Nanostructured polymer films with metal-like thermal conductivity

Yanfei Xu1,3, Daniel Kraemer1,4, Bai Song1,5, Zhang Jiang2, Jiawei Zhou1, James Loomis1, Jianjian Wang1,6, Mingda Li1,7, Hadi Ghasemi1,8, Xiaopeng Huang1,9, Xiaobo Li1,10 & Gang Chen1

Due to their unique properties, polymers—typically thermal insulators—can open up opportunities for advanced thermal management when they are transformed into thermal conductors. Recent studies have shown polymers can achieve high thermal conductivity, but the transport mechanisms have yet to be elucidated. Here we report polyethylene films with a high thermal conductivity of 62 Wm⁻¹ K⁻¹, over two orders-of-magnitude greater than that of typical polymers (~0.1 Wm⁻¹ K⁻¹) and exceeding that of many metals and ceramics. Structural studies and thermal modeling reveal that the film consists of nanofibers with crystalline and amorphous regions, and the amorphous region has a remarkably high thermal conductivity, over ~16 Wm⁻¹ K⁻¹. This work lays the foundation for rational design and synthesis of thermally conductive polymers for thermal management, particularly when flexible, lightweight, chemically inert, and electrically insulating thermal conductors are required.

References:
1 Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA. 2 Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA. 3 Present address: Department of Mechanical and Industrial Engineering, University of Massachusetts Amherst, Amherst, MA 01003, USA. 4 Present address: Modern Electron, Bothell, WA 98011, USA. 5 Present address: Department of Energy and Resources Engineering, Peking University, Beijing 100871, China. 6 Present address: Advanced Cooling Technologies, Inc., Lancaster, PA 17601, USA. 7 Present address: Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA. 8 Present address: Department of Mechanical Engineering, University of Houston, Houston, TX 77004, USA. 9 Present address: 2205 W Olive Way, Chandler, AZ 85248, USA. 10 Present address: State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, 430074 Wuhan, Hubei, China. Correspondence and requests for materials should be addressed to G.C. (email: gchen2@mit.edu)
from soft robotics, organic electronics to 3D printing and artificial skin, polymers continue to infiltrate modern technologies thanks to their unique combination of properties not available from any other known materials. They are lightweight, durable, flexible, corrosion resistant, and easy to process, and hence are expected to offer significant advantages over traditional heat conductors, such as metals and ceramics. However, application of polymers in thermal management has been largely hampered by their low thermal conductivities (~0.1 Wm$^{-1}$ K$^{-1}$). To date, metals and ceramics remain the dominant heat conductors.

The fact that polyethylene (0.2–0.5 Wm$^{-1}$ K$^{-1}$) is composed of a backbone of carbon-carbon bonds similar to those in diamond, one of the most thermally conductive materials (above 1000 Wm$^{-1}$ K$^{-1}$), encourages research in thermally conductive polymers. Importantly, atomistic simulations have suggested that an individual crystalline polyethylene chain can achieve very high—possibly divergent—thermal conductivity, in agreement with the non-ergodic characteristics of one-dimensional conductors discussed by Fermi et al. However, the experimental measurement of such theoretically high thermal conductivities remains elusive. By increasing the crystallite orientation and crystallinity, the thermal conductivity of polymers can increase considerably, such as polyethylene nanofibers (~104 Wm$^{-1}$ K$^{-1}$). Although exceptionally conductive, these measured values are still much lower than the numerical predictions for bulk single-crystalline polyethylene (~237 Wm$^{-1}$ K$^{-1}$). There is no precise mechanism that accounts for the deviation of experimental and theoretical values. And the main factors that govern the thermal conductivity in these fibers remain poorly understood. It is generally known that such materials are not perfect crystals, but instead semicrystalline polymers containing mixed crystalline and amorphous regions. Translating the remarkably high thermal conductivity seen in simulation as well as in polyethylene nanofibers into a scalable polymer presents a major challenge in synthesis. Overcoming this challenge will broadly expand the scope of nanofiber use in thermal management, since practical applications require large areas or volumes of materials. Recently, Ronca et al. reported stretched ultra-high molecular weight film with thermal conductivity as high as 65 Wm$^{-1}$ K$^{-1}$, measured using a commercial laser-flash system, and Zhu et al. reported thermal conductivity of fibers as high as 51 Wm$^{-1}$ K$^{-1}$ by further processing of commercial spectra fibers using an electrothermal method. These reports show the potential of achieving high thermal conductivity in macroscopic samples. However, the structural property relationship has yet to be further elucidated.

