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# EFFECT OF GERMANIUM CONCENTRATION ON THE LATTICE THERMAL CONDUCTIVITY FOR SiGe ALLOY NANOWIRES

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## ABSTRACT

The thermal conductivities of $Si_{1-x}Ge_x$ nanowires (NWs) synthesized with Ge concentrations of 0.0%, 0.4%, 4%, and 9% and different diameters were measured from 0 to 450 K using Debye-Callaway model modified to include both longitudinal and transverse phonon modes explicitly has been developed to describe the lattice thermal conductivity for SiGe alloy nanowires as a function of nanowire diameter, alloy concentration, and temperature, obtaining a satisfactory quantitative agreement with experimental results. The model uses nanowires boundary, defects and umklapp and normal phonon-scattering parameters that scaled in a consistent manner with Debye temperature and phonon velocity from their related melting point. The results indicated that the weaker diameter dependence of the thermal conductivity recently observed in $Si_{1-x}Ge_x$ nanowires ($x<0.1$), as compared to pure Si nanowires. The calculations present in the full range of alloy concentrations, ($0 \le x \le 1$), which may serve as a basis for comparison with future experiments on high alloy concentration nanowires.

*Keywords: Lattice thermal conductivity; Debye-Callaway model;* $Si_{1-x}Ge_x$ *alloys*

## INTRODUCTION

Recently, the thermal properties of semiconductor nanowires (NWs) have become the subject of significant attention both for basic science and practical applications. Numerous theoretical calculations[1–3]predicted a significant decrease of the phonon thermal conductivity $K_{th}$of inorganic semiconductor NWs compared to the bulk(Callaway 1959; Holand 1963) value due to modification of phonon dispersion relation and increased boundary scattering at submicron length scales. A significant reduction is likely to create problems in thermal management of high speed and high power nanodevices. Conversely, low $K_{th}$ could be advantageous for nanoscale solid-state energy conversion devices[4].

Understanding thermal transport at the nanoscale is a very important problem. With the continued miniaturization (Shrunk) of the feature size in microelectronic devices, the heat dissipation per unit area will increase which is the major limiting issue for future technological development[5] and also all physical properties such as thermal, electrical, and optical characteristics become noticeably different from those at the bulk system. At the same time, nanostructuring can lead to materials with desirable thermoelectric transport properties, a fundamental one being their thermal conductivity[6].

The measurement of the thermal conductivity at the nanoscale is a challenging experiment. The first measurements of thermal transport of semiconductor NWs were carried out by D. Li *et al.* [7,8]. They measured the thermal conductivity of a few individual Si NWs grown by the vapour liquid solid (VLS) method. The results obtained by D. Li *et al.* [7] showed a strong suppression of the thermal conductivity of the Si NWs as compared with bulk Si. Furthermore, a dependence of the thermal conductivity with the NWs diameter was revealed; the smaller the NW diameter the lower the thermal conductivity.

The thermal conductivity of NWs is not only suppressed by the reduced diameter, but, A. Hochbaum et al. showed that the surface corrugation of the NWs is a major issue for the reduction of the thermal conductivity[9].

Using the thermal bridge device developed by Li et al., the thermal conductivity of other semiconductor NWs has been measured [8,10-14]. All the NWs measured, independently of their nature display a dramatic reduction of the thermal conductivity with the diameter, clearly revealing that the thermal conductivity of NWs is limited by the NWs dimensions. Wingert et al. carried out thermal conductivity measurements on Ge NWs [14], showing that a critical diameter of the NW, the thermal conductivity follows a linear dependence with temperature, in agreement with the behavior of the 22nm Si NW measured in [11,13]. Therefore, this phenomenon can be considered as an intrinsic property of the NWs with reduced dimension, suggesting changes in the phonon transport when phonon confinement effects start to be relevant.The thermal conductivity of NWs is not only suppressed by the reduced diameter, but, A. Hochbaum *et al.* showed that the surface corrugation of the NWs is a major issue for the reduction of the thermal conductivity[9].

Starting from 1960, thermal characterization of solids revealed that the most of the high there conducting materials are adamantine (Diamond like) compounds such as diamond, SiC, AlN, GaN, SiGe, Silicon etc.[14,15]. These compounds are used for compact semiconductor devices, where as efficient heat removal can be crucial to give good performances. To see if is possible to increase the device substrate ability in the thermal management, experimental investigations have been recently done on some of these materials (diamond, silicon and also germanium[16-21]) to establish the role on the phonon conductivity of defects, impurities, and grain boundary and son on.

