

Thermal fluctuations, stress relaxation, and actuation in carbon nanotube networks

S. V. Ahir and E. M. Terentjev

Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom

S. X. Lu and B. Panchapakesan

Department of Electrical and Computer Engineering, University of Delaware, Newark, Delaware 19716, USA

(Received 15 June 2007; revised manuscript received 24 August 2007; published 30 October 2007)

Do carbon nanotubes behave like polymers? The answer appears to be—some do and some do not, depending on the number of walls. Analyzing the behavior of carbon nanotube networks, as found in sheets of “bucky paper,” provides an intriguing insight into the characteristics of nanotubes, noninvasively deducing the fundamental response of individual tubes from the average characteristics of the collective. We report stress relaxation experiments on single- and multiwalled carbon nanotube networks and also present their reversible photomechanical actuation response. Experimental similarities between multiwalled nanotube networks and a “sticky” granular system are observed, while single-walled tubes display entropic behavior akin to a polymer network. In both cases, photostimulated actuation is orders of magnitude larger than thermal expansion predictions. The analogy between single-walled tubes and entropically driven polymer chains, and between multiwalled tubes and granular networks, suggests a paradigm for theoretical and experimental analysis.

DOI: [10.1103/PhysRevB.76.165437](https://doi.org/10.1103/PhysRevB.76.165437)

PACS number(s): 61.46.-w, 62.20.-x, 81.07.De

I. INTRODUCTION

The way in which links of an entangled network interact with each other is not a fully understood problem in the natural sciences. While much success has been gained from studying polymeric systems and treating them as thermally diffuse statistical objects, understanding of granular media is still in its infancy.¹ Whether carbon nanotubes should be treated as thermal statistical objects exploring their conformational space or as completely nonthermal overconstrained systems analogous to granular matter has, until now, remained unclear. Carbon (in graphite, graphene, and nanotube form) is unique because of the stability of the delocalized π system coupled with the σ -bonded honeycomb lattice. The thermal- and photoactuation response of such systems is still poorly understood but improved mastery could reap great technological benefit in areas as diverse as biotechnology² and optical telecommunications.^{3,4}

Large photomechanical actuation of polymer–multiwalled nanotube (MWNT) composites has been recently demonstrated.^{6–8} Evidence suggests that the degree of tube alignment within the polymer matrix directly influences the magnitude and the direction of actuation of nanocomposites and that the mechanism of actuation is due to the contraction of locally aligned polymer chains around the tubes, causing them to buckle.⁹ The focus of our present work is on pure nanotubes, how they interact with each other, and how this interaction changes with temperature or under light. Zhang and Iijima first demonstrated that single-walled nanotube (SWNT) fibers can bend under light.⁵ Only a few studies have been reported regarding the thermal expansion of nanotubes.^{10–13} There are a number of theoretical ideas on possible nanotube-light interaction mechanisms^{14–17} and nanotube thermal expansion,^{18–21} some of which contain conflicting predictions regarding whether nanotubes expand or contract in their long (z) axis when heated.

Tube networks such as nanotube films, commonly known as “bucky paper,”²² offer a platform on which to test nano-

tube behavior under stress, heat, and light, Fig. 1. The fundamental question we are asking is whether the tubes perform as elastic (or plastic) rods, or are they able to explore their conformational space like thermally fluctuating polymer chains. The secondary question is about the nature of linkages in nanotube networks. A few important studies have appeared in the literature but have not yet answered these primary questions.^{23,24} One should not confuse this issue with the volume of successful literature describing the mechanical response of nanotubes, such as their static Young’s modulus.^{25–27} There remains very little literature exploring the dynamic-mechanical properties of nanotube networks either under stress or when heat or light stimulus is applied. To this end, we present extensive experimental data on the mechanical relaxation of SWNT and MWNT films when a relatively large stress is applied. We find that all nanotube networks show very strong stress relaxation, which may raise questions about ever reaching the equilibrium in overconstrained systems and also the values of elastic moduli reported in the literature. There is, however, a fundamental difference between MWNT and SWNT networks: the mechanical response of the former is totally independent of temperature, suggesting a granularlike viscoelasticity. Con-

