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STUDY OF THIN-FILM SI/GE SUPERLATTICE THERMOELECTRIC MATERIALS

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Abstract : Superlattices consist of alternating solid thin layers of different materials stacked periodically. The lattice mismatch and electronic potential differences at the interfaces and resulting phonon and electron interface scattering and band structure modifications can be exploited to reduce phonon heat conduction while maintaining or enhancing the electron transport. This article focuses on a range of materials used in superlattice form to improve the thermoelectric figure of merit.

Key words : thermal conductivity, thermoelectricity. PACS : 73.61.-r

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INTRODUCTION

Superlattices, which are periodic structures made of alternating solid thin layes with thickness ranging from tens to hundreds of angstroms, were theoretically predicted and proposed in 1970s Superlattices are generally divided in to two categories : long-period suplerlattices p<1, and short –period superlattices, p>1, where p is the phonon mean free path and l is the superlattice period. Ideas in using superlattices to improve the thermoelectric figure of merit (ZT) through the enhancement of electronic conductivity and reduction of phonon thermal conductivity were first discussed in workshop by M.S. Dresselhaus, T. Harman and R. Venkatasubramaniam[1]. Subsequent publications from Dresselhaus's group on the quantum size effects on electrons drew wide attention and inspired intense research, both theoretical and experimental, on the thermoelectric properties of quantum wells and superlattices[2].

Superlattices are anisotropic. Different mechanisms to improve ZT along directions both parallel and perpendicular to the film plane have been explored. Along the in-plane direction, potential mechanisms to increase ZT include quantum size effects that improve the electron performance by taking advantage of sharp features in the electron density of states [2], and reduction of phonon thermal conductivity through interface scattering [3]. Along the cross-plane direction, one key idea is to use interfaces for reflecting phonons while transmitting electrons [4], together with other mechanisms, such as electron energy filtering[5] and thermionic emission[6], to improve electron performance. These mechanisms have been explored through a few superlattice systems whose consistent materials have reasonably good thermo electric properties to start with, V-V materials such as Si/Ge[7], with the most impressive results obtained in Si/Ge superlattice,.

The large ZT improvements observed in this superlattice shattered the ZT~1 ceiling that persisted until the 1990s, opening new potential applications in cooling and power generation using solid state devices. Much research is needed in materials, understanding, and devices to further advance superlattice thermoelectric technology.

Materials And Properties.

The work on superlattice-based thermoelectric material, mostly focussed on perfect layer systems. The current thin-film device technologies for V-V compounds use either on or the other of these two material systems. Si/Ge superlattice systems have been studied for their thermoelectric properties. Si/Ge and Si/SiGe alloy superlattices have shown a large reduction in thermal conductivity compared with that of homogeneous alloys in the cross-plane direction,[7], while in the in-plane direction, thermal conductivity values are comparable with that of the homogeneous alloy with equivalent composition to the superlattices[9].

Characterization of Thermoeletric Properties

Thermoelectric properties measurements in many cases have been the bottleneck in the development and understanding of superlattice-based materials. Because of anisotropy, all thermoelectric properties, including the seebeck coefficient α, electrical conductivity σ, and thermal conductivity K, should be measured in the same direction, and, ideally on the same sample. Along the in-plane direction, thermal conductivity is usually the most difficult parameter to measure. However, the substrate and the buffer layers can also easily overwhelm the Seebeck coefficient and electrical conductivity measurements. The need to isolate the properties of the film from those of the substrate and the buffer layer often influences the choice of the substrate and the film thickness in the growth of superlattices. In the cross-plane direction, the 3ω method and the pump-and-probe method are often used to measure the thermal conductivity of superlattices[10]. However, measuring the seebeck coefficient and the electrical conductivity in the cross plane direction can be even more challenging.

Current Understanding

Experimental results so far have shown that the thermal conductivity reduction was mainly responsible for ZT enhancement in the superlattices. Theoretical studies on the thermal conductivity have been carried out[8]. These models generally fall into two different camps.

The first group treats phonons as incoherent particles and considers interface scattering as the classical size effect that is analogous to the Casimir limit at low temperature in bulk materials and Fuchs-Sonderheim treatment of electron transport.[11-13]. These classical size effect models assume that interface scattering is partially specular and partially diffuse, and can explain experimental data for superlattices in the thicker period limit.

The other group of models is based on the modification of phonon modes in superlattices, considering the phonos as totally coherent[14,15]. In superlattices, the periodicity has three major effects on the phonon spectra : (1) phonon branches fold, owing to the new periodicity in the growth direction; (2) mini-band gaps form; and (3) the acoustic phonon in the layer with a frequency higher than that in the other layer become flat or confined because of the mismatch in the spectrum, comparison with experimental data, however, shows that the group velocity reduction alone is insufficient to explain the magnitude of the thermal conductivity reduction perpendicular to the film plane, and it fails completely to explain the thermal conductivity reduction along the film plane[15,16]. The reason is that the lattice dynamics model assumes phase coherence of the phonons over the entire superlattice structure and does not include the possibility of diffuse interface scattering, which destroys the perfact phase coherence picture. Partially coherent phonon transport models can capture the trend of thermal conductivity variation in both the in-plane and the cross-plane directions over the entire thickness range[17,18].

Applications

Superlattice thermoelectric technology is also now being actively considered for the thermal management of hot-spot and transistor off-state leakage current in advanced microprocessors. Besides cooling applications, superlattice based thermoelectric devices can also be used for power-conversion applications.

Superlattices with high ZT can improve the performance of in-plane devices. For sensor applications, thermal by pass through the substrate must be minimized by removing the substrate, transferring the superlattice film to another low-thermal conductance substrate, or depositing the film directly on a low-thermal conductivity substrate.

One big question regarding superlattice based thermoelectric coolers and power generations is their stability and reliability. These devices operate under high heat and current fluxes, and both thermo-and electromigration are of great concern. At this stage only a little work has been done. However, the high-temperature reliability of superlattice materials has not been studied.

Summary And Research Needs

For a long time, the miximum ZT for all bulk materials was limited to ZT=1, and as a consequence, applications have been limited to niche areas. Progress made in superlattice-based thermoelectric materials show that ZT=1 is not a theoretical limit. With the availability of high-ZT materials, many new applications will emerge. The progress made also calls for more effort in materials development, theoretical understanding, and device fabrication, concurrent with the pursuit of practical applications of these materials.

Continuous improvement in ZT for different materials in different temperature ranges are needed. In addition to reducing the phonon thermal conductivity, the principle of increasing ZT through quantum confinement of electrons should be exploited, including the exploration of one dimensional nanowires and nanowire superlattices[19]. Further reductions in thermal conductivity may be possible in a periodic superlattices. Similar effects that lead to a reduction in phonon thermal conductivity may be observed in other nanostructures that are more amenable to mass production. In addition to materials development, theoretical studies are needed to further understand the electron and phonon thermoelectric transport, Particularly quantitative tools capable of predicting thermoelectric transport properties are needed. While ZT has reached high values in superlattices, devices made of these materials have not reached the best performance of bulk thermoelectric coolers, due to difficulties in electrical contacts, heat spreading, materials matching, and fabrication.

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