Thermal Conductivity of the Crystalline Silicon

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Amelia Carolina Sparavigna

(Department of Applied Science and Technology, Politecnico di Torino)

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Abstract

In this paper we discuss and compare theoretical and experimental data of thermal conductivity in crystalline Silicon (natural and enriched). We focus in particular on the role that the isotopic mass defects have in determining the value of this conductivity.

Keywords: Solid State physics, Materials Science, Thermal Conductivity, Crystalline Silicon, Isotopic Mass Defects, Phonon Scattering.

Introduction

In the theoretical study of the thermal conductivity in non-metallic solids, a widely used approach is that of solving the Boltzmann equation of the phonon thermal transport to obtain the related conductivity [1-3]. Many researchers are using methods of solution based on the relaxation time approximation, of which we proposed a simplified discussion in [4,5]. Other researchers prefer the use of methods which are involving more specific atomic models [6-12]. In general, all the used methods are able of describing the behavior of the thermal conductivity, when they are involving a proper formulation of the normal and umklapp phonon-phonon scattering and of the other scattering mechanisms due to the presence of defects in the crystal.

If we consider a solid where only phonons are acting in the thermal transport, the sample turns out to be like a box containing them. At very low temperatures, we have a few phonons interacting only with the walls of the box. As the temperature increases, the population of phonons increases too and consequently their thermal conductivity. However, with a further increase of temperature, also the interactions among phonons become relevant. Therefore, after the thermal conductivity has reached a maximum, its value starts decreasing with a further increase of the temperature.

Among the processes which are influencing the values of the thermal conductivity we find the scattering of phonons due to the presence of isotopic mass defects. Actually, the natural crystals of several elements are composed by different isotopes; that is, in these crystals with natural composition

we have the sites of the crystalline lattice occupied by atoms of the same element having different masses. This disorder of the lattice reduces the thermal conductivity.

For an experimental study of the thermal conductivity without the isotope effect, the sample needs to be made of an isotopically enriched material. Among the materials which have received specific enrichment for the analysis of thermal conductivity we find diamond-like Carbon, Silicon and Germanium [13-17]. As resulting from experiments, the isotopic purity of the solid is strongly influencing the thermal conductivity. This is important for technological applications of these materials, such as for a better comprehension of the thermal transport.

Here we discuss the case of crystalline Silicon. Specifically, we discuss some of the literature on it, which is providing experimental data. Theoretical works had been proposed in the past by the author, who solved the phononic Boltzmann equation in crystals in which different isotopes are present. Let us start for these theoretical works on the thermal conductivity of Silicon.

An iterative approach

In [18,19] we proposed an iterative approach to the solution of the Boltzmann equation. In the calculation of the thermal conductivity, we inserted competing scattering mechanism due to isotope impurities and the three-phonon collisions, without resorting to the systematic use of the relaxation time approximation. The model that we used was not so specific as in the following works [6,7], but it was able to give a good agreement with experimental data for silicon, here shown in the Figure 1.

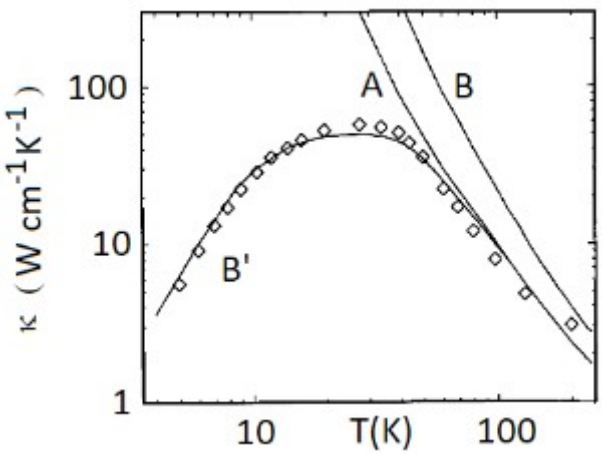

Figure 1: Thermal conductivity of Silicon as a function of temperature at room pressure. The curves A and B are the results of calculations proposed in [19], with and without isotopic mass defects, in the case of absence of the boundary scattering. This scattering is considered for curve B'. The experimental points are from Holland and Neuringer [20].

Let us note that the results given in the Figure 1 had not been obtained by using adjustable parameters. Actually, the use of adjustable parameters is rather common in the methods involving the relaxation time approximation. A criticism to this use was made by Srivastava too [21].

In [19] we have also considered the isotope effect in Germanium. And for it too we observed the

relevant role of the purity of the sample. For our further discussion on Silicon, let us extrapolate from the Figure 1 the thermal conductivity at 200 K. For the natural Silicon it is of 2.2 W/cm-K and for the pure sample, it is of 3.8 W/cm-K.