We have been engaged in scaling up the high thermal conductivity of individual nanofiber to more macroscopic films. Here, we report a thermal conductivity measurement of 62 Wm$^{-1}$ K$^{-1}$ in polyethylene films (Fig. 1). The thermal conductivity in our film outperforms that of many conventional metals (304-stainless steel ~15 Wm$^{-1}$ K$^{-1}$) and ceramics (aluminum oxide ~30 Wm$^{-1}$ K$^{-1}$). Motivated by the theoretically large thermal conductivity of single-crystal polymers, we fabricate thermally conductive polymer films with an emphasis on minimally entangling and maximally aligning the chain, rather than solely pursuing a high crystallinity. We further uncover the thermal transport mechanisms through the combination of structural analysis, determined by high-resolution synchrotron X-ray scattering, and a phenomenological thermal transport model. We find that the film actually consists of nanofibers with crystalline and amorphous regions along the fiber and that the amorphous regions have remarkably high conductivity (~16 Wm$^{-1}$ K$^{-1}$), which is central to the high thermal conductivity (~62 Wm$^{-1}$ K$^{-1}$). Increased control over amorphous morphology is a promising route toward achieving thermal conductivities approaching theoretical limits.

### Results

**Polymer processing.** We start with commercial semi-crystalline polyethylene powders (Fig. 1a), which feature randomly oriented lamellar crystallites (lamellae) dispersed in an amorphous chain network (Fig. 1d). We dissolve the powder above its melting temperature in decalin, allowing the initially entangled chains to disentangle (Fig. 1d). This greatly reduces the entanglements for the subsequent processing. Afterward, the hot solution is extruded through a custom-built Couette-flow system, which imparts a shear force on the polymer chains and led to further disentanglement. To maintain the disentangled structure, the extruded solution flows directly onto a liquid nitrogen-cooled substrate. Some segments of the polyethylene chains fold back into thin lamellae upon drying, while others remain disordered albeit less entangled (Fig. 1d). Finally, the as-extruded films (Fig. 1b) are mechanically pressed and drawn inside a heated enclosure using a continuous and scalable roll-to-roll system. Heating allows the disentangled polymer chains to move more freely and facilitates alignment along the draw direction (Fig. 1c, d).

**Microscale and nanoscale morphology of polymers.** In order to track the evolution of polymer structures, we imaged the as-purchased powders, the extruded films and films of various draw ratios (final length/initial length) using scanning electron microscopy (SEM, Fig. 1e–j). The powder consists of porous particles with an average size of ~100 μm (Fig. 1e). After extrusion, the film surface appeared isotropic with randomly distributed microflakes (Fig. 1f). During drawing, the film self-organized into a clear fibrous texture along the draw direction. The diameters of the fibers comprising the film reduced as the draw ratio increased, which led to a smoother and denser texture (see Fig. 1g and Fig. 1h). We further tore a x70 film apart to explore the detailed internal structures where individual fiber can be clearly observed (Fig. 1i, j), and multiple interior fibers with smaller diameter ~8 nm were also seen (Supplementary Fig. 3).

**Thermal conductivity measurements.** To study the thermal properties of these polyethylene films, we employed two distinct experimental schemes: a home-built steady-state system (Fig. 2a) and a widely-adopted transient method called time domain thermoreflectance (Fig. 2c, Supplementary Notes 2 and 3). On the steady-state platform, we measured heat current as a function of temperature difference across a sample film (Fig. 2b, Supplementary Fig. 1a and Table 1). We investigated the systematic errors including on radiation and parasitic heat losses, and carefully minimized measurement errors (see Supplementary Note 2, Supplementary Figs. 1 and 2). To validate the steady-state measurement accuracy, we tested several control samples including 304-stainless steel foil (S. Steel 304), Zylon fibers, Dyneema fibers, Sn films, and Al films, our measured thermal conductivity values are in general agreement with established values.

