

REPORT ON 6TH U.S.-JAPAN JOINT SEMINAR ON NANOSCALE TRANSPORT PHENOMENA.SCIENCE AND ENGINEERING

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The objective of this U.S.-Japan joint seminar series is to provide a cross-disciplinary and international forum for discussing and identifying outstanding science and technology issues in the area of nanoscale thermophysics and energy conversion and to foster collaboration among researchers in these areas. The first of this seminar series, championed by the late Professors Chang-Lin Tien and Kunio Hijikata, was held in Kanazawa, Japan, in June of 1993. Subsequent meetings have been held every three years, alternating venues between the United States and Japan. The Sixth U.S.-Japan Joint Seminar on Nanoscale Transport Phenomena - Science and Engineering was held in Boston, Massachusetts, July 13.16, 2008, and was organized by Professors Gang Chen from MIT, Fushinobu Kazuyoshi from Tokyo Institute of Technology, Shigeo Maruyama from Tokyo University, and Pamela Norris from University of Virginia. Nearly 100 scientists participated in the seminar. (The agenda of the seminar is attached at the end at this report[14].) The seminar included keynote sessions and invited sessions, as well as a dedicated poster session of selected presentations from an open call for papers. All papers presented in the regular sessions, the invited sessions, were upon invitation by the organizers. Invited sessions used a mixed form of communication: each speaker gave a 5-minute summary of his work followed by a 30-minute poster session of just the papers summarized orally, and then these speakers came back to the podium, serving as panelists to answer questions regarding their papers and session themes. This format offered good opportunities for the presenters to discuss their work with the participants. Reports for each session were summarized by session chairs. Following is a brief summary of the sessions.

OPENING RECEPTION AND PRESENTATION

K. Okazaki gave a big picture talk on the evening of July 13, 2008, pointing to the urgency of global warming and the roles of nanoscale science and technology to address the challenges. Examples in CO₂ sequestration and catalysis were given. Okazaki's talk drew lots of discussion, including concerns about the long-term stability of proposed CO₂ sequestration methods.

SESSION 1: KEYNOTE SESSION

The first keynote session included talks by M.S. Dresselhaus [1] and S. Maruyama [2]. The speakers addressed very important directions to be noted by this continuing seminar series. Dresselhaus discussed the need for materials research to confront the challenge of the explosive energy demand increase. The world energy needs will reach 33 TW by 2050. Nanostructured materials are important for energybased applications due to (1) their unique physical phenomena, (2) their higher surface area, and (3) the independent control of each parameter that is not available in conventional bulk. Dresselhaus focused on solar energy, such as solar electricity (PV), solar fuel (biomass), and solar thermal, and thermoelectricity related issues. The importance of having a roadmap for the R&D, like Moore's law, which has served as the guiding role to the semiconductor electronics industry, was pointed out. The talk concluded with the challenges we face including the need for doubling and tripling the energy supply by 2050 and 2100, respectively, and of introducing

young researchers to the R&D arena. The increasing importance of the role to be played by the thermal and heat transfer community and the importance of nanoscale transport phenomena for nanostructured materials research was highlighted.

Maruyama discussed transport phenomena in nanostructured materials, mainly carbon nanotubes (CNTs), and the challenges to be faced by the thermal and heat transfer community. The talk started with various applications, such as Li ion battery electrodes and promising research seeds in future nanoelectronics. The focus was primarily on theoretical approaches. It was shown through molecular dynamic (MD) calculations of CNT thermal conductivity that the conductivity is predicted to increase with increasing nanotube length but it eventually reaches a constant value[15]. It was pointed out that although the calculation requires large computational resources, it was essential to perform MD calculations for different CNT lengths that were neglected in earlier works. The intrinsic and extrinsic thermal boundary resistance in CNTs and the characteristics of single-walled nanotubes in a matrix, which are crucial to evaluating the transport properties, were also discussed. Overall, the speakers in this keynote session provided very important messages. That is, the seminar and the community can contribute, by means of nanoscale transport phenomena, to tackling important global issues, such as the challenges in energy-based applications. This seminar series is expected to play a strategic role in discussions of up-to-date theoretical and experimental activities for the purpose of continuously introducing young researchers to the issues and to establishing potential collaborations for our continuing cooperation.

SESSION 2: TRANSPORT IN NANOSTRUCTURES

The second session included eight presentations focused on transport in nanostructures. Theoretical and experimental approaches for exploring transport phenomena in nanostructures were introduced and discussed, including their applications for thermoelectric devices, nanomanufacturing, and phase change memory devices. Broido [3] explained the theoretical techniques describing phonon transport based on Boltzmann and nonequilibrium Green's function calculation without any adjustable parameters. The interatomic constants are calculated using first-principle density functional perturbation theory. The obtained thermal conductivities show good agreement with the intrinsic data of bulk silicon and germanium.

