Final Report

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Tailoring Thermal Radiative Properties with Doped-Silicon Nanowires

(Supplement title: A Theoretical and Experimental Study of Near-Field Thermal Radiation for Noncontact Refrigeration and Thermal Rectification)

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Program Scope

Aligned doped-silicon nanowire (D-SiNW) arrays form a hyperbolic metamaterial in the mid-infrared and have unique thermal radiative properties, such as broadband omnidirectional absorption, low-loss negative refraction, etc. A combined theoretical and experimental investigation will be performed to characterize D-SiNW arrays and other metamaterials for tailoring thermal radiative properties. Near-field thermal radiation between anisotropic materials with hyperbolic dispersions will also be predicted for potential application in energy harvesting. This project will allow the measurement data to be confirmed by additional verifications and detailed uncertainty analysis for archival publication. This project aims at realizing D-SiNW-based metamaterials for thermal radiation control as well as for near-field imaging in the mid-infrared region. Other metamaterials, such as graphene, multilayered structures, and gratings or the combination will also be considered.

Summary of Major Accomplishments

A new kind of anisotropic metamaterial with a hyperbolic dispersion in a broad infrared region has been proposed and demonstrated based on aligned doped-silicon nanowire (D-SiNW) arrays. D-SiNW-based metamaterials have unique thermal radiative properties, such as broadband omnidirectional absorption whose width and location can be tuned by varying the filling ratio and/or doping level. Furthermore, high figure of merit (FOM) can be achieved in a wide spectral region, suggesting that D-SiNW arrays may be used as a negative refraction material with much less loss than other structured materials, such as layered semiconductor materials. We have also shown that D-SiNWs and other nanostructures can significantly enhance near-field thermal radiation. The study of near-field radiative heat transfer between closely spaced objects and the electromagnetic wave interactions with micro/nanostructured materials has become an emerging multidisciplinary field due to its importance in advanced energy systems, manufacturing, local thermal management, and high spatial resolution thermal sensing and mapping. We have performed extensive study on the energy streamlines involving anisotropic metamaterials and the applicability of the effective medium theory for near-field thermal radiation. Graphene as a 2D material has attracted great attention in nanoelectronics, plasmonics, and energy harvesting. We have shown that graphene can be used to tailor the transmittance, reflectance, and absorptance of nanostructured materials. Furthermore, graphene can be used to enhance near-field coupling to increase the phonon tunneling probability. We have performed analysis of near-field thermophotovoltaic devices with backside reflecting mirror and with tungsten gratings. We have predicted a large enhancement of electroluminescent refrigeration at a separation distance down to 10 nm due to near-field thermal radiation effect. A heat flux measurement system is developed to measure the near-field radiation in vacuum. We have fabricated doped Si plates separated by sparsely distributed posts to create a 200-800 nm vacuum gap. Our measurement results demonstrate that 11 times enhancement of near-field thermal radiation between parallel doped-Si plates with a lateral dimension 1 cm by 1 cm.

With this DOE project, the PI's group has published 32 journal papers [1-32] and two book chapters [33,34] as listed under publications. These works reflect a significant contribution to advance the fields of nanoscale thermal radiation and optical metamaterials research. The PI has been invited to give nearly 10 seminars each year by other institutions internationally as well as to deliver a large number of invited, plenary, and keynote speeches at international conferences and workshops. The PI was Lead Guest Editor for several special issues in this field published in J. Heat Transfer and J. Quant. Spectrosc. & Radiat. Transf. The PI Zhang served as the Chair for the *2nd International Workshop on Nano-Micro Thermal Radiation* (NanoRad2014), Shanghai, China, June 2014 (over 90 attendees from nine countries or regions). He was the General Chair of the *5th ASME Micro/Nanoscale Heat & Mass Transfer International Conference*, January 2016, Singapore (with some 300 participants). PI Zhang has been elected APS Fellow (2015-16) and received 2015 ASME Heat Transfer Memorial Award. The students supported by or participated in this research have received several awards and recognitions, including travel grant, best poster award, best thesis award, invited talks, etc. Selected research results and findings are highlighted in the next section.

Major Technical Contributions

Doped-Si Metamaterials for Negative Refraction and Near-Field Radiative Heat Transfer

We have shown that low-loss, all-angle negative refraction can be achieved in the infrared region using doped-silicon nanowire (D-SiNW) arrays [1]. The structure is illustrated in Fig. 1a while negative refraction is shown in Fig. 1b in which the D-SiNW array of 20 μm is in the region $-10\ \mu m < z < 10\ \mu$m. Potential applications include wideband absorber [2], near-field radiative transfer enhancement, thermal imaging, and designing flat lenses and collimators. The mechanism for low-loss is explained by using loss-enhanced transmission, along with impedance matching and the absence of resonances. The figure of merit (FOM), defined as the ratio of the real to the imaginary part of k_z, is plotted in Fig. 1c. The FOM increases with wavelength and is higher for heavily doped Si (more loss) and can exceed 100 in the mid-infrared region [1].