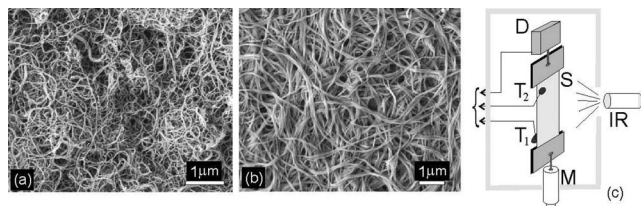


FIG. 1. Networks of carbon nanotubes. (a) SEM image of MWNT mat. (b) SWNT mat, under similar magnification, highlighting the tube bundles. (c) Scheme of the experiment, which controls the temperature (T_1, T_2) and strain via a micrometer (M) and measures the exerted force via a dynamometer (D), in ambient relaxation or on irradiation.

versely, SWNT networks reveal an effective rubber modulus that is linearly dependent on temperature, indicating the dominant role of entropic elasticity. Additionally, photomechanical experiments were carried out on MWNT/SWNT films. The actuation response is orders of magnitude larger than predicted by leading theories describing the local properties of the lattice. Again, we find a dramatic contrast: stretched SWNT mats contract, when heated or irradiated by light, further cementing the notion that networks of SWNTs could, in fact, be treated as cross-linked polymers. In contrast, the stretched MWNT networks further expand under the same conditions of irradiation. The actuation mechanism in both warrants much further investigation. The results have potentially widespread consequences for many researching carbon nanotubes, granular systems, and polymer physics.

II. METHODS

A. Preparation

Both SWNT and MWNTs were purchased from Nanolab, Inc. SWNT diameter is 1–1.5 nm, length $>10\ \mu\text{m}$, and purity $>90\%$. MWNT diameter is 15–45 nm, length is 5–20 μm , and purity $>95\%$. The fabrication of both SWNT and MWNT films follows the same procedures.

Tubes were first dispersed in isopropyl alcohol to form a uniform $\sim 0.1\ \text{mg/ml}$ suspension after 30 h ultrasonication at approximately 30 W power. At such a low power, we expect no significant tube breakdown to occur and, indeed, extensive scanning electron microscopy (SEM) imaging found no change in tube length after sonication. The nanotube suspensions were then vacuum filtered through a mixed cellulose ester filter to produce carbon nanotube films. The thickness of the films was controlled by the amount of solution used in filtration. The resulting sheets were rinsed with isopropyl alcohol and de-ionized water and then dried at $75\ ^\circ\text{C}$ for 2 h to remove the remaining solvents, after which the sheets were peeled directly off the filter to leave free-standing carbon nanotube films, Figs. 1(a) and 1(b).

Although the main results below are reported for the films sonicated for 30 h, we also made samples with only 2 h and over 2 week sonication. There was a visible decrease in the average SWNT bundle size with the length mixing, but the photoactuation response showed no difference at all.

B. Testing procedure

Experiments were conducted with the dynamometer (Pioden Controls Ltd.) housed in a custom made thermal-control box. Distance between the clamps was controlled using a micrometer with $\pm 0.001\ \text{mm}$ accuracy. The rig was calibrated with weights to give a direct measure of stress and (fixed) strain. When required, the cold light source (Schott KL1500 LCD) was positioned $\sim 20\ \text{mm}$ from the sample. The peak power density, at 675 nm and this distance to the sample surface, was determined as $1.5\ \text{mW/cm}^2$, cf. Fig. 1(c).

To standardize the results across all samples, preliminary checks were undertaken to accurately find the zero-strain natural length L_0 of each sample, for every experimental run.

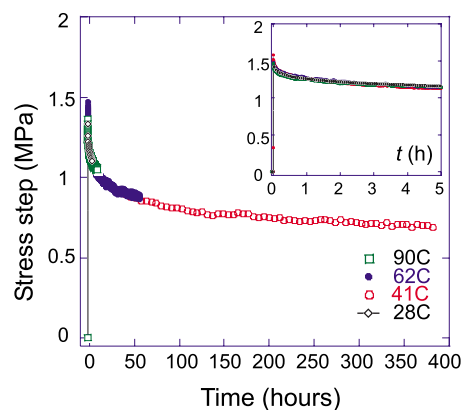


FIG. 2. (Color online) Stress relaxation of a MWNT mat kept at fixed length, after a step strain of 0.2%. Different curves obtained in the same experiment at 28, 41, 60, and $90\ ^\circ\text{C}$ are color coded. The inset shows the relaxation during the first 5 h.

