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First-principles molecular dynamics investigation of the atomic-scale energy transport: From heat conduction to thermal radiation

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ABSTRACT

First-principles molecular dynamics simulation based on a plane wave/pseudopotential implementation of density functional theory is adopted to investigate atomic scale energy transport for semiconductors (silicon and germanium). By imposing thermostats to keep constant temperatures of the nanoscale thin layers, the initial thermal non-equilibrium between the neighboring layers is established under the vacuum condition. Models with variable gap distances with an interval of lattice constant increment of the simulated materials are set up and statistical comparisons of temperature evolution curves are made. The equilibration time from non-equilibrium state to thermal equilibrium state of different silicon or/and germanium layers combinations are calculated. The results show significant distinctions of heat transfer under different materials and temperatures combinations. Further discussions on the equilibrium time are made to explain the simulation results. As the first work of the atomic scale energy transport spanning from heat conduction to thermal radiation, the simulation results highlight the promising application of the first-principles molecular dynamics in thermal engineering.

1. Introduction

As the integrated circuits (ICs) are being miniaturized, complementary metal–oxide-semiconductor (CMOS) has stepped into the fabrication era of 22-nm for the central processing unit (CPU) in 2011 by Intel [1]. Transistor size and structure plays a crucial role in fulfilling the Moore's Law [2]. With the continuing improvement of manufacturing technology, the execution efficiency is also being improved. The 22-nm processors, which are known as 3D Tri-Gate transistor using three gates wrapped around the silicon channel in a 3D structure, bring unprecedented computational performance, and accompany tremendous heat generation. Hence, the atomic scale thermal and thermoelectric transport should be considered with the miniaturization process. Understanding the atomic scale thermal characterization and the control of materials species, interfaces, and structures is of the prime importance. In addition, the International Technology Roadmap for semiconductors (ITRS) 2006 Front End Process Update indicates that equivalent physical oxide thickness will not scale below 0.5 μm, which is approximately twice the diameter of a silicon atom. Consequently, to probe thermal transport would produce far reaching influence to the future development of thermal management of ICs.

In the manufacturing process, it is inevitable to leave gaps/defects inside the transistor packages at the length of the atomic scale. Thermal radiation coexists with thermal conductance that occurs at such confined geometrical structures. Due to the ubiquitous gaps in synthetic materials, especially in the fabricated ICs, understanding and predicting the heat transfer between two bodies separated by a distance of atomic scale have been a key issue in both theoretical and application points of view [3]. In addition, the thermodynamics properties of nanoscale thin films could significantly differ from that of the bulk materials [4]. Thermal energy transport is no longer a pure conceptual phenomenon but has become a crucial topic in the field of thermal management of micro-electro-mechanical systems (MEMS) and nano-electro-mechanical systems (NEMS) [5].

Because of the existence of temperature gradient in the materials, the difference of vibrational energies between adjacent atoms causes them to collide with each other; this leads to diffusive transfer of kinetic energy through particles inside the materials. Heat transfer occurs spontaneously from materials at higher temperatures to that at lower temperatures. Energy carriers, such as phonons [6] and free electrons [7], play crucial rules in solid state heat conduction. As for the thermal radiation, heat transfer is realized by electromagnetic radiation generated by the motion of charged particles in the matter. The spectrum emissive power of blackbody radiation is a well-understood physical phenomenon that depends on the object's temperature and obeys Planck's law. The Stefan–Boltzmann's law can be used to obtain emissive power of blackbody surface. However, it is only true for objects distances that are sufficiently large compared with the thermal wave length

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Nomenclature

$\boldsymbol{a}$	lattice vector, Bohr = 0.52917720859 Å
$\boldsymbol{b}$	reciprocal lattice vector, Bohr^{-1}
c_0	speed of light in vacuum, m/s
E	atomic-scale energy, J
E_{ions}	interaction energy of the bare nuclear charges, Ha = 27.21138386 eV
E^{KS}	Kohn–Sham energy, Ha
E_{xc}	exchange functional, Ha
f_i	integer occupation number
f_G^{PW}	plane wave basis
$\boldsymbol{g}$	a vector with triple of integer values, Bohr
$\boldsymbol{G}$	reciprocal space vector, Bohr^{-1}
$\boldsymbol{h}$	lattice matrix
$\boldsymbol{k}$	wave vector
M	atomic mass, a.u. = 0.00054857990943*a.m.u*
n	index of refraction
$\boldsymbol{R}$	atomic position vector, Bohr
$\boldsymbol{R}^N$	union of all the atomic position vectors
s	coordinates scalar
T_s	kinetic energy of a non-interacting system, Ha
U	potential, Ha
V_{ext}	external potential, Ha
V_H	Hartree potential, Ha