Following the methods given in [6,7], the effect of isotopes on the thermal conductivity of Silicon was investigated again in [22]. Actually, in [6], [7] and [22], we applied the iterative method on a revised model of the solid: we used a crystal described by its real Brillouin zone and Hamiltonian to determine properly the three-phonon normal and umklapp processes.

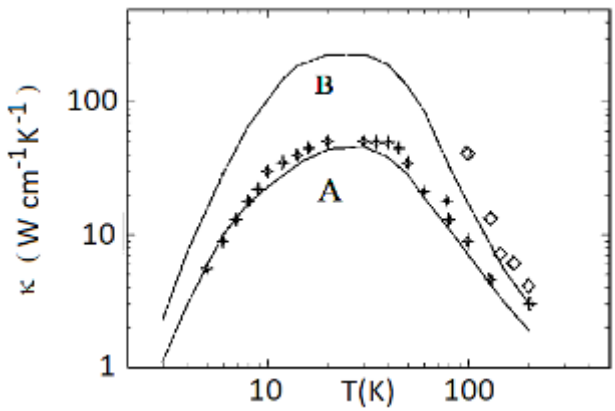

Figure 2: Thermal conductivity of Silicon as a function of temperature at room pressure. The curves A and B are the results of calculations proposed in [22], with and without isotope scattering. The experimental points are from Refs. 20 and 23.

Again, for the further discussion, let us extrapolate from the Figure 2 the thermal conductivity at 200 K. For the natural Silicon it is of 2. W/cm-K and for the pure sample, it is of 3. W/cm-K .

As we can see from the values obtained in [19] and [22], we have that the ratio of the thermal conductivity of enriched and natural Silicon, at 200 K, is 1.72 in the case of [19], and it is 1.5 in the case of [22]. It means that, using a more realistic mode, the effect of the isotopic mass defects is reduced. From [22], we estimate that the thermal conductivity at 200 K is reduced of 30% at the most.

Experimental results

At the time we investigated the use of the iterative approach to solve the Boltzmann equation, we had not the opportunity of having a service, such as Google Scholar or other web supports, to find easily the literature on the subject. As the reader can see, we had at our disposal only the access to a few experimental data for Silicon. Now, let us examine the further data that we find on the web today.

First, we consider the paper by Capinski, Maris, Bauser, et al. [24]. The experimental data are given in the Figure 3.

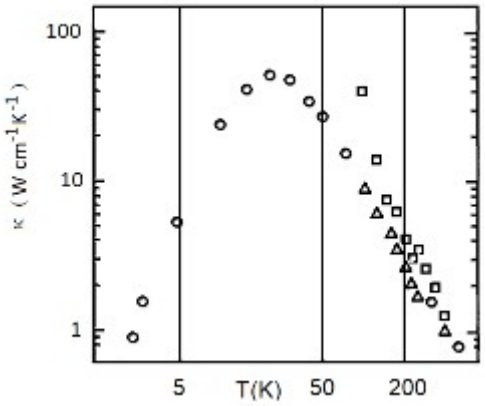

Fig. 3: Thermal conductivity of isotopically pure ^{28}Si (squares) and Si of natural isotopic abundance (triangles) [24]. The circles are the data from Ref.25.

The data from Ref.24 tell us that, at 200 K, the thermal conductivity of the natural Silicon is of about 2.8 W/cm-K and that of the isotopically pure Silicon of about 4 W/cm-K . The ratio of the thermal conductivity is 1.42. The isotope scattering is reducing the thermal conductivity of 30%. These results seem in good agreement with the previously given theoretical data.

In 2000, new data on the thermal conductivity of Silicon had been published [26]. Here in the Figure 4, we give some of the experimental points.

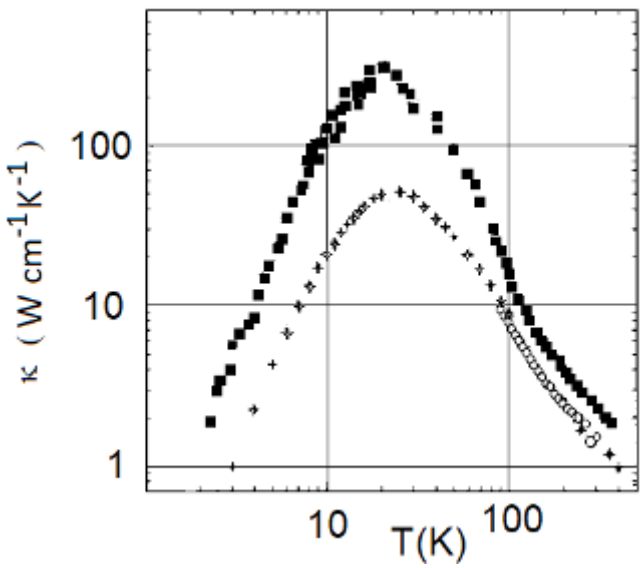

Fig. 4: Thermal conductivity of isotopically pure ^{28}Si and Si of natural isotopic abundance. Some data from Ref.26. The open dots represent the measurements on sample SIN1 of natural silicon. As told in [26] the “plus” symbols denote the “standard” curve for natural Si, as given by [27].

