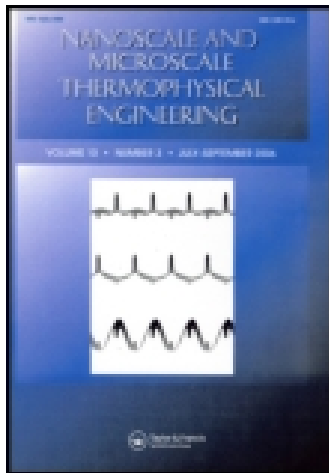


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## Nanoscale and Microscale Thermophysical Engineering

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### Evaluating Broader Impacts of Nanoscale Thermal Transport Research

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## EVALUATING BROADER IMPACTS OF NANOSCALE THERMAL TRANSPORT RESEARCH

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*The past two decades have witnessed the emergence and rapid growth of the research field of nanoscale thermal transport. Much of the work in this field has been fundamental studies that have explored the mechanisms of heat transport in nanoscale films, wires, particles, interfaces, and channels. However, in recent years there has been an increasing emphasis on utilizing the fundamental knowledge gained toward understanding and improving*

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*device and system performances. In this opinion article, an attempt is made to provide an evaluation of the existing and potential impacts of the basic research efforts in this field on the developments of the heat transfer discipline, workforce, and a number of technologies, including heat-assisted magnetic recording, phase change memories, thermal management of microelectronics, thermoelectric energy conversion, thermal energy storage, building and vehicle heating and cooling, manufacturing, and biomedical devices. The goal is to identify successful examples, significant challenges, and potential opportunities where thermal science research in nanoscale has been or will be a game changer.*

**KEY WORDS:** nanoscale thermal transport, broader impacts, nanotechnology, nanomaterials, heat transfer

## INTRODUCTION

Because of its high relevance to societal needs, heat transfer research has had a long history spanning over 300 years. The formulation of Newton's law of cooling [1] and Fourier's law [2] in 1701 and 1822, respectively, established the foundations for studying convection and conduction, two of the three heat transfer modes. In comparison, comprehensive understanding of the third heat transfer mode, thermal radiation, began with the discovery of Planck's law in 1900 [3], which played an important role in the development of quantum physics, a scientific revolution that has profoundly impacted both basic science and modern technologies. Though it was known that the three foundational laws in heat transfer could break down at small length or timescales, the need for a paradigm shift was largely absent until the rapid expansion of research and development activities in nanotechnology over the past two decades. The recent advances in top-down patterning and bottom-up synthesis methods have led to the fabrication of nanoscale and atomic scale particles, wires, films, and channels for applications in information, medical, and energy technologies. As one notable example, the printed line widths of ultra-large-scale integrated circuit devices are now as small as tens of nanometers. The characteristic length of these and some other devices and functional materials is now comparable to or shorter than the mean free path or wavelength of the heat carriers, which include molecules, photons, electrons, phonons, and magnons. As such, these new problems cannot be adequately treated by the three classical heat transfer laws. This circumstance has stimulated the emergence and expansion of the research field of nanoscale thermal transport over the past two decades.

This multidisciplinary research field has benefited from several major breakthroughs in experimental techniques in the broader nanoscale science and technology research community, including scanning probe microscopy [4, 5], nanofabrication, ultrafast lasers, and spectroscopy. In addition, it has taken advantage of the rapid development in large-scale parallel computing. These new capabilities have enabled a number of remarkable feats, which could have been deemed unfeasible two decades ago. For example, several experimental techniques have been invented to probe heat conduction in devices and materials as small as atomic scale structures such as individual wires, films, and interfaces or as fast as the femtosecond scale which is comparable to the relaxation time of the energy carriers [6–11]. In addition, first-principle theoretical computation has been able to predict thermal properties of a number of materials without the use of adjustable fitting parameters [12]. Though these accomplishments are impressive and the research activities in nanoscale thermal transport have reenergized basic research in heat transfer, which has often been perceived as a mature discipline, there have been increasing questions about the broader impacts of basic research in nanoscale thermal transport on technological applications.

In this opinion article, we attempt to evaluate the broader impacts of basic research in nanoscale thermal transport on the heat transfer discipline, workforce development, and technologies that address societal needs. Instead of providing a comprehensive coverage, we highlight several examples where nanoscale thermal transport research has already made or will potentially make a positive impact. These examples include heat-assisted magnetic recording (HAMR), phase change memories, thermal management of microelectronics, solid state thermoelectric energy conversion, heating and cooling of buildings and vehicles, localized heating-assisted nanomanufacturing, and nanoparticles for thermal therapeutics. The goal is to clarify the technological and societal relevance of nanoscale thermal transport research and to suggest future directions for this rapidly evolving field.

## FUNDAMENTAL HEAT TRANSFER DISCIPLINE

### Conduction

Heat conduction is a traditional cornerstone of nanoscale thermal transport research. Some highlights are discussed in a recent survey [13], reviews [14–16], and a focused volume [17]. The workhorse techniques for measuring thermal transport in thin films and across interfaces are thermorefectance [16] and electrothermal methods [6, 18]. Remarkable measurements of individual nanowires and nanotubes use microfabricated bridges [19, 20] and sometimes integrate atomic-level imaging [21–23]. Other experimental accomplishments have involved phonon mean free path spectroscopy [24–27], atomically thin two-dimensional (2D) materials [11, 28, 29], potential phonon coherence [30, 31], ultralow thermal conductivity [23, 32], scanning thermal probes [33, 34], and X-ray and neutron scattering techniques [35–37].

Conduction modeling has also seen great advances. One highlight is first-principles calculations of the thermal conductivity with no free parameters [12, 38–40], which also include interface resistance calculations using Green's functions [41, 42]. Insightful real-space simulations using molecular dynamics (MD) and wavepackets are now routine [43–46]. Conceptual advances enable both rigorous and intuitive understanding of increasingly sophisticated experiments such as mean free path spectroscopy [47, 48] and the sometimes dramatic effects of anisotropy [32, 49, 50].

The outlook for continued fundamental advances in nanoscale heat conduction is quite promising. Both computational and analytical innovations are needed to advance to larger and more complicated unit cells. A truly robust first-principles framework would be agnostic to the choice of the size of the unit cell. For example, in silicon a robust calculation should give the same thermal conductivity whether using the primitive unit cell with a two-atom basis, a conventional unit cell with an eight-atom basis, or even a much larger unit cell with  $10^6$  atoms in the basis. However, the latter is not possible with current first-principles schemes due to the tremendous increase in computational time for larger unit cells. Furthermore, existing first-principles algorithms are based on the phonon Boltzmann equation, which invokes the concept of a mean free path [12, 38]. Because the phonon mean free path cannot be smaller than its wavelength, and the smallest wavelength is larger than the unit cell, the Boltzmann equation is not appropriate for strong scattering within a single unit cell. This represents a serious limitation for modeling both complex thermoelectric materials, which may have short mean free paths but tens or even more than one hundred atoms in the primitive unit cell, and amorphous materials which, strictly speaking, have an infinitely large unit cell. Theoretical advances provide hope for much

needed cross-validations between theory and experiment without any free parameters and for problems ranging from thermal phonon coherence to thermal boundary conductance. Nonlinear and actively controlled heat conduction is another opportunity, including thermal diodes and switches [51–55]. Unconventional carrier types such as magnons are also of interest [56–60]. Finally, following the tremendous advances of the last decade [61–63], experimentalists will continue to push spatial and temporal resolution limits.

### Convection and Phase Change Heat Transfer

Nanoscale thermal science research can make important contributions to our fundamental knowledge of convection and liquid–vapor phase change, with impacts in medical diagnosis and therapy [64, 65] and thermal management [66, 67]. Improved numerical methods that integrate small-scale mechanistic models into larger scale models of fluid flow and heat transfer are recommended to advance understanding of boiling phenomena [68]. One such small-scale modeling approach is MD simulation (e.g., [69]), which captures subcontinuum features of solid–liquid interfacial heat transfer and phase change. MD addresses deficiencies arising from inadequate specification of boundary conditions in continuum models and thus may enable better descriptions of interfacial phenomena such as hydrodynamic slip at the solid–liquid interface and contact angle [70]. Additionally, it has the potential to resolve disagreements between experiments and macroscopic heat transfer models on solar steam generation in nanofluids [71] and may provide insight into their increased critical heat flux [72, 73]. An important deficiency is the limited length scales (maximum only approximately nanometer) that can be treated. To extend molecular-scale insight to the length scales of interest in engineering practice, multiscale techniques that treat interfaces atomistically while treating other regions with less computationally expensive continuum approaches should be pursued further. Examples include hybrid approaches that couple atomistics with computational fluid dynamics [74–76] and atomistics with fluctuating hydrodynamics [77, 78].