The imposed fixed extensional strain was then calculated as $\varepsilon = (L - L_0)/L_0$, with L provided by the micrometer reading. After such a fixed prestrain was applied to each sample, readings of stress were recorded to verify that the material was equilibrated in its prestrained state, and then the stimulus (additional strain, heat, or light, depending on the type of experiment undertaken) was applied. Data were continuously logged via a personal computer acquisition card with high temporal resolution.

III. STRESS RELAXATION

First of all, let us examine stress relaxation of nanotube mats, after a fixed step strain is applied, cf. Fig. 1(c); this is a classical isostrain experiment in viscoelastic medium. Figure 2 shows the details of very long (375 h) relaxation of stress in the MWNT film, after a step of 0.2% extensional strain is applied at $t=0$. A series of identical experiments for a range of temperatures is also presented in Fig. 2. The inset magnifies the evolution for the first 5 h. The curves are remarkably identical for the range of temperatures tested, implying that the magnitude and mechanism of stress relaxation are indistinguishable. Importantly, we observe that if the relaxation is stopped at any time and a fresh step strain is imposed, the response modulus reproduces the original high value, so we are not looking at the slow degradation or disentanglement of the tube network. The relaxing stress does not reach an expected equilibrium plateau but instead appears to relax indefinitely. This is akin to natural rubber which often demonstrates indefinite relaxation, for instance, due to the sliding cross-links.²⁸ This also suggests that in many experiments on carbon nanotubes, the mechanical equilibrium might be a deceptive target; this may be a simple explanation of a vast variation of values for the Young modulus reported in the literature. The values of (relaxing) Young's modulus are shown in Fig. 3 and it is clear that depending on the rate of applied deformations, one could obtain a very different response. Our results suggest that researchers have not yet fully recognized the time dependent (viscoelastic) characteristic of the stress relaxation.

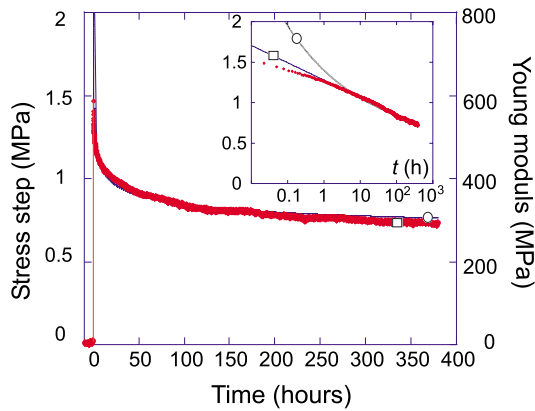


FIG. 3. (Color online) The analysis of the long relaxation in a MWNT mat carried out at 41 °C. The data are fitted by the power law $\Delta\sigma(\text{MPa})=0.5+0.9t^{-0.2}$ and a logarithmic decay $\Delta\sigma=1.3-0.2\ln(t)$. The inset plots the same data on a $\ln t$ axis, indicating that logarithmic relaxation (labeled by \square) is a much closer fit than the power law (labeled by \circ).

The detailed data analysis in Fig. 3 suggests a very slow but remarkably large amplitude of stress relaxation. The inset illustrates the same data plotted on the logarithmic time axis, which highlights how the best power-law fit deviates from the data more significantly, while the logarithmic relaxation given by $\Delta\sigma(\text{MPa})\approx 1.3-0.2\ln t$ fits the experimental results almost perfectly after the first ~ 1 h of relaxation. Such slow dynamics is very rare in physics and resembles the finding in overconstrained randomly quenched systems. It is found, for instance, in the relaxation of the angle of repose in a sandpile²⁹ or in polydomain nematic elastomers.³⁰ In each case, it is the network of quenched mechanical constraints that leads to the exponential increase in the activation barrier as the equilibrium approaches and a logarithmic relaxation as a result.