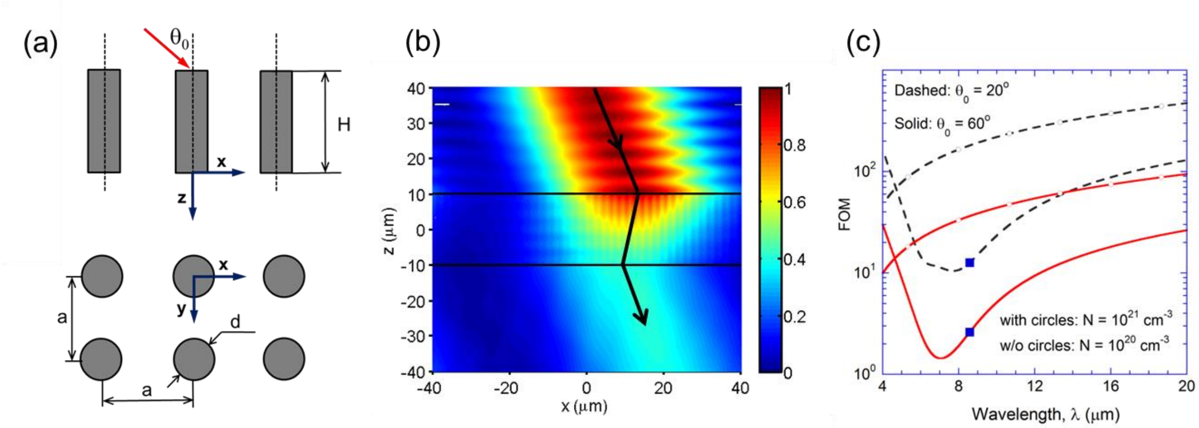

Fig. 1. (a) Schematic of D-SiNW array; (b) Illustration of negative refraction at air-D-SiNW array interfaces; (c) FOM versus wavelength for two doping levels with filling ratio of 0.05 [1].

We have also predicted broadband tunable mid-infrared absorption based on D-SiNWs in the far field [2] and enhanced near-field thermal radiation with graphite (naturally hyperbolic) and carbon nanotubes (CNT) [3]. Nanostructured materials can also enhance radiative transfer in the near field, especially with hyperbolic metamaterials made with doped Si. We have considered a number of different nanostructures containing and compared their capability in enhancing near-field thermal radiation [12]. As illustrated in Fig. 2, for nanowires and nanoholes, the heat transfer coefficient, *h*, can be enhanced by over an order of magnitude more than that for bulk doped Si counterpart and several orders of magnitude greater than that between blackbodies in the far field. The analysis presented in this work may benefit the design of nanostructures for applications such as more efficient noncontact thermal management and energy conversion.

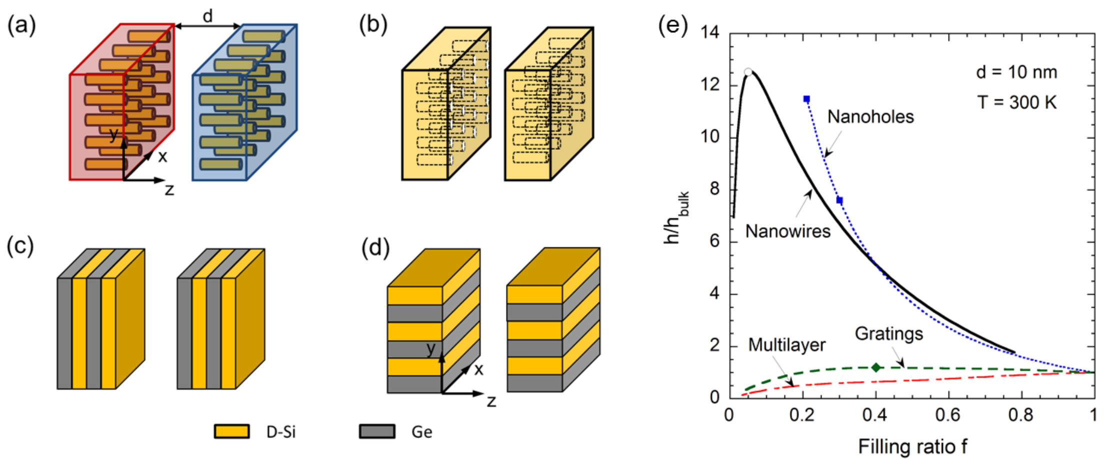

Fig. 2. (a) Schematic of near-field radiation between (a) D-SiNWs, (b) D-SiNHs, (c) multilayers, and (d) 1D gratings separated by a vacuum gap with distance *d*; (e) Predicted near-field heat transfer coefficient (*h*) normalized to bulk doped Si for the four structures with varying filling ratio [12].

