Experimental Core Deformation of Silicon from Powder X-ray Diffraction

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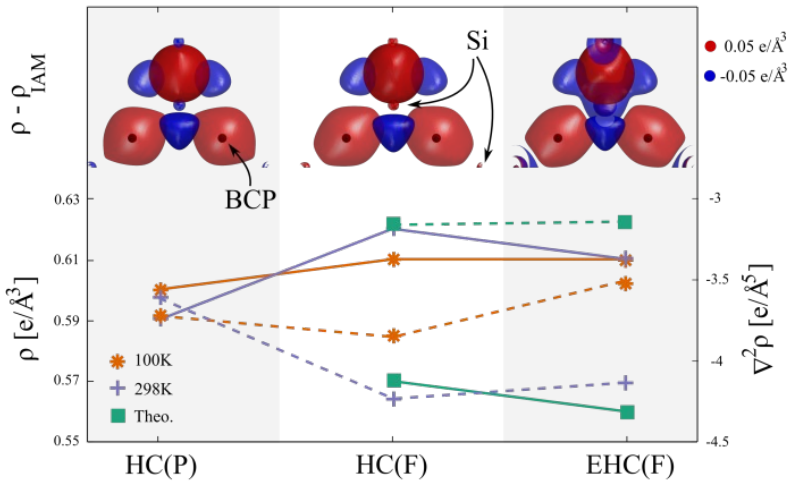
Crystalline silicon is arguably the most studied extended network structure; the electron density (ED) and structure factors have been determined and redetermined numerous times. All the proposed ED models agree on the covalent characteristics of the valence density; however, the agreement does not extend to the core ED. The study of Spackman suggests an inner-shell deformation, while Deutsch argues that such deformation is accompanied by a second displacement parameter controlling the inner density.[1,2] The major obstacle in verifying these models has been the limited data range. Based on a benchmark study on diamond, synchrotron powder diffraction has recently been established as a viable tool for probing the inner ED. [3,4]
Here, we present the modelling of the core density of crystalline silicon based on data up to $\sin(\theta)/\lambda = 1.7$ Å^{-1}. The valence density is in perfect agreement with previous results, validating the use of structure factors obtained from powder diffraction. The core ED is modelled explicitly in the framework of the extended Hansen-Coppens (HC) multipolar model [5,6], revealing an increase in the core ED. The core ED of the Si atom thereby contracts upon formation of covalent bonding in crystalline silicon. The atomic displacement parameter was determined from a Wilson plot, containing no clear evidence of a second displacement parameter.



[1]: M. A. Spackman, Acta Cryst. A **42,** 271 (1986)

[2]: M. Deutsch, Phys Rev B **45**, 646 (1992)

[3]: M. R. V. Jørgensen *et al.* IUCrJ **1**, 267 (2014)

[4]: N. Bindzus, *et al.* Acta Cryst A **46**, 39 (2014)

[5]: N. Hansen and P. Coppens, Acta Cryst A **34**, 909 (1978)

[6]: A. Fischer, *et al.* J Phys Chem A **115**, 13061 (2011)