Advanced experimental techniques will also be essential to probe fundamental liquid–vapor phase change processes [66]. In a recent special issue of *Nanoscale and Microscale Thermophysical Engineering*, current experimental challenges in boiling [68], condensation [79], thin film evaporation [70], metrology [80], and fabrication [81] of micro- and nanostructured phase change surfaces were reviewed. Combined diagnostic measurements for multiple quantities including temperature, heat flux, and velocity are needed [80]. Efforts to increase spatial, temporal, and temperature resolution should be pursued. A variety of complementary diagnostics including electrothermal, optical, photothermal, and photoacoustic methods are promising in this regard, as are high-speed video, particle image velocimetry, microfabricated platforms, and single bubble generation/measurement devices [80].

### Radiation

Radiative thermal transport in the near field—that is, in the regime where the spatial separation between a hot and a cold surface is less than the Wien’s thermal wavelength—has potential [82–85] for achieving radiative heat fluxes that are orders of magnitude larger than those predicted by far-field radiative heat transfer calculations that are based on Planck’s law. Near-field radiative effects are expected to hold promise for several technological applications in energy conversion and thermal management [86–90]. In order

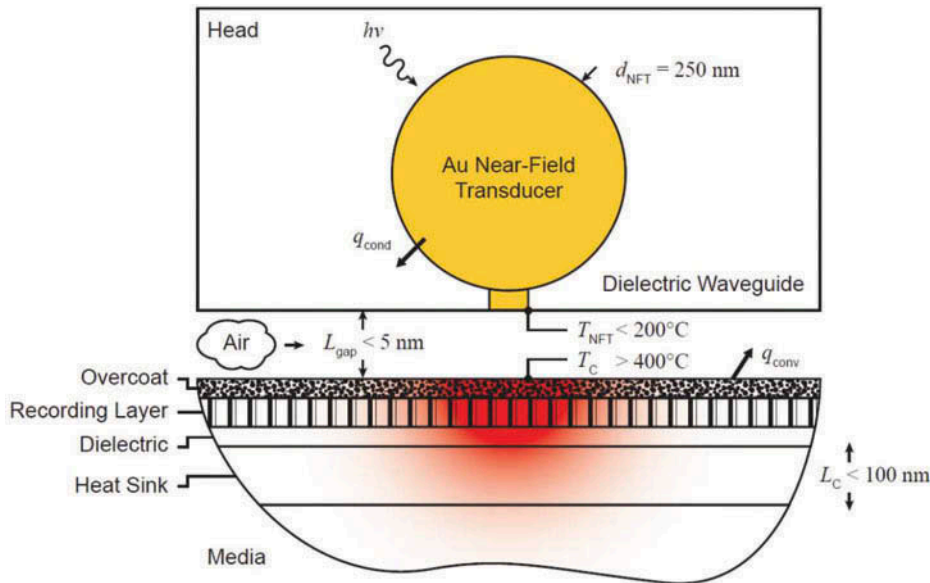
to quantitatively describe near-field radiative heat transfer, Polder and Van Hove [91] developed a framework by employing the formalism of fluctuation electrodynamics [92]. Several researchers have used this approach to computationally understand near-field heat transfer between metals [91], dielectrics [93, 94], and doped semiconductors [89]. Such studies suggest several intriguing possibilities, including near-field thermal rectification [88, 89], near-field thermal modulation using phase change materials [86, 87], and enhancement of near-field radiative transport by creating nanoporous surfaces [95].

In comparison to the considerable number of theoretical studies, experimental work on near-field thermal radiation is rather limited due to the experimental challenges. Specifically, to date all of the quantitative measurements [11, 96–99] of near-field heat transfer between parallel plates have been limited to micrometer-sized gaps. In order to circumvent the experimental difficulties involved in making parallel-plate measurements, researchers have exploited the sphere–plane geometry [82–84] or the tip–plane geometry [100] where it is possible to attain nanoscale gaps. Further, recent experiments [85] have also managed to probe the spectral characteristics of near-field radiation using near-field scanning probe techniques. Insights obtained from such experiments indeed confirm the basic validity of the theoretical predictions. However, there is a strong need for novel approaches to probing near-field radiative heat transfer to test numerous computational predictions that have remained unverified. These experiments are key to realizing the potential of near-field thermal phenomena in technological applications such as thermophotovoltaics, near-field-based thermal management, high-resolution calorimetry, and others. A detailed examination of the progress, impacts, and future opportunities of basic research in near-field thermal radiation is offered in an opinion paper published in this special issue [101]. In addition, the following section on heat-assisted magnetic recording describes a vivid example of the technological impact of this and other topics in nanoscale thermal transport research.

## HEAT-ASSISTED MAGNETIC RECORDING

Scientific understanding and measurement tools developed in the field of nanoscale thermal transport are enabling the development of HAMR [102] as a next-generation technology for data storage and will continue to play a role as this technology matures in the next decade. In conventional magnetic recording technology, the position of the magnetic bit of information in the recording media is determined by the spatial configuration and timing of a magnetic field applied to the recording layer by the pole piece of the read–write head. The size of the magnetic bit is currently limited to about 50 nm, because for state-of-the-art recording layers smaller bits of magnetic information do not have sufficient long-term stability against thermal fluctuations that randomize the information [103, 104]. Growth in storage density in the past decade has been truly remarkable, but significant further increases in storage density are not possible using current technology because any increase in the hardness of the recording layer—and therefore thermal stability of the layer—will render the recording layer impossible to write using any feasible magnetic field strength.

HAMR circumvents these problems by applying a nanoscale temperature excursion to write data in high-coercivity material at sub-50-nm bit size [102], a striking example of the engineering of thermal transport at the nanoscale to enable revolutionary advances in information technology. A schematic depiction of HAMR technology is presented in Figure 1 [105]. Issues of thermal science and engineering at the nanoscale are encountered



**Figure 1** Schematic diagram of heat-assisted magnetic recording (HAMR). The Au thin-film “near-field transducer” (NFT) is a part of the read/write head. Near-infrared light from a diode laser is coupled into the NFT by a dielectric waveguide. Electromagnetic radiation emitted from the protrusion at the bottom of the NFT is used to heat a nanoscale region of the recording media. The heated region is depicted as a reddened area in the figure. The gap between the head and the recording media is approximately 5 nm. The recording media is made of several layers optimized to control microstructure and the magnetic and thermal transport properties.

in both the read/write head and the media. In the head, a critical issue is thermal management of the near-field transducer (NFT) that is used to focus near infrared light from a laser diode into a  $<25 \times 25 \text{ nm}^2$  region of the recording media. Absorption in the Au NFT produces substantial heating that must be efficiently dissipated into the supporting materials to limit the temperature excursion below  $200^\circ\text{C}$ , above which the NFT slowly fails as surface diffusion alters the shape of the transducer. Detailed understanding of the temperature field in the read/write head is also essential for controlling the head media spacing that is currently  $<10 \text{ nm}$  and will need to decrease further to achieve storage densities envisioned in a fully implemented HAMR technology. Better methods for determining the temperature of the NFT with high time resolution during optical excitation are needed.

For conventional recording media and processes, thermal energy limits the recording density because thermal energy sets a lower limit on the size of a magnetic bit that is fully stable and can maintain its magnetic state for many years. In the HAMR media and recording process, thermal energy becomes a useful tool for transiently reducing the magnetic hardness so that the magnetization of the heated region can be altered by the magnetic field of the pole-piece. In HAMR media, the dimensions and precise placement of the bit of magnetic information are controlled by spatial and temporal evolution of the temperature field in the media that is produced by heating by the NFT. The media moves past the head at a speed on the order of  $10 \text{ m/s}$ , and the desired bit size is on the order of  $10 \text{ nm}$ . Therefore, the temperature excursion of the media must be on the order of  $1 \text{ ns}$ . Lateral heat flow smoothens out the lateral temperature gradient and produces undesired lateral fluctuations in location of the magnetic bit. Fortunately, composite recording media