The MWNT mat has many tubes in contact with each other at multiple junctions along their contour length. In the junction regions, pristine nanotubes attract via short-range intertube van der Waals potentials, enhanced over the typical granular or colloid levels due to the high polarizability of graphene. The primary van der Waals minimum occurs at short distances (<2 nm) with a depth of $\sim 40k_B T$ for SWNTs³¹ and up to an order of magnitude higher in MWNTs. This attraction quickly tends to zero at a distance ≥ 2.5 nm.^{31,32} This confirms the basic mechanism of network binding in nanotube mats, as well as the fact that the tube strands between junctions are relatively interaction-free. The large nonthermal stress relaxation suggests that junctions are sliding under stress. It is possible to estimate the activation energy of this sliding, if the final equilibrium stress level could be unambiguously determined (which we could not do even after 375 h). This nonthermal nature of isolated MWNT segments between junctions makes it conceptually difficult to talk about, for instance, the tube persistence length ℓ_p , cf. Fig. 1(a), which is a very common notion in literature. Persistence length has a clear statistical meaning in fluctuating polymer chains or filaments,^{33,34} however, one would not use this parameter to describe, for instance, a crumbled ball of copper wire or a force network in a granular pile. Indeed, in

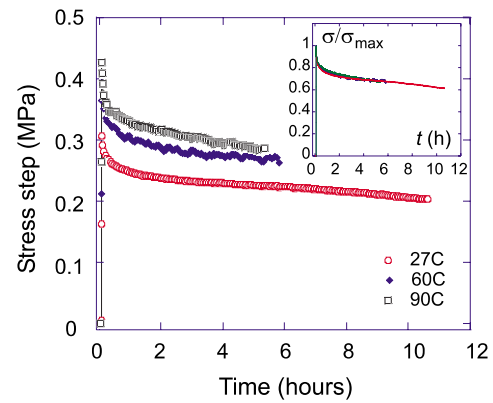


FIG. 4. (Color online) Stress relaxation of a SWNT mat kept at fixed length, after a step strain of 0.2%. The jump in stress is different at each temperature. However, the inset displays the normalized data $\Delta\sigma/\Delta\sigma_{\text{max}}$, indicating the universal relaxation mechanism, not altered with temperature.

its dynamic-mechanical response, the MWNT mat is close to a network of elastic rods or the sticky granular system, still poorly understood³⁵ but a good candidate for experimental studies of the glass and jamming transitions. These ideas are consistent with the notion of rotating bonds between nanotubes suggested by Hough *et al.*³⁶ Whitten *et al.*³⁷ also raise the idea of intertube junctions, in their case able to slip in solution.

We now discuss the response of SWNT mat in an identical step-strain experiment at different temperatures. Figure 4 provides the results of such an experiment. These results are in marked contrast with the MWNT response. At any stage of relaxation, the stress in a stretched SWNT network increases as the temperature is increased. In Fig. 5, the Young modulus is plotted as a function of temperature (in all cases, isostrain conditions are maintained). Each line in the graph represents the linear fit to a set of stress measurement taken at a particular time of relaxation. The classical feature of rubber elasticity is clearly reproduced here, with the effective Young modulus $Y\propto k_B T$. This result implies the entropic nature of SWNT network: for SWNTs, unlike MWNTs, thermal fluctuations are, in fact, significant. We now can talk

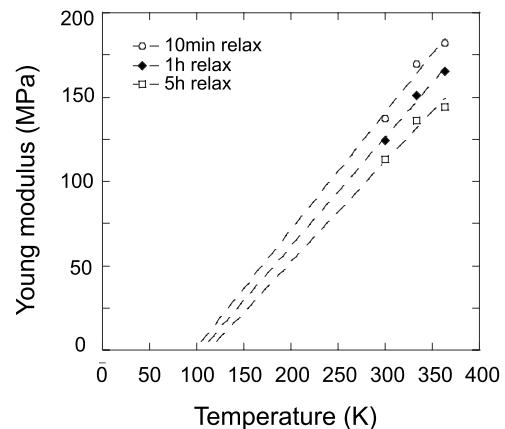


FIG. 5. The variation of Young's modulus in a SWNT mat with temperature measured at three separate times of relaxation.

about the statistics of conformations and, in particular, the persistence length of SWNT, ℓ_p . This length can still be very high [in the range 20–70 μm (Ref. 38)], if one considers an ideal nanotube, but in reality, defects contribute to decreasing ℓ_p . One may be tempted to conclude that SWNTs can be treated as thermally diffuse statistical objects, much like polymer chains.