It was postulated that graphene sheets covering broadband hyperbolic substrates can further improve near-field photon transport by extending the number of tunneling evanescent modes [16]. Figure 3a illustrates two graphene-covered D-SiNW arrays separated by a vacuum gap. The near-field transmission coefficient contour is shown in Fig. 3b in terms of the angular frequency and parallel wavevector component β, divided by the wavevector in free space k_0. In the calculation, the temperatures of both the emitter and receiver are near room temperature, the doping concentration of the SiNWs is 10^{20} cm^{-3} with a filling ratio of 0.02, the chemical potential of graphene is 0.3 eV, and the vacuum spacing d is 200 nm. The transmission coefficient is close to unity across a broad frequency range up to 1.5×10^{14} rad/s and a large wavevector space up to $20k_0$. All the photons emitted in this regime will be absorbed, which gives a blackbody behavior in the near field. As a result, the heat transfer coefficient of this hybrid structure exceeds that for plain D-SiNWs arrays or suspended graphene sheets alone [16]. Similar results were also predicted for graphene-covered CNT arrays [20].

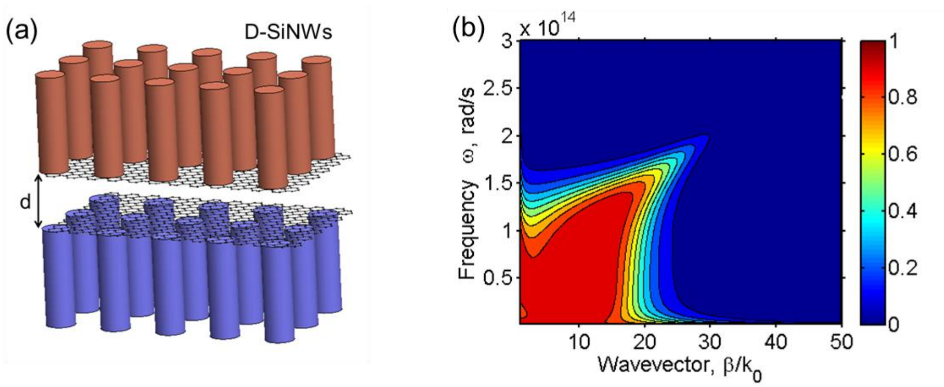

Fig. 3. (a) Schematic of near-field radiative heat transfer between graphene-covered semi-infinite doped-Si nanowires separated by a vacuum gap d. (b) Photon tunneling probability for p-polarization, showing near-unity tunneling probability in a broad frequency and wavevector region [16].

Metasurfaces and Gratings for Near-Field Enhancement

Artificial nanostructures have attracted much attention in the field of both photonics and phononics due to their unprecedented radiative and thermal properties beyond the capabilities of bulks. Recently, we have used exact formulations including scattering theory and the Green's function method to calculate near-field radiation between periodic 1D, 2D, and 3D structures. It was found that patterning thin doped-Si films into 1D and 2D metasurfaces could increase the radiative heat flux by more than an order of magnitude in a certain thickness range [24]. The underlying mechanism lies in the excitation of hyperbolic modes for broad frequency region and k-space. Furthermore, if a single layer of graphene sheet were patterned into a ribbon array, the dispersion of graphene plasmons would become hyperbolic. Extremely high-k evanescent waves can couple with hyperbolic graphene plasmons and, subsequently, a giant enhancement of the near-field radiative heat flux [25].

Metasurfaces, planar metamaterials with subwavelength thicknesses, have some peculiar advantages over conventional metamaterials, such as less volumetric propagation loss, relative easy fabrication, and compatible integration with other nanodevices. As shown in Fig. 4, patterning the film into 1D metasurface can enhance near-field thermal radiation for all practical volume filling ratios. The gap distance is set to 100 nm, the thickness of metasurface is 400 nm, and T_1 = 310 K and T_2 = 290 K. Interestingly, while the 2D metasurface yields a radiative heat flux higher than that of thin films at moderate filling ratios, it does not support a heat flux as high as that of the 1D metasurface. The underlying mechanism of enhanced thermal radiation of metasurfaces lies in the support of hyperbolic dispersions leading to broadband high local density of states [24].

Graphene ribbons have been extensively studied for their unique optical properties. Here we theoretically calculate the near-field radiative heat transfer between two parallel arrays of graphene ribbons. As shown in Fig. 5, the heat flux between graphene ribbons can be 15.3 times that between suspended graphene sheets at d = 15 nm and is more than 3 folds when d increases to 100 nm [25]. Here, T_1 = 310 K and T_2 = 290 K. This giant enhancement may offer possible benefits to energy harvesting of thermal radiation by increased heating rate capability, potential thermal management enhancements, and augmented noncontact temperature measurement. The underlying mechanism is due to the excitation of hyperbolic graphene plasmons featured with open dispersion relations and thus infinite density of states. The agreement between the numerically exact calculations with EMT is good especially for large gap spacings.