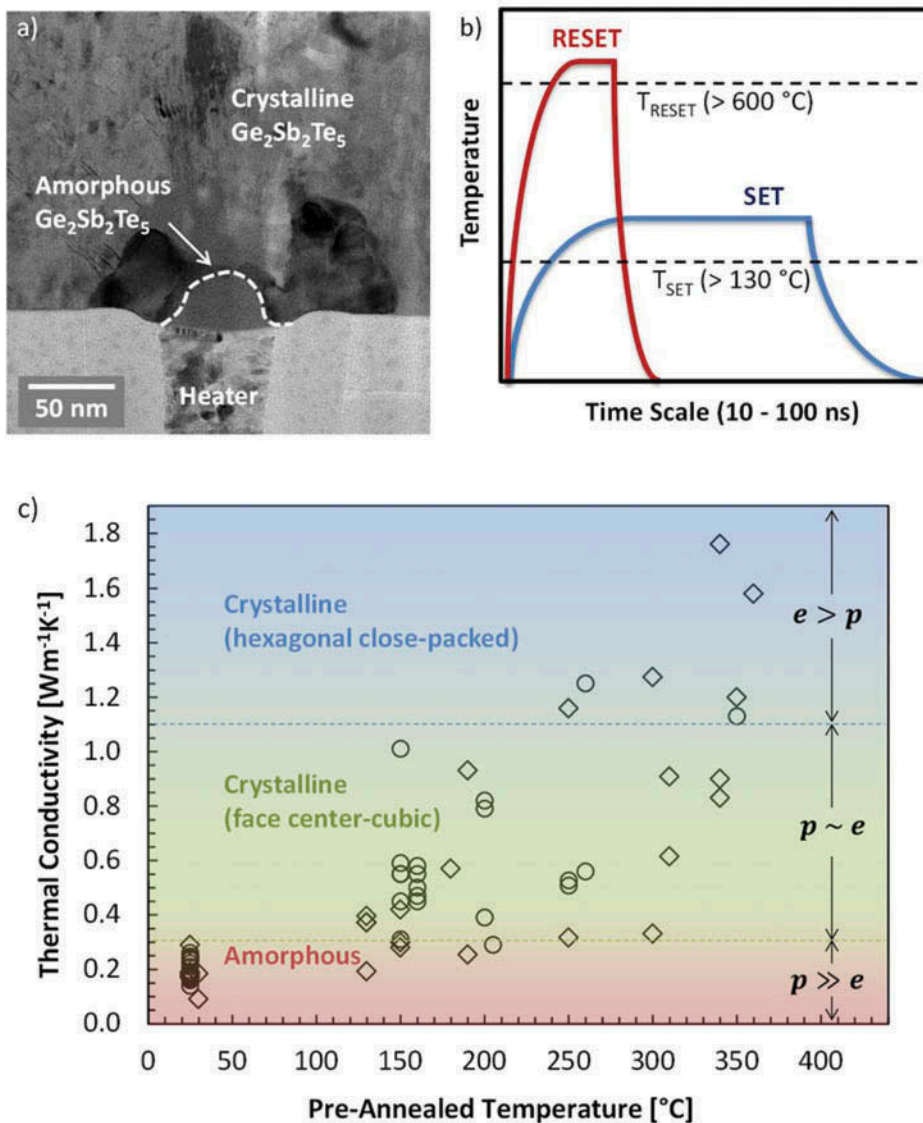
have small lateral thermal conductivity because of high densities of interfaces between the ferromagnetic grains (often a modification of FePt) and the nonmagnetic phase (often diamond-like carbon) that separates the grains. However, precise values for the lateral thermal conductivity of the recording layer are not yet known and will require further advances in experimental methods. Heat flow in the direction perpendicular to the surface of the media is relatively easily characterized using, for example, methods based on time-domain thermorefectance (TDTR) [106, 107] and are important for understanding the lifetime of the temperature excursion as heat moves from the recording layer through the various layers of the media that are needed to control the microstructure of the recording layer. There are engineering tradeoffs in the thermal design: better confinement of heat in the recording layer reduces the amount of optical power needed to heat the recording layer, but better confinement of heat increases the duration of the temperature excursion. Ideally, materials that would dynamically change their thermal transport properties during the temperature excursion could provide futuristic technology where engineers can separately optimize the energy deposition and heat dissipation in the recording media.

The advanced experimental techniques and modeling approaches that have been developed by the academic community in the past decade—for example, TDTR [106, 107]—are being used by the industry leaders to characterize and thereby engineer the thermal properties of the HAMR recording media and recording head. A group at Western Digital recently reported a study of thermal properties of HAMR recording media using TDTR [108]. A time-domain thermorefectance measurement setup is also in operation at the Seagate Technology research development facility in Minnesota.

## PHASE CHANGE MEMORIES

The nanoscale heat transfer community has had a significant impact on the thermal characterization and detailed engineering of materials and devices for phase change memory (PCM) technology. PCM is a thermally based data storage that requires temperature transient-driven phase transitions in a chalcogenide material, as shown in Figures 2a and 2b. This technology has been commercialized by Samsung and Micron [109, 110] and researched extensively by companies including IBM and Intel. In contrast to other microelectronic technologies, in which heat removal is a secondary but critical part of component design, thermal transport plays a central role in all figures of merit for PCM, including data writing rate and spatial density, energy consumption, and bit stability. Though it is not yet clear whether PCM will outpace flash memory, this technology has found several niche applications and offers great potential for universal memory applications [111–113]. Here we summarize the impacts of nanoscale thermal transport research in PCM technology development by considering (1) basic thermal properties characterization using electrical and optical approaches, (2) interpretation of electrothermal transport physics, and (3) innovative device designs.

The PCM research community has leveraged and extended the TDTR and  $3\omega$  methodologies to reveal a variety of the fascinating thermal transport phenomena that occur in the phase-change chalcogenides [114–130]. The TDTR methodology has proven useful for extracting coupled (and even decoupling [125]) film and interface properties. The  $3\omega$  measurement approach has been applied in the framework of novel experimental geometries to characterize the in-plane thermal conductivity [126] and the impact of rapid heating events [127]. Thermal transport studies of PCM materials are challenging due to complex impacts of the several material phases that can coexist depending on temperature



**Figure 2** (a) Cross-sectional transmission electron micrograph (TEM) of phase change memory devices, which store data bits through thermally induced phase transitions in  $\text{Ge}_2\text{Sb}_2\text{Te}_5$ . (b) Schematic illustration of the programming pulses, which require extreme temperature transients, which involve complex electrothermal phenomena. (c) Room-temperature thermal conductivity data for  $\text{Ge}_2\text{Sb}_2\text{Te}_5$  films [114–130]. The circles are obtained from the  $3\omega$  measurements [114, 115, 119–121, 126, 127] and the diamonds are obtained from the TDTR measurements [116–118, 122–125, 128–130]. Annealing at elevated temperatures induces phase transitions in  $\text{Ge}_2\text{Sb}_2\text{Te}_5$  and dictates the variation in the thermal conductivity, which is associated with varying contributions of electrons ( $e$ ) and phonons ( $p$ ).

history, geometry, neighboring materials, and process details [131, 132]. Rapid temperature transients and gradients can produce phase impurities, which have a strong influence on thermal transport [126, 127, 133], as well as electrical [134], thermoelectric [135], and mechanical properties [136]. An additional (and fundamentally enticing) complexity is

the strongly differing roles of electron and phonon transport depending on the material phase, as shown by the thermal conductivity data in Figure 2c. Though atomic vibrations are responsible for heat conduction in the amorphous phase, electron heat conduction can become important in at least one of the crystalline phases [116, 128]. The acoustic properties of the crystalline phases are very similar, and the difference in the thermal conductivity is attributed to the electron contribution, which is also in good agreement with predictions using the Wiedemann-Franz law and separate measurements of electrical properties [116, 128]. Heat conduction across interfaces with surrounding passivation and electrode materials ( $\text{SiO}_2$  [121, 123],  $\text{ZnS}:\text{SiO}_2$  [114],  $\text{TiN}$  [125, 128, 130],  $\text{Al}$  [129]) is governed by acoustic properties and interfacial disorder [125, 130].

The impact of the nanoscale heat transfer community extends to both practical and breakthrough improvements in the design of PCM devices. The thermal transport properties identified in the past studies allow complex device simulations accounting for phase impurities [133], thermal boundary resistance [137], and thermoelectric heating effects [138]. Precise knowledge of thermal transport phenomena and the resulting distributions of temperature, electric field, and current density have made important contributions to the improvement of existing designs and the discovery of breakthrough geometries. One example is multibit technology, which provides multiple levels of resistance (and hence overcomes the density limits posed by traditional binary storage) through thermally distributed phase transition regions [139–142]. The advancements in our understanding of transport properties have played a pivotal role in the improved accuracy of the simulation work that has been pursued by the broader research community on this topic. The same is true for the general topic of device optimization and engineering—thermal transport studies in PCM materials have enabled a new level of precision in device engineering by corporate leaders in the area that is sustaining the continued improvement of performance metrics for the technology.

