computing such as matched filtering, where the goal is to maximize the signal-to-noise ratio by cross-correlating an incoming waveform with a time-reversed template [81]. The phased array can be configured to produce a sharp peak only when the incident pulse matches a target amplitude profile. A specific wave-speed distribution can be achieved by tailoring various design parameters, and with further adjustment of the receiver medium, a desired focal point can be realized (see details in SM [30]). The result is a passive, mechanical matched filter that amplifies the desired waveform while flattening and attenuating all others [82].

In essence, these programmable mechanical wave transport systems can unlock mechanical signal processing and computation by mechanically encoding and manipulating information directly through amplitude- and precompression-dependent wave propagation, rather than through electronic logic. In this way, operations such as thresholding, filtering, gating, and analog pattern mapping, that are normally implemented using data- and power-intensive digital electronics, can be realized intrinsically in the material via deterministic, rate-independent constitutive memory, allowing real-time computation without active control, sensing, or external power. Furthermore, such VACNT-enabled functional devices can potentially be incorporated into physical adjoint optimization schemes [83] by adopting appropriate state-navigation protocols, despite the inherently hysteretic and path-dependent response of VACNT foams. In contrast, achieving such integration is challenging in conventional hysteretic materials, where additional time-dependent effects (viscoelasticity) or irreversible deformation (plasticity) prevent deterministic access to and reuse of constitutive states.

VII. DISCUSSION AND CONCLUSION

We demonstrated, for the first time, constitutive return point memory in an elastically recoverable VACNT foam. In contrast to contemporary geometric bistability-based memory

in metamaterials, RPM is the material's ability to remember deformation history in the stress-strain hysteresis—analogue to magnetic RPM. Unlike other RPM-like phenomena, the constitutive RPM of VACNT foams is intrinsic and stable: It does not drift with material aging as in unfolded crumpled sheets [13], does not rely on amplitude-specific training to encode the memory of a configurational state as in amorphous solids [28,29], and is not coupled to irreversible plastic deformation as in elastoplastic systems [27]. We developed a novel frictional damping model that reveals constitutive RPM originates from the memory of loading history encoded in nanoscale stick-slip frictional interactions. We showed that the interplay between RPM and frictional interactions impart dual tunability to the VACNT's dynamic modulus—enabling stiffening via precompression and softening via dynamic strain.

We leverage the tunable dynamic modulus to experimentally demonstrate stress-wave propagation in a periodic VACNT array waveguide with rigid interlayers, with wave speed independently adjustable by dynamic input amplitude and precompression—a capability uniquely enabled by the robust RPM effects in VACNTs. Arranging these individual waveguides in radial symmetry creates a VACNT-enabled phased array, where wave-speed tunability is used to achieve phase contrast in wave propagation. This allows individual stress waves to focus at a certain input amplitude while diffusing and attenuating at higher amplitudes, enabling amplitude-dependent shock limiters.

Additionally, the fatigue resistance [31,32] and thermal stability [33], together with their superamphiphobic nature [84], render RPM in VACNTs resilient and reliable under repeated loading and extreme environments [85,86]. These attributes position VACNTs as an enabling material for next-generation adaptive shock absorbers, ultrasound imaging, vibration isolation, analog signal processing, and a wide range of real-time analog computing applications.

Because of limitations imposed by viscoelastic fading memory effects and plastic deformation, the unique properties of VACNTs cannot easily be replicated in traditional material systems. However, fundamental insights from the DDSSF model suggest that similar behavior can be achieved in nanostructured materials whose dissipative response is governed by nanoscale friction rather than viscoelasticity—including boron nitride nanotubes, fibrous biological materials, and nanoarchitected materials with engineered frictional dissipation.