The thermal conductivity of a crystalline solid-solution is significantly smaller than the thermal conductivity of a pure crystals because high-frequency phonons are strongly scattered by deviations from the perfect periodicity of the crystal[22]. This property of semiconductor alloys has been applied for nearly 50 years in improving the efficiency of semiconductors used in thermoelectric cooling, heating, and power generation[23]. In modern high speed and high-power electronics, however, the reductions in thermal

conductivity created by mass-disorder are detrimental to the operation of the device.
Among thermoelectric semiconductors, SiGe is one of the most important thermoelectric materials because it has shown relatively high thermoelectric efficiency at high temperatures compared to other semiconductors[24]. Thermal conductivity of bulk SiGe alloy is much smaller than that of pure Germanium and the pure Silicon[25]. SiGe based devices are being widely used for spacecraft and space station applications due to their excellent chemical and mechanical stability[24,26-28].The principal object of this note to show that a simple, phenomenological theory of lattice thermal conductivity[12] is capable of giving a reasonably good account of experimental results concerning the effect of varying the concentration of the Ge on the conductivity of $Si_{(1-x)}Ge_x$.

H. Kim et al. showed the effect of composition on the thermal conductivity of semiconductor NWs. They measured the thermal conductivity of alloyed $Si_{1-x}Ge_x$ NWs with a small Ge fraction [29]. Even for very diluted alloys the thermal conductivity of the NWs showed a dramatic reduction compared to pure Si NWs. Thereafter, H. Kim et al. grew rough $Si_{1-x}Gex$ NWs and showed the combined effect of the alloy composition and the surface roughness [30], which permits additional reductions of the thermal conductivity.Thus, opening almost unlimited possibilities for designing nanodevices by its thermal conductivity, which is very attractive for thermoelectric devices. It is only very recently that the thermal conductivity of homogeneous SiGe nanowires with $0< x <0.1$ have been reported, obtaining remarkable reductions in $K_{th}$ below the bulk alloy value.
Finally, we have presented the theoretical dependence of SiGe alloy nanowire thermal conductivity as a function of diameter, temperature, and Ge fraction based on Debye-Callaway model which permit rather a good fitting to the experimental data available.

## CALCULATION OF THERMALCONDUCTIVITY

The Callaway-Holland formalism has been successfully used for the description of the thermal conductivity of bulk semiconductors [31,32]; therefore, it must be conveniently adapted to the nanoscale requirements. It is well known that one of the main consequences of the reduced dimension of the NWs is the appearance of quantum confinement effects; in particular, base on the kinetic theory, the lattice thermal conductivity in general is represented by

$$K_{th} = \frac{1}{3}\sum C_v(\omega) v_g \lambda(\omega) \qquad \ldots(1)$$

Following the approach used by Asen-Palmer[21], who calculated the lattice thermal conductivity for some of group IV and III-V semiconductors using the Debye-Callaway formalism. The lattice thermal conductivity can then be expressed as a sum over one longitudinal ($K_L$) and two degenerate transverse ($K_T$) phonon branches[16 $K_{th} = K_L + 2K_T \quad \ldots(2)$

Where

$$K_{L(T)} = K_{L_1(T_1)} + K_{L_2(T_2)} \qquad \ldots(3)$$

The general expression for the lattice thermal conductivity, which includes an additional term for the normal processes, can be found[33]. The partial conductivities $K_{L_1(T_1)}$ and $K_{L_2(T_2)}$ are the usual Debye-Callaway terms and are given by[21]

$$K_{L_1(T_1)} = \frac{1}{3} A_{L(T)} T^3 \int_0^{\theta_{L(T)}/T} \frac{\tau_C^{L(T)}(x) x^4 e^x}{\tau_N^{L(T)}(x)(e^x - 1)^2} dx \, , \qquad \ldots(4)$$

$$K_{L_2(T_2)} = \frac{1}{3} A_{L(T)} T^3 \frac{\left[\int_0^{\theta_{L(T)}/T} \frac{\tau_C^{L(T)}(x) x^4 e^x}{\tau_N^{L(T)}(x)(e^x - 1)^2} dx\right]^2}{\int_0^{\theta_L(T)/T} \frac{\tau_C^{L(T)}(x) x^4 e^x}{\tau_N^{L(T)} \tau_R^{L(T)}(x)(e^x - 1)^2} dx} \qquad \ldots(5)$$