Several papers in the session employed classical MD techniques to study heat conduction in nanostructures. MD simulations can reduce the computational load compared to ab initio calculations, allowing the investigation of complicated but practical nanometer-scale systems. Using MD simulations, Shiomi and Maruyama [4] investigated diffusive-ballistic phonon transport in CNTs. The study revealed that the thermal conductivity of a CNT is sensitive to the environment due to the modedependent phonon boundary scattering but less sensitive to atomic vacancy. A series of studies on mass transport and phase change of water inside CNTs was presented, attracting interest. Miyazaki [5] carried out both Boltzmann transport equation calculations and MD simulations to investigate the reduction mechanism of the thermal conductivity of nanoporous materials. The results show that the most important factor is an artificially reduced phonon mean free path because the phonon group velocity is reduced by nanoholes. In the poster session, he also discussed parallel computation for MD simulations using a CELL method and fabrication processes for nanoporous thermoelectric materials using a nanoporous polymer substrate.

Using MD simulations, Lukes [6] addressed phonon scattering by nanoparticles embedded in a solid. The obtained spectral-directional dependence offers new insight into boundary scattering mechanisms of acoustic phonons at the rough surface of a nanoparticle inclusion in a medium.

Lukes's conclusions can have potential connections to Dames's [7] analysis on thermal rectification in nanocomposites with nonsymmetric nanoparticle inclusions. Dames developed a novel thermal rectification scheme showing that thermal rectification does not violate the second law of thermo-dynamics, because the net heat current is zero at thermal equilibrium. With the presence of a temperature gradient, phonon emissions at the two end contacts become nondiffusive. In this case, the asymmetric scattering phase function of embedded asymmetric nanoinclusions results in a thermal diode effect. In thermoelectric devices, charge carriers and phonons drift in two opposite directions; therefore, an embedded nanostructure can be engineered that allows phonon transport in the forward bias direction of the diode and electron transport in the reverse bias direction of the diode. This mechanism can potentially lead to enhancements of the thermoelectric figure of merit.

The connections between the MD work, experiments, and industrial applications were discussed, along with challenges in the computational load and the degree of complication in the models. In the panel discussion, future challenges of the link between the first principle approaches and MD simulation were promoted. There were questions on the advantages/disadvantages of calculating thermal conductivity using equilibrium and nonequilibrium MD simulations. Interpretations of the temperature jumps and challenges in tuning thermostats to achieve suitable phonon distributions were also discussed.

Recent progress in several experimental studies attracted interest from the audience. Chen [8] discussed thermal and thermoelectric transport measurements of silicon nanowires. While the thermal conductivity of silicon nanowires grown by the vapor.liquid.solid method shows clear diameter dependence, this is not the case for rough silicon nanowires fabricated by electrochemical etching. The thermal conductivity of a rough 50-nm-diameter nanowire is suppressed by the amorphous limit. This large suppression cannot be explained by existing theories if only diffuse phonon surface scattering is taken into account. The electrical conductivity and Seebeck coefficient measured using a different method on a different rough nanowire were found to be similar to the bulk values. This is because the electron mean free path and wavelength in highly doped silicon are much smaller than the 50-nm nanowire diameter. Combining the three transport properties measured separately on two different nanowires using two different methods would result in the thermoelectric figure merit (ZT) being about 100 times larger than the bulk value.

King [9] discussed applications of heated silicon atomic force microscopy (AFM) tips for metrology and nanomanufacturing. He suggested that very high-temperature gradients on the order of 10 K/nm can be achieved at the tip-sample contact. It was shown that the local Seebeck coefficient could be measured for the heated tip. The spatial resolution and the Seebeck coefficient sensitivity of this and other local probing techniques were also discussed in the panel. It was suggested that the spatial resolution is limited by the size of the heated zone in the sample, which is in turn limited by the fundamental length scales, including electron and phonon mean free paths, and the tip radius, as found in similar measurements conducted using etched tungsten tips in a scanning tunneling microscope. Because the Seebeck coefficient is less than 1 mV/K in typical semiconductors, a high tip-sample temperature difference is needed to obtain a thermovoltage signal that is larger than the electrical potential resolution of 100 mV for common electrical AFM techniques such as Kelvin force probe microscopy. The heated AFM tip has been employed for several other metrology applications such as mapping thermal conductivity and phase transformations in polymers at high spatial resolution. It was also successfully demonstrated that the heated AFM tip can be utilized as a versatile nanosolder gun to pattern different materials on different substrates.

Goodson [10] discussed thermal transport issues in phase change memory devices, whose electrical resistance can be cycled between two states using pulse electrical heating and cooling that results in reversible transformation between the amorphous and crystalline phase. Since power consumption of this device is limited by the electrical heating rate needed to achieve the phase transformation temperature, low thermal conductivity thin-film coatings such as layered films are desirable for this application. Optimization of the device design and performance requiring detailed thermal transport measurements and nanoscale modeling provides new research opportunities.