However, this conclusion has to be taken together with the well-established bundled nature of SWNT assemblies, very different from the coiled polymer chains, cf. Fig. 1(b), and the separated nature of clearly nonthermal MWNTs. As single-walled tubes are flexible enough to be thermally excited, they assemble in highly aligned bundles held by van der Waals forces. However, these bundles must be dynamic in the sense that their range of conformations is explored under thermal motion of continuously bonding and debonding flexible tubes: the corresponding entropy would then account for the temperature-dependent modulus (analogous to polymer networks where the coiled chains between junctions explore their conformational freedom). The MWNT strands are much more rigid and not able to bend under thermal excitation so that the structure of their mats, Fig. 1(a), is entirely dependent on preparation history. The large amplitude of stress relaxation and the continuing decrease of the modulus even after a very long time once again demand a careful evaluation of the Young modulus data reported in the literature.

Another key finding points to the difference between entropic polymer and entropic SWNT bundles. The inset in Fig. 4 shows the normalized stress relaxation, rescaled by $\Delta\sigma/\Delta\sigma_{\text{max}}$, helping to clarify the long-time relaxation mechanism of SWNT films. The normalized curves collapse onto each other, suggesting that the mechanism of long stress relaxation in SWNT films is the same regardless of temperature, just like in MWNT films. This is not the case for a cross-linked polymer network where relaxation is a diffusion-controlled process and hence its rate varies with T leading to the famous time-temperature superposition. Non-thermal relaxation in nanotube networks suggests that the main mechanism is different. We believe that it is related to the sliding of junctions between nanotubes, the motion of which is dominated by friction as discussed. The rate of long-time normalized relaxation of stress is much faster in SWNT networks; this is in line with the idea of sliding junction as the binding energy is certainly proportional to the nanotube dimensions.

IV. PHOTOACTUATION

The response of nanotubes to near-IR light is now discussed. The use of a cold light source is an effective means by which to remotely transfer energy to the system quickly. The photoactuation experiments were conducted using an identical method to that outlined in Ref. 39, cf. Fig. 1(c), with the results presented in Figs. 6 and 7.

The first of the two graphs shows the MWNT film response. The significant drop in stress indicates that the sample uniaxially expands its underlying natural length on irradiation. The expansion is fully reversible, as the sample

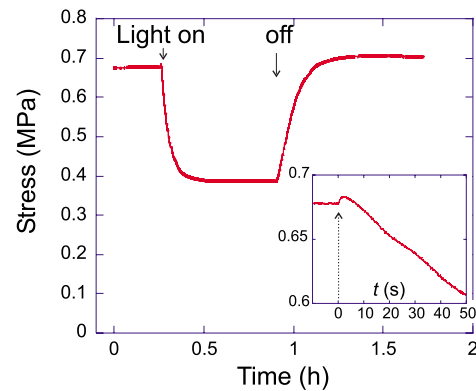


FIG. 6. (Color online) Photomechanical actuation of a MWNT mat recorded at fixed, prestrained, and equilibrated sample length. The inset shows the initial contractive stress response of the film during the first few seconds when the light source is switched on, illustrating the initial peak.

returns to its original stressed state on removing the source of external energy. This is a very important observation, eliminating many possible mechanisms based on, e.g., degradation, induced defects, or enhanced junction sliding, which would all be irreversible. Characteristically, the kinetics of this photomechanical response is very slow, although at least 1–2 orders of magnitude faster than the ambient stress relaxation, Fig. 2. There is a small but significant and reproducible contraction in the initial seconds after the light source is switched on, highlighted in the inset of Fig. 6. Similarly, when the light is switched off, the same magnitude peak (in opposite direction) is observed, which further suggests that this phenomenon is no artifact of experimentation but rather a significant, albeit relatively small, and short-lived event. This feature is recalled when we examine and contrast the response of SWNT film to irradiation, Fig. 7. Clearly, SWNTs contract under IR radiation, leading to the increasing stress on the constrained sample. The effect is also fully reversible and its relatively fast kinetics are illustrated in the inset to Fig. 7.