The factor (1/3) comes from the three contribution branches of vibration spectrum. In Eqs.(4) and (5), $(\tau_N)^{-1}$ is the scattering rate for normal phonon processes, $(\tau_R)^{-1}$ is the sum of all resistive scattering processes (except normal process), and $(\tau_C)^{-1} = (\tau_N)^{-1} + (\tau_R)^{-1}$, with superscripts $L$ and $T$ denoting longitudinal and transverse phonons, respectively. The quantities $\theta_L$ and $\theta_T$ are Debye temperatures appropriate for the longitudinal and transverse phonon branches, respectively, and

$$A_{L(T)} = \frac{k_B^4}{2\pi^2 \hbar^3 v_{L(T)}} \quad \text{and} \quad x = \frac{\hbar\omega}{k_B T} \qquad \ldots(6)$$

here $\hbar$ is the Planck constant, $k_B$ is the Boltzmann constant, $\omega$ is the phonon frequency and $v_{L(T)}$ are the longitudinal (transverse) acoustic phonon velocities, and has a form $(v_b^{-2})_{L(T)} = (1-x)v_{b,Si}^{-2} + x(1-x)v_{b,Ge}^{-2}$ respectively.

The resistive scattering rate is the sum of scattering rates due to phonon-phonon umklapp scattering (anharmonic, $\tau_U$), point defect scattering (isotopes of different mass and alloy, $\tau_M$), scattering from the boundaries ($\tau_B$) and normal three phonon scattering ($\tau_N$). According Matthiessen's rule, the combination of relaxation due to longitudinal as well as transverse phonons and they are given by:

$$(\tau_R^{L(T)})^{-1} = (\tau_U^{L(T)})^{-1} + (\tau_M^{L(T)})^{-1} + (\tau_B^{L(T)})^{-1} + (\tau_N^{L(T)})^{-1} \qquad \ldots(7)$$

The relaxation rate due to the three phonon Umklapp scattering rate for longitudinal and transverse phonons in terms of the dimensionless variable ($x$), is given by [16,34]:

$$\tau_U^{-1}(x) = B_U \omega^2 T e^{-\theta_D/3T} \text{ with } B_U^{L(T)} \approx \frac{\hbar \gamma_{L(T)}^2}{M v_{L(T)}^2 \theta_{L(T)}^D} \qquad \ldots(8)$$

where M is the average mass of an atom in the crystal, *v* is phonon velocity, and γ is the Grüneisen anharmonicity for longitudinal and transverse, respectively.

For most processes, the scattering rate depends on the frequency. Normal processes can then have an important influence by transferring energy between different modes and thus preventing deviation much from the equilibrium population. The approach of Asen-Palmer et al.[21], which has been used by Morelli et al.[16] for Si and Ge is also used similarly like umklapp process as follows:

$$[\tau_N^{L(T)}(\omega)]^{-1} = B_N^{L(T)}\omega^a T^b \qquad \dots (9)$$

With $$B_N^{L(T)}(a,b) \approx \left(\frac{k_B}{\hbar}\right)^b \frac{\hbar\gamma_{L(T)}^2 V^{(a+b-2)/3}}{M v_{L(T)}^{a+b}} \qquad \dots (10)$$

where $B_N^{L,T}$ is coefficient of Normal processes, and *a,b* are adjustable parameters. In present work, its considered the cases (a,b)=(2,3) and (a,b)=(1,4) for longitudinal and transverse phonons, respectively, and V is the volume per atom in the crystal.

The important effect where is taken into account in present work is point defect scattering rate has a fourth-power dependence on frequency that is common for Rayleigh scattering. The relaxation rate for the mass-difference scattering is calculated from the relation[33,35]:

$$\tau_M^{-1} = x(1-x)I\omega^4 \qquad \dots (11)$$

and I is the impurity strength parameter, which at least is the sum of two terms:

$$I = I_{iso} + I_{imp} \qquad \dots (12)$$

The first term of Eq.(12), $I_{iso}$ being due to the scattering by distribution of isotopes of the elements in the compound, and is given by :

$$I_{iso} = I_{iso}^L + I_{iso}^T \qquad \dots (13)$$

where

$$I_{iso}^{L(T)} = \frac{V_o}{4\pi v_{L(T)}^3}\Gamma_M \qquad \dots (14)$$

for longitudinal and transverse modes, respectively, $\Gamma_M$ is the measure of the strength of the mass-difference scattering and is defined as