SESSION 3: KEYNOTE SESSION

In his keynote address, Ohara [11] reported studies on the energy transfer mechanisms at liquid-solid interfaces revealed by MD simulations. By modeling chain molecules such as alkane, realistic interfaces such as lipid bilayers are simulated in addition to simple liquid-solid interfaces. By decomposing the MD heat flux term into several components, various energy transfer mechanisms can be examined. Energy transfer through lipid bilayer membranes was characterized by thermal resistances within and between carbon chains. The analysis led to the understanding of the thermal boundary conductance chain-length dependence. It was also shown that the liquid molecular motion perpendicular to the solid surface is predominantly important for the liquid-solid interface boundary conductance. The effect of a self-assembled monolayer (SAM) film at the interface was also stressed. A fivefold increase in thermal boundary conductance was predicted when adding a SAM layer at the liquid-solid interface. It was discussed that the value itself is still slightly different from the experimental estimation by Cahill et al. [12]. Even though several issues have to be addressed for the direct comparison of experiments, an important contribution to the study of thermal boundary resistance by MD is well recognized. Technical issues, such as the choice of the potential function and the slightly higher temperature used, were also addressed. Finally, the challenge of designing a liquid-solid interface with smaller thermal boundary resistance, such as SAM layers, was discussed with further design through MD simulations possible. Though water is still the best liquid with virtually the highest thermal conductivity, design of a liquid system with better thermal conductivity is the challenging task at hand.

Majumdar presented a keynote that began with the big picture: the energy crisis the world is now facing and two approaches to the problem. Though most of the solutions presented at the conference were focused on developing new energy conversion processes to increase the current supply of available energy, the problem can also be addressed by decreasing the dependence on energy by developing more efficient devices. Nano transport phenomena play an important role in developing new technologies that increase the conversion efficiency and decrease the waste heat in many applications. Following this general challenge to the community, Majumdar discussed how research in thermal transport phenomena has encountered insurmountable limits, creating barriers for the design of new devices and systems. Examples include the alloy limit of thermal conductivity in crystalline materials, the amorphous limit of thermal conductivity, critical heat flux in boiling heat transfer, etc. His presentation then focused on understanding the fundamental nature of these limits. Invariably, these limits relate to insights about the length scales of the phenomena, which often occur in the nanoscale regime. These limits can be overcome by using nanostructures that are cleverly designed to exploit unique features that cannot be accessed by macroscopic structures. For example, experiments have shown that the thermal conductivity can be reduced below the alloy limit by a factor of five to six, that the amorphous limit can be broken, and that the critical heat flux in boiling can be increased by a factor of two. He challenged the audience on whether these "limits" can be

pushed even further: what are the true physical limits and how far are we from those limits, and what new technologies can open up by breaking these limits?

SESSION 4: ENERGY CONVERSION

Seven speakers participated in this session, presenting their research results in the area of energy conversion. The main topics discussed are relevant to applications in fuel cell and thermoelectric technologies with both experimental and modeling work.

Hone [13] presented experimental work on CNTs, including their electrical, mechanical, and electromechanical properties. The measured electromechanical response is consistent with theory without any additional free parameters. The mechanical properties of individual suspended graphene sheets were measured by AFM nanoindentation and resulting values are explained by the intrinsic strength of the carbon-carbon bonds.

Fushinobu [14] presented a multiscale analysis to predict macroscale polymer electrolyte fuel cell (PEFC) behavior that focuses on the electrochemical reactions and the transport phenomena. The model predictions of current density in the fuel cell are consistent with experimental results, particularly at low current density. One of the salient features of the model is the calculation from first principle analysis of the oxygen reduction reaction (ORR) on the cathode catalyst. Inoue carried out both numerical calculations on the chemical reactions of methanol on Pt catalysts and experimental work to understand the proton transport mechanisms in a membrane for a direct methanol fuel cell (DMFC). The main dehydrogenation process was modeled by ab initio MD and the calculated chemical reaction process is consistent with experimental results. Experimental results show that the flow in a nanochannel can be explained by both a rarefied gas flow and a surface diffusion flow. These experimental results can be applied to characterize the proton transport properties in the ion-permeable membrane of a DMFC. Shao-Horn [15] investigated the correlation between the size-dependent activity of Pt nanoparticle catalysts and the surface atomic and electronic structures. The experimental approach employs a technique that can control the Pt particle size. High-resolution spectroscopy and TEM measurements show that the surface structure and the valence band of Pt nanoparticles change with particle size.

Ram [16] presented a model for the optimization of thermoelectric generators. He pointed out that the nondimensional thermoelectric figure of merit of the material (ZT) does not mirror the system-level power conversion efficiency, and that thermal impedance matching plays a major role in designing power generation systems. Ren [17] presented experimental results on the figure of merit of nanostructured bismuth antimony telluride alloys. A peak ZT value of 1.4 was reported at 100 °C; the improvement is due to the reduction in thermal conductivity. ZT remains high over a wide temperature range, from 1.2 at room temperature to 0.8 at 250 °C. This trend makes nanostructured bismuth antimony telluride alloys useful for both cooling and power generation applications, and it paves the way for the use of nanocomposites in developing high-performance, low-cost thermoelectric materials. Vashaee [18] presented transport models analyzing recent experimental results on the thermoelectric properties of nanocomposites. There is a reduction of the bipolar thermal conduction due to grain boundary scattering that may contribute to the reduction in thermal conductivity in bismuth antimony telluride alloys. The models predict that an enhancement in ZT requires that the grain size must be comparable to the effective phonon mean free path so that the thermal conduction is reduced but must be much larger than the charge mean free path to preserve the charge carrier mobility.