## THERMAL MANAGEMENT OF MICROELECTRONICS

Thermal phenomena influence the performance and reliability of microelectronic devices and systems across a range of regimes from the package to transistors and interconnects. Thermal challenges have been amplified in modern systems as the industry moves toward three-dimensional (3D) chip stacks and lightweight, mobile platforms with minimal options for removing heat. Nanoscale transport can be important in all of these regimes, including for packaging, where novel material systems often involve nanoscale inclusions and complex interfacial phenomena. The nanoscale heat transfer community has contributed in a variety of ways to the engineering of microelectronic devices and systems, including (1) metrology development, (2) advanced simulation strategies at the device level, and (3) invention of promising composite media for packaging.

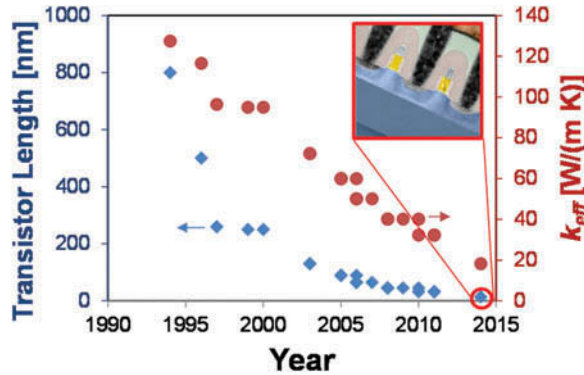
Microelectronic devices integrate materials with widely varying properties and feature sizes. Substantial developments in metrology over the past few decades have allowed quantification of the thermal properties of the individual materials comprising the device (ranging from the silicon device layer to thermal interface materials and packaging materials), interface thermal resistances between materials, and spatial mapping of device- and package-level temperatures, all of which are crucial for predicting and understanding thermal transport within microelectronic devices. A range of techniques including the thermorefectance [143–146], infrared microscopy [147–149],  $3\omega$  and related techniques

[150], and photoacoustic methods [151] have contributed strongly to the precise measurement of these properties. These methods have demonstrated the effective reduction of the thermal conductivity of silicon at the nanoscale, when the feature size [152] or the dimensions of the hot spot [153] are on the order of the phonon mean free path. Dielectric films, necessary for electrical insulation within microelectronic devices, have been evaluated from theoretical and experimental perspectives [154–158] aided largely by the development of the  $3\omega$  method [150]. Beyond the thermal conductivity, evaluation and understanding of the interfacial thermal resistance between the adjacent layers are often a limiting factor for reducing device temperatures. Thermoreflectance characterization of metal–dielectric interfaces has demonstrated the complexity of the electron–phonon interactions at the interfaces [159]. Specifically, at these interfaces, the electrons and phonons can depart significantly from equilibrium, a situation that can be resolved in some cases using a two-temperature model [160]. Beyond overall measurements of thermal interface resistance, spatially resolved techniques [161–163] provide further insight into the nonuniformity of the interfaces and can also identify regions of the interface suffering from delamination and voiding.

In addition to materials characterization, years of thermal transport studies have shed light on the temperature distributions both within individual transistors and within complete packages. Infrared thermography and thermoreflectance thermal imaging [164, 165] provide noncontact, micrometer- to submicrometer-resolution thermal images of operating devices, and atomic force microscopy (AFM)-based thermal mapping [7, 166] yields even higher resolution thermal maps. Compact thermal simulation methods [167–173] allow rapid design optimization and provide a framework for implementing dynamic thermal management strategies. In modern transistors, subcontinuum effects and complex electron–phonon interactions impact the temperature profiles. Over the past two decades, the impact of ballistic effects and electron–phonon interactions on device temperatures and performance has been demonstrated by detailed multicarrier phonon device simulations [174–177].

Nanocomposites are useful for microelectronics packaging replacing traditional thermal interface materials (TIMs) and potentially as novel phase-change energy storage materials within the package. Two strategies for developing high thermal conductivity, mechanically flexible nanocomposites TIMs include (1) adding high thermal conductivity nanoparticles as fillers to polymeric TIMs and (2) development of aligned films of nanowires and carbon nanotubes. Adding small weight percentages of carbon nanotubes or graphene nanoplatelets enhances the thermal conductivity of polymeric TIMs [178]. Aligning the nanoscale filler along the direction of heat flow can further enhance the thermal transport across the interface. Thus, vertically aligned carbon nanotube forests have been proposed for thermal interface applications for decades [179]. The highest quality lab-scale demonstrations show that aligned carbon nanotube forests can outperform metallic solders [180, 181], with the advantage of mechanical compliance [182] necessary to accommodate mismatch in thermal expansion. More recently, polymeric nanowire arrays [183] demonstrated low thermal resistances comparable with the best conventional thermal greases. Beyond TIM applications, transient spikes in power dissipation could be absorbed using phase change nanocomposite materials [184]. For encapsulated micro- or nanoparticles, the shell remains solid while the core absorbs energy during the melting process [185].

Current trends toward extreme power devices, portable and wearable electronics, and 3D logic/memory integration will require further development of thermally aware chip and



**Figure 3** Evolution of critical transistor length scale and a corresponding estimate of the associated effective thermal conductivity of the silicon domains in those devices at room temperature. Heat conduction is impaired by a variety of mechanisms as dimensions fall far below the mean free paths of the dominant phonons. Three-dimensional FinFET designs (as shown in the inset scanning electron micrograph as courtesy of International Business Machines Corporation, © International Business Machines Corporation [186]) are the latest advances as we approach the 14 nm node, and the accurate simulation of these devices requires phonon scattering considerations that have emerged, at least in part, from the nanoscale heat transfer community over the past two decades. Data for transistor length scale are from Zhirnov [315]. The corresponding thermal conductivity is estimated using the thermal conductivity integral approach with the critical transistor length scale as the limiting dimension [152].

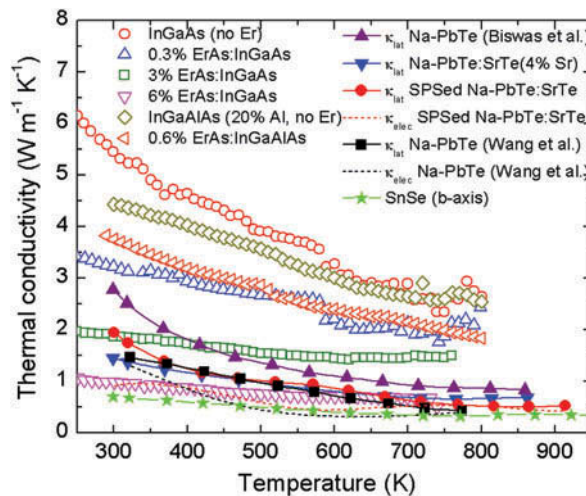
packaging designs. As an example, Figure 3 shows the evolution of transistor sizes in the past decades and estimates the corresponding decreasing size-dependent thermal conductivity of silicon. Novel 3D FinFET designs [186] push the transistor technology to the 14 nm node and merit further thermal design considerations. Additionally, the development of GaN high-electron-mobility transistors was limited by thermal challenges including significant self-heating due to high power densities and limited cooling strategies [187]. Precise characterization illustrated the importance of not only improving the heat spreading in the substrate (using SiC and diamond) but also minimizing the interfacial thermal resistances between the substrate and the GaN device layers [187, 188]. Integrating these detailed material and interface measurements with device simulations allows for design optimization before device fabrication.

## SOLID STATE THERMOELECTRIC ENERGY CONVERSION

More than 90% of primary energy is first converted to heat, but only 12% is transformed to end-use applications [189]. Currently, there are no commercially available large-scale waste heat recovery systems. Solid-state thermoelectric devices can directly convert heat to electricity and vice versa. The conversion efficiency of thermoelectric devices is directly related to the thermoelectric figure of merit,  $ZT = S^2\sigma T/\kappa$ , where  $S$  is the Seebeck coefficient,  $\sigma$  is the electrical conductivity,  $T$  is the absolute temperature, and  $\kappa$  is the thermal conductivity of the thermoelectric material [190]. If  $ZT$  is larger than 3–5, the thermoelectric device can be competitive with traditional mechanical energy conversion systems. The improvement in the efficiency of thermoelectric energy conversion has been slow because all the material properties comprising  $ZT$  are mutually coupled, and it is extremely difficult to enhance one property without affecting another. In a bulk material, it is very important to optimize the doping density for a large  $ZT$  because there

is a trade-off between the Seebeck coefficient and the electrical conductivity. Over the past few decades, several strategies have been proposed to enhance the Seebeck coefficient. One earlier idea is based on quantum confinement effect in low-dimensional materials [191, 192]. A recent work has examined the limitations of such effects in low-dimensional structures [193]. Other approaches are based on carrier energy filtering by planar or nonplanar heterostructure barriers [194, 195], band convergence with alloying [196], and resonant states by impurities [197]. The modulation doping scheme has been recently proposed to enhance the electrical conductivity [198]. However, recent breakthroughs for the enhancement of thermoelectric materials beyond  $ZT \sim 1$  have been achieved mostly by reducing the lattice thermal conductivity in nanostructured materials.