$$\Gamma_M = \sum_i f_i (1 - \frac{M_i}{\overline{M}})^2$$

Here, $f_i$ is the fractional concentration of the isotopes of mass $M_i$ and $\overline{M} = \sum_i f_i M_i$ is the average atomic mass, while for binary compound composed of two different elements, A and B and is defined as

$$\Gamma_M(AB) = 2\left[\left(\frac{M_A}{M_A+M_B}\right)^2 \Gamma(A) + \left(\frac{M_A}{M_A+M_B}\right)^2 \Gamma(B)\right] \qquad \ldots (15)$$

The value of $\Gamma_M(AB)$ for SiGe is calculated from isotope of Si (92% of $^{28}$Si, 4.6% of $^{29}$Si, 3.1% of $^{30}$Si) and Ge (20.5% of $^{70}$Ge, 27.4% of $^{72}$Ge, 7.8% of $^{73}$Ge, 36.5% of $^{74}$Ge, 7.8% of $^{76}$Ge) and is equal to 5.87 x $10^{-4}$, and the factor 2 is referred to the fact that AB is a binary compound[16].

The second term of Eq.(12), $I_{imp}$, is the strength parameter due to scattering caused by impurities and is given by the relation:

$$I_{imp} = I^L_{imp} + I^T_{imp} \qquad \ldots(16)$$

where

$$I^{L(T)}_{imp} = \frac{3V_o^2 S^2}{\hbar v^3_{L(T)}} N_{imp} \qquad \ldots(17)$$

where S is the scattering factor which usually has a value close to unity[34] and $N_{imp}$ is the impurity concentration. However, in bulk crystals, impurity concentration up to $10^{17}$cm$^{-3}$ has no effect on lattice thermal conductivity [36].

In the present work, the phonon-boundary scattering rate is assumed to be independent on both temperature and frequency, and can be written as[33]:

$$(\tau_B^{L(T)})^{-1} = \frac{v_{L(T)}}{d} \qquad \ldots(18)$$

where d is the effective diameter of the sample (d=2R for cylindrical with radius R, d=$L_C$ for rectangular, where $L_C$ =1.12ab, a,b is side length). The value of d will be adjusted slightly for each crystal in order to fit the $T^3$ dependence of the thermal conductivity at the lowest temperatures. So, the expression for the phonon-boundary scattering used for description of the K(T) has a form

$$(\tau_B^{L(T)})^{-1} = v_{L(T)}\left(\frac{1}{L_C}\frac{(1-P)}{(1+P)} + \frac{1}{\ell}\right) \qquad \ldots(19)$$

Where $l$ is sample length in the direction of heat flow and is equal to 5 x $10^{-6}$m, and $P$ is probability of scattering on surface and its value varies from zero to one.

In order to calculate lattice thermal conductivity for SiGe nanowires, the crystal size dependent parameters should be taken care of. These parameters are phonon group velocity $v_g$, Debye temperature $\theta_D$, surface roughness $P$, and lattice dislocation which is regarded as defects impurity $N_{\mathrm{def}}$. An explicit analytic expression is obtained[21,33,34]. When the system is assumed to be isotropic, the average group velocity is proportional to the crystals characteristic Debye temperature. In this case, if the system is considered to be isotropic, then[37];

$$\theta_D \propto \frac{2h}{\pi k_B}\left(\frac{3N_A}{4\pi V}\right)^{1/3} v_g \qquad \dots (20)$$

where $N_A$ is Avogadro number, and $V$ is the volume per atom. Eq.(20) is assumed to be valid for nanoscale size crystals and if $L$ is the size dimension of the nanostructure denoted by n, such as the diameter of nanowires or the thickness of thin films, and the superscript $b$ for ($L\rightarrow\infty$) denotes the corresponding bulk limit, then Eq.(20) for both the bulk and nanowire will give the size dependence of both the phonon group velocity and the Debye temperature in the form,

$$\frac{v_g^n}{v_g^b} = \frac{\theta_D^n}{\theta_D^b} \qquad \dots (21)$$

where $v_g^b$ is bulk group velocity, $v_g^n$ is the nanowire group velocity, $\theta_D^b$ is bulk Debye temperature and $\theta_D^n$ is the nanowire Debye temperature.

