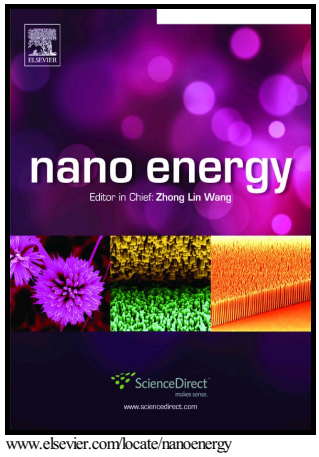
Lone-pair electrons induced anomalous enhancement of thermal transport in strained planar two-dimensional materials

Guangzhao Qin, Zhenzhen Qin, Huimin Wang, Ming Hu



www.elsevier.com/locate/nanoenergy

PII: S2211-2855(18)30357-4
DOI: https://doi.org/10.1016/j.nanoen.2018.05.040
Reference: NANOEN2748

To appear in: *Nano Energy*

Received date: 24 March 2018
Revised date: 6 May 2018
Accepted date: 17 May 2018

Cite this article as: Guangzhao Qin, Zhenzhen Qin, Huimin Wang and Ming Hu, Lone-pair electrons induced anomalous enhancement of thermal transport in strained planar two-dimensional materials, *Nano Energy,* https://doi.org/10.1016/j.nanoen.2018.05.040

Lone-pair electrons induced anomalous enhancement of thermal transport in strained planar two-dimensional materials

Guangzhao Qin,[1] Zhenzhen Qin,[2] Huimin Wang,[1] and Ming Hu[1, 2, 3, a)]

[1)] *Institute of Mineral Engineering, Division of Materials Science and Engineering, Faculty of Georesources and Materials Engineering, RWTH Aachen University, Aachen 52064, Germany*

[2)] *Aachen Institute for Advanced Study in Computational Engineering Science (AICES), RWTH Aachen University, Aachen 52062, Germany*

[3)] *Present address: Department of Mechanical Engineering, University of South Carolina, Columbia, SC 29208, USA*

(Dated: 17 May 2018)

Strain engineering is one of the most promising and effective routes towards continuously tuning thermal transport due to the flexibility and robustness. However, previous studies mainly focused on quantifying how the thermal conductivity (κ) is modulated by strain, while the fundamental understanding on the electronic origin has yet to be explored. In this paper, we establish a microscopic picture of the lone-pair electrons driving strong phonon anharmonicity in two-dimensional (2D) materials beyond the traditional three-dimensional (3D) systems. We provide solid explanation for the unexpectedly up to one order of magnitude enlarged κ of a class of planar monolayers with bilateral tensile strain applied, which is in sharp contrast to the strain induced κ reduction in graphene despite their similar planar structures. The anomalous positive response of κ to tensile strain is attributed to the attenuated interaction between the lone-pair electrons and the bonding electrons of neighboring atoms, which reduces phonon anharmonicity and leads to the enlarged κ. Our study uncovers the electronic origin of the strain modulated thermal transport, which would have great impact on future investigations related to energy nanotechnologies and applications.

Keywords: strain, two-dimensional, thermal transport, first-principles, lone-pair electrons

I. INTRODUCTION

Effective manipulation of heat conduction plays a key role in high-performance thermal management for future nano energy technologies. Phonons play the dominant role in thermal transport in semiconductors[1–3]. Fundamental insight into phonon transport is of great significance to the efficient manipulation of heat flow, which is one of the appealing thermophysical problems with enormous practical implications related to energy, such as electronic cooling, thermoelectrics, phase change memories[3,4], thermal devices (diodes, transistors, logic gates)[5], *etc.* Thermal transport can be manipulated with rationally designed methods or engineered material process, as well as suitable geometry shape control and material composition, such as nanostructuring[6–8], doping[9], introducing porosity[10], applying external field[11], and physical/chemical processing[12–18]. Besides, efficient thermal modulation can also be realized by strain engineering[19–21]. Since it is flexible and robust, strain engineering has become one of the most promising and effective routes towards continuous tunability. Moreover, under realistic conditions in many systems, synthetic materials and nanoscale devices usually contain residual strain after fabrication[22]. However, previous studies mainly focused on quantifying how the thermal conductivity is modulated by mechanical strain[19,21,23–31], while the fundamental understanding of the electronic origin has yet to be explored.

Two-dimensional (2D) materials with graphene as a representative have been intensively studied for many years for their promising applications in lots of fields[3]. Among these, monolayer hexagonal boron nitride (h-BN) with a wide band gap (∼5.0-6.0 eV) offers alternative solutions compensating to the gapless graphene, which establishes the key role of 2D nitrides in advancing the development of next generation nano-electronics[32]. Beyond h-BN, hexagonal aluminum and gallium nitrides (h-AlN and h-GaN) have been also successfully fabricated in experiments recently[33–36] and received extensive attention due to their fascinating properties[36–41]. Thus, the thermal transport properties of these 2D group III-nitrides (h-BN, h-AlN, h-GaN), especially their response to the external mechanical strain, are of great interest for designing energy related novel devices with desirable thermal transport properties in terms of high performance thermal management.

In this paper, we performed a comparative study of thermal transport in 2D group III-nitrides (h-BN, h-AlN, h-GaN) and graphene based on first-principles calculations. The lattice thermal conductivity (κ) of 2D group III-nitrides is unexpectedly enlarged by one order of magnitude with bilateral tensile strain applied, which is in sharp contrast to the strain induced κ reduction in graphene. Considering that such enhancement was for a long time thought to be intrinsic and unique only for non-planar 2D materials, such as silicene and phosphorene, it is very intriguing to observe the anomalous

a)Electronic mail: minghu@cec.sc.edu