Separate studies on heating the film using the ambient temperature change on the samples reveal that the actuation

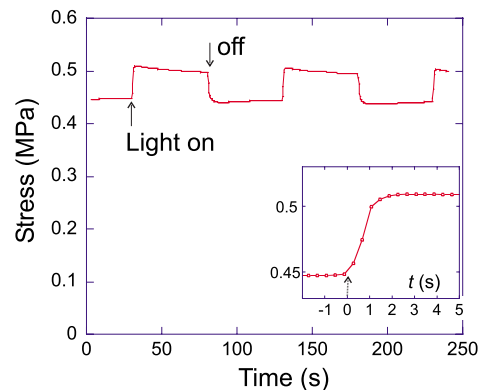


FIG. 7. (Color online) Photomechanical actuation of a SWNT mat, in the same conditions. In contrast to MWNT network, this sample is contracting on illumination. The detailed onset kinetics, highlighted in the inset, matches well the compressed exponential result reported in Ref. 9.

stress is around half that of the stress achieved using light. The comparable responses suggest that the mechanism responsible for nanotube actuation might be thermally based, but the heating efficiency is much higher on infrared photon absorption than through the exchange with atmosphere. However, since the rate of the light-off relaxation is evidently the same as the light-on rate, for both materials, it cannot be purely due to ambient cooling of the samples: clearly, photon absorption has a direct effect on the mechanical state of nanotubes.

One could explain SWNT contraction using a variety of models. Literature is divided as to whether SWNTs expand or contract in their z axis. Depending on the model used, such effects can have some orientational dependence; however, in all cases, the strains induced are extremely small. The first idea that we must consider in view of our earlier findings is the effect of rubber band contraction on heating, which is due to the increasing weight of conformational entropy. As this is a significant factor in the description of single-walled tubes and their bundles, one expects as in classical thermodynamics that $(\partial f/\partial T)_x = (\partial S/\partial x)_T$, with x the stretching and f the corresponding force. This basic consequence of entropic elasticity is almost completely independent of what actual graphene lattice does microscopically.

Anharmonicity of interatomic potentials causes thermal expansion or contraction in most lattice systems.⁴⁰ It is dependent on the balance between phonon modes and respective Grüneisen parameters.¹⁹ Axial thermal contraction of an individual (10,10) nanotube at room temperature has been shown in simulations.²⁰ The x-ray scattering experiments by Maniwa *et al.*¹² register an average lattice constant expansion of $\sim 0.75 \times 10^{-5} \text{ K}^{-1}$. For our *bulk* system, using these values, we cannot explain our actuation strains of $\sim 0.1\%$ by solely lattice potential effects, unless the extremely high local heating is assumed on photon absorption (the mean temperature change across the sample was only 20°). The magnitude of contraction seen in Fig. 7 is more than an order of magnitude greater than predicted by thermal expansion ideas. A definitive measurement of individual nanotube photoresponse would involve irradiating a tube in a setup similar to single-chain AFM studies.⁴¹ The difference between the atomic scale theory (where only short segments of nanotubes are considered) against macroscopic assemblies of nanotubes is highlighted through these results.

V. SUMMARY

Networks of carbon nanotubes may be the first system that exhibits metallic, semiconducting, and polymerlike properties within one material and apparently demonstrate a reversible light-induced actuation. The stress relaxation of both MWNT and SWNT films have been studied over short

and long time scales and at different temperatures. Both nanotube networks continually relax their stress (creep), which we assign to slippage (but not breakage) between nanotube junctions within the entangled network. The MWNT network has an effective modulus that is temperature invariant but has a strong (viscoelastic) time dependence. This nonthermal system is most similar to a transient network of elastic rods or a “sticky” granular network. It might well prove useful as a generic structure to experimentally probe granular matter. Conversely, and surprisingly, the SWNT networks demonstrate a significant entropic response, with the (relaxing) modulus depending linearly with temperature, just like in cross-linked polymer chains. We associate this effect with the dynamic nature of SWNT bundles, where continuous thermal rearranging of tubes provides the conformational entropy.

