Photomechanical actuation in polymer–nanotube composites

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For some systems, energy from an external source can trigger changes in the internal state of the structure, leading to a mechanical response much larger than the initial input. The ability to unlock this internal work in a solid-state structure is of key importance for many potential applications. We report a novel phenomenon of photo-induced mechanical actuation observed in a polymer-nanotube composite when exposed to infrared radiation. At small strains the sample tends to expand, when stimulated by photons, by an amount that is orders of magnitude greater than the pristine polymer. Conversely, at larger applied pre-strain, it will contract under identical infrared excitation. The behaviour is modelled as a function of orientational ordering of nanotubes induced by the uniaxial extension. It is thought that no other materials can display this continuously reversible response of so large a magnitude, making rubber nanocomposites important for actuator applications.

t is the ability of certain actuator materials to change their dimensions upon application of a given stimulus, such as heat, electric voltage or light, that makes them such attractive systems to study. Actuators with differing characteristics and mechanisms have been widely adopted by industry to fill a variety of technological requirements¹. Some actuators have a one-way response, whereas others are based on an equilibrium, reversible response to the given stimulus. Some (smart) materials exhibit a latent ability to actuate under the right conditions, such as shape-memory alloys² or liquid-crystal elastomers³. Other systems require the blending of two (or more) distinctly different materials to impart a new physical response leading to the actuation process. The work presented here focuses on the second class of equilibrium mechanical actuation using carbon nanotubes embedded in a polymer matrix.

The sometimes complex behaviour of multiwalled carbon nanotubes (MWCNTs) has heralded some interesting insights into the possibility of designing nanoelectromechanical (NEMS) systems⁴. The actuator properties of individual bending MWCNTs under an applied electric field have been studied experimentally⁵. Their torsional actuator properties have also been reported^{6,7}. It is interesting to note that all these studies focus on individual MWCNTs and not a collection of tubes, nor their composite in a continuous elastic matrix. Little information is available on the actuation properties of nanotube assemblies or the effect they can have on material actuation properties when embedded in a polymer.

A few studies have recently appeared in the literature that attempt to look at the actuation behaviour of polymer–nanotube composites^{8–10}. However, important as they are, many of these studies have concentrated on accentuating the already present features of the host matrix by adding nanotubes. The nanotubes, in essence, exaggerate actuator behaviour by either improving electromechanical responses or heating the sample more efficiently because of their inherent high conductivity¹¹. Particularly new and important is the case of electrically stimulated mechanical contraction of MWCNTs embedded in liquid-crystal elastomers¹². The work of Courty *et al.*¹² was unique in that it details a novel electro-actuator response due to the presence of MWCNTs that otherwise would not occur in that system.

The present work introduces a new but simple polymercomposite system (comparing with nematic elastomer composites of ref. 12), which, by the presence of MWCNTs, produces a

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Figure 1 Scheme of apparatus, X-ray scattering image and the azimuthal intensity scan. a, The sample (S) is clamped in the frame with its length (for example, prestrain) controlled by the micrometer (M) and the exerted force measured by the dynamometer (D). Thermocouples (T_1 and T_2) are placed in front and behind, close to the sample surface. The actuation is provided by the infrared light source uniformly illuminating the sample. The whole rig is enclosed in the thermally controlled compartment. A similar setup is used when taking wide-angle X-ray measurements. **b**, The X-ray image showing key scattering reflexes; the outer ring corresponding to 3.4 Å is the signal from the multiwall nanotubes. The inner ring is indicative of the correlation length of mesh size, see text. The arrow shows the direction of the aligning strain. **c**, The typical azimuthal intensity variation, $I(\beta)$, at a scattering angle corresponding to the outer (MWCNT, 3.4 Å) ring. The data are fitted by the theoretical model of ref. 17.