As draw ratio was increased, film thermal conductivity along the draw direction was improved dramatically, reaching 62 Wm$^{-1}$ K$^{-1}$ at x110 (Figs. 2b and 3a). Notably, we saw no sign of saturation in thermal conductivity (Fig. 3a, b will be discussed below), which suggested more room for further conductivity enhancement beyond...
×110 draw ratio. Recent atomistic simulations further corroborate this expectation.

Two-color time-domain thermoreflectance (TDTR) experiments were conducted to study transient heat conduction in the films and to further validate the steady-state results (Fig. 2b). We fabricated a 150-μm-thick laminate consisting of 100 layers of ×50 films and carefully microtomed a cross-section (roughness ~10 nm, Supplementary Fig. 6 and Note 2) perpendicular to the draw direction. Representative thermoreflectance signals are reported in Fig. 2d, from which we extracted an average thermal conductivity of 33.6 Wm⁻¹K⁻¹ (Fig. 3a) along the draw direction. The TDTR results agree well with values obtained using the steady-state system (Fig. 3a). The successful demonstration of 100-layer laminate with such high thermal conductivity implies potential scalability not only along the drawing direction, but also in the thickness direction. In addition, we have investigated film thermal stability, obtaining <5% thermal conductivity variations before and after annealing (24 h at 80 °C).

Atomic scale and nanoscale structure investigation. To reveal correlation between such a high thermal conductivity and structure, we quantitatively investigated the structure at both atomic scale and nanoscale by high-resolution wide-angle and small-angle X-ray scattering. Representative scattering patterns are reported in Fig. 4a, from which we extracted a correlation length of ~100 nm along the draw direction. The nanoscale structure is further investigated using scanning electron microscopy (SEM) and atomic force microscopy (AFM). Representative SEM images are reported in Fig. 4b, from which we extracted a microstructure of oriented crystallites interconnected by aligned amorphous chains. The nanoscale structure is further investigated using high-resolution transmission electron microscopy (HRTEM) and electron diffraction. Representative HRTEM images are reported in Fig. 4c, from which we extracted a nanoscale structure of oriented crystallites interconnected by aligned amorphous chains.
small-angle synchrotron X-ray scattering (Fig. 4 and Supplementary Note 4). Wide-angle X-ray scattering (WAXS) measurements were used to determine the crystallite orientation and crystallinity. Comparisons between the as-extruded and drawn films show a clear transition from concentric rings characteristic of polycrystalline samples to short arcs (×110), which eventually become discrete spots (×110), suggesting improved alignment of the crystallites (×2.5). The orientation order parameter quickly increases from zero for the as-extruded films to nearly saturated value for perfectly aligned crystals at a draw ratio as low as ×2.5 (Fig. 4d). The thermal conductivity of the ×2.5 films (4.5 W m⁻¹ K⁻¹) was over 10 times larger than the as-extruded ones (0.38 W m⁻¹ K⁻¹, Fig. 3). We therefore expect the excellent alignment of the crystallites to be responsible for the limited thermal conductivity enhancement at very low draw ratio, which is consistent with the conventional strategies to improve the thermal transport in polymers.

However, after ×10 draw ratio where the orientation factor nearly saturates, we observed an additional 10-fold thermal conductivity enhancement to the 62 W m⁻¹ K⁻¹ (×110), which...
that during the stretching the crystallinity clearly suggested other enhancement mechanism. We noticed Measured and computed thermal conductivities for the polymer rate at low draw ratios (below ×10) and then steadily grew to over 90% in ×110 films (Fig. 4d, Supplementary Fig. 9f and Note 4). Contrary to the past work that emphasized on crystallinity-dependence of the thermal conductivity, the weak growth rate of crystallinity at high draw ratios is clearly not sufficient to account for the dramatic boost of the thermal conductivity, and there is even no sign of saturation of the conductivity as the draw ratio increases (Fig. 3).