There have been studies about the optimal embedded nanoparticle size to minimize the lattice thermal conductivity of a solid. Mingo et al. [199] showed that 5–10 nm nanoparticles in SiGe alloys can reduce the lattice thermal conductivity from  $\sim 10$  to  $1\text{--}2 \text{ W m}^{-1} \text{ K}^{-1}$  over a wide temperature range. Less than 1% volume fraction of nanoparticles was necessary to make such a large reduction in the thermal conductivity when the nanoparticle size was optimized. In general, the lattice thermal conductivity decreases as the nanoparticle volume fraction increases in a material. As an example, when the ErAs nanoparticles of 2–4 nm in diameter were randomly distributed in InGaAs, the lattice thermal conductivity was lowered continuously with increased percentage of the nanoparticles as shown in Figure 4 [200]. However, the electron mobility was also reduced by the nanoparticles, which is detrimental to the thermoelectric energy conversion. Hence, the optimal  $ZT \sim 1.3$  at 800 K was found when 0.6–0.8% ErAs was introduced [201].



**Figure 4** Temperature-dependent lattice thermal conductivity of selected recent nanostructured thermoelectric materials: InGaAs with embedded ErAs nanoparticles (open symbols, Kim et al. [200]), Na-doped PbTe:(SrTe) series (filled triangles, inverted triangles, and circles, Biswas et al. [202]), Na-doped PbTe with optimized synthesis condition (filled squares, Wang et al. [218]), and SnSe crystal along the b-axis [215]. For the Na-doped PbTe:(SrTe) series, subsequent reduction of the lattice thermal conductivity is shown from no SrTe (filled triangles) to with 4% SrTe (filled inverted triangles) and to the sample prepared by spark plasma sintering with 4% SrTe (filled circles). For comparison with their lattice counterparts, the electronic thermal conductivities are also shown for the spark plasma-sintered Na-doped PbTe:(SrTe) (red dotted line, Biswas et al. [202]), and the Na-doped PbTe (black dotted line, Wang et al. [218]).

At elevated temperatures, near 600 K or higher, several PbTe-based nanostructured materials have shown high performance. Recently, a  $ZT \sim 2.2$  has been reported for the spark plasma-sintered Na-doped PbTe:SrTe, and the  $ZT$  enhancement was attributed to the all-scale hierarchical material structures that significantly reduced the lattice thermal conductivity [202]. By combining the atomic substitutions with Na impurities, the embedded SrTe nanocrystals, and the mesoscale grain boundaries created by spark plasma sintering, a wide range of phonon mean free paths could be effectively suppressed to achieve very low lattice thermal conductivity, about  $0.5 \text{ W m}^{-1} \text{ K}^{-1}$  at 900 K, as shown in Figure 4. One should note that typically the impact of increased phonon-defect and phonon-boundary scattering decreases as the temperature increases beyond the peak thermal conductivity value because the Umklapp processes start to dominate. It is worth investigating why in Na-doped PbTe:SrTe the enhanced  $ZT$  is only observed at very high temperatures compared to the structure without nanograins.

Another approach to reducing phonon thermal conductivity is found in skutterudites and clathrates that have complex cage-like crystal structures with voids in which “rattler” atoms are inserted, with the corresponding thermal conductivity reduction believed to be caused by some combination of resonant scattering by local rattling modes, increased Umklapp scattering, and/or group velocity reduction due to avoided crossings [35, 203]. Recently, an ab initio study found that coexistence of two stable phases in a partially filled skutterudite ( $\text{Ba}_x\text{Co}_4\text{Sb}_{12}$ ) can also be responsible for the very low thermal conductivity via enhanced phonon scattering at interfaces between the two phases [204]. Beyond 1000 K, SiGe has long been known to be a good thermoelectric material.  $ZT$  close to 1.3 at 1200 K was reported in nanostructured SiGe synthesized by hot pressing and ball milling [205]. The impact of the superlattice structure on lattice thermal conductivity has been attributed to various effects, including modification of the phonon spectrum (e.g., zone folding and band gap formation) [206] and phonon localization, diffuse or specular scattering of phonons at interfaces due to acoustic mismatch, and scattering of phonons at defects (e.g., dislocations from lattice mismatch) [207]. It is still unclear to what extent phonon behavior is coherent in these systems, which is a requirement for phonon spectrum modification. A low cross-plane thermal conductivity of  $0.22 \text{ W m}^{-1} \text{ K}^{-1}$  at 300 K was estimated for the  $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$  thin-film superlattice [208] using an extrapolation of the lattice contribution to thermal conductivity as a function of carrier concentration. This result needs to be further studied. Other examples include PbTe/PbTe<sub>0.75</sub>Se<sub>0.25</sub>, Si/Ge, and short-period AlAs/GaAs superlattice [209]. It has been claimed that rough silicon nanowires can have  $ZT \sim 0.6$  at room temperature and  $\sim 1$  at lower temperatures [210, 211]. Interestingly, recent studies analyzing a wide number of samples did not observe a clear correlation between the thermal conductivity and the nanowire diameter, suggesting that statistical characteristics of the surface roughness need to be taken into account [212–214]. The highest reported  $ZT \sim 2.6$  [215] to date is surprisingly for a binary material, undoped SnSe single crystal along the  $b$  axis, without complex alloying or nanostructuring. This high  $ZT$  is attributed to the extremely low thermal conductivity, which is lower than the value obtained in a recent theoretical calculation for the  $b$  axis [216]. In addition, the highest  $ZT$  reported in a recent work for doped polycrystalline SnSe is only about 0.6 [217], because the lattice thermal conductivity of the polycrystalline sample was found to be even higher than that reported earlier for the SnSe single crystal.

As the lattice thermal conductivity has been significantly lowered by phonon engineering, the thermal conduction by charge carriers may become a dominant thermal conduction mechanism in a solid. The electronic thermal conductivity is proportional to

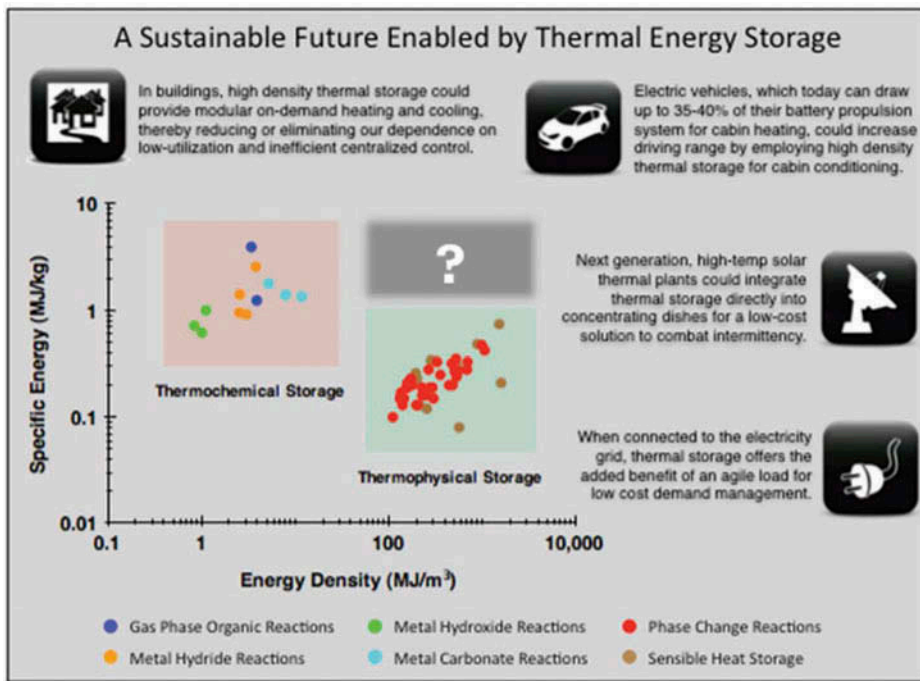
the electrical conductivity due to the Wiedemann-Franz relation. Typically thermoelectric materials are highly doped to achieve a high electrical conductivity, which results in a fairly high electronic thermal conductivity, comparable to the lattice thermal conductivity at high temperatures as shown in the Na-doped PbTe alloys in Figure 4. One can lower the doping level to reduce the electronic thermal conductivity. However, if the doping level is lowered greatly, the contribution from the minority carriers becomes nonnegligible, so that the detrimental bipolar thermal conductivity is added to the total thermal conductivity [218]. There are recent proposals to use heterostructure barriers to selectively suppress the transport of minority carriers with selective filtering of electrons or holes at the heterointerfaces [219]. Surprisingly, when the bipolar thermal conductivity is effectively suppressed, a very low doping level could be utilized to achieve further enhancement of the figure of merit, and a  $ZT$  higher than 3 is predicted for p-type PbTe at 900 K. It will be interesting to study if minority carrier blocking could be already in play in the PbTe nanocomposite with  $ZT \sim 2.2$  [202]. It is anticipated that the future in thermoelectric energy conversion will be promising only when the nanoscale thermal transport for both phonons and charge carriers can be simultaneously optimized.