mechanical response to the infrared (IR) irradiation. Remarkably, we obtain both a compressive and an expansive response mode depending on the external uniaxial strain applied to the composite sample. We assert that this behaviour is dependent on nanotube orientation within a homogenous polymer matrix. Multiwalled nanotubes are uniformly dispersed in polydimethylsiloxane (PDMS) at concentrations of 0.02, 0.5, 1, 4 and 7 wt%. A schematic of the apparatus is given in Fig. 1a. For characterization purposes, the linear mechanical responses of our nanocomposites have been tested for different nanotube loadings in the crosslinked PDMS matrix. As the concentration of MWCNTs is increased from zero to 4 wt% loading, the rubbery network becomes stiffer and the Young modulus Y increases by a factor of two. This is expected and in line with literature findings^{13,14}. A detailed account for subtle variations in measured moduli can be garnered if consideration is given to the polymer-nanotube interface and relaxation of local stress in the composites, to be published elsewhere.

It is important to characterize nanotube alignment quantitatively. Wide-angle X-ray diffraction is used as a method to determine the average tube orientation as a function of increasing applied uniaxial strain. Figure 1b shows characteristic features of the diffraction halos. This example is for a 7 wt% sample, initially non-aligned, stretched by $\varepsilon = 0.33$ (33%). The Bragg peak around 3.40 Å corresponds to the (002) scattering plane, which describes the inter-shell spacing periodicity within the multiwall tubes (see for example ref. 15).

A question must arise about the bright scattering ring corresponding to the length scale ~7.5 Å. This is an interesting feature and is exactly the same in the pristine PDMS rubber prepared in the same batch. In the pristine PDMS network, with no solvent, the only X-ray contrast arises from the difference in crosslink distribution. A very clear scattering length must be an indication of crosslink density fluctuations (in other terminology called clustering). As the extensive theory of this phenomenon suggests¹⁶, at the given chain lengths and crosslinking density our PDMS network is well below the 'crosslink saturation threshold' and the correlation length of clustering should be of the order of mesh size. The length scale of ~7.5 Å is very accurately this size and, accordingly, we believe this scattering to be produced by small-scale crosslink density fluctuations (for comparison, same conditions

of scattering from a non-crosslinked PDMS melt showed no such reflection). These should not affect macroscopic properties, or even the local MWCNT embedding.

As the applied uniaxial strain increases, the 3.4 Å (MWCNT) ring develops an increasing azimuthal bias ($I(\beta)$ in Fig. 1c) indicating the orientational ordering of tubes. For instance, at the pre-strain value of $\varepsilon = 0.6$ (60%) this induced order Q reaches as high as ~0.29. As a comparison, samples that had been accidentally pre-stressed during preparation are shown to have a very low orientational order parameter, $Q \le 0.005$. Therefore, composites with no significant initial alignment, on subsequent stretching reached much higher values of induced orientational order. We assume this is due to a much more rigid network surrounding the tubes and attempting to deform affinely, thus imposing significant orientational bias, than in a loosely crosslinked gel under similar deformation. Furthermore, the change in orientation on stretching is reversible, that is, equilibrium. The details of this study of nanotube reorientation.

There is an issue, well argued in the literature^{18,19}, about whether a truly isotropic nanotube dispersion can be obtained. Regarding the tubes as rigid rods with extremely high aspect ratio, well dispersed in an amorphous medium, the Onsager transition to the sterically enforced orientational ordering could start at very low concentrations. We have as yet observed no clear indication of truly nematic liquid-crystalline architecture in our system, although this could be due to a number of factors including matrix viscosity at sample preparation stage.

The intriguing response of our nanocomposite samples to IR is shown in Fig. 2, presenting the data on stress measured in the 1 wt% sample, initially non-aligned. It is important to spend some time and fully appreciate the information provided in this complex plot. At the start of experiment the sample has a 2% pre-strain ($\varepsilon = 0.02$) applied to it, and has been allowed to mechanically equilibrate. Let us follow the first data set, which represents the 2%-strain line (the lowest curve in Fig. 2). The plot shows the raw data of measured stress as a function of time of the experiment. The initial stress reading is simply the measure of Young's modulus $Y \approx 1.15$ MPa, and this value of stress is transferred as an open symbol to the plot in Fig. 3. At a certain moment of time the IR light source is



