Predicting crystal structures by random searching

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We predict crystal structures by relaxing randomly chosen starting configurations within density functional theory (DFT). We combine this with constrained searches incorporating knowledge derived from the random searches and other sources. Applications to various systems are described, including hydrogen[1], silane[2], and aluminum hydride[3].

Our study of solid hydrogen includes harmonic proton zero point motion[1] and leads to a radical revision of the DFT phase diagram up to 400 GPa. The most stable phases remain insulating to very high pressures, in agreement with experiment, and we find a strong candidate for phase III.

We predict that the equilibrium structure of silane is insulating up to about 200 GPa[2]. Our stable structure in the pressure range 50-250 GPa has very symmetry, each Si site is equivalent and each H site is equivalent, and it is held together by electron-deficient three-center-two-electron bonds. This structure has been observed in a recent x-ray diffraction study[4].

For aluminum hydride we find a transition from the insulating low-pressure alpha phase to an insulating layered structure and then to a metallic phase[3]. Our metallic structure has been observed in a recent x-ray diffraction study[5].

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