These observations convinced us that unknown factors other than the crystalline phase play the crucial roles especially at high draw ratios. We therefore resorted to the structures of the amorphous region for clues. Quantitative analysis of small-angle X-ray scattering (SAXS) intensity profiles along the draw direction reveals two humps at scattering vectors that differ by a factor of two (Fig. 4e and Supplementary Note 4), indicating a periodic structure with a repeating unit consisting of alternating crystalline and amorphous phases (Fig. 4d, Supplementary Figs 10 and 11)33. This picture agrees with the widely-accepted lamell-like structural model for stretched polyethylene2. The displacement of the humps toward smaller scattering vectors with increasing draw ratio indicates that the period length grows with drawing. Normalized electron density profiles further reveal the relative lengths of crystalline and amorphous regions in each unit (Fig. 4f inset). Specifically, the amorphous fraction decreases with increasing draw ratio (Fig. 4f), consistent with the increasing trend of crystallinity (Fig. 4d) and film thermal conductivity (Fig. 3a). We have also converted the WAXS data in Fig. 4d into amorphous to superlattice total length ratio and plotted them in Fig. 4f to further illustrates the consistency between the SAXS and WAXS data (see Supplementary Note 4 for more details). However, decreasing the fraction of amorphous region alone cannot account for such observed ultrahigh conductivity, because amorphous phase is simply too thermally resistive6.

Discussion

To provide further evidence of the dominant role of amorphous region, a phenomenological one-dimensional thermal transport model is developed (Supplementary Note 5). Based on the structural parameters obtained in WAXS and SAXS, the crystalline and amorphous regions are randomly mixed in the extruded films. Upon stretching, aligned fibers consisting of alternating crystalline and amorphous regions are developed in the interior of the film. The average fiber diameter was estimated to be ~10–50 nm nanometers (Fig. 1i, j, Supplementary Figs. 3c and 11), justifying the use of a one-dimensional model $k = \frac{1 - \eta}{k_c + \eta/k_a}$ for the axial thermal conductivity. Here $\eta$ is the amorphous fraction in a periodic unit, and can be fitted from the SAXS analysis (Fig. 4f and Supplementary Note 5), while $k_c$ and $k_a$ are the thermal conductivities of the crystalline and amorphous regions, respectively. The crystalline thermal conductivity $k_c$ depends on the crystallite size due to phonon scatterings at laterally boundaries as well as the length direction. Huang et al.20 used first-principles to calculate thermal conductivity of 1D polyethylene chain and bulk crystals, and they also discussed size based on diffuse phonon scattering at boundaries. We choose to use their thermal conductivity data for 1D chain as a function of the chain length as values of the crystalline region, since diffuse boundary scattering could be too severe an assumption due to the weak interaction between the crystalline and amorphous phases and possibility continuity of polymer molecules from the crystalline to the amorphous region. Combined with our measured total thermal conductivity, these yield the amorphous thermal conductivity $k_a$ as the draw ratio (Fig. 3b). We do caution that this estimation is subject to uncertainties in $k_a$ and believe that our estimation of the amorphous region thermal conductivity represents a lower bound (Supplementary Note 5). It is clearly seen from Fig. 3b that the
experimentally measured high thermal conductivities at higher draw ratios suggest a high $k_a$ (~5.1 Wm$^{-1}$ K$^{-1}$ at ×50 and 16.2 Wm$^{-1}$ K$^{-1}$ at ×110 versus typical 0.3 Wm$^{-1}$ K$^{-1}$). In other words, the amorphous region after drawing is no longer composed of random disordered chains, but rather has developed some degrees of the orientation order with more extended and aligned chains. This is also consistent with our experimental observation that the isotropic amorphous diffusing ring gradually disappeared from ×10 to ×110 (WAXS, Fig. 4b and Supplementary Fig. 12), and consistent with the Raman study by Zhu et al. on further stretched Spectra fiber$^{17}$. The extracted high thermal conductivity of the amorphous region with some molecular
Fig. 4 Structural characterization using synchrotron X-ray scattering. a Illustration of the experimental setup and the orthorhombic unit cell of crystalline polyethylene. The incident beam is perpendicular to the drawn direction. The lattice constants were obtained as \( a = 7.42, b = 4.95, c = 2.54 \text{ Å} \), where \( c \)-axis is the chain direction. b Wide angle X-ray scattering (WAXS) patterns from the \( 1 \times 10 \) and \( 1 \times 110 \) films. Characteristic Bragg scattering by the \( (h0k) \) and \( (hk0) \) plane groups were observed. The \( (hk0) \) group appears perpendicular to the draw direction. c Small angle X-ray scattering (SAXS) patterns from the \( 1 \times 10 \) and \( 9 \times 90 \) films, which clearly show an isotropic-to-anisotropic transition. d First-order orientation parameter and the effective crystallinity obtained from WAXS. e Scattering intensity linecuts of the SAXS patterns along the draw direction. Two humps appeared at scattering vectors that differ by a factor of two, suggesting a periodic structure with a repeating unit consisting of alternating crystalline and amorphous phases (Supplementary Note 4). The humps moved toward a smaller \( q \) with increasing draw ratio, indicating an increase in the period length. f The fraction of amorphous region in one periodic unit as a function of draw ratio (Supplementary Fig. 14). The blue circles were directly extracted from the SAXS data, while the shaded zone marked the range (±40%) of fitted data which were used in the one-dimensional thermal model (Supplementary Note 5). The red squares recast the crystallinity data in Fig. 4d. Inset is the normalized electron density profile obtained from SAXS analysis (Supplementary Note 4).