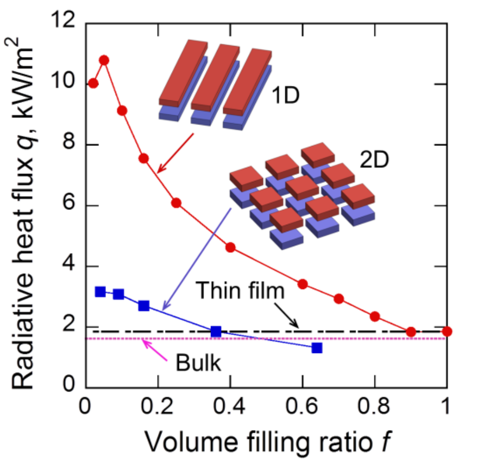

Fig. 4. Predicted near-field radiative heat flux between metasurfaces [24].

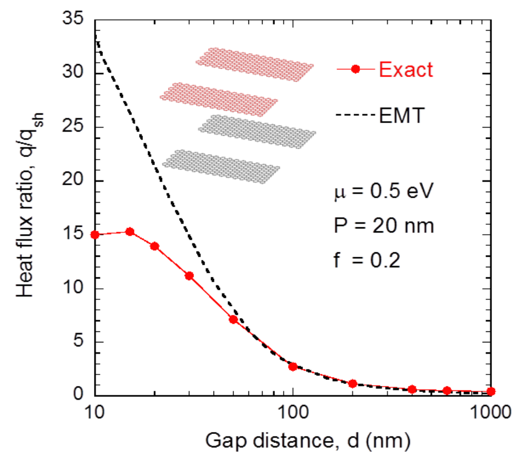

Fig. 5. Enhancement of radiative heat flux of graphene ribbon over graphene sheets [25].

Near-field thermal radiation and Casimir interaction induced by quantum mechanical electromagnetic fluctuations have received much attention in recent decades. Based on the scattering theory, our group has demonstrated simultaneously enhanced energy transport and suppressed momentum exchange are demonstrated by patterning doped-silicon surfaces in the near field [26]. The grating structure is shown in Fig. 6a, with the calculated near-field radiative fluxes (6b) and Casimir forces (6c).

It is found that the radiative heat flux between doped-silicon gratings exceeds that between planar surfaces and can be one or even two orders of magnitude as high as what is predicted by the geometry-based Derjaguin's proximity approximation (PA); see Fig. 6b. The underlying mechanism is interpreted as due to the excitation of broadband hyperbolic modes which facilitate photon tunneling, especially when the period is small. This is confirmed by comparison of the results from the scattering theory with those from the effective medium theory (EMT). The Casimir force, which may cause stiction and even failure of mesoscopic devices, is reduced with the grating structures as predicted by both the scattering theory and PA. As shown in Fig. 6c, depending on the separation distance, PA may over- or under-predict the Casimir force [26].

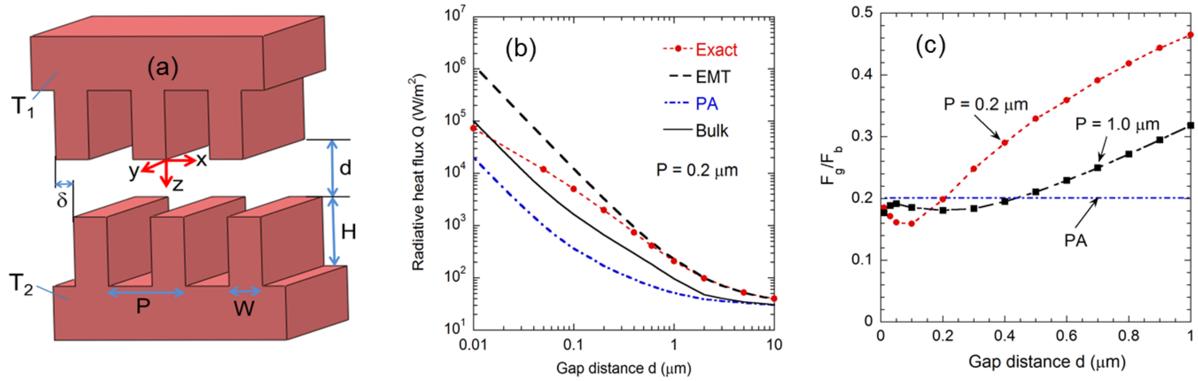

Fig. 6: (a) The doped-silicon grating structure; (b) Radiative heat flux versus gap spacing; (c) Ratio of the Casimir force of doped-Si gratings F_g to that of bulk doped silicon F_b for different period [26]. EMT = effective medium theory; PA = proximate approximation.