In the panel discussion, the mechanisms responsible for the reduction of the bipolar

thermal conductivity component and the effects of temperature and doping concentrations on thermoelectric performance were discussed. A challenge was issued to develop a technique for employing electron beam microscopy platforms to provide easier and faster measurements of thermoelectric properties. The panel provided insight into the present temperature and density of states measurement using electron beam microscopy. While scanning probe-based systems may be available, their spatial resolution is not yet sufficient. The discussions on thermoelectrics emphasized the need for modeling the figure of merit of nanocomposites in order to assess the upper limits of potential figures of merit and guide synthesis efforts. The need to find a simple expression for the system-level figure of merit for thermoelectric power generators was emphasized. The importance of size and crystalline orientation of Pt nanoparticles on catalyst activity was discussed and challenges related to the experimental characterization of surface structure of small nanoparticles were raised. The potential application of CNTs as catalysts for fuel cells, particularly the importance of crystalline defects, was proposed.

SESSION 5: KEYNOTE SESSION

The keynote presentation by X. Zhang discussed optical metamaterials and nanoplasmonics. Recent theory predicts a new class of metastructures made of engineered subwavelength entities - meta "atoms" and "molecules" - which enable unprecedented electromagnetic properties that do not exist in nature. Zhang gave examples of artificial plasma, artificial magnetism, and a surface plasmon superlens developed by his group that focuses far below the diffraction limit. He also demonstrated that metamaterials can have a profound impact on a wide range of applications such as nanoscale imaging, nanolithography (X-ray wavelength at optical frequency), integrated nanophotonics, and biosensing. In the follow-up question-and-answer session, Zhang addressed a number of questions from the audience, including the resolution limit of the super lenses, applications of superlenses in biosensing, and his commercialization plan of nanomanufacturing using the plasmonic lens array.

Volz's [19] keynote addressed coherence effects on heat transfer in nanostructures. First his theoretical predictions for the effects of phonon coherence on thermal transport in 3D photonic crystals were discussed. When 3D photonic crystals consist of a cubic network of nanoparticles embedded in a solid matrix, the thermal conductivity is predicted to decrease by one order of magnitude due to the decrease of the phonon group velocities and the reduction in the phonon mean free path. The size effect on the contact resistance was also discussed. When the contact cross section and the body size are decreased, the various phenomena that appear were explained, including the bottleneck effect in the cross section and multireflections in the small body. MD simulation results were shown to describe the contact cross-section effects. The case of the contact between a nanowire of sub-10-nm diameter with a plane surface was examined and the governing mechanisms for contact resistance at low temperature were discussed.

SESSION 6: LIGHT MATTER INTERACTION

Seven presentations were given in this session on light matter interaction, covering the spectrum from temperature-dependent fluorescent to photonic structures to nanoscale thermal radiation, with length scales from atomic to microns.

Borca-Tasciuc's presentation [20] was perhaps the most controversial, engendering the most discussion. The main objective of the work was to use quantum dots (QD, ZnS passivated CdSe) tethered to 1.4-nm Au nanoparticles as a thermometer. The fluorescence of the QDs was

calibrated with temperature to the position of the wave-length of peak emission. The main point of interest (and controversy) is that when the QD is tethered to the Au via Au-NH-CO-QD (where the carbon is double bonded to the oxygen), the temperature measured is, ~ 3 °C higher than for untethered QDs and Au nanoparticles in a borate buffer solution at a pH of 7. It was postulated that the linker served as a molecular heat pipe, transporting excess heat from the Au nanoparticle to the QD. As many audience members noted, this result is unexpected for radio wave frequency-heated solutions, since the 1.4-nm Au particle would seemingly be too small for any significant eddy current heating. A proposed potential E&M heating mechanism is that the Au-QD system has some magnetic moment, particularly if aggregation occurs. Other postulates were offered as to the cause of the remarkable result. The mechanism behind the experimental observation is an open question.

Hopkins [21] presented a fundamental study on the effects of inter- and intraband transitions on thermophysical property measurements of metal films on dielectric substrates. Using a two-temperature model along with band theory, it was shown that the d-shell interband and the s to d-shell intraband excitation of transition metals (Cu in this case) impact the electronic heat capacity and the coupling coefficient G. A new two-temperature model is developed that depends on both the density of states of the materials and the incident wavelengths of the optical excitations. The nonlinear responses of G and heat capacity change the temperature predictions several orders of magnitude over classic linear theory. The main result showed that the strongest component of the nonlinear response came from the d band interband transitions in the density of states rather than primarily the intraband transitions from s to d.

Lee and Narayanaswamy [22, 23] both discussed thermal radiative phenomena at the nanoscale. They showed both experimentally and theoretically that the spectral and directional thermal emissions off a flat surface could be shifted from a Planckian distribution to a sharp spectral emission, depending on the angle the surface is observed. A multilayer construct is used that starts with a thin (50-nm) Ag layer, topped by a dielectric stack of 150-nm-thick Si_3N_4 and 150-nm-thick SiO_2 , repeated three times. The resulting photonic structure converts the underlying isotropic broadband thermal emission from the Si wafer to the spectral directive emission. Narayanaswamy [23] presented measurements of the near-field radiative emission from the surface to a sphere mounted on an atomic force microscopy (AFM) cantilever. Using this tool, electromagnetic forces (Maxwell stress tensor and Poynting vector) were measured to discern some interesting near-field effects that arise from thermal emission on Casimir and van der Waal forces, as well as departure from Planck's distribution in the near-field.

