Defect Characterization in Cubic Boron Arsenide and Boron Phosphide via Aberration-Corrected Low-Voltage STEM

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As transistors inside chips become more compact, heat generation and management have become a limiting factor for high performance electronic devices[1,2]. One approach towards better heat management is to synthesize high thermal conductivity materials. Diamond, the highest thermal conductivity material, possesses room temperature thermal conductivity up to 2,200W $m^{-1}K^{-1}$, but its application is limited by the high synthesis cost and small thermal expansion coefficient, which makes it incompatible with CMOS devices. Cubic boron arsenide (BAs), a III-V compound, is predicted to possess room temperature thermal conductivity slightly lower than that of the diamond[3]. Its thermal expansion coefficient is calculated to be compatible with Si, and millimeter scale growth has been demonstrated[4]. However, experimentally synthesized cubic BAs has so far exhibited much lower thermal conductivity than theoretically predicted[4]. A likely reason for the low measured thermal conductivity of BAs is the presence of intrinsic defects introduced during crystal growth.

In this work, we use electron microscopy to examine the structure and possible defects in cubic BAs synthesized using two different methods[5,6]. The room temperature thermal conductivity of both samples is measured to be 900~1000W $m^{-1}K^{-1}$, 70% of the theoretical value[7]. In bright and dark-field TEM images of the sample synthesized by F. Tian *et al*[5], we observed grains extended hundreds of nanometers to few micrometers, separated by sharp grain boundaries running parallel to the crystal surface. By performing selected area electron diffraction (SEAD) and aberration-corrected low-angle annular dark field scanning transmission electron microscopy (ADF-STEM) at 80 kV, we identified the lattice within neighboring grains to have mirror symmetry, forming twin boundaries in between. However, similar defects are not observed in BAs samples synthesized using method by S. Li *et al.*[6], despite similar measurement results of their thermal conductivity. Such findings imply that the measured thermal conductivity in these samples is not limited by twin boundaries, but rather point defects such as impurities and vacancies. Moreover, by using a low 80 kV accelerating voltage, monochromated imaging with <0.3 eV energy spread, and drift-corrected frame averaging, we are able to image the lattice at high resolution while limiting knock-on damage of light elements such as B and P.

Another III-V compound, cubic boron phosphide (BP), is also predicted to possess high thermal conductivity reaching half of that of BAs[8]. We conduct STEM studies on isotopically pure BP crystals synthesized by S. Li *et* al[9], with a measured room temperature thermal conductivity of 540 W $m^{-1}K^{-1}$, approaching the theoretical value of 580 W $m^{-1}K^{-1}$. No signatures of large scale structural defects or point defects are observed. Our findings will provide insight into synthetic methods to improve crystal quality, in order to produce BAs and BP crystals with ultra-high thermal conductivity[10].

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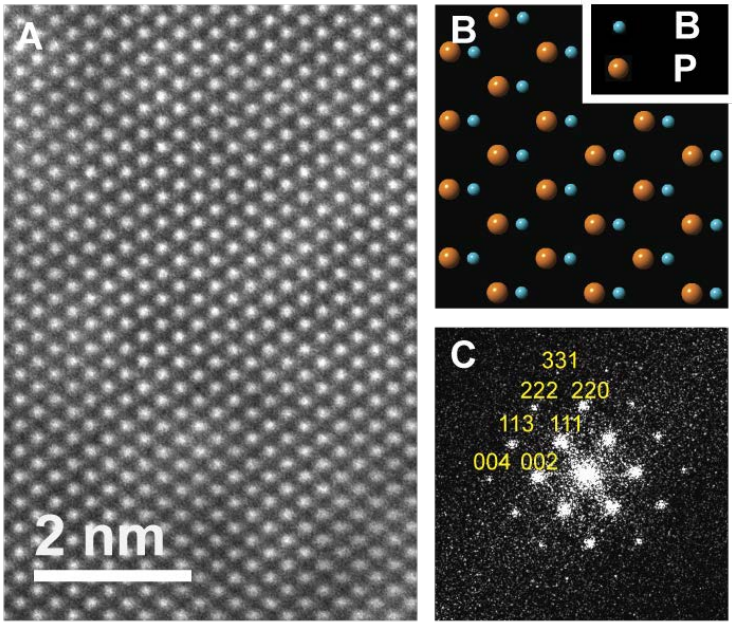

Figure 1. ADF-STEM imaging of BP crystal (A) Annular dark field STEM image of a BP crystal along the [110] zone axis. **(B)** Cartoon of BP crystal structure along the [110] zone axis. **(C)** Windowed Fourier transform of the STEM image in **(A)**. Reproduced from Reference 9.