Quantum levitation enabled by repulsive Casimir force has been desirable due to the potential exciting applications in passive-suspension devices and frictionless bearings. We have investigated the Casimir interaction between two nanostructures separated by an intervening fluid is investigated. While stable levitation is achievable when one substrate and a thin film coated on another substrate are separated by a dielectric fluid, the levitation position depending on the film thickness is fixed and not tunable. We have theoretically shown dynamically tunable stable levitation based on the configuration of dissimilar gratings separated by an intervening fluid using exact scattering theory [28]. The levitation position is insensitive to temperature variations and can be actively tuned by adjusting the lateral displacement between the two gratings. We have demonstrated the possibility of applying quantum Casimir interactions into macroscopic mechanical devices working in a noncontact and low-friction environment for controlling the position or transducing lateral movement into vertical displacement at the nanoscale; this may have practical applications in both microscopic and macroscopic devices.

Measurement of Near-Field Radiative Transfer between Flat Plates

For energy harvesting applications, increasing the surface area and shrinking the gap spacing are both critical in order to achieve high radiative heating rates. While tremendous progress has been made in recent years toward experimental realization, measurements between planar surfaces with square-centimeter-sized areas at deep submicron gap distances are still quite challenging due to difficulties in controlling the gap spacing. Here, we report measurements of heat transfer near room temperature between two 1 cm by 1 cm doped-Si parallel plates, separated by a vacuum gap from about 200 nm to 780 nm. The measured strong near-field radiative transfer is in quantitative agreement with the theoretical prediction based on fluctuational electrodynamics. The largest measured radiative heat flux is 11 times as high as the blackbody limit for the same hot and cold surface temperatures. Our experiments have produced the highest radiative heat transfer rate observed to date across submicron distances between objects near room temperature [31].

A heat flux measurement system is developed to measure the near-field radiation in vacuum as shown in Fig. 7a. The sample is placed in the middle and consists of two 1 cm^2 doped-silicon pieces with a sparse array of SiO_2 micropillars. The pillars fabricated on one plate support the other by spring loading forces to maintain the necessary gap spacing, as shown in Fig. 7b.

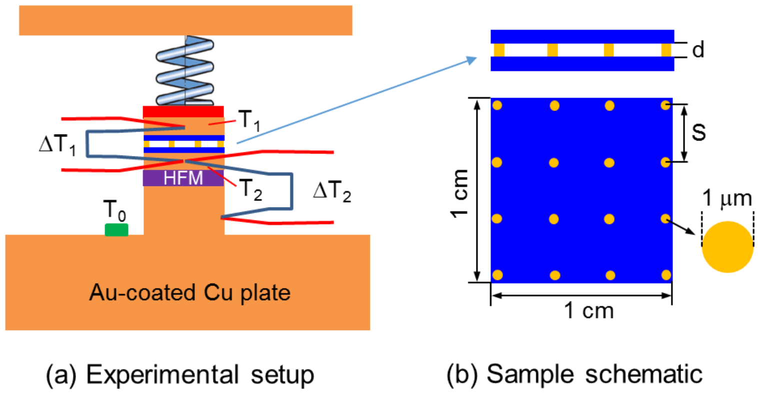

Fig. 7. Schematics of the experimental setup for measuring near-field radiative heat transfer between flat plates and the structure of the sample. (a) The measurement stage that contains a stack of layers below the spring, namely, a heater, a Cu plate, the sample containing two doped-Si plates separated by a gap, another Cu plate, and a heat flux meter (HFM) mounted on a Cu heat sink. (b) The sample made of two doped-Si pieces separated by a submicron gap using SiO2 posts, where S is the distance between adjacent posts [31].

Figure 8a displays q_{rad} for three different gap spacings versus the temperature difference ΔT between the two doped-Si plates. The isolated solid symbols represent the measured results with uncertainty bounds indicated by the error bars. The dashed lines are from the fluctuational electrodynamic calculation with the shaded region indicating the uncertainty bounds. The large enhancement in nanoscale thermal radiation is attributed to the excitation of coupled surface plasmon polaritons (SPPs). The uncertainty of the theoretical calculation is mainly due to the determination of the gap spacing. Different ΔT is created by controlling the power provided by the DC power supply. However, due to radiation from the top of the heater and conduction through the spring, not all the heating power flows down across the sample. For example, with a 265 mW power provided by the DC power supply, about 191 mW (134 mW due to radiation and 57 mW due to conduction) passes through the sample, creating a ΔT = 16.5°C across its 200 nm gap. Figure 8b illustrates the results for radiative heat transfer coefficient h_{rad} for 14 measurements at different gap spacings. The measured data are presented as filled symbols with error bars, while the calculated values at the same temperature and gap spacings are shown as open circles. Each shape of the solid symbols represents a different sample under specified applied forces. The solid (red) line represents the calculated results using the average T_H and T_L (i.e., 318.5 K and 302.3 K, respectively) of all measurements and plotted as a function of gap spacing. The two dotted lines represent the calculation uncertainty bounds. The horizontal dashed line is the blackbody limit using the average T_H and T_L. Note that h_{rad} increases as d decreases, reaching a value of 81.2 $W/m^2 \cdot K$ that is about 11 times that of the blackbody limit at the same emitter and receiver temperatures. The experience gained and the facilities mentioned can be employed with modifications to study near-field energy conversion devices.

