STRUCTURAL DISORDER AND THERMAL TRANSPORT IN NANOSCALE SOLIDS

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An Abstract of a Thesis Submitted to the Graduate

Faculty of Rensselaer Polytechnic Institute

in Partial Fulfillment of the

Requirements for the Degree of

DOCTOR OF PHILOSOPHY

Major Subject: Materials Science and Engineering

The original of the complete thesis is on file
in the Rensselaer Polytechnic Institute Library

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Troy, New York

June 2006
(For Graduation August 2006)

ABSTRACT

We use a combination of molecular dynamics simulations and vibrational mode analysis to study thermal transport in nanostructured solids. We will describe how increasing degree of structural disorder affects localization and polarization of phonons (thermal vibration waves) and their ability to carry heat in nanocrystalline silicon and chemically functionalized carbon nanotubes.

The role of disorder due to the presence of grain boundaries is exposed in our studies of nanocrystalline silicon. Our vibrational mode analysis demonstrates that the vibrations that carry most of the heat in small grain ($\lesssim$ 3 nm) structurally heterogeneous nanocrystalline silicon are almost identical in nature to those in homogeneous amorphous silicon, where the majority of the vibrations are delocalized and unpolarized. Consequently, the principal thermal conductivity mechanism in such a nanocrystalline material is the same as in the amorphous material. With increasing grain size, the vibrational modes become progressively more like that of a crystalline material; this is reflected in a *crossover* in the mechanism of thermal transport to that of a crystalline material.

The role of point defect disorder is studied in connection with chemical functionalization of carbon nanotubes. These studies were motivated by the use of carbon nanotubes as thermal transport enhancing nanofibers in composites. We demonstrated that functionalization decreases the interfacial resistance between the

tube and the matrix, however, at the expense of the decrease of the intrinsic carbon nanotube conductivity caused by phonon scattering by defects. Remarkably, at the defect concentration of $\sim$ 1 % of functionalized C atoms, the thermal conductivity decrease saturates - further increase in the defect density does not lower thermal conductivity. To understand this behavior we use vibrational mode analysis of chemically functionalized single-walled carbon nanotubes to analyze the role of point defects on phonons. We demonstrate that majority of the vibrations in functionalized tubes are delocalized but are not polarized. Only low-frequency, long-wavelength phonons preserve their polarization in the presence of increasing density of point defects. Consistently, molecular dynamics simulations of low frequency phonon propagation show relatively weak scattering from point defects. These low frequency phonons are likely to be responsible for the saturation of thermal conductivity decrease in the presence of increasing defect concentration.