Taguchi [24] also presented a method for thermometry using quantum dots. In this thermometry method, quantum dots were immobilized onto silica surfaces with a silanol linker. A nanowire heater made of permalloy was patterned on the silica and the fluorescence emission was calibrated in situ using a Peltier heater and a thermocouple. Then, using a microscope, surface temperatures were recorded with up to 10-nm resolution using a near-field fiber that excited and captured the emission.

J. Xu [25] and X.F. Xu [26] both presented near-field scanning optical microscopy (NSOM) results of photon transport through and from materials. Both used "bow-tie" apertures to enhance the NSOM signals. J. Xu showed that photons moving through plasmonic structures with patterned sub-diffraction-sized holes through a thin metal film can constructively and destructively interfere with plane waves that move through the film itself. The incident plane wave interaction with the plasmonic structure leads to the perhaps counterintuitive result that the phase shifts between the light going through the holes versus the metal film cause interference that is not sensitive to the

hole size, but which is strongly dependent on the film thickness. X. Xu used femtosecond pulse shaping to excite materials with impulses at the same frequency as the thermal phonon vibrational frequency of the material being probed with a bow-tie NSOM. With this technique, it is possible to both start and stop phonons propagating in a material. With the ability to excite coherent oscillations, impulsive stimulated Raman spectroscopy of the material can be collected through the NSOM. He was able to show adatoms within a sputtered material matrix with nanometer spatial resolution. This application is an example of ultrafast temporal excitation being linked with nanoscale resolution to determine nanoscale material composition.

SESSION 7: KEYNOTE SESSION

Matsumoto's [27] keynote presentation discussed microbubbles and nanobubbles. It was first shown that both sea fish and river fish can live together in water involving nanobubbles. Matsumoto then discussed the validity of applying the Young-Laplace equation to nanobubbles and whether there is a bubble diameter dependence for surface tension. From MD simulations, it was concluded that the equation can be applied even to nanobubbles and that the surface tension can be considered constant. The initial stage of bubble nucleation was then discussed. From the Young-Laplace equation, the inner pressure of a nanobubble must be extremely high; therefore, he posed a question: Is it possible to create nanobubbles on a heated surface? From MD simulation, he concluded that nanobubbles can emerge even on a perfectly flat plate. Finally, he discussed the surface charge of microbubbles. He showed simple experimental results that microbubbles tend to have negative charges, but the reason for these negative charges has not yet been clarified. During the discussions, the condition of heating in the MD simulation and the origin of microbubble negative charge were discussed. Some of the most important points are what are the essential differences between microbubbles and nanobubbles and why can both sea fish and river fish live together in water involving nanobubbles. To answer these questions, further research is needed.

Mahan [28] described his unpublished work on the development of an analytical theory of heat transfer between crystals and liquids. Transverse phonons in liquids are highly damped and all prior theories have ignored the contributions of the transverse modes to heat transport. The new theory solves the boundary conditions for phonon reflection and transmission using the pair-distribution functions in the liquid, the pair distribution function between the crystal and the liquid, and the matrix elements of the liquid-solid interaction potentials as inputs. During discussions the following points were addressed: (1) Can the strength or form of the interface interaction be tuned to maximize heat transfer, or is heat transfer a monotonic function of the strength of the solid-liquid interaction? (2) Since the theory is linear, can we estimate the importance of anharmonic or inelastic channels for heat transport at solid-liquid interfaces? (3) How appropriate is it to use the language of "phonons" to describe the vibrational modes of a liquid? (4) Do we need to consider the diffuseness of the transition between solid and liquid? (5) And since most liquids are polyatomic, can the theory be extended to polyatomic liquids?

SESSION 8: SOLID-LIQUID INTERFACES

Eight presentations were given in this session on solid-liquid interfaces. The talks covered mass transport through nano/microchannels [29,34] or through nano-fabricated surface [35] and heat transfer enhancement by nanostructured surfaces [36,37] or by fluids with nanoparticles and nanodroplets [38], with a central focus on molecular sensing and heat transfer enhancement.

The spatial scale of "channels" and "pores" widely ranges from ~1 nm for single DNA

identification to $\sim 100\ \mu\text{m}$ for cell separation. Controlling/manipulating surface properties is essential for all the proposed devices. Manipulating the surface charge of channels results in rectification of ion transport [29, 33], which leads to new devices such as fluidic "transistors" and efficient proton exchange membranes. Coating channel surfaces with receptors brings a new cell separation technique without cumbersome cell labeling [30,32]. Manipulating fluids on nanostructures and nanowire arrays [35] is promising for controlling wetting fluid transport and heat transfer. However, the energy input budget and the side effects need to be studied. MD simulation is still the dominant tool for studying heat and mass transport in nanochannels and on nanostructured surfaces [36, 37]. Both reducing the liquid-solid interfacial resistance and increasing the evaporation heat transfer rate on nanostructured liquid-solid interfaces have been attempted, but the length scales simulated are far from the fabricated structures that are currently studied experimentally. Nanofluids, which are fluids containing nanoscale solid particles, have been studied for around a decade since their inception. Heated discussions seem to conclude that the overall gain in heat transfer performance from these nanofluids could be negligible because of the enhancement of thermal conductivity due to the increase of viscosity. "Nanoemulsion fluids" [38], which are a suspension of liquid droplets instead of solid particles, could be very promising for energy storage due to their enhanced heat capacity. Nonlinearity due to hydrodynamic interactions between droplet/droplet and droplet/base liquid seems responsible for the good performance.