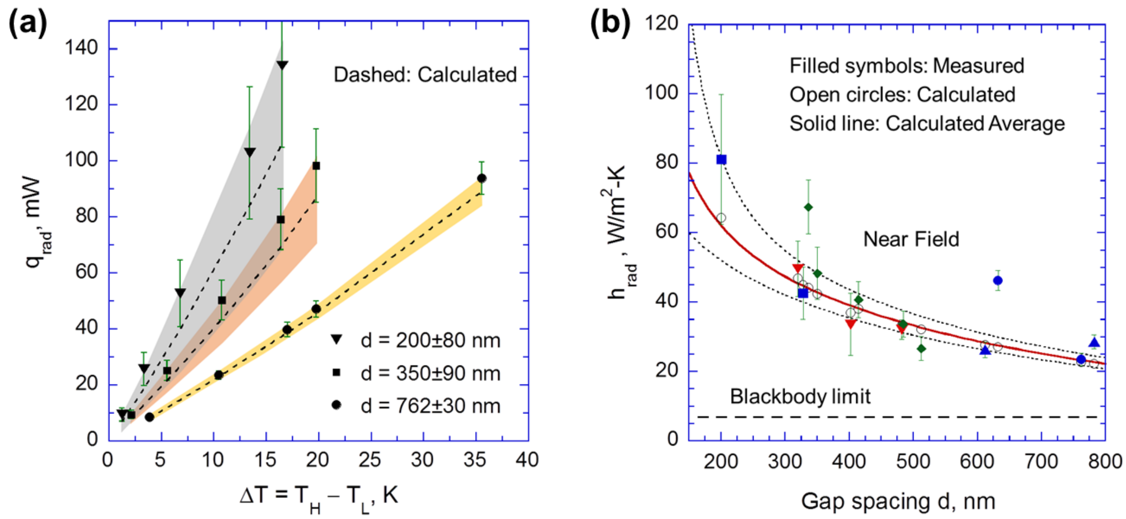

Fig. 8. (a) Radiative heat transfer rate for three different gap spacings as a function of the temperature difference. (b) Radiative heat transfer coefficient for 14 measurements at different gap spacings with ΔT ranging from 15.2 K to 19.2 K. Each symbol is for the same sample under different applied pressures. The results show about one order of magnitude enhancement of near-field radiative transfer down to 200 nm spacing [31].

Near-Field Thermophotovoltaics and Luminescent Refrigeration

Direct heat-to-electricity conversion can be achieved through thermophotovoltaic (TPV) systems, which have been demonstrated for energy harvesting in the far field and investigated in the near field. The basis is to use the photocurrent generated by incident photons from a high-temperature emitter that creates electron-hole pairs in the TPV cell. A TPV system has some technological advantages since it requires no moving parts, is pollution-free, and can utilize a variety of heat sources from solar energy to waste heat. However, the major challenge in conventional TPV technology is the low power throughput and conversion efficiency. Near-field thermal radiation, in which the radiative heat transfer can be increased to exceed the far-field blackbody limit predicted by Planck's law, holds great potential in enhancing the TPV performance by bringing the TPV emitter in close proximity to the cell within nanometer vacuum gaps.

We have theoretically modeled near-field TPV systems by considering a backside reflector and by reengineering the surface to reduce the recombination velocity [17]. The envisioned TPV system is illustrated in Fig. 9a, and the predicted efficiencies based on fluctuation-dissipation theorem show a significant enhancement by the backside mirror as shown in Fig. 9b. The enhancement is as much as 35% when T_H = 1250 K. The mirror reflects (i.e., recycles) long-wavelength photons back to the emitter, since these photons can only create thermal effect if absorbed by the PV cell materials and cannot generate electron-hole pairs. The quantitative study performed here on the important parameters that affect the performance of near-field TPV cells may facilitate the realization of such devices for practical applications. We are also considering using gratings to enhance near-field TPV performance.