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A. G. conducted the quasistatic compression, stress relaxation, and DMA experiments on VACNT foams. A. G. and B. M. carried out the wave propagation experiments and developed the frictional damping model. B. M. performed the analysis for phased array system and wave coalescence. N. J. conducted preliminary quasistatic compression and stress-relaxation experiments. K. C. synthesized the VACNT samples. A. G., B. M., and R. T. analyzed the results and drafted the manuscript, with input from all authors. A. G. and B. M. contributed equally. R. T. supervised the study.

The authors declare no competing interest.

DATA AVAILABILITY

The data and code that support the findings of this article are openly available and their access links are provided in the manuscript.

APPENDIX A: CVD SYNTHESIS

We synthesize VACNT foams using a floating catalyst thermal chemical vapor deposition process, with toluene as the carbon source and ferrocene as the catalyst precursor. Our tCVD setup consists of a 1100-mm-long fused quartz glass tube (Technical Glass Products 50 × 55) enclosed with custom-made sanitary fittings attached at both ends. For synthesis, the glass tube is placed inside a tube furnace (Carbolite Gero CTF 12/75/700) maintained at a temperature of 1100 K. A 20 × 20-mm *p*-type silicon wafer substrate is positioned in the furnace's peak heating zone (12 in. from the inlet) to facilitate CNT growth. Steel fittings are then attached at both ends, and argon gas is flowed at 760 sccm from the inlet to create an inert atmosphere inside the tube at 1 atm pressure. The gas exits through a glass bubbler connected at the outlet. Once the air inside the enclosure is fully replaced with argon, hydrogen gas is introduced at 40 sccm, making the carrier gas mixture 5% hydrogen and 95% argon. Hydrogen enhances catalyst activity by reducing amorphous carbon formation, resulting in purer CNTs. The flow of the carrier gas mixture is regulated using individual mass flow controllers (MKS G-series GE50 A). To initiate the synthesis, a feedstock solution of 0.8 g ferrocene in 80 ml toluene ($[w/v] = 0.01$ g/ml) is supplied at 0.8 ml/min using a glass syringe and syringe pump (NE-1000) connected to the inlet. The CNTs grow vertically aligned on the substrate, reaching a height of approximately 1.5–3 mm. Once the feedstock solution is depleted, the furnace is switched off, and the hydrogen flow is stopped while argon continues to flow, allowing the furnace to cool down to approximately 700 K. At this temperature, the substrate with a 1–3-mm-thick VACNT film is retrieved and prepared for mechanical characterization. The film is separated and cut into smaller pieces:

A biopsy punch is used to cut circular samples, and a microtome blade is used for square samples. The density of the samples typically ranges between 0.13 and 0.26 g/cm³.

APPENDIX B: UNIAXIAL QUASISTATIC COMPRESSION

We conducted quasistatic compression and stress-relaxation experiments using a commercial load frame, an Instron Electropulse E3000, equipped with a 250 N load cell. From a CVD-synthesized VACNT film measuring $\sim 20 \times 20$ and 1.59 mm in thickness, we cut a square cross-section sample with dimensions of 5.09×5.08 mm. First, we preconditioned the pristine sample by applying cyclic ramp loading at a strain rate of 0.01 s^{-1} for 5 cycles, recording both the loading and unloading responses. On the preconditioned sample, exhibiting stable stress-strain behavior, we performed cyclic quasistatic compression at various strain rates.

APPENDIX C: STRESS-RELAXATION EXPERIMENTS

For consistent comparison, we performed the stress-relaxation experiments on the same sample used for quasistatic characterization. To measure stress relaxation for various values of compressive strain, we applied ramp strain loading at a strain rate of 0.05 s^{-1} , which was chosen because it is slow enough to reduce overshoot in strain due to the load frame's inertia but fast enough to quickly reach the desired strain level. We applied the strain up to the desired compression strain value and held it constant for 30 min. Since the majority of stress relaxation in viscoelastic materials occurs within the first 20–30 min, a duration of 30 min is sufficient to ascertain the occurrence of any relaxation. Additionally, since we found that VACNT foams do not exhibit any relaxation, extending the relaxation period will not provide any new information.