## HEATING AND COOLING OF BUILDINGS AND VEHICLES

Currently, buildings and vehicles consume 70 quads/year (1 quad =  $1.055 \times 10^{18}$  J) out of 100 quads/year of primary energy used in United States. Cooling and heating of buildings alone accounts for 15 quads/year of primary energy [220]. If all other heating and cooling needs in buildings (e.g., refrigerators, hot water) are included, this number jumps to more than 20 quads/year [220]. Similarly, cooling of vehicles consumes more than 7 billion gallons of gasoline/year [221]. For internal combustion vehicles, heating can be provided from the heat dissipated in the radiator and is thus free; however, for electric vehicles that is not the case. Electric vehicles draw as much as 35–40% of their electrical battery capacity for cabin heating/cooling [222]. In internal combustion engine vehicles the fuel efficiency and pollution are significantly impacted by the temperature of the engine at the start [223]. Nanoscale thermal science has a very significant role to play in increasing the energy efficiency of buildings and vehicles by developing various technologies such as better thermal storage, thermal diodes, radiative emitters, and membranes.

High-energy-density thermal storage can provide modular on-demand heating and cooling, thereby reducing or eliminating the dependence on low-utilization inefficient centralized air conditioning. Another major impact of high-energy-density thermal energy storage could be in its use as an onboard thermal battery in electric vehicles to provide heating and cooling of the cabin. Conventional combustion engines vehicles could also benefit by using thermal storage to avoid cold engine starts to improve average fuel efficiency by 10% or more and reduce tailpipe emissions of unburned fuel [223]. Despite all these technological promises, there is a fundamental challenge. Thermal storage with both high volumetric and gravimetric energy density does not exist, as shown in Figure 5 [224]. Recent advances in materials such as metal organic frameworks or zeolites [225], which are inherently nanostructured, can lead to increased energy density by utilizing the binding energy and adsorptivity of adsorbents. High energy density must also be complemented by high thermal conductivity for thermal power delivery and charging time reduction of the storage system. High-thermal-conductivity, low-density nanostructured materials such as ultrathin graphite foams can be used as fillers in thermal storage materials to enhance the charging and discharging rates [226].





**Figure 5** Material-level theoretical volumetric and gravimetric energy densities for leading thermal storage systems are plotted, illustrating the distinct advantages of thermochemical and thermophysical approaches. Note that the energy densities have been calculated for a wide range of temperatures. As a point of reference, the active materials in a state-of-the-art lithium ion battery have volumetric and gravimetric energy density of roughly 5,000 MJ/m<sup>3</sup> and 1.3 MJ/kg; no existing thermal energy storage material has comparable performance. The inset text describes the potential impacts of a next-generation high-density thermal storage system.

Vapor compression systems are the most dominant technology for air conditioning of buildings. Because a conventional vapor compression system can only provide sensible cooling, it reduces the temperature to the dew point so that the relative humidity of the supply air is  $\sim 100\%$ , which results in the condensation of the water from the humid air. This significantly increases the work input into the cooling system. Sometimes depending on the relative humidity and the temperature, the dew point temperature can be very low, which in turn requires reheat to reach the desired indoor temperature. This leads to significant decrease in the effective coefficient of performance of the cooling system, which increases the cost to consumers. One approach is to separate the dehumidification step from the cooling step. For example, dehumidification can be provided by membranes that are pervious to water molecules and impervious to air molecules [227]. Manipulation of thermal and mass transport in nanostructured membranes provides exciting opportunities for both fundamental and applied research.

With the rapid advancement in metamaterials, which are artificial composite materials with microscopic geometries organized to engineer new properties, it is becoming possible to manipulate thermal energy transport in ways that were not previously possible. Some of these advancements can have significant impact on cooling and heating energy requirements. For example, recently it has been reported that nanostructured metal-dielectric photonic structures can provide passive cooling of buildings by emitting strongly

in the mid-infrared range to deep space [228]. It was shown that cooling power in excess of  $40 \text{ W/m}^2$  was possible even in direct sunlight, and similar concepts have been used to passively cool by more than  $40^\circ\text{C}$  at night [229]. Recently, thermal cloaking and inverters have also been reported [230]. Is it possible to use these ideas to thermally shield a building or a vehicle to reduce the cooling and heating needs? Despite the difficulty in predicting the future, basic research in nanostructured thermal materials and devices has the potential to significantly impact future energy technologies by increasing the efficiency of heating and cooling, which plays a significant role in our energy ecosystem.

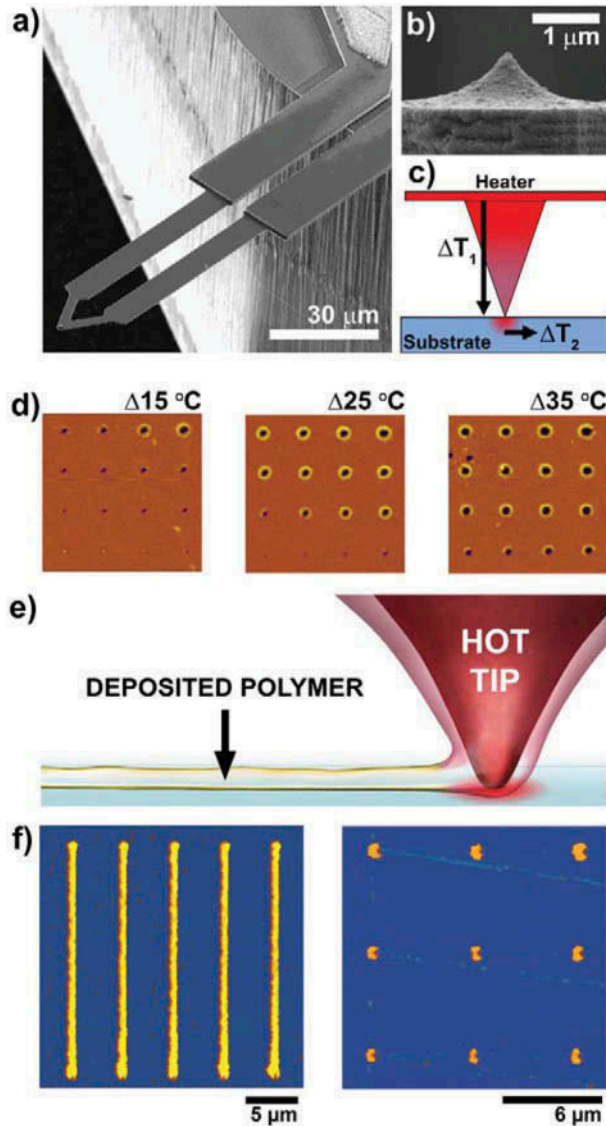
## HEAT ASSISTED NANOMANUFACTURING

Nearly every manufacturing process requires spatial and temporal control of temperature and heat flows. Nanometer-scale control of temperature and heat flows offers unique opportunities and challenges for nanomanufacturing. Recent research uses nanometer-scale heated probe tips for local temperature control in order to control manufacturing processes and local tuning of material properties. Figure 6 provides an overview of this research. Figure 6a shows an AFM probe with an integrated solid state heater and Figure 6b shows a sharp tip for local sample interaction [231–233].

A heated AFM tip can generate a large temperature increase at the interface between the tip and a substrate; in some cases, this temperature increase can reach several hundred degrees Celsius, which is sufficient to cause thermophysical changes such as melting [234] or thermochemical reactions [235, 236]. The substrate near the heated tip can have a large temperature gradient, which can reach  $10^8 \text{ K m}^{-1}$ . When such a large temperature gradient is applied to a liquid, temperature-induced gradients in surface tension can cause surface tension-driven flows [237].

When the hot AFM tip is in contact with a surface, two large temperature gradients develop: one at the tip-sample contact where the temperature rise extends roughly 20 nm past the tip-substrate interface and the second along the length of the tip from the cantilever heater to the surface [234, 238]. These temperature gradients can drive thermocapillary flows (Figure 6c), even when the temperature rise is only a few degrees [239]. Figure 6d shows pits formed in a molecular glass thin film using a heated tip, where changes in surface tension induced by the surface temperature gradient determines the final pit shape. Thermocapillary-driven nanolithography shows promise as an efficient, high-resolution lithographic method compatible with many materials.