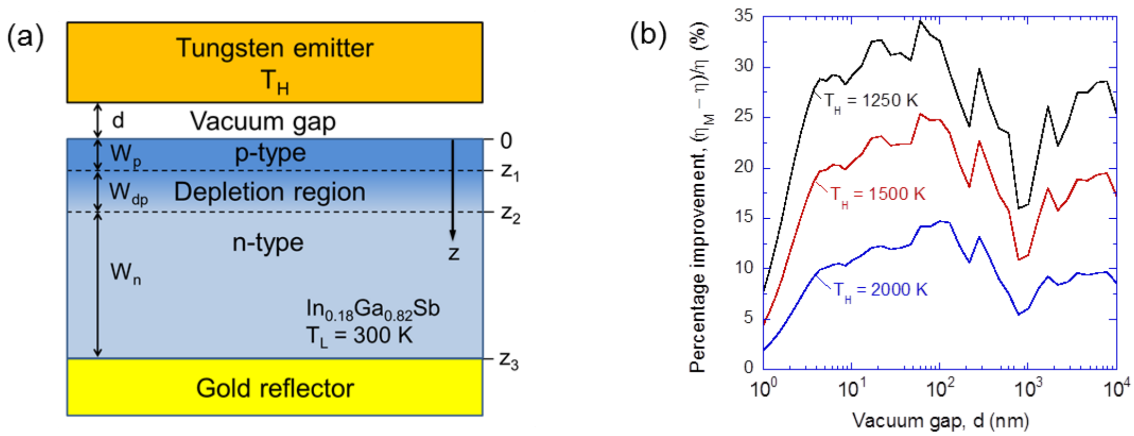

Fig. 9. (a) Schematic of the near-field TPV system with a gold mirror as the back reflector. (b) The percentage improvement in conversion efficiency by adding a mirror compared to no mirror [17].

We have also performed calculations based on the rigorous scattering theory and showed that, if the emitter is made of nanostructures as shown in Fig. 10a, the power output can be further increased as shown in Fig. 10b [32]. It is found that the power output can be increased by 40% while improving the efficiency from 29.9% to 32.0% with a selected grating emitter as compared to the case of a flat tungsten emitter. Reasons for the enhancement are found to be due to the enhanced energy transmission coefficient close to the band gap. This work shows a possible way of improving NFTPV and sheds light on how grating structures interact with thermal radiation at the nanoscale.

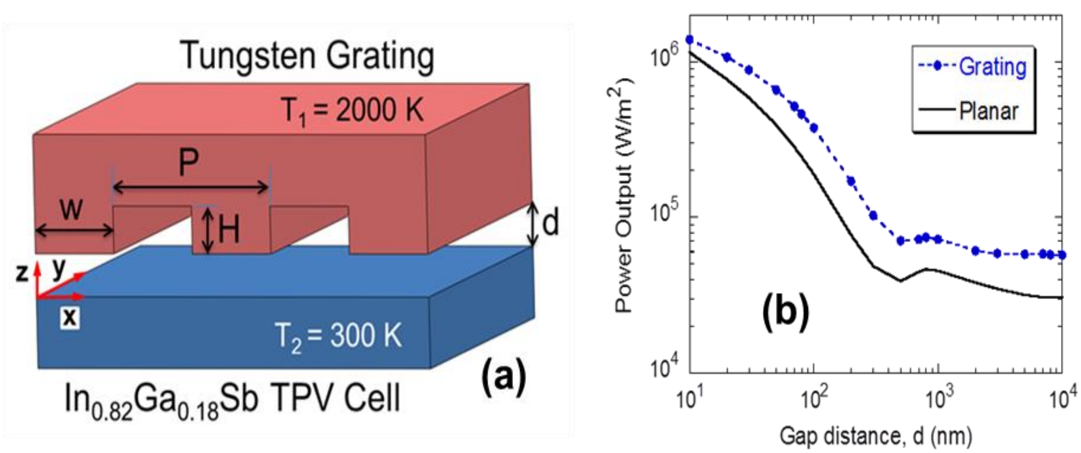

Fig. 10. Schematic of the near-field TPV system with a tungsten grating emitter (upper) and the calculated power output (lower) for cases with and without grating (planar) [32]. Even without grating, near-field effects can enhance the power output over far-field for more than 30 times when *d* = 10 nm.

Electroluminescent refrigeration, though theoretically proposed half a century ago, is rarely reported due to the lower refrigeration power per unit area and the requirement of extremely low nonidealities. Liu and Zhang [30] have developed a model that can take into account the position dependence of the chemical potential of photons in the diode based on fluctuation-dissipation theory combined with dyadic Green's function approach for a multilayered system. The system is schematically shown in Fig. 11a, where the lower portion is a p-i-n junction that is biased with a voltage V. As a result, the photons emitted, including propagating waves, evanescent waves, and ultrahigh wavevector (k) modes, have a nonzero chemical potential of qV at frequencies higher than the bandgap. Subsequently, more energy is emitted from the intrinsic region even though its temperature is lower than the top plate (T_2). By employing dedicated choices of materials, we show that both near- and far-field refrigeration can be achieved for certain range of temperature differences and the refrigeration rate can be improved by orders of magnitude by operating at the nanoscale. As shown in Fig. 11b, by operating the device in the near-field regime with a vacuum gap down to 10 nm, the refrigeration rate may be enhanced by 2000-fold over the far-field scenario. Furthermore, photon tunneling through evanescent waves can increase the tolerance of non-intrinsic nonradiative recombination to 31.6%. Thus, the achievable cooling temperature against the ambient T_2 = 300 K extends from 284.2 K to 270.6 K. Note that for 470 nm < d < 830 nm, as shown in Fig. 11b, the conventional thermal radiation at longer wavelengths from the Ge plate to the diode that overwhelms the luminescent refrigeration, resulting in a loss of the refrigeration effect. This study opens a route to greatly enhance electroluminescent refrigeration for solid-state noncontact thermal management.