POSTER SESSION

In addition to the invited keynotes and sessions, an evening poster session was dedicated to selected presentations from an open call for papers. Nineteen posters were presented covering the development and characterization of new materials, understanding material interfaces including solid/liquid interfaces, and nanofluidics.

For the development of thermoelectric devices, Wang et al. [39] reported a first-principles study on thermoelectric properties of three lanthanum chalcogenides: La_3S_4 , La_3Te_4 , and La_3Se_4 . The results showed that all three materials exhibit semimetallic behavior with a small overlap in the conduction and valence bands, and the electronic band structures and density of states suggest a high ZT value. Minnich et al. [40] developed a numerical code to calculate the electrical and thermal properties of bulk and nanocomposite thermoelectric materials using the Boltzmann equation under the relaxation approximation. The numerical results were in good agreement with the experimental data obtained in both bulk and nano-SiGe materials. Muto et al. [41] presented an experimental system to directly measure the thermoelectric conversion efficiency of a single thermoelectric element. This method achieved a more accurate ZT value than that obtained by measuring the electrical conductivity, the Seebeck coefficient, and the thermal conductivity.

In the nanotube area, Cronin et al. [42] investigated thermal transport across single-walled CNTs using a microfabricated membrane structure with integrated temperature sensors and Raman spectroscopy. A nanotube was suspended on a microfabricated temperature sensor to monitor the heat provided by the laser on the nanotube at varying power levels, and the Raman g band shift determined the temperature of the CNT. Estrada et al. [43] investigated the thermal and mechanical coupling between CNTs and their dielectric environment. A measurement platform with sub-mK temperature resolution was developed and used to investigate the properties of aligned CNT arrays and random CNT networks. MD simulations were also performed to understand the CNT-SiO₂ thermal interface conductance. Stewart et al. [44] modeled thermal transport through boron nitride nanotubes using a new first principles thermal transport approach. The thermal conductance

calculated from the phonon transmission of the isotropically pure boron nitride nanotube was fairly consistent with the measured values, and an independent scattering calculation accounted for the reduction in thermal conductivity due to 10 B isotopes. Teweldebrhan et al. [45] experimentally studied the thermal conductivity of graphene suspended over a trench on a silicon wafer. The thermal conductivity of graphene measured by the noncontact Raman optical technique was in the range of 3,080 - 5,150 W/mK. The measured values were explained theoretically using calculated acoustic phonon dispersion in graphene and available data for Gruneisen parameters.

The thermal conductivity of other novel materials and interfaces was also investigated. Henry et al. [46] studied the thermal conductivity of single polyethylene chains using MD simulations. Though bulk polyethylene is typically known to be a thermal insulator, the thermal conductivity of an individual chain module was predicted to be as high as 300 W/mK with a maximum conductance of 10^9 W/K using both the Green-Kubo and modal decomposition methods. The results show promise for polymers to be engineered to achieve high thermal conductivities for various applications. Schmidt et al. [47] used a pump-probe method to measure the thermal conductivity in the directions along and perpendicular to the Si/Si_{0.7}Ge_{0.3} superlattice film. The in-plane thermal conductivity was found to be about ten times higher than the cross-plane thermal conductivity. Son et al. [48] measured the interface thermal resistance between gold and tungsten nanowires and their substrates. Joule heating thermometry techniques with a movable AFM tip were used to probe the spatial distribution of the interface thermal resistance. Furthermore, photothermal measurements were performed to study the interface thermal resistance between CNTs and the substrate and a heat conduction model was developed to support these measurements. Budaev et al. [49] developed an analytical acoustic model to predict the interface thermal resistance between dissimilar materials. The model suggested that the heat-carrying phonons obeyed the introduced principle of heat radiation instead of the commonly used Sommerfeld radiation condition. This model provides more channels for coupling between phonon propagation in contacting media than is allowed by the conventional acoustic model.

New fabrication methods for novel materials have also been developed. Duong et al. [50] developed a new fabrication technique for high volume fraction nanocomposites of vertically aligned CNT forests. Enhancements of the nanocomposites' mechanical, thermal, and electrical properties were measured. A Monte Carlo simulation was utilized to understand the effects on the effective thermal conductivity of CNT orientation, weight fraction, and interface thermal resistance between the polymer and CNTs. Moghaddam et al. developed a controlled method to create self-assembled functionalized nanoporous silicon for silicon-based CMOS compatible proton exchange membrane (PEM) fuel cells. This method eliminates challenges associated with attaching a Nafion membrane to silicon fuel cell structure, which can fail over time due to significant volume change of the polymer membrane. A new method was developed to create nanoporous silicon membranes using an anodization process. The membranes created were 20 μ m thick and had pores with diameters ranging from 5 to 7 nm.