Nanotube modulus variation with temperature has been theoretically studied but with mixed results in the literature: some papers show a decrease in modulus with increasing temperature,⁴² while others demonstrate no change. Related to our work is the experimental result by Arinstein *et al.*⁴³ on the temperature dependence of modulus obtained from single polymer nanofibers that show the following: (1) the temperature dependence is strongly influenced by fiber diameter and (2) larger diameter fibers display an entropic-dominated response. Our experiments demonstrate that the same results cannot be transferred from polymer nanofibers to CNTs—larger diameter nanotubes do not demonstrate entropic behavior.

Exposure of the MWNT network to near-IR light causes a fully reversible photomechanical expansion, almost four decades larger than what would be expected through simple lattice thermal expansion-contraction arguments. On-off hysteresis is also negligible. Better aligned MWNT systems such as those found in fibers unambiguously demonstrate nanotube contraction in the long (z) axis. In contrast, SWNT networks, upon exposure to IR light, contract in the direction of prestrain. This work reported an indication of entropic-type contraction in such a system. Single-walled carbon nanotube films do behave like cross-linked polymer chains. Should it be possible to cross-link the individual SWNTs chemically, one could very well create a pure SWNT elastomer with some intriguing, fully reversible, properties.

ACKNOWLEDGMENTS

The authors wish to thank Sam Edwards, D. Tománek, J. Gornall, K. Channon, A. Craig, and R. Vaia for many useful discussions. This work was carried out with the support of the EPSRC and a CASE award from Makevale Ltd. at the University of Cambridge, and the NSF Career Award No. ECS 0546328 at the University of Delaware.