Figure 2 Response to IR radiation at different values of pre-strain. Raw data on stress measured at fixed sample length of a 1 wt% nanocomposite (different pre-strain curves labelled on the plot). A fixed pre-strain is applied to each given sample, which is then allowed to relax for a minimum of 10 minutes. Readings of stress are taken for two minutes and then the IR source is switched on to full intensity. After 15 minutes of exposure, the light source is switched off and further relaxation data is collected. This protocol is repeated for the same sample but at a different applied pre-strain ε after it has been equilibrated. Each sample is tested under a range of applied pre-strains between 2% and 40%.

switched on and the stress reading changes. In this case (for 2% prestrain) the change is downwards, meaning that the sample's natural length has expanded on actuation. The new (IR-on) stress reading is transferred to the plot in Fig. 3 as a filled symbol corresponding to the applied pre-strain of 2%. After a period of constant irradiation, during which the stress reading remains stable, the light source is switched off—and the stress reading returns to its original value. This experiment is then repeated with the same sample pre-strained at different values, up to 40%, as shown by the sequence of stressreading curves, Fig. 2, and the corresponding pairs of stress-strain points, with and without IR stimulation, in Fig. 3. A question of possible sample degradation may be asked: we have deliberately conducted this type of experiment in a random sequence of prestrain/equilibration cycles. The evidently consistent trend proves the reversibility of the sample state.

The first main conclusion we make is the unambiguous and significant photo-actuation response of PDMS nanocomposites. However, the immediate next question is this photo-mechanical response changing sign at a certain level of uniaxial pre-strain. In other words, equilibrium or weakly stretched composites show reversible expansion on irradiation, whereas highly stretched ones show a much stronger tendency to contract (hence the increase in the measured stress). This is the central result of this paper and we shall spend some time discussing and analysing it. There are two main questions: what is the mechanism of photo-mechanical response, and why does it change its macroscopic signature on sample extension?

Figure 4 summarizes the actuation effect by plotting the magnitude of stress step in the IR-on state, at different levels of pre-strain and for samples with increasing MWCNT loading. Although this is not explicitly measured in our (isostrain) experiment, we can directly calculate the change of the underlying natural length of the samples on actuation. This is shown on the right axis of the same plot, explicitly illustrating the regions of expansion and contraction. Remarkably, all samples with different nanotube loading appear to have a crossover



Figure 3 Stress–strain variation under IR irradiation. The data from Fig. 2 for 1 wt% nanocomposite, highlighting the opposite direction of actuation at different pre-strains, evident in Fig. 2, and the crossover at $\varepsilon^* \approx 0.1$ (10%).

at the same point, around 10% pre-strain (the increase in the effect amplitude with loading is to be expected).

There is no noticeable change in the reading of stress, Fig. 2, with time after the IR source is switched on. This means that the heat transfer from the irradiated sample face into its bulk plays no significant role in the mechanism of mechanical actuation. The effect is highly reproducible, over many cycles of irradiation, which leads us to conclude no degradation due to non-radiative photon decay takes place in the nanocomposite samples. The effect has also been observed in a different polymeric matrix using a crosslinked side-chain polysiloxane nematic polymer. For comparison, the pristine PDMS rubber in the same experiment shows a very minor stress response, two orders of magnitude smaller than in Fig. 2, which we attribute entirely to the sample temperature change on IR irradiation. The change in temperature by IR heating is unavoidable and reaches $\Delta T \sim 15$ °C maximally, in our setting. This highlights an important question as to whether the response is due to the photon absorption or the plain heat transfer. Although not presented in detail here, we have studied the mechanical response purely due to the temperature change. The temperature results are an order of magnitude smaller than in the case of IR-stimulation. The conclusion we reach is that such an effect does exist (that is, the MWCNT-loaded composite has a stronger mechanical response than a pristine polymer on the same change ΔT) but its value is insignificant in comparison with the direct IR-photon absorption mechanism.

