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Theoretical Model for Predicting Superconductors and New Solid Structures

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Using only the atomic numbers and the atomic masses of the constituent atoms of a solid, it is now possible to predict[1] which crystal structure from a group of candidate structures is likely to be the most stable at a given pressure. It is also possible to predict other properties of the solid and the likelihood that it will be superconducting.[2,3]

The method used is based on quantum theory and relies on the use of high-speed, large-memory computers. Recently, the theory has been used to predict that at high pressures silicon will become a hexagonal superconducting metal. In fact, two hexagonal forms were studied, simple hexagonal and hexagonal close-packed, and both were predicted to be superconducting. Experimental verification came from Grenoble, France,[2] and the University of California at Berkeley.[3]

The theoretical approach is based on a conceptually simple "pseudopotential" model[1] of a solid where the atomic valence electrons are separated from the remaining "core" electrons and nuclei of the atoms. The positive cores or ions, each composed of a nucleus and core electrons, are put in a periodic array characteristic of the solid structure, and each core is assumed to be unchanged when the solid is formed from the atoms. In contrast, the valence electrons form a sea of free electrons which can interact with the lattice of cores and form the bonds characterizing the solid. The valence electron-core interaction is modeled by a pseudopotential which can be computed from the atomic properties using only the atomic numbers.

To predict which structure is most stable at a given pressure or volume, the total energy of the model solid is computed for each choice of structure for varying volumes. The minimum energy structure at a given volume is the most likely choice for the system and its characteristics such as lattice constants, compressibilities, cohesive energies, vibrational spectra, and a host of other properties can be predicted.

Silicon has served as the prototype system to test the method. At atmospheric pressure, the properties of the semiconducting diamond structure have been examined in detail and are reproduced to a high degree of accuracy by the theory. At higher pressures, Si becomes metallic first in a white tin structure around 100 kilobars and then in the simple hexagonal and hexagonal close-packed structures at approximately 150 and 400 kilobars respectively. Experimentally, these pressures can be achieved using diamond anvil cells[4] composed of opposing flat surfaces of diamonds surrounded by a steel gasket. When the sample is placed in a medium such as a soft powder between the diamonds (which are pressed together mechanically), the pressures near the sample are fairly hydrostatic, and megabar pressures can be obtained. Small chips of ruby are placed in the cell, and then their pressure-dependent fluorescence is used to monitor the cell pressure, X-ray measurements through the diamond give the crystal structure of the sample, and electronic leads are introduced into the cell to determine the superconducting transitions.

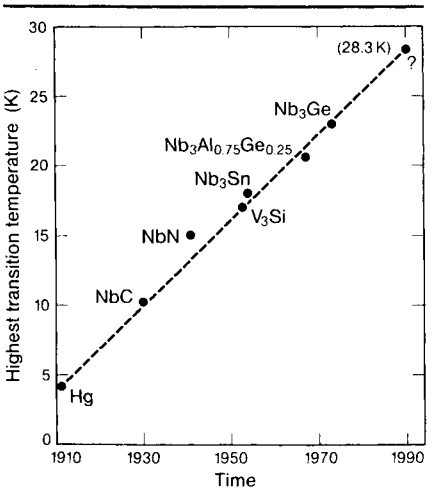

Figure 1. The highest transition temperature superconductors vs. time.

The pressure dependence of T_c is predicted to be the highest near the structural transitions from simple hexagonal to white tin or hexagonal close-packed, and these effects are observed in the measurements. There appears to be a close connection between the structural phase change and T_c, and there is hope that a study of this connection may lead to higher T_c superconductors.

Although the underlying physics governing superconductivity is explained by the Nobel Prize theory of Bardeen, Cooper, and Schrieffer,[5] predicting the existence of superconductivity or evaluating T_c using this theory is difficult. An accurate and detailed knowledge of the non-superconducting solid is necessary to compute T_c. Although it was possible to use the theory to predict that certain classes of materials, like doped degenerate semiconductors, would be superconducting,[6] it did not help much in the search for new superconductors. It is even difficult to give an accurate prediction[7] of the maximum T_c, and researchers often relied on extrapolations of the highest T_c achieved as a function of time (as shown in Figure 1) to inspire explorations. However, it now appears possible that the theory can help. A possible approach is to change the theoretical parameters to maximize T_c subject to the restriction that the crystal structure remains stable. It is likely that metastable phases will play an important role, and these can also be explored using the present theory.

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