In the area of fluidics, Li et al. [51] used MD simulations to examine nanoscale fluid surface interactions. The simulations were used to investigate three cases: (1) gas-nanoparticle collisions, (2) gas transport in nanochannels, and (3) particle transport in nanochannels. The first case indicated that diffuse scattering caused by gas adsorption occurs on the particle surface. For gas transport in nanochannels, the thermal vibrations have an important role in gas diffusion when the wall binding energy is strong. Finally, in the third study, the particle mobility was found to vary depending on the competing effects of the surface and fluid drag. Takada et al. [52] reported a new

fabrication technique for hollow poly-lactic acid microcapsules using microbubbles as templates. By keeping thermodynamically stable uniform-sized microbubbles inside a solution droplet, dissolving poly-lactic acid and then slowly drying the solvent, uniform-sized hollow microcapsules of 3 μm in diameter and 500 nm in thickness were obtained. Valencia et al. [32] reported novel synthesis of polymeric nanoparticles for drug delivery applications using microfluidic rapid mixing. The experimental results showed that the average diameter and the diameter distribution of nanoparticles could be tuned by controlling the flow rate. These results can provide the foundation for a microfluidic platform for controlled synthesis of nanoparticles. Sen et al. [31] demonstrated a simple nanofluidic Coulter counter system with feedback control that enables two measurements on the same molecule of DNA. Compared to existing systems, this approach was expected to be more applicable for sizing a variety of particles and molecules. Duan et al. [53] studied ion transport in 2-nm-deep nanochannels formed by controlled surface etching on a silicon wafer followed by anodic bonding with a Pyrex substrate. The results demonstrated that surface charge dominates ion transport at concentrations up to 100 mM. It was also found that there is no clear ion dependence on conductance at low concentrations. Tatsumi et al. [54] reported MD study of ion transport in 2-nm-diameter silica nanopores. The model silica pores were partially modified by a hydrophobic surface functional group to modify the surface dipole moment. The calculated results revealed that the surface dipole moment affects the ion transport inside the nanopores.

Finally, Prasher et al. [55] provided an overview of various nanomaterials that can potentially influence current electronic systems. Examples were nanostructured thermoelectric materials, nanothermal interfacial materials, and nanofluids.

SESSION 9: INDUSTRIAL PANEL

Representatives from Seagate Technology, Denso, Intel, and the Office of Naval Research gave brief presentations highlighting their nanotechnology needs, concerns, and opportunities. The following were major points of interest. Prasher summarized Intel's view that cooling technologies should not require any changes in the chip design. Therefore, it is not anticipated that microchannel cooling will be adopted. A possible upcoming technology is the use of localized cooling by positioning thermoelectrics directly over the hot spots. Nakamura highlighted Denso's technology development of personal air conditioning units employing thermoelectric devices that utilize waste heat. Hipwell [56] emphasized Seagate's continuing need to improve metrology techniques and their interest in near field imaging. Spector of the Office of Naval Research summarized the current technology trend of utilizing all electric sources in new generation ships and aircraft, thus placing great emphasis on the need for drastic reductions in the size of the cooling components. A panel discussion prompted many questions from the audience with an important message surfacing for students in this field. Experience has shown industry that, generally speaking, nanoscale thermal transport students are quite comfortable working with other disciplines and are often able to come up with unique solutions by applying their knowledge to an analogous area. When asked what skills industry requires for new engineers, the overwhelming answer was passion and curiosity.

SESSION 10: KEYNOTE SESSION

Cahill [12] began his keynote presentation by discussing recent advances in time-domain thermoreflectance measurements. This technique is based on the known temperature dependence of the cap material's reflectivity. By resolving the thermoreflectance on the picosecond timescale, the thermal conductance of thin films can be determined with acute accuracy. This is a noncontact

method that eliminates the effects of thermal contact resistance. This technique was employed to study the amorphous limit of the thermal conductivity. Cahill presented the ultralow thermal conductivity of W/Al₂O₃ and WSe₂ nanolaminates, with measurements as low as 0.04 Wm⁻¹ K⁻¹. The material's high interface density reduces its thermal conductivity by almost two orders with respect to the corresponding single-crystal material and a factor of six lower than the calculated amorphous limit. These results show that nanostructures can reach a lower thermal conductivity than that of the bulk limit.

Yamamoto's [57] keynote was a comprehensive review of phonon transport in CNTs. At low temperatures, quantum transport is important and conductance is universal, independent of geometry and chirality, and insensitive to defects. Near room temperature, however, the phonon mean free path becomes comparable to the tube length. Therefore, transport becomes length dependent and can be explained by considering scattering at the ends. This work gave another angle to examine the extensive MD simulation work carried out in Maruyama's group and other groups on CNTs.

SESSION 11: EMERGING EXPERIMENTAL AND NUMERICAL TECHNIQUES

The final session of the conference featured seven presentations on emerging experimental and numerical techniques. The experimentalists described a range of new measurement methods with excellent temperature sensitivity and spatial and temporal resolution and presented results for various nanostructures including nanotubes, nanowires, thin films, and nanoscale hot spots. The numerical work featured first principles calculations of the electron and phonon relaxation times in low-dimensional carbon nanostructures, in excellent agreement with experiments.