We believe the reason behind the change of actuation direction on increasing sample extension is due to the nanotube alignment. A simple affine model of induced orientational order gives the biased probability distribution of tube axes

$$P(\theta) = \frac{\lambda^{3/2}}{(\cos^2\theta + \lambda^{3/2}\sin^2\theta)^{3/2}},$$
 (1)

with uniaxial stretching factor $\lambda = 1 + \varepsilon$ (see Supplementary Information for derivation). This corresponds to the data in Fig. 1c and predicts the orientational order at relatively low pre-strains: $Q \approx \sqrt[3]{5\varepsilon}$. At the crossover point, $\varepsilon^* \approx 0.1$, giving $Q \approx 0.06$. Let us

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Figure 4 Summary of IR response. The magnitude (in kPa) of exerted actuation stress (the height of steps in Fig. 2), as a function of pre-strain, for samples with increasing MWCNT loading. The right *y* axis shows the corresponding actuation stroke: the change in natural length L_0 on IR irradiation.

now apply the same ideas about the induced orientational bias and averaging of the (hypothetical) individual nanotube response.

We assume this individual response is, in essence, a contraction because this is what the better-aligned composite response is. It is easy to imagine why this could be for an initially rod-like tube: on photon absorption it may generate kink-instabilities, thus decreasing its net length. Let us simply assume that each nanotube, on IR stimulus, undergoes a contraction by a factor $\Delta < 1$ (certainly proportional to radiation intensity, which was kept constant in our work), accompanied by a transversely isotropic volume conserving expansion $1/\sqrt{\Delta}$. This means that a local strain is created with the principal axes along the current nanotube orientation (at angle θ to the macroscopic z axis, see Fig. 5a)

$$\Lambda (IR) = \begin{pmatrix} 1/\sqrt{\Delta} & 0 & 0\\ 0 & 1/\sqrt{\Delta} & 0\\ 0 & 0 & \Delta \end{pmatrix}.$$
 (2)

The projection of this local strain on the macroscopic axis of sample extension (and force measurement) is $\lambda_z(IR) = \Delta \cos^2\theta + (1/\sqrt{\Delta})\sin^2\theta$. When we average this local contribution with the probability of finding the nanotube at this orientation, $P(\theta)$, it will produce an estimate of the effective stroke of actuation ($\lambda_z - 1$) or, if multiplied by the corresponding Young modulus, the exerted stress of Fig. 4. Such a model is very crude indeed, ignoring a large number of undoubtedly important and delicate factors of continuum elasticity and nanotube morphology. However, it is elastically self-consistent and has only one parameter Δ that carries all the underlying complexity of the nanotube problem in it.

The orientational averaging is straightforward:

$$\langle \lambda_z \rangle = \int_0^\infty [\Delta \cos^2 \theta + (1/\sqrt{\Delta}) \sin^2 \theta] P(\theta) \frac{1}{4\pi} \sin \theta \, d\theta \, d\varphi$$
$$\approx \frac{1}{3} \left(\Delta + 2/\sqrt{\Delta} \right) - \frac{2}{5} \varepsilon (1/\sqrt{\Delta} - \Delta).$$
(3)

Although the integral above has a full analytic form, it is more



Figure 5 Scheme of local and macroscopic strains, and the prediction of the actuation model. a, Scheme illustrating how the distortion (kinking or undulation) of an individual tube, lying at an angle θ to the alignment axis, projects on the *z* axis to contribute to the macroscopically uniaxial strain, equation (2). **b**, The result of theoretical modelling based on orientational averaging of local deformations from each nanotube, equation (3); the dashed line shows the linear approximation at small pre-strain ε . Nanotube contraction factor is chosen $\Delta = 0.8$, as suggested by the crossover strain value $\varepsilon^* \approx 0.1$.