Lindemann's proposition of melting criterion, known to be valid for small particles, states that a crystal will melt when the root mean square displacement (MSD) of atoms in the crystal exceeds a certain fraction of the inter-atomic distance[38]. From that of the relationship between the melting point and the Debye temperature of crystals can be determined. From the Einstein specific heat theory, the square of the characteristic temperature is proportional to the melting point $T_m$ of the crystal:

$$\theta_D = Const.\left(\frac{T_m}{MV^{2/3}}\right)^{1/2} \qquad \dots (22)$$

where $M$ is the molecular mass. By dividing the left and right hand side of Eq.(22) for nanowires by the same equation for bulk, one obtains[39],

$$\frac{\left(\theta_D^n\right)^2}{\left(\theta_D^b\right)^2} = \frac{T_m^n}{T_m^b} \qquad \dots (23)$$

where $T_m^b$ is the melting temperature for the bulk, and for Si is equal to 1690K and for Ge=1210K[40],while for SiGe is equal to [$T_m^b$=1690-279.6x] and $T_m^n$ for nanowires.In the nanoscale range, relation (23) gives a size dependent Debye temperature $\theta_D^n$ calculated from the samples melting point. Then the size dependent melting point[25]:

$$\frac{T_m^n}{T_m^b} = \exp\left(-\frac{2(S_m - R)}{3R(r_n/r_b - 1)}\right) \qquad \ldots (24)$$

For low-dimensional crystals, it is clear that $r_b$ in Eq.(24) should be dependent on *d*, and is given by:

$$r_b = (3 - d_s)h \qquad \ldots(25)$$

where $d_s$ is structure dimension, $d_s$ = 0 for nanocrystals, 1 for nanowires and 2 for thin films, and *h* is the diameter of a spherical volume occupied by a crystal lattice and its value is equal to 0.3338nm for Si[41] and for Ge is 0.381[**?**], while for SiGe ($h_{SiGe}$=0.3338+0.172x)nm.

Table1: Parameter values used for calculation

| Parameters | Formula |
|---|---|
| Lattice constant (nm) | a = $0.002733x^2 + 0.01992x + 0.5431$ |
| Atomic Mass(g/mol) | M = $28.09(1-x) + 72.61x$ |
| Density(Kg/m$^3$) | $\rho = 2329 + 3493x - 499x^2$ |
| Melting point(K) | $\theta^L = 586 – 253x$<br>$\theta^T = 240 – 90x$ |
| Atomic diameter(nm) | $h_{Si}$ =0.3338<br>$h_{Ge}$ =0.351<br>$h_{SiGe}$ = 0.3338 + 0.0172$x$ |
| Atomic Volume (nm$^3$) | $V_o = a^3/8$ |

## RESULT AND DISCUSSION

A big constraint for the applications of NWs in which the thermal conductivity plays a major role is the lack of experimental data over a large range of diameters and temperatures, or in its defect the absence of a model supplying such information. In view of the difficulties inherent to thermal conductivity measurements at the nanoscale, and the variety of diameters and Ge concentration, the development of a model capable of predicting the thermal conductivity in a wide range of diameters and Ge concentration acquires a capital interest.The current development, towards smaller and faster devices, forces the developers to look into other materials than silicon. A natural choice is a $Si_{1-x}Ge_x$alloy (*x* denotes the relative Ge concentration in the alloy), which is relatively easy and cheap to incorporate into existing standard Si processes[43]. $Si_{1-x}Ge_x$has the same lattice structure as Si, but its lattice constant increase with increasing *x*.

In comparison between the experimental data and present work for four NWs using a modified Callaway-Holland formalism (Debye-Callaway models) is shown in Figure (1), it can be seen that rather good quantitative agreement is obtained for three types of NWs with Ge concentration x=0.004 and 0.09, especially at temperature 300K and above, which is better than the result present in Ref.[**35**]. While for the NWs with *x*=0.04 and D=344nm, where there is a big deviation with experimental data. This may be due to the fact that K is very sensitive to Ge concentration *x*. There is a significant decrease of lattice thermal conductivity especially at smaller diameter D and high Ge concentration. The latter comes from the result of increased umklapp, boundary and defect (alloy or dislocation) which is the most important effect taken into account in present work, and also due to modification of phonon dispersion due to the spatial confinement.