The temperature gradient along the length of the tip can drive molten polymer flow from the cantilever tip onto the surface, a technique known as thermal dip-pen nanolithography (tDPN; Figures 6e–6f) [240, 241]. tDPN can be used to directly write nanometer-scale features out of many materials, including thermoplastic polymers, polymer-nanoparticle composites, and metals [242–245]. The deposited features can be as small as about 10 nm and as large as several micrometers. Recent advances in the fabrication of diamond tips allows for extremely low tip wear, such that the tip can scan over a surface for several meters [246, 247]. The mass flow rate from the tip onto the substrate, as well as the geometry of the final deposited structure, depend on the magnitude of the temperature gradient, Laplace pressure due to the tip curvature, and the solidification of the molten material [234]. Adjusting the heater and surface temperatures controls the geometry of the written structures. Recently, tDPN has been used to fabricate electronic devices from graphene nanoribbons and nanometer-scale mechanical resonators [248, 249].



**Figure 6** (a) Scanning electron microscope image of a heated atomic force microscope cantilever. (b) Closeup of the nanometer sharp tip at the end of the heated cantilever, with an ultrananocrystalline diamond coating to reduce wear. (c) Schematic of the temperature gradients: one along the length of the tip and one extending radially away from the tip–surface contact. (d) Thermocapillary transport of an organic thin film as a function of tip temperature and dwell time. (e) Schematic of thermocapillary mass transport from a hot tip to a cold surface. (f) Polyethylene features written with a heated tip during tip scanning or tip dwelling.

A cold tip on a hot surface also provides for nanometer-scale temperature control and the potential for nanomanufacturing. One example is a cold surface in close proximity with a hot polymer substrate, where the temperature gradient between the surfaces creates a fingering instability in the polymer and causes the polymer to flow toward the cold surface [250]. Lithographically patterned protrusions from the cold surface locally enhance

the temperature gradient, causing the polymer to selectively flow toward the protrusions [251]. This technique has been used to create polymeric micropillars, gratings, and optical structures [252, 253].

Nanometer-scale heating elements can also generate temperature gradients and thermocapillary flows in thin organic films, such as paraffin, pentacene, polystyrene, poly(methyl methacrylate), and molecular glasses [239, 254–256]. Such organic films are commonly used for nanolithography. In one example of thermally controlled nanolithography, carbon nanotubes were embedded in a thin film of a molecular glass. Joule heating of the electrically conducting nanotubes resulted in a temperature rise in each of these nanotubes and a temperature gradient near each of these nanotubes. The molecular glass near the heated nanotubes flowed away from the nanotubes, due to the temperature-induced softening of the molecular glass, as well as the gradient in surface tension. Under the same conditions, semiconducting nanotubes did not become hot, and the molecular glass near these nanotubes did not soften and flow. Thus, the heating conditions could be used to select either the conducting or the semiconducting nanotubes for fabrication [237]. The purification of semiconducting nanotubes is a key challenge for the field of nano-electronics, so the ability to isolate semiconducting nanotubes using heat is particularly exciting. The technique could be extended to other nanometer-scale hotspots generated; for example, at defects in carbon nanotubes or carbon nanotube junctions [257].

Future research into nanomanufacturing with nanometer-scale temperature control should focus on developing manufacturing processes that are inexpensive, facile, robust, and scalable. Atomic force microscope cantilever arrays are well suited for process scale up, and a number of examples exist in the recent literature for heated tips [258, 259]. Improved designs of heated tip arrays to better control thermocapillary mass transport would lead toward a deposition system capable of patterning structures made from a wide variety of organic materials, in registry with one another, in a robust and scalable fashion. Furthermore, most nanometer-scale heaters are joule heated, which requires electrical contact to the structures, complicating the lithographic process. Selectively heating structures without making physical contact, such as with electromagnetic radiation, would provide a simple, fast, and highly property-dependent route toward wafer-scale thermocapillary lithography.

## NANOPARTICLES FOR THERMAL THERAPEUTICS

Nanoparticle heating in therapeutic and preservation-based biomedical applications is an emerging area with transformative potential. Specifically, model inorganic nanoparticles, such as gold and magnetic (i.e., iron oxide) nanoparticles, can be functionalized to distribute, target, bind, and/or internalize in tissues, vessels, and specific cells for both cancer and regenerative medicine applications. The advantages of laser gold nanoparticle (NP) heating for subcellular to whole tissue applications are the speed and intensity of heating (typically several orders of magnitude higher than iron oxide); however, this gold NP technique has a disadvantage relating to the depth of penetration of light (typically less than a centimeter) into tissues [260]. For this reason, magnetic nanoparticles have also been pursued, because they have the ability to interact with physiologically innocuous deep penetrating electromagnetic fields (several centimeters) to provide heating in a number of new applications. Further, these iron oxide nanoparticles can be targeted to cancer or even bacterial cells to induce destruction [261–263]. Questions as to the extent to which these nanoparticles produce heating localized on the nanoscale versus bulk and whether this

produces enhanced therapeutic effects persist in the field [263, 264]. Answering these questions will provide new information that guides improved design and use of nanoparticles in a growing number of heating-based biomedical applications.

### Magnetic Nanoparticle Heating

At lower radio frequencies, uniform magnetic fields can penetrate through the entire body without significant interference or attenuation [265], thereby providing constant and uniform heating within deep-tissue NP deposits for the timescales (10–60 min) required for bulk tissue applications, such as cancer destruction or warming of vitrified tissues. Thus, magnetic fluid hyperthermia continues to be of considerable interest for cancer treatment and controlled warming of vitrified systems for regenerative medicine applications [266–268]. For this reason, it is imperative to identify biocompatible, reproducible, and high magnetic heating particles. Once these have been identified and characterized in the lab, they will need to be more fully characterized in partnership with the National Characterization Lab (<http://ncl.cancer.gov>).

On the heating side, a recent study showed that industrially produced superparamagnetic particles (single domain crystals  $\text{Fe}_3\text{O}_4 < 20$  nm) are more efficient heaters than ferromagnetic particles (multidomain  $> 20$  nm) on a per mass Fe per particle mass basis [269]. Interestingly, based on theory, additional heating is expected by more tightly controlling the superparamagnetic iron oxide size. Specifically,  $\text{Fe}_3\text{O}_4$  NPs smaller than  $\sim 20$  nm generally exhibit a single magnetic domain and demonstrate superparamagnetic behavior [270, 271]. Thus, heating due to relaxation losses by Néelian (rotation of the magnetic moments within the NP) and Brownian (due to viscous effects as the entire NP aligns with the external magnetic field) mechanisms are present. These losses depend on NP size, concentration, magnetic characteristics, shape, and crystalline structure and have been theoretically described but can vary significantly depending on the actual particle [272].

More recently it has also been demonstrated that the aggregation state of the superparamagnetic particle dramatically reduces the specific absorption rate of these particles [273]. Heating rates relevant to hyperthermia and regenerative medicine applications generally require  $\text{Fe}_3\text{O}_4$  NP concentrations on the order of 1 mg/mL in tissue [262, 269]. This is a difficult concentration to reach by intravenous injection followed by passive biodistribution, thereby often necessitating interstitial injection [269, 274].

One important question is to what extent these “bulk” heating techniques are also nanoscale in their thermal impact. More specifically, to what extent are nanoparticles measurably “hotter” than bulk during the heating process? Importantly,  $\text{Fe}_3\text{O}_4$  radio frequency (RF) heating requires milligram Fe/g tissue for bulk heating, whereas laser-excited gold nanoparticles can achieve similar heating at microgram Au/g tissue [260, 269]. Continuum analysis and scaling suggest that gold nanoparticle heating at appropriate fluences ( $\text{W}/\text{cm}^2$ ) can support  $\geq 1^\circ\text{C}$  at the nanoparticle surface [275, 276] and has been used to drive numerous applications from drug delivery to whole tumor heating, as recently reviewed [260]. However, scaling the heating for iron oxide NPs under clinically relevant fields (100s kHz and 10–20 kA/m) and loading suggests that only a very small fraction of a degree temperature increase would occur at the NP surface [262, 277, 278]. This controversy has been fed by a number of studies that have reported indirect (nonthermal) measurements that suggest magnetic nanoparticle surface heating in excess of 10s of degrees Celsius above bulk under applied alternating magnetic fields [279–282]. This conflicts strongly with scaling of RF

heat generation in bulk-loaded magnetic nanoparticle systems that suggest only a small fraction of degrees Celsius temperature rise at the nanoparticle surface relative to their surroundings, particularly in cellular systems [262, 269, 277, 278]. This seeming paradox remains unresolved in part due to the difficulty of directly measuring temperature at the nanoscale and our ability to perform the measurement noninvasively on live cells.