transparent to give its limit at small imposed pre-strain ε . This shows the key point: at very low pre-strain, $\varepsilon \rightarrow 0$, the average actuation stroke of the disordered nanocomposite is a positive $(\lambda_z - 1)$, that is, the expansion of its natural length. However, above the threshold pre-strain ε^* this average deformations becomes negative, that is, the contraction. It is easy to find

$$\varepsilon^* = \frac{5(2 - \Delta^{1/2} - \Delta)}{6(1 + \Delta^{1/2} + \Delta)},\tag{4}$$

so that the prediction would be to observe the crossover at $\varepsilon^* \approx 0.1$ if the nanotube response factor $\Delta \approx 0.8$, that is, on IR-irradiation the nanotube itself contracts overall by ~20%. The value is higher than one might expect, considering reports in the literature of nanotube strains of only 1–2%. However, as Fig. 5a indicates, our proposition is not that of the lattice strain of nanotube walls but a contortion of the tube as a whole. Although this has not yet been directly observed and reported in the literature, a similar effect of resonant undulation has been seen (in simulation²¹ and in experiment⁵) in response to distortion beyond the linear regime. Although in our system the tubes respond under totally different conditions, embedded in an elastic matrix under strain and absorbing the IR photons, the overall distortion factor of 20%, suggested by the model fit, is perhaps not altogether unreasonable.

Figure 5b plots the full (non-expanded) result of orientational averaging of actuation stroke $(\langle \lambda_z \rangle - 1)$ from equation (3) to illustrate the points discussed here. The qualitative behaviour (as summarized in Fig. 4) is reproduced here almost exactly, including the magnitude of the predicted actuation stroke (that is, the ratio $L_0(\text{IR})/L_0(0)-1$). Therefore, it is very likely that the orientational feature of the effect, with its change of actuation direction at a critical level of induced alignment, is captured correctly, whereas much more work is required to understand the individual nanotube response to IR radiation generating the phenomenological factor Δ used in this analysis.

The strength of photo-actuator response, at a given radiation intensity, is of the order of tens of kilopascals. Translated into the stroke, this corresponds to actuation strains of 2–4%. As expected, the response increases at higher nanotube loading. Similar (thermal actuation) behaviour is also observed when the samples are heated by the same amount, but this has an order of magnitude lower amplitude.

Understanding the nature of the actuator mechanisms in this system certainly warrants further theoretical and experimental investigation. Many questions remain completely unclear, in particular, the response of an individual nanotube, embedded in a polymer matrix, to IR photons. It is also not clear what the effect would be if different types of nanotube were used, that is, smaller multiwall diameters, single-wall tubes and so on. Future investigations may also want to address the issue of using another host matrix and confirm its relatively neutral role in the actuation mechanism.

With actuating materials already used in such widespread applications, from micromanipulators to vibration control, the discovery of a structure that can respond to stimulus in both directions may open new possibilities and could mean an important new step towards finding applications for nanotube-based materials above and beyond improvements in existing carbon-fibre technologies.

METHODS

Multiwalled nanotubes (Nanostructured & Amorphous Materials) are used with purity verified (using scanning electron microscopy, SEM) as >95%. These nanotubes were not surface-modified at any time during processing.

The polymer matrix is PDMS (polydimethylsiloxane) Sylgard 184 silicone elastomer from Dow Corning, USA. In pristine condition this crosslinks with hydrosilane crosslinker, the curing agent supplied with Sylgard 184 by Dow Corning, forming a uniform solvent-free elastomer. We have verified (with SEM on microtomed and on freeze-fractured surfaces) that the resulting polymer network is pure crosslinked PDMS with no filler particles, as sometimes is the case with supplied elastomer mixes.

The nanotube–polymer composites are fabricated by first carefully weighing the desired quantity of nanotubes and PDMS polymer. Calculations of weight percentage take into account the crosslinker, to be later used in the mixture. The viscous fluid is sheared using an Ika Labortechnik mixer for a minimum of 24 hours.