APPENDIX D: DYNAMIC MECHANICAL ANALYSIS

We conducted broadband DMA experiments using a custom-built dynamic mechanical analyzer, consisting of a piezoelectric actuator (Physik Instrumente P841.10) and a dynamic force sensor (PCB Piezotronics 208C01). The actuator's motion is controlled by a position servo control module and an amplifier (Physik Instrumente E509 and E505, respectively). The actuator is attached to a fixed steel block, while the force sensor is mounted on a linearly displaceable steel block facing the actuator. To perform DMA measurements, the preconditioned sample is first attached to the actuator tip and then precompressed to a desired strain between the actuator and the force sensor by adjusting the force sensor's block using a micrometer head. Once the sample is precompressed, we apply 20 cycles of sinusoidal strain with the desired amplitude and frequency

by supplying a voltage waveform to the actuator using an analog output device (National Instruments, NI-9269). The force sensor records the force output via a signal conditioner (PCB 482C15) and an analog input device (NI 9215). After collecting the 20-cycle signal, the first 5 and last 5 cycles are removed, and the average of the middle 10 cycles is taken to obtain the hysteresis response. From this hysteresis response, the dynamic modulus and loss factor are measured through amplitude and frequency sweeps.

While our custom DMA setup enables high-frequency measurements up to $f = 1 \text{ kHz}$, the available amplitude range decreases with frequency, being limited to $7.6 \text{ }\mu\text{m}$ for $f < 100 \text{ Hz}$ and reducing to $2 \text{ }\mu\text{m}$ as the frequency approaches 1 kHz . However, the frequency-independent behavior of VACNTs allows us to measure the dynamic modulus and loss factor as functions of amplitude at low frequencies, with the results applicable to all frequencies. Thus, we conducted amplitude sweep experiments at $f = 10 \text{ Hz}$ for amplitudes up to $7.6 \text{ }\mu\text{m}$. For further higher amplitudes, up to $20 \text{ }\mu\text{m}$, we used the Instron ElectroPulse E3000 load frame, as described above, at a frequency of 0.02 Hz , leveraging the frequency-independent behavior.

APPENDIX E: WAVE PROPAGATION EXPERIMENT

We constructed the periodic waveguide using 17 VACNT foam samples, each with an average thickness of 2.06 mm and a diameter of 5 mm , alternately layered with 17 aluminum cylinders measuring 8 mm in length and 6 mm in diameter [Fig. S5(a) [30]]. We selected VACNT samples with similar densities and stress-strain responses to ensure uniform precompression across all layers. Before assembling the waveguide, we preconditioned each foam sample up to 50% strain. We used an aluminum cylinder of identical dimensions as a projectile, launching it with a custom-built gas gun powered by an electric air compressor. The projectile's design produced a wavelength comparable to the structural periodicity, placing wave propagation in the short-wavelength regime where dispersion dominates. By adjusting the compressor pressure, we controlled the projectile's velocity. Notably, increasing the amplitude of the incident stress pulse did not significantly alter its frequency content, allowing us to isolate amplitude effects on wave propagation for signals with similar spectral characteristics.

To capture the time evolution of the traveling stress pulse, we mounted strain gauges (Kyowa KSPB-1-350-E4) on the sixth and 14th aluminum interlayers. These gauges were configured in a quarter-bridge circuit and balanced using individual signal conditioning amplifiers (Vishay 2310B). Signals were recorded using an NI cDAQ-9174 chassis and an analog input module (NI 9215), operated through MATLAB's data acquisition toolbox. We measured the projectile speed (v_i) just before impact using a PASCO wireless smart gate (PS-3225). To calculate the effective

wave speed (v_s), we divided the known separation distance between the strain gauges—measured after applying pre-compression—by the time delay [Fig. S5(b) [30]] between the strain gauge signals—measured using the FWHM middle-point method. For each precompression level, we repeated the experiment for every input velocity 3 times to test repeatability.

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