### **Laser Heating of Gold Nanoparticles**

A recent review highlights the use of laser heating of gold nanoparticles for biomedical applications at the subcellular, cellular, and tissue levels [260]. Example applications include gene activation or silencing, molecular surgery, nanoparticle or endosomal drug release, protein denaturation, single cell destruction, and photothermal therapy of cancer [260]. All of these applications rely on NP type, concentration, distribution, and aggregation state to create local heat generation [260, 278]. This heat generation is relatively well understood for idealized conditions and, as already stated, can exceed RF heating of  $\text{Fe}_3\text{O}_4$  particles by several orders of magnitude for readily accessible lab or clinical conditions [278, 283–285]. However, under real conditions the NP distribution is not homogeneous; for example, it is often higher around blood vessels [286–289]. Further, in the case of NP aggregation, the absorption may be red-shifted and enhanced under optical excitation [275, 278]. In addition, optical properties such as the dielectric constant that determines the absorption cross section ( $C_{\text{abs}}$ ) are hydration and temperature dependent and thus change during heating. These nonideal factors result in different optical properties from the idealized conditions and thus require further study, especially in biological systems (i.e., solutions, gels, cells, and tissues). Though aggregation dramatically reduces the RF heat generation in  $\text{Fe}_3\text{O}_4$  [273], similar studies in gold or gold-decorated systems remain to be performed in phantoms, cells, and tissues.

### **Nanoscale and Nanoparticle Thermometry**

With the advent of new methods to heat biomaterials at the nanoscale and particularly with nanoparticles, new temperature measurement techniques are increasingly desirable. These measurements can provide detailed thermal information around nanoparticles as well as assess thermal lesion during the tissue and/or cell treatments. Although many nanoscale temperature measurement techniques such as scanning thermal microscopy have been developed or are under development as reviewed elsewhere [16], its measurement in biological and physiological environments has not been well established and is an area of active research. One attractive approach is based on the use of fluorescence nanomaterials [290–293] because of its compatibility with aqueous environments. In addition, it can be easily integrated with other biomedical optical techniques for noninvasive temperature measurements. As an example, quantum dot (QD) has been used for thermometry because it has been known that the wavelength of their peak emission and the fluorescence intensity of QDs vary considerably with temperature [294, 295]. Using these temperature-dependent fluorescence characteristics, a QD-mediated thermometry technique has been developed to provide intraoperative guidance of thermal surgeries [296–298]. In addition to surgical guidance, the thermometry has been extended to measure nanoscale as well as subcellular thermal features [290, 293, 299, 300]. Although this technique can address many challenges of micro- and nanoscale temperature measurements for biomedical applications, further research is warranted when it combines with nanoparticle-mediated heating

modalities. Recent studies have reported the observation of strong near-field interaction between plasmonic nanoparticles and QDs, which substantially increase QD's quantum yield [301, 302]. However, detailed mechanism of this near-field interaction and its implication is still not well understood. In order to accurately measure temperatures around nanoparticles, the significance of this near-field interaction should be further elucidated.

## WORKFORCE DEVELOPMENT

The nanoscale thermal transport community has produced an exceptionally large number of researchers with doctoral degrees who have impacted both academia and industry, in the United States and abroad. As one metric to quantify this impact, one may examine the doctoral and postdoctoral trainees from a sampling of well-established laboratories in the nanoscale transport community. Such an analysis of six representative leading groups reveals that over the last decade alone, this community has created at least 50 new university faculty members in a range of institutions. At least an equal number have gone on to work in companies like Intel, IBM, AMD, Freescale, Applied Materials, Seagate, Tesla Motors, Ford, Alphabet Energy, Romny Scientific, and GMZ Energy and national laboratories such as Lawrence Livermore and Sandia.

The contributions of this large number of researchers have impacted a variety of areas including thermal metrology, scanning probe microscopy, semiconductor fabrication, thermal management of electronics and batteries, thermoelectric materials, and novel high-density memory. It should be noted that the community has resulted in at least 15 from the groups sampled above leading women researchers at academic institutions and industrial labs. Further, students from this community have also become leading researchers in a variety of areas other than thermal transport, such as nanofluidics, cellular mechanics, and surface science and engineering.

The strong impact of the community on both academic and industrial research can be traced to the broad and deep student training it has provided in both science and engineering. Specifically, the community has extensively developed graduate courses on nanoscale heat and energy transport. This commitment to education is also reflected in the multiple graduate-level textbooks they have written in this area [303–306]. It is also noted that members of the community have developed online courses that are accessible to a broad range of researchers in academia and industry [307–309].

Finally, we note that the nanoscale transport community has provided abundant leadership to the broader academic and scientific community in multiple roles, with more than 15 active researchers in the field having served as chancellors or presidents of universities, college deans, departmental heads, program directors at funding agencies, and directors of governmental agencies.

## SUMMARY AND OUTLOOK

These discussions show that basic research in nanoscale thermal transport over the past two decades has made a positive impact not only in the fundamental heat transfer discipline but also on workforce development and technologies that address the societal needs. As two of the notable examples, the metrology techniques invented by this research community have helped the development of heat-assisted magnetic recording and phase change memories, two technologies that were still at the early stage of research two decades ago and are now entering the marketplace. In another example, the recent findings of increased

thermoelectric figure of merit in PbTe-based alloys has led to the adoption of these materials in the radioisotope thermoelectric power source of the Mars Science Lab rover Curiosity [310], which was successfully landed on the Red Planet on 6 August 2012.

In addition, nanoscale thermal transport research has the potential to make even broader impacts by focusing on use-inspired basic research relevant to a number of grand challenges. As one example, the high heat flux and hot spots in semiconductor devices have become the major barriers for the microelectronic industry to follow Moore's law to reduce the size and increase the density and performance of transistor devices. In addition, the emergence of flexible mobile electronic devices has given rise to a different set of thermal management challenges associated with the low thermal conductivity of flexible substrates and space constraint of these devices. Recent and future breakthroughs in experimental and computational research of heat spreading materials, microfluidics, and phase change heat transfer can potentially enable enhanced thermal management solutions, which are needed for the microelectronic industry to get back on track with Moore's law and introduce flexible devices and other new products.

In addition to the relatively old and difficult problem of thermal management of electronic devices, nanoscale thermal transport research is starting to make an impact on other grand challenge problems. For example, though there remain numerous unsolved problems associated with the mechanical strength, thermodynamic stability, abundance of the constituent elements, cost, and toxicity of new thermoelectric materials, opportunities exist for the use of some of the new materials for fabricating cost-effective vehicle thermoelectric waste heat recovery devices at a scale commensurate with the global vehicle manufacturing enterprise. In comparison, a  $ZT$  value of larger than 10 would be required to allow standalone solar thermoelectric generator to achieve the conversion efficiency of a solar Rankine cycle [311]. However, there could be still an opportunity to considerably increase the conversion efficiency of concentrated solar power systems of a high concentration ratio with a thermoelectric topping cycle, although such a topping cycle has been shown to be ineffective at a concentration ratio below 100 [312]. Moreover, research in selective solar absorbers based on nanofluids [313] and other nanostructured materials may enable the development of combined photovoltaic and concentrated solar power systems with both a high conversion efficiency and storage capability. In addition, research in localized and ultrafast heating methods is poised to make a significant impact on advanced manufacturing technologies, whereas the investigation of functional nanoparticles for thermal therapeutics provides a promising route to developing treatments of cancers and other diseases. Therefore, nanoscale thermal transport research can potentially lead to game-changing approaches to addressing some of the 14 grand challenges for engineering identified by the U.S. National Academy of Engineering [314], including making solar energy economical and engineering better medicine.

In this opinion article, we suggest that the focus of nanoscale thermal transport research should be on use-inspired basic research relevant to grand challenges faced by society. However, it would be a mistake to deemphasize the roles of either curiosity-driven pure basic research, which may have high potential to advance understanding of fundamental heat transfer processes but low potential for creating useful technologies in the near-term, or applied thermal engineering research and developments, which aim to create useful devices and systems instead of advance scientific understanding. Curiosity-driven inquiries, such as pushing the limit in thermal transport measurements down to the atomic and single molecular levels, are indispensable for the continuous generation of new ideas and tools for use-inspired heat transfer research. In addition, successful development



and demonstration of functional devices and systems to address societal needs, especially those that capitalize on the advances and breakthroughs in basic nanoscale thermal transport research, will become increasingly critical for justifying the relevance of this rapidly evolving field.

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