Crosslinker is added to the mixture after 24 hours. The ratio of cross-linker to PDMS is 1:10, according to Sylgard 184 specification, ensuring negligible sol fraction after preparation of a pristine network. The sample is then further sheared for another 30 seconds before being placed in vacuum for five minutes, at all times remaining at ambient temperature to ensure little crosslinking reaction takes place in this time. This suitably removes the air cavities that unavoidably form during processing. It is then deposited in a specially designed reactor (centrifuge compartment with PTFE film strip lining its inner wall) and placed in a centrifuge at 5,000 r.p.m. (about 4,000g) and 80 °C. At this temperature the PDMS crosslinking is much faster.

The quality of nanotube dispersion is monitored throughout the processing with the use, initially, of optical microscopes and later with a high-resolution SEM (HRSEM, Phillips XL 30 series) as aggregate sizes reduce well below optical resolution. We find that a shearing regime lasting 24 hours is suitable in removing nanotube aggregates due to the inherently high viscosity of the host polymer. Samples are identified by the wt% of MWCNTs mixed with the PDMS. Most experiments have been conducted on the 0, 0.02, 0.5, 1, 4 and 7 wt% MWCNTs in PDMS elastomer films. In all cases the sample dimensions were kept approximately the same, 30×1.5 mm, with the film thickness 0.2 mm, so that the area exposed to radiation was ~0.45 cm² and the cross-section area for calculating stress was ~3 × 10⁻⁶ m².

A 25g Dynamometer (from Pioden Systems) was housed in a custom-made thermal-control box with an open front end. The device, together with two independent thermocouples, outputs data though a DAQ card to a PC, see Fig. 1a. Distances between the clamps was controlled by using a securely fitted micrometer with ± 0.001 mm accuracy. The rig is calibrated with weights to give a direct measure of stress and strain. The light source (Schott KL1500 LCD, maximum intensity at ~675 nm, 702 µW cm² at 1 m distance) was positioned 20 mm \pm 0.02 mm from the sample surface. Calculation suggests that the total power of light delivered to the sample was of the order of 0.008 W. Additionally, the technique used to measure temperature involves placing the thermocouples on the sample surface itself. Hence, the ~15 °C increase in temperature is truly a measure of temperature change across the surface. This is not a microscopic measure of temperature throughout the sample, however, the same techniques have been repeated for every experimental run with the thermocouples manually placed at differing regions over the sample and the same relative change is always recorded.

Standardization across all samples is carried out through pre-experimental checks involving accurately finding the zero strain value of each experimental setup and calibrating the test rig from this point. Readings of stress are taken for two minutes, to verify that the material is equilibrated in its 'natural' pre-strained state, and then the IR source is switched on to full intensity. After 15 minutes of exposure, the light source is switched of further relaxation data are collected for the remaining 15 minutes. This protocol is then repeated for the same sample but at a higher applied pre-strain ε . Each sample is tested under a range of applied pre-strains between 2% and 40% (0.02 $\leq \varepsilon \leq 0.4$).

As a comparison the same experiment is carried out on the 1 wt% MWCNT sample, but the IR source is replaced by a mica-insulated heater (Minco Products) mounted approximately 10 mm away from the sample.

Wide angle X-ray diffraction measurements were carried out on a Phillips PW1830 wide angle X-Ray generator (WAXS) using Cu_{ka1} radiation (=1.54 Å), running at 40 kV and 40 mA. Azimuthal curves were generated as detailed in classical literature, as well as in more recent treatment²⁰ specifically for nanotubes. The azimuthal intensity variation of the arcs at the scattering angle corresponding to MWCNT (002) layer periodicity of 3.4 Å allows calculation of the degree of orientation.

Relative intensity along the azimuthal arcs, $I(\beta)$ (see Fig. 1c), is the signature of the orientational distribution function; when approximated as a Legendre polynomial series in cos β , it gives a direct measure of the (Herman's) orientational order parameter, in this context often called S_{ab} or

 $Q \equiv \langle P_2 \rangle = \frac{1}{2} \langle 2 \rangle cos^2 \beta \rangle$ -1), where the averaging is performed with $I(\beta)$ as the distribution function. The more advanced method of orientational order parameter analysis²¹ gives similar values. See Supplementary Information for details.

