Realizing the Ultrahigh Thermal Conductivity and Ambipolar Mobility of Cubic Boron Arsenide (c-BAs)

Gavin Lindsay Literature Seminar Dec 1, 2022

Semiconductor materials are essential components of everyday devices including solar cells, photodiodes, and the transistors that are fundamental components of computer processors. The steady decrease over time in the sizes of transistors and other integrated circuit elements, as embodied in Moore's Law, has led to a significant new problem, which is that today's integrated circuits evolve considerable heat per unit volume.[1] Overheating can be avoided by more efficiently dissipating the heat or by utilizing semiconductor materials with faster response times to decrease the required power. [2] I will introduce a new semiconductor material that fulfills both of these roles.

Two key properties of semiconductor materials are thermal conductivity and ambipolar mobility. Thermal conductivity is rate at which heat is transferred in a material, and ambipolar mobility which is a measure of the mobility of electrons and holes in a material. Efficiently dissipating heat increases the lifetime of semiconductor devices by decreasing the stresses from thermal expansion.[3] Increasing the mobility of the charge carriers can increase the output or decrease the power required to maintain the same output.

Current semiconductor devices in use have low thermal conductivities and ambipolar mobilities. Of common semiconductor materials, SiC has the highest room temperature thermal conductivity of around 500 W mK^{-1} whereas Si, the most used semiconductor, has a room temperature thermal conductivity of only 150 W mK^{-1}.[4] Diamond has a much higher thermal conductivity of about 2000 W mK^{-1} but it is an insulator. Very few semiconductors have thermal conductivities near diamond as seen in figure 1. High thermal conductivities are generally seen in materials with small atoms and strong bonds. This correlation predicts that cubic boron arsenide (c-BA) should have a relatively low thermal conductivity, owing to the large size and relatively weak bonds formed by arsenic.

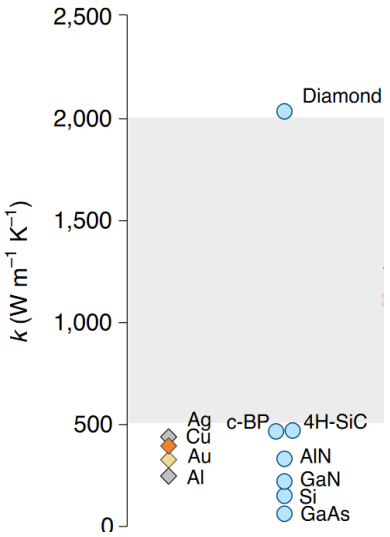

Figure 1. Common thermal conductivities of materials showing the large gap between semiconductors and diamond.

However, these predictions do not include the effects of phonon-phonon scattering. This phenomenon plays a key role in an approach called band gap engineering: high thermal conductivities should be seen in materials that have large phonon band gaps, owing to a reduction in inelastic phonon scattering.[4] In 2013, computational modeling that included the effects of phonon-phonon scattering suggested that c-BA should have an unusually high thermal conductivity of over 2,000 W mK^{-1} owing to the large band gap between

the acoustic and optical phonons.[5] Including four-phonon scattering, which is often considered negligible, lowers the calculated conductivity to 1,260 W mK^{-1}, but this value is still much larger than those of most other semiconductor materials.[6] Scattering of phonons off defects in the crystal also decreases the thermal conductivity, and attempts to verify the thermal conductivity of c-BA experimentally yielded a value of only 200 W mK^{-1} about a tenth of the theoretical value.[7]

In addition to scattering off other phonons and defects, phonons and electrons can scatter off one another. This scattering greatly affects electron and hole mobilities. Recent computations have predicted that c-BA should have high ambipolar mobilities.[8]

In 2018, Fei Tian et al. experimentally verified the high thermal conductivity of c-BA.[9] c-BA seed crystals were synthesized by chemical vapor transport (CVT), and those with the highest purity were cleaned and used to grow larger high-quality single crystals by CVT. The local thermal conductivity was determined by time- and frequency-domain thermoreflectance studies to be 1,300 W mK^{-1} at room temperature, in agreement with the four-phonon computational prediction.

The bulk thermal conductivity was then measured by two methods that determined the temperature gradient across the single crystal when in contact with a heat source and sink. Both methods gave values of around 800 W mK^{-1}. The bulk value is somewhat smaller than the local value measured by thermoreflectance, most likely due to scattering off defects in the crystal, but, even so, the bulk thermal conductivity is still more than five times greater than that of Si.

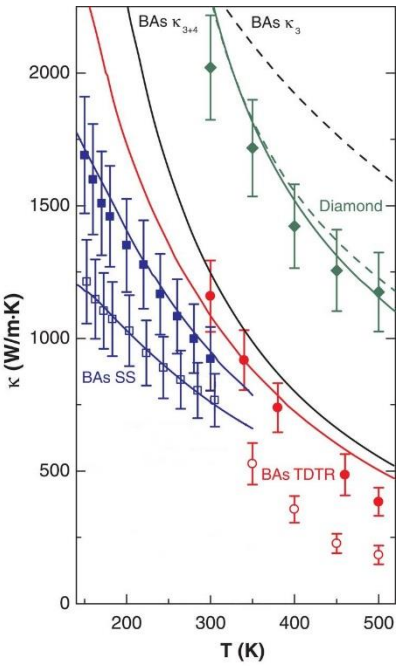

Figure 2. Theoretical thermal conductivity for diamond (green) and c-BA (black) with 3 (dashed) and 3+4 (solid) phonon scattering and experimental data for two different crystals (filled and unfilled symbols) for TDTR (red) and thermocouple (blue) data with their fits (lines).

The ambipolar mobility of c-BA was measured in 2022 by Jungwoo Shin et al.[10] An optical transient grating was used to measure both the local thermal conductivity and the ambipolar mobility. By varying the seed crystal, doping, and precursor material for the CVT synthesis, the effect of impurities was also explored. c-BA was measured to have an ambipolar mobility of 1,600 $cm^2\ V^{-1}\ s^{-1}$ that is close to the theoretical prediction of 2,100 $cm^2\ V^{-1}\ s^{-1}$, and a thermal conductivity of 1,200 W mK^{-1} in agreement with the previous paper. Charged and neutral impurities were shown to decrease both thermal conductivity and ambipolar mobility, with charged impurities having a larger detrimental effect.

These two papers confirm the high thermal conductivity and ambipolar mobility of c-BA. The experimental values validate the phonon band gap engineering approach for high thermal conductivity materials and the computational predictions of the properties. Although these

properties are promising, the use of these materials is limited by the synthetic procedure which is long and produces crystals with large variations in the properties. In addition, impurities decrease both values, but would be needed to p and n dope c-BA for use as an active semiconductor material. Pure c-BA may still be of interest as a replacement for pure Si as the backboard for other p and n doped semiconductor materials. However, the higher ambipolar mobility and thermal conductivity of c-BA make it a great candidate for a new semiconductor material.

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