Shi [58] described an experimental apparatus that measures the longitudinal thermal conductivity, electrical conductivity, and Seebeck coefficient of 1D and 2D nanostructures. Recent improvements to the apparatus include implementing a thermal four-point probe to mitigate the effects of thermal contact resistance and enhancing the capabilities for the TEM imaging of the sample after it has been mounted on the measurement platform, which gives atomic-level information about sample geometry and surface conditions. Measurements were shown for various materials, including CNTs, nanowires, and thin films, covering a broad range of phonon and electron wavelengths and mean free paths. Takahashi [59] presented a novel method to measure the thermal conductivity of amorphous carbon nanostructures. MEMS thermal sensors were developed based on a direct-current heating method. The amorphous carbon was directly deposited on a Pt nanofilm using electron beam-induced deposition (EBID), and the thermal conductance change of the combined Pt nanofilm/amorphous carbon nanodeposit was measured. A two-dimensional numerical simulation was also presented, and the uncertainty of this method was calculated at less than 7%. The results indicated the applicability of this technique for accurate understanding of the thermal properties of nanostructures deposited by NEMS (nano electro mechanical systems) techniques.

Nakabeppu [60] presented a novel nano-calorimeter device (chip-calorimetry) fabricated by an integrated MEMS technique. The probe included a heater, thermocouples, and a thermopile sensor. Experimental measurements were demonstrated on the chip-calorimeter using cantilever-type probes. Time DTA (differential thermal analysis) was performed for samples in the microgram to nanogram range. Mass measurement using the mechanical resonance of the cantilever probe was shown with an estimated resolution of 5 ng. Measurements were conducted with spherical particles for calibration and indium particles for calorimetry. The new analysis method is promising for analyzing very small samples that are important in nanotechnology. Yang [61] described a new

method for nanoscale thermal measurements based on timeresolved diffraction of soft X-rays. The sample was a sapphire substrate micropatterned with various nickel diffraction gratings. The Ni lines are heated by an 800-nm wavelength pump beam. The 30-nm wavelength probe beam was produced using higher harmonic generation. The diffraction efficiency of the probe beam was very sensitive to the thermal expansion of the Ni lines, resulting in an excellent temporal and spatial resolution measurement of the substrate's transient thermal response. The measurements of gratings with smaller line widths exhibited a clear signature confirming the transition from diffusive to ballistic phonon transport.

Pipe [62] presented a new optical probe enabling highly resolved optical imaging beyond the diffraction limit of light. The novel probe consists of a photodiode and organic LEDs monolithically integrated on the tip of a scanning probe. The probe is fabricated by a micro/nanomachining technique including vacuum thermal evaporation and FIB milling processes. The signal-to-noise ratio (SNR) was calculated as 17, and there was plenty of room to improve the spatial resolution. This technique of scanning probe microscopy using monolithically integrated organic devices may become a powerful tool for material characterization of biological samples.

Pop described several experimental and theoretical studies of heat generation and dissipation in CNTs. Microfabrication methods have been developed to routinely measure the high-bias I-V curves of individual nanotubes, which exhibit remarkably different behavior depending on their chirality (metallic vs. semiconducting). A membrane-based method to measure the thermal conductivity of arrays of CNTs, which may be well-aligned or random networks, is currently being developed. MD simulations of heat transfer between a CNT and SiO₂ substrate were also presented, suggesting a line conductance of approximately $0.17 \text{ W m}^{-1} \text{ K}^{-1}$, which is consistent with experimental observations and a thermalization length (characteristic fin length) of approximately 200-300 nm. Bonini [63] presented first-principles calculations of the fundamental relaxation times in CNTs and graphene. The calculations used density functional theory to predict both electron-phonon and phonon-phonon relaxation times. The calculations have no adjustable parameters and are consistent with experimental measurements of the frequencies and temperature dependence of the Raman peaks and their line widths. These results led to a deeper understanding of the anharmonic response and inelastic relaxation mechanisms for both electrons and phonons in low-dimensional graphitic materials.

CLOSING DISCUSSION

The closing session discussed important questions arising from the three-day meeting. It is recognized that new materials and theories are needed as a guide to materials selection. Measurements and characterization tools are of critical importance. Transport in complex and nanostructured materials is an important area, particularly the influence of anharmonic coupling, boundaries and contacts, and electron-phonon-photon interactions. For electron-phonon interactions, polymers are of special interest given that electrons and phonons are strongly coupled due to molecular deformation. It is recognized that high throughput measurement techniques are particularly desirable; for example, how to accelerate the characterization of nanowires, how to measure electrical properties using noncontact methods, how to experimentally determine electronic contributions to thermal conductivity, and how one can do spectroscopy on electrons and phonons. Suggestions using fluorescent materials that depend on local electrical field for noncontact measurements were proposed. In addition to high throughput, accurate and repeatable characterization of thermal transport in nanostructures is also challenging. Questions were raised on

liquid transport: How can one develop a simple kinetic equivalent theory? Japan strives for a large scale introduction of fuel cells in 2020, but first breakthroughs are needed in lowering cost and increasing reliability.

It was announced that the next U.S.-Japan seminar will be held in Japan with Professors S. Maruyama, K. Fushinobu, L. Shi, and J. Lukes cochairing the meeting.

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