Mechanical testing has been conducted on the same dynamometer rig, Fig. 1a, finding the quasistatic stress–strain relationship on uniaxial stretching narrow strips of PDMS nanocomposites. In the linear regime of small strains this identifies the equilibrium Young's modulus, from $\sigma = Y\epsilon$.

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References

- Huber, J. E., Fleck, N. A. & Ashby, M. F. The selection of mechanical actuators based on performance indices. Proc. R. Soc. A 453, 2185–2205 (1997).
- 2. Battacharya, K. Microstructure of Martensite (Oxford Univ. Press, UK, 2004).
- 3. Warner, M. & Terentjev, E. M. Liquid Crystal Elastomers (Oxford Univ. Press, UK, 2003).
- Cumings, J. & Zettl, A. Low-friction nanoscale linear bearing realized from multiwall carbon nanotubes. *Science* 289, 602–604 (2000).
- Poncharal, P., Wang, Z. L., Ugarte, D. & de Heer, W. A. Electrostatic deflections and electromechanical resonances of carbon nanotubes. *Science* 283, 1513–1516 (1999).
- Williams, P. A. et al. Torsional response and stiffening of individual multiwalled carbon nanotubes. Phys. Rev. Lett. 89, 255502 (2002).
- 7. Fennimore, A. M. et al. Rotational actuators based on carbon nanotubes. Nature 424, 408-410 (2003).
- Landi, B. J. et al. Single wall carbon nanotube-nafion composite actuators. Nano Lett. 2, 1329–1332 (2002).
- Koerner, H., Price, G., Pearce, N. A., Alexander, M. & Vaia, R. A. Remotely actuated polymer nanocomposites—stress-recovery of carbon-nanotube-filled thermoplastic elastomers. *Nature Mater.* 3, 115–120 (2004).
- Tahhan, M., Truong, V. T., Spinks, G. M. & Wallace, G. G. Carbon nanotube and polyaniline composite actuators. *Smart Mater. Struct.* 12, 626–632 (2003).
- 11. Naciri, J. et al. Nematic elastomer fiber actuator. Macromolecules 36, 8499-8505 (2003).
- Courty, S., Mine, J., Tajbakhsh, A. R. & Terentjev, E. M. Nematic elastomers with aligned carbon nanotubes: New electromechanical actuators. *Europhys. Lett.* 64, 654–660 (2003).
- Gorga, R. E. & Cohen, R. E. Toughness enhancements in poly(methyl methacrylate) by addition of oriented multiwall carbon nanotubes. J. Polym. Sci. B 42, 2690–2702 (2004).
- 14. Harris, P. J. F. Carbon nanotube composites. Int. Mater. Rev. 49, 31-43 (2004).
- Charlier, J. C. & Michenaud, J. P. Energetics of multilayered carbon tubules. *Phys. Rev. Lett.* 70, 1858–1861 (1993).
- 16. Panyukov, S. V. & Rabin, Y. Statistical physics of polymer gels. Phys. Rep. 269, 1-131 (1996).
- Deutsch, M. Orientational order determination in liquid-crystals by X-ray diffraction. *Phys. Rev. A* 44, 8264–8270 (1991).
- Somoza, A. M., Sagui, C. & Roland, C. Liquid-crystal phases of capped carbon nanotubes. *Phys. Rev. B* 6308, 081403 (2001).
- 19. Islam, M. F. et al. Nematic nanotube gels. Phys. Rev. Lett. 92, 088303 (2004).
- Jin, L., Bower, C. & Zhou, O. Alignment of carbon nanotubes in a polymer matrix by mechanical stretching. *Appl. Phys. Lett.* 73, 1197–1199 (1998).
- Yakobson, B. I. & Bradec, C. J. & Bernholc, J. Nanomechanics of carbon tubes: instabilities beyond linear response. *Phys. Rev. Lett.* 76, 2511–2514 (1996).

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Competing financial interests

The authors declare that they have no competing financial interests.