Greek

ε^{KS}	extended Kohn–Sham energy function, Ha
δ_{ij}	orthonormality relation, $\langle\varphi_i\|\varphi_j\rangle$
μ_i	fictitious mass, a.u.
Ω	volume of the unit cell, Ha
θ_i	angle of incidence, °
θ_c	critical angle, °
ω	frequency of the incident energy, Hz
$\mathcal{L}$	Lagrangian form of energy, Ha
Ψ	electronic wave function
$\mathcal{H}_e$	Hamiltonian of the electron, Ha
ϕ	electronic orbital
λ_{tw}	thermal wave length, m

λ_{tw} given by Wien's law. The typical wavelengths involved in thermal radiation are in the order of microns [8]. For the heat transfer with an interfacial distance of angstroms to nanometers, the radiation mechanism may change dramatically [9]. When two bodies are separated by a distance that is comparable or much shorter than the dominant emission wavelength, the validity of the classical radiative transfer equations is challenged, as the wave nature of thermal radiation needs to be taken into account.

Electronic charge and thermal energy transport in nanometer scale and ultra-short time scale (from femtoseconds to nanoseconds) have drawn attentions from scholars and have been studied extensively in the past decades. For heat conduction, a unified constitutive equation covering the transport behaviors of diffusion, phonon electron interaction and pure phonon scattering was proposed by Tzou in 1995 [10]. Jia et al. [11] performed a numerical study of heat flux of a nanofluid system showed the thermal conductivity increases with the nanoparticle volume fraction. Different empirically predefined inter-atomic potentials were used to study the thermal dynamical properties of water by Mao [12]. For thermal radiation, a bimaterial atomic force microscope cantilever was used by Narayanaswamy et al. [13] to obtain the "heat transfer-distance" curves between a sphere and a substrate. Their results showed that the enhancement of heat transfer exceeded that predicted by Planck's blackbody radiation theory. The surface phonon polariton was the factor that caused thermal fluctuations of the electromagnetic field. Two parallel glass surfaces were employed by Hu et al. [14] to measure the radiative heat flux across the micro gaps. Volokitin and Persson [15] studied heat transfer between the two parallel semi-infinite bodies separated by subwavelength distance via electromagnetic interaction. As it was reported in the literature, with the interfacial distance decreasing, the heat transfer increased dramatically. But the physical mechanism behind such increase still remains to be undiscovered. Kittel et al. [16] reported a measurement to obtain the heat transfer rate with the distance between microscope tip and sample. However, the measured results differed distinctly from the divergent behavior predicted by standard macroscopic fluctuating electrodynamics. They interpreted the results in terms of finite microscopic correlations inside the materials.

Comparing with experimental investigation, theoretical reasoning and numerical simulation can obtain results under extreme conditions that experiments cannot achieve. Molecular dynamics (MD) is a computer simulation of physical movements of atoms and molecules. The history of MD can date back to the mid-1950s when the first computer simulations on simple systems are performed [17]. In classical MD [18], with the help of the predefined empirical potential function, force field interaction between atoms, molecules, and larger clusters, can be obtained and used to calculate their motions with time. Consequently, trajectories of these molecules and atoms can be calculated via numerically solving the Newton's equations of motion. However, due to the dependence of predefined potential function to the forces acting on atoms and molecules, the accuracy and application are restricted by the available potential functions. In addition, to tackle the problems, we are facing on thermal transport issues in atomic scale via a comprehensive simulation of the thermal radiation involving electromagnetic fluctuation, for the shortcoming that classical MD fails to take dynamical electrons impact into account. The implementation of first-principles MD simulation [19] offers us an effective alternative. In the first-principles approach, the forces acting on nuclei can be directly computed from surrounding electronic structure calculation via the density functional theory, and then trajectories of motion can be generated.

The first-principles based simulations of atomic scale interfacial thermal transport have not been widely investigated and documented. Luo and Lloyd [20] pioneered the first-principles non-equilibrium MD simulation in different combinations of multi-thin-layers under thermal gradient. Later, Koker [21] extended the equilibrium MD approach to obtain the thermal conductivity based on the kinetic conductivity relation, with phonon lifetimes, group velocities, and heat capacities by combining the first-principles MD and lattice dynamics. The thin layers in Ref. [20] were placed close to each other and no distance between the neighboring layers. As aforementioned, the existence of interfacial gaps is unavoidable in reality. In this work, the first-principles MD simulation of the atomic-scale energy transport via heat conduction and thermal radiation in spatial and temporal perspective is carried out. Transport phenomena occurring between silicon and/or germanium layers with gaps are investigated. Silicon is chosen because it serves as a principal component of most semiconductor devices, most importantly in the ICs and microchips. On the other hand, germanium is an important semiconductor material being used in transistors, solar cells [22] and other electronic devices. In addition, the silicon–germanium alloys are rapidly becoming