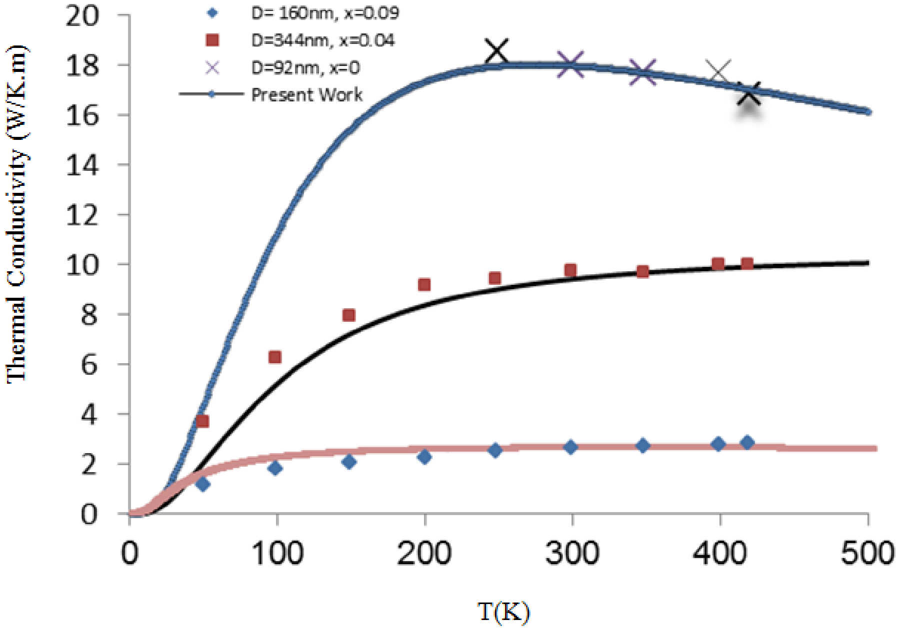

Figure 1: Thermal conductivity K versus temperature T for $Si_{1-x}Ge_x$ NWs. X is the germanium concentration and D is the NWs diameter. The lines show calculation results using Eq.(1), the symbols represent experimental results from ref.[30]

Alloy scattering is a powerful scattering mechanism and it is a dominant in the SiGenanowires as compared to Si and Ge nanowires where it is absent. The alloy scattering isthe result of atomic mass difference between the Si and Ge and the change of springconstant representing the bonding between atoms in SiGe. This alloy scattering effectsmoothens out the peak in the thermal conductivity curve of the SiGe nanowires which isfound in Si and Ge at around 100K and 80 K respectively[16]. This is due to the fact that alloyscattering is more dominant than phonon-phonon Umklapp scattering[7]. The phonon-phononUmklapp scattering sets in at 180K for Si and 101K for Ge[44]. Using linear approximation ofthe ‘Umklapp scattering set-in’ temperature, Umklapp scattering is supposed to occur at140K for the SiGe nanowires. A possible explanation for the shape of the curves is asfollows. Before the Umklapp scattering comes into play, that is before 140K the boundaryscattering is dominant and the thermal conductivity increases with temperature. After 140K,the phonon frequencies are higher and the dominant alloy scattering comes into play alongwith the less dominant phonon-phonon Umklapp scattering.9, because, it is related to the very fast frequency dependence of alloy scattering.  alloy scattering blocks high frequency phonons very effectively, but it is totally transparent to low frequency phonons. Thus, at higher temperatures,the boundary scattering effect gets relaxed and the balancing act between the increase ofalloy scattering and decrease of boundary scattering keeps the curve almost constant at hightemperatures. Thus, the thermal conductivity of an alloy is dominated by low frequency phonons with very long mean free paths, whereas in nonalloys *K* contains contributions from a larger range of frequencies with shorter mean free paths on average[35].

Introducing a boundary therefore affects the thermal conductivity of an alloy already at rather large values of *D*, whereas for that same *D* there is little effect on *K* of a nonalloy. In the limit of very small *D*, however, boundary scattering dominates over alloy and anharmonic scattering at all frequencies. In that regime, *K*of Si and SiGe become similar, because the effect of ‘bowing’ introduced by alloy scatteringdisappears[35].

Finally, we concluded that the thermal conductivity of $Si_{1-x}Ge_x$ NWs as a function of diameter, temperature and alloy concentration. As well as the lattice thermal conductivity decreased with increasing diameter, thus it can be seen that as the concentration is increase the thermal conductivity of $Si_{(1-x)}Ge_x$is increased as shown Figure (1). Thus results obtained the weak dependence on diameter is reported and rather a good agreement with the experimental data.

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