- ¹S. R. Nagel, in *Soft and Fragile Matter*, edited by M. E. Cates and M. R. Evans (IOP, University of Reading, Berkshire, 2000).
- ²N. W. S. Kam, M. O'Connell, J. A. Wisdom, and H. J. Dai, *Proc. Natl. Acad. Sci. U.S.A.* **102**, 11600 (2005).
- ³S. Tatsuura *et al.*, *Adv. Mater. (Weinheim, Ger.)* **15**, 534 (2003).
- ⁴A. G. Rozhin, Y. Sakakibara, S. Namiki, M. Tokumoto, H. Kataura, and Y. Achiba, *Appl. Phys. Lett.* **88**, 051118 (2006).
- ⁵Y. Zhang and S. Iijima, *Phys. Rev. Lett.* **82**, 3472 (1999).
- ⁶H. Koerner, G. Price, N. A. Pearce, M. Alexander, and R. A. Vaia, *Nat. Mater.* **3**, 115 (2004).
- ⁷S. V. Ahir and E. M. Terentjev, *Nat. Mater.* **4**, 491 (2005).
- ⁸S. X. Lu and B. Panchapakesan, *Nanotechnology* **16**, 2548 (2005).
- ⁹S. V. Ahir and E. M. Terentjev, *Phys. Rev. Lett.* **96**, 133902 (2006).
- ¹⁰S. Bandow, *Jpn. J. Appl. Phys., Part 2* **36**, L1403 (1997).
- ¹¹Y. Maniwa *et al.*, *Phys. Rev. B* **64**, 241402(R) (2001).
- ¹²Y. Maniwa *et al.*, *Phys. Rev. B* **64**, 073105 (2001).
- ¹³R. B. Pipes and P. Hubert, *Compos. Sci. Technol.* **63**, 1571 (2003).
- ¹⁴M. Verissimo-Alves, R. B. Capaz, B. Koiller, E. Artacho, and H. Chacham, *Phys. Rev. Lett.* **86**, 3372 (2001).
- ¹⁵E. Piegari, V. Cataudella, V. M. Ramaglia, and G. Iadonisi, *Phys. Rev. Lett.* **89**, 049701 (2002).
- ¹⁶V. Perebeinos, J. Tersoff, and P. Avouris, *Phys. Rev. Lett.* **94**, 086802 (2005).
- ¹⁷S. B. Cronin *et al.*, *Phys. Rev. Lett.* **96**, 127403 (2006).
- ¹⁸N. Mounet and N. Marzari, *Phys. Rev. B* **71**, 205214 (2005).
- ¹⁹P. K. Schelling and P. Keblinski, *Phys. Rev. B* **68**, 035425 (2003).
- ²⁰Y. K. Kwon, S. Berber, and D. Tomanek, *Phys. Rev. Lett.* **92**, 015901 (2004).
- ²¹H. Jiang, B. Liu, Y. Huang, and K. C. Hwang, *J. Eng. Mater. Technol.* **126**, 265 (2004).
- ²²R. H. Baughman *et al.*, *Science* **284**, 1340 (1999).
- ²³L. Berhan, Y. B. Yi, A. M. Sastry, E. Munoz, M. Selvidge, and R. Baughman, *J. Appl. Phys.* **95**, 4335 (2004).
- ²⁴U. Vohrer, I. Kolaric, M. H. Haque, S. Roth, and U. Detlaff-Weglikowska, *Carbon* **42**, 1159 (2004).
- ²⁵P. Poncharal, Z. L. Wang, D. Ugarte, and W. A. de Heer, *Science* **283**, 1513 (1999).
- ²⁶M. M. J. Treacy, T. W. Ebbesen, and J. M. Gibson, *Nature (London)* **381**, 678 (1996).
- ²⁷M. F. Yu, O. Lourie, M. J. Dyer, K. Moloni, T. F. Kelly, and R. S. Ruoff, *Science* **287**, 637 (2000).
- ²⁸S. M. Clarke, F. Elias, and E. M. Terentjev, *Eur. Phys. J. E* **2**, 335 (2000).
- ²⁹H. M. Jaeger, C.-h. Liu, and S. R. Nagel, *Phys. Rev. Lett.* **62**, 40 (1989).
- ³⁰S. M. Clarke and E. M. Terentjev, *Phys. Rev. Lett.* **81**, 4436 (1998).
- ³¹L. A. Girifalco, M. Hodak, and R. S. Lee, *Phys. Rev. B* **62**, 13104 (2000).
- ³²R. Yerushalmi-Rozen and I. Szleifer, *Soft Matter* **2**, 24 (2006).
- ³³T. P. J. Knowles, J. F. Smith, A. Craig, C. M. Dobson, and M. E. Welland, *Phys. Rev. Lett.* **96**, 238301 (2006).
- ³⁴J. R. Blundell and E. M. Terentjev, *J. Phys. A: Math. Theor.* **40**, 10951 (2007).
- ³⁵E. I. Corwin, H. M. Jaeger, and S. R. Nagel, *Nature (London)* **435**, 1075 (2005).
- ³⁶L. A. Hough, M. F. Islam, P. A. Janmey, and A. G. Yodh, *Phys. Rev. Lett.* **93**, 168102 (2004).
- ³⁷P. G. Whitten *et al.*, *Carbon* **43**, 1891 (2005).
- ³⁸R. Duggal and M. Pasquali, *Phys. Rev. Lett.* **96**, 246104 (2006).
- ³⁹S. V. Ahir, A. M. Squires, A. R. Tajbakhsh, and E. M. Terentjev, *Phys. Rev. B* **73**, 085420 (2006).
- ⁴⁰C. Li and T.-W. Chou, *Phys. Rev. B* **71**, 235414 (2005).
- ⁴¹T. Hugel, N. B. Holland, A. Cattani, L. Moroder, M. Seitz, and H. E. Gaub, *Science* **296**, 1103 (2002).
- ⁴²C. L. Zhang and H. S. Shen, *Appl. Phys. Lett.* **89**, 081904 (2006).
- ⁴³A. Arinstein, M. Burman, O. Gendelman, and E. Zussman, *Nat. Nanotechnol.* **2**, 59 (2007).