orientation is much higher than that of oriented polyethylene fibers grown in a template19, despite theoretical prediction of higher thermal conductivity of polyethylene than polyethylene in crystal form34.

In summary, we have developed a scalable manufacturing process for producing polymer films with metal-like thermal conductivity. Unlike conventional approaches focusing on crystalline phase in polymers that can only marginally increase the thermal conductivity, we engineered the none-crystalline chain through disentanglement and alignment and achieved remarkably high thermal conductivity. The past few years have witnessed a surge in the interest of using polymers for thermal management and energy conversion. We believe that the high thermal conductivity achieved in these polymer films, with their unique combination of characteristics (light weight, optical transparency, chemical stability etc.) will play a key role in many existing and unforeseen applications. Of course, polyethylene itself has limitations in the temperature range it can cover. We foresee that further improvement of the thermal conductivity of the persistent amorphous phase will be the key developing the next generation of heat-conducting polymers, in polyethylene and beyond.

Methods

Fabrication of thermally conductive polyethylene films. See Supplementary Note 1 for more details on solution preparation, extrusion, and drawing process. The draw ratios were obtained as the ratio of final to initial film length, with ~20% uncertainty.

Thermal conductivity measurements. Direct measurement of the electrical heating power \( (P_\text{el}) \) as a function of temperature difference \( (T_S - T_C) \) across a sample film was performed (Fig. 2a, Supplementary Fig. 1 and Supplementary Tables 1, 2 and Note 2)35. Briefly, \( T_S \) (303 K, hot clamp temperature) was kept constant via feedback control of \( T_C \) while \( T_C \) (cold clamp temperature) was reduced to create a small temperature difference (up to 10 K) by systematically increasing the thermolectric cooling power. Multiple measurements of \( P_\text{el} \) were performed at a given temperature difference once the system had reached steady state (Supplementary Note 2). Subsequently, the slope of the linear fit yields, according to Fourier’s law and after correction for thermal shunting, the film thermal conductance \( (G) \) which further gives the film thermal conductivity \( (k) \) as \( k = G \cdot L/A \) (Supplementary Fig. 1b). Here, \( L \) and \( A \) are respectively the film length and cross-sectional area (Supplementary Tables 1 and 2), which were measured using a suite of tools including a micrometer, optical microscope and profilometer (Supplementary Fig. 2). Special effort was taken to minimize the thermal radiation exchange and to ensure that the reported thermal conductivity is conservative even if any residual radiation exists (Supplementary Note 2). The data that support the conclusions of this study are available from the corresponding authors on reasonable request.

Data availability

The data that support the findings of this study are available from the corresponding authors on reasonable request.

Received: 30 August 2017 Accepted: 21 March 2019
Published online: 16 April 2019
3. Rus, D. & Tolley, M. T. Design, fabrication and control of soft robots. *Nature Mater.* **536**, 266–268 (2016).

4. Chen, A., Liu, J. & Bao, Z. Pursuing prosthetic electronic skin. *Nature Mater.* **15**, 937–950 (2016).

5. Heeger, A. J. Semiconducting and metallic polymers: the fourth generation of polymeric materials (Nobel Lecture). *Angew. Chem. Int. Ed.* **40**, 2591–2611 (2001).