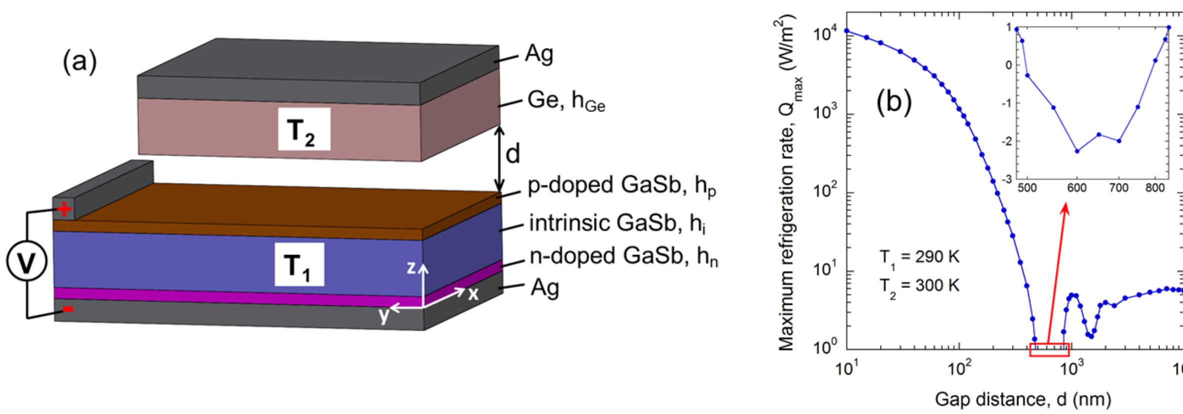

Fig. 11. (a) Schematic of a forward-biased *p-i-n* GaSb diode separated with a Ag substrate with Ge deposited on top with a gap distance of d. The temperature of the diode is T_1, and is assumed to be lower than that of the top object. In all calculations, $h_p = h_n = 0.1$ μm, $h_i = 1$ μm, $h_{Ge} = 500$ μm, $T_2 = 300$ K. Unless specified, $T_1 = 290$ K. (b) The maximum refrigerate rate from the biased diode versus the gap distance d. Significant enhanced refrigeration rate can be achieved at nanometer distances. Detailed can be found in [30].

List of Publications (Sponsored by this DOE-BES grant)

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List of Participants and Human Resources Development

The project supported PI Professor Zhuomin Zhang for one summer month during 2014-2017. The PI oversaw the research, supervised students, as well as helped prepare papers and documentations.

Dr. Xianglei Liu, a Ph.D. student, has been working on this project since 2014. He is mostly supported by this project except during 2015-2016 academic year when he was supported as a Graduate Teaching Assistant. He has carried out extensive theoretical studies related to micro/nanostructured materials for both near and far field thermal radiation and radiative properties. He defended and graduated in May 2016. He was one of the ten recipients of the Georgia Tech Sigma Xi Best Ph.D. thesis award for 2016. He is currently hired as a Professor at the Nanjing Aeronautics and Astronautics University, Nanjing, China.

Dr. Jesse Watjan, a Ph.D. student, has been supported by this project to work on near-field radiation measurements between doped silicon plates. He had previously been supported as a Graduate Teaching Assistant for two years. He also performed modeling of near-field TPV with tungsten gratings and showed the enhancement in power output and efficiency. He received a Ph.D. degree in May 2016 and is currently hired as a Research Engineer at the Knolls Atomic Power Laboratory in New York.

Dr. Bo Zhao, a Ph.D. student, has been supported by Georgia Tech teaching assistant as well as partially by the National Science Foundation. He has collaborated with the other two students on modeling near-field and far-field radiative heat transfer, especially related to graphene and periodic grating structures. He also collaborated extensively on the near-field thermal radiation measurements with Jesse Watjen. After graduating in December 2016, he has been a post-doctoral fellow at Stanford University.

Dr. Trevor Bright, graduated in Fall 2013, was supported as a postdoctoral fellow from March 2014 through July 2014, and thereafter land a research position in Aerospace Corporation. During the period of time while working as a postdoc, he co-authored two papers on hyperbolic metamaterials (especially metallodielectric multilayers) for near-field thermal radiation.

Dr. Zihao Zhang, a Ph.D. student, has been supported by Georgia Tech teaching assistant as well as partially by the National Science Foundation. He has collaborated with Dr. Liu on modeling near-field radiative heat transfer, especially related to graphene and carbon nanotubes from 2013-2015. He is currently an Assistant Professor at the University of North Texas (UNT).