Let us consider the data from Ref.26 at 200 K. The thermal conductivity of the natural Silicon is of about 2.3 W/cm-K and that of the isotopically pure Silicon of about 4 W/cm-K (Figure 3). In [26], the authors discussed the result in the following manner. “In principle, the thermal conductivity enhancement,

which is most prominent at the maximum of kappa, where umklapp processes are frozen out, should vanish rapidly at higher temperatures. However, we find an almost constant enhancement in kappa of about 60% between 100 and 300 K and estimate this effect to persist up to at least 400 K. This observation also contradicts the predictions of a recent calculation for the thermal conductivity enhancement in isotopically pure silicon". The authors are referring to article [28].

The behavior at high temperature

Data published in [26] had been discussed in the work by Inyushkin in [29]. He noted, about the phonon free path at low temperatures, that a "tremendous discrepancy" between theoretical and experimental values exists, and it "cannot be rationalized in the framework of the current views of heat conduction in crystals in the boundary scattering regime". Actually, also the lack of results for sample SIN1 (natural Silicon) at low temperatures was stressed.

The authors of [26] considered Inyushkin's analysis and reconsider their results. "In view of the interest, in this problem" [29], "we have recently remeasured the same and other similar samples with a different apparatus around T = 300 K. We consistently found an increase in the k of ^{28}Si with respect of that of ^{nat}Si of about 10 %, whereas the values of this enhancement measured below 100 K are the same as those in the original data" [30].

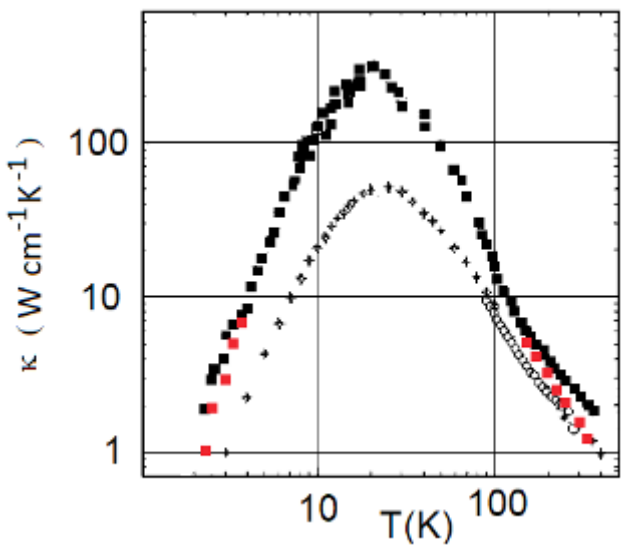

Fig. 5: The same as in the Figure 4 with the red squares representing the correction reported in [30] for the data of pure ^{28}Si.

The authors, in the new measurements, found the thermal conductivity of the natural Silicon the same as before, but the "values reported earlier for ^{28}Si are systematically 90 W/m-K higher than the present ones". "This difference … below 100 K … reduces the enhancement … reported earlier for ^{28}Si from 60% to about 10%. The reason for the discrepancy is not known, but we believe the new data to be more reliable".

Therefore we can revise accordingly the data of the Figure 4, obtaining Figure 5.

Let us consider the value of the thermal conductivity of the enriched Silicon at 200 K. From the Figure 4, we have a value of 400 W/m-K: if we reduce it of 90 W/m-K, we obtain a value of 3 W/cm-K (Figure 5). Let us note that this value is in good agreement with the result of the theoretical model in [22]. From the plot, the conductivity of natural Silicon seems being 2.2 W/cm-K. The ratio of the conductivity of enriched and natural Silicon seems being 1.36. Then, the enrichment increases, at 200 K, the conductivity of 36% .

Discussion

Let us consider again the results given in [22]. In the Figure 6, we can see side by side the theoretical results given in the Ref.22 and the experimental data given in the Figure 5, with the corrections for the enriched sample (red squares).