6. Sperling, L. H. Introduction to polymer science. (John Wiley & Sons, Inc., Lehigh University Bethlehem, Pennsylvania 2006).

7. Peacock, A. Handbook of polyethylene: structures, properties, and applications. (CRC Press, Taylor & Francis Group, Exxon Chemical Company, Baytown, Texas, 2013).

8. Balandin, A. A. Thermal properties of graphene and nanostructured carbon materials. *Nat. Mater.* **10**, 569–581 (2011).

9. Henry, A. & Chen, G. High thermal conductivity of single polyethylene chains using molecular dynamics simulations. *Phys. Rev. Lett.* **101**, 235502 (2008).

10. Fertig, E., Pasta, J. & Ulam, S. Studies of nonlinear problems. Report No. LA-1940 (Los Alamos Scientific Lab., N. Mex., United States, 1955).

11. Poulaert, B., Chielens, J. C., Vandenhende, C., Issi, J. P. & Legras, R. Thermal conductivity of highly oriented polyethylene fibers. *Polym. Compos.* **31**, 148–151 (2010).

12. Choy, C. L., Fei, Y. & Xi, T. G. Thermal conductivity of gel-spun polyethylene fibers. *J. Polym. Sci. Part B Polym. Phys.* **31**, 365–370 (1993).

13. Fujishiro, H., Ikebe, M., Kashima, T. & Yamanaka, A. Thermal conductivity and diffusivity of high-strength polymer fibers. *Ipn. J. Appl. Phys.* **36**, 5633–5637 (1997).

14. Chen, S., Henry, A., Tong, J., Zhong, R. & Chen, G. Polyethylene nanofibers with very high thermal conductivities. *Nat. Nanotechnol.* **5**, 251–255 (2010).

15. Wang, X., Ho, V., Segalman, R. A. & Cahill, D. G. Thermal conductivity of high-modulus polymer fibers. *Macromolecules* **46**, 4937–4943 (2013).

16. Ronca, S., Igarashi, T., Forte, G. & Rastogi, S. Metallic-like thermal conductivity in a lightweight insulator: solid-state processed ultra high molecular weight polyethylene tapes and films. *Polymer (Gould)* **123**, 203–210 (2017).

17. Zhu, B. et al. Novel polyethylene fibers of very high thermal conductivity enabled by amorphous restructuring. *ACS Omega* **2**, 3931–3944 (2017).

18. Kurabayashi, K. & Goodson, K. E. Impact of molecular orientation on thermal conduction in spun-coated polyimide films. *J. Appl. Phys.* **86**, 1925–1931 (1999).

19. Singh, V. et al. High thermal conductivity of chain-oriented amorphous polyethylene. *Nat. Nanotechnol.* **9**, 384–390 (2014).

20. Wang, X., Kaviani, M. & Huang, B. Further improvement of lattice thermal conductivity from bulk crystalline to 1-D chain polyethylene: a high-yet-finite thermal conductivity using first-principles calculation. https://arxiv.org/abs/1701.02428 (2017).

21. Wang, X., Kaviani, M. & Huang, B. Phonon coupling and transport in individual polyethylene chains: a comparison study with the bulk crystal. *Nanoscale* **9**, 18022–18031 (2017).

22. Chen, H. et al. Thermal conductivity of polymer-based composites: fundamentals and applications. *Prog. Polym. Sci.* **59**, 41–85 (2016).

23. Loomis, J. et al. Continuous fabrication platform for highly aligned polymer films. *Technology* **02**, 189–199 (2014).

24. Sweet, J. N., Roth, E. P. & Moss, M. Thermal conductivity of Inconel 718 and 304 stainless steel. *Int. J. Thermophys.* **8**, 593–606 (1987).

25. Arpaci, V. S., Kao, S.-H. & Selamet, A. *Introduction to Heat Transfer*. (Pearson, University of Michigan, USA, 2000).

26. Smith, D. E., Babcock, H. P. & Chu, S. Single-polymer dynamics in steady shear flow. *Science* **283**, 1724–1727 (1999).