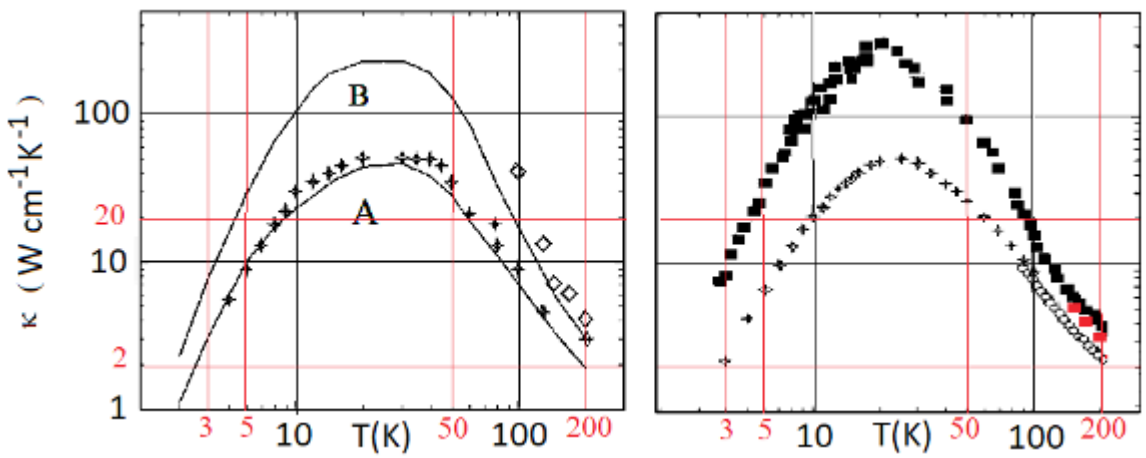

Figure 6: On the left, the theoretical curves from [22], on the right the experimental data [26,30]. The red lines had been added to help the reader in the comparison.

We can discuss the comparison. First of all, I have removed the experimental data of the enriched Silicon below 3 K. These data depend on the boundary scattering and therefore on the size of the sample. For the enriched sample, the specific value of the thermal conductivity at the peak is different, however the thermal conductivity obtained from theory between 3 K and 10 K and between 30 K and 200 K is in good agreement with experimental data. For the natural sample, above 10 K, the overall behaviors are in agreement. Moreover, the theoretical curve is in agreement with the open circles from [26].

New data

In [31], the researchers measured the thermal conductivity of isotopically enriched ^{28}Si "independently in three laboratories by high precision experiments on a total of four samples of different shape and degree of isotope enrichment in the range from 5 to 300 K with particular emphasis on the range near room temperature. The results obtained in the different laboratories are in good agreement with each

other. They indicate that at room temperature the thermal conductivity of isotopically enriched ^{28}Si exceeds the thermal conductivity of Si with a natural, unmodified isotope mixture by 10±2%. This finding is in disagreement with an earlier report by Ruf et al.". The results given in [31] are in agreement with the data from [32].

Let us note that the results for natural Silicon from [31], at 200 K, are rather different from data given in [24], and also from [26] and [30].

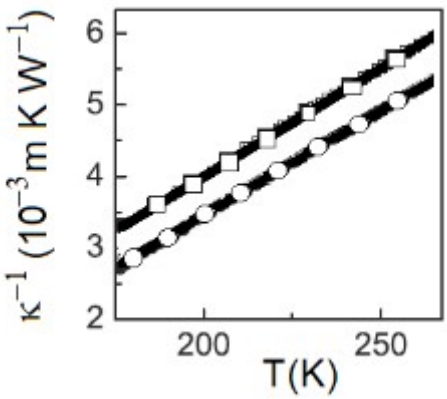

Fig.7: Data from [31].

From the Figure 7, at 200 K, the conductivity is comprised between 2.5 and 2.9 W/cm-K. The ratio of the conductivity is 1.16. Actually, the new measurements had increased the value of the thermal conductivity in the natural sample of 16%.

Let us stress this fact, that it is a good set of measurements on the natural sample which is relevant for any model of the phonon scattering by the isotopic mass defects. For what concerns the calculation in [22], the given model based on a real representation of the crystalline lattice and of the scattering processes produces values of the thermal conductivity in good agreement for the pure crystal. For the crystal with natural isotopic mass defects, the formula used for the scattering produced by these defects is giving, at 200 K, a conductivity 25% lower than the new experimental value. In the case of Diamond and Germanium, that were studied in [22] too, the agreement between theory and experiments is good on all the range of temperature.

Conclusion

In this paper we have presented some theoretical curves and experimental data on the thermal conductivity of crystalline Silicon. Let us stress that a good overall agreement of the theoretical curves with experimental values exists, in particular for enriched Silicon [22]. The theoretical curves had been obtained from a realistic model, without the use of adjustable parameters. For what concerns the natural Silicon, the most recent data here considered give a thermal conductivity at 200 K which is about 10% larger than the previously determined value. It would be interesting to understand the origin of this difference in the experimental values.

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