27. Peterlin, A. Drawing and extrusion of semi-crystalline polymers. *Colloid Polym. Sci.* **265**, 357–382 (1987).

28. Kraemer, D. & Chen, G. A simple differential steady-state method to measure the thermal conductivity of solid bulk materials with high accuracy. *Rev. Sci. Instrum.* **85**, 023108 (2014).

29. Paddock, C. A. & Esley, G. L. Transient thermoreflectance from thin metal films. *J. Appl. Phys.* **60**, 285–290 (1986).

30. Cahill, D. G. et al. Nanoscale thermal transport. *J. Appl. Phys.* **93**, 793–818 (2003).

31. Schmidt, A. J., Chen, X. & Chen, G. Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermoreflectance. *Rev. Sci. Instrum.* **79**, 114902 (2008).

32. Deutsch, M. Orientational order determination in liquid crystals by X-ray diffraction. *Phys. Rev. A.* **44**, 8264–8270 (1991).

33. Feigin, L. A. & Svergun, D. I. Structure analysis by small-angle X-ray and neutron scattering. (Springer, Princeton Resources, Princeton, New Jersey, 1987).

34. Zhang, T., Wu, X. & Luo, T. Polymer nanofibers with outstanding thermal conductivity and thermal stability: fundamental linkage between molecular characteristics and macroscopic thermal properties. *J. Phys. Chem. C* **118**, 21148–21159 (2014).

**Acknowledgements**

The authors acknowledge support from Department of Energy/Office of Energy Efficiency & Renewable Energy/Advanced Manufacturing Program (DOE/EEREAMO) under award number DE-EE0005756 (for the fabrication platform and prior to 2/2016); the MIT Deshpande Center (9/2016–8/2017 for market studies), and U.S. Department of Energy (DOE)–Basic Energy Sciences (Award No. DE-FG02-02ER45977, for fundamental understanding of structure-property relationship since 7/2017). This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357. The authors thank T. Sanchez and N. Liu from MIT undergraduate research opportunities program for sample preparation; G. Ni for the photographt help; W. Dinatle and L. Wu for SEM discussions; C. Marks and D. Bell for microscopic discussions at the Center for Nanoscale Systems, Harvard University; N. Thoppy for his participation in the DOE project; S. Huberman, L. Meroesah and V. Chioyian for domain-temperature thermoreflectance discussions; C. Settens (CMSE, MIT); H. Li (Northeastern University), M. Minas (Northeastern University), S. Bälling (Columbia University), and M. Terban (Columbia University) for providing X-ray diffraction testing and discussions; G. Ni, J. Tong, S. Borotika, T. Cooper, Y. Huang, Q. Song, and L. Weinstein for manuscript discussions. S. Huberman, V. Chioyian, J. Mendoua, L. Meroesah, G. Ni, T. Cooper for the discussion and proofreading, and Jionghui Zhang and Professor Baoding Huang (The Hong Kong University of Science and Technology) for original simulation results on thermal conductivity (along the chain direction) as a function of the 1D polyethylene chain. We would like to note that our manuscript has been posted on arXiv: Yanfei Xu et al. Nanostructured polymer films with metal-like thermal conductivity. https://arxiv.org/abs/1708.06416 (2017).

**Author contributions**

The materials were fabricated by Y.X. The material thermal properties were characterized by D.K., B.S., Y.X. and J.Z. The material structures were characterized by Y.X. and Z.J. Structural and thermal modelling were performed by Z.J. and I.Z., respectively. Custom-built continuous production platform was built by J.L. J.W., M.L., H.G., X.H., X.L. participated in different phases of this project and contributed to the discussion and understanding of the materials. The manuscript was written by Y.X., B.S. and Z.J. with comments and inputs from all authors. G.C. directed the research.

**Additional information**

Supplementary Information accompanies this paper at https://doi.org/10.1038/s41467-019-09697-7.

**Competing interests:** A US Patent No. 9,109,846 has been granted and another patent application has been filed. Continuous fabrication platform for highly aligned polymer films.

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**Journal peer review information:** Nature Communications thanks Joseph Feser and the other anonymous reviewer(s) for their contribution to the peer review of this work. Peer reviewer reports are available.

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