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CALCULATING AND PREDICTING THE PROPERTIES OF MATERIALS

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INTRODUCTION

Using microscopic theory, it is now possible to predict the structures and properties of real materials from first principles. This breakthrough was brought about by recent advances in the theory of the electronic structure of solids and by the availability of supercomputers. Many researchers in the field believe we are at the beginning of an era where we will be capable of designing materials with desirable properties using quantum theory and computers.

PRESENT RESEARCH AND OBJECTIVES

The determination of the structural, electrical, and mechanical properties of solids from the constituent atoms has been a longstanding problem which has challenged solid state physicists ever since the invention of quantum mechanics in the 1920's. There were both conceptual and computational difficulties. For example, to obtain the vibrational properties (e.g., phonon frequencies) of a crystal, a precision of one part in ten million is needed for the energy of each atom in systems which typically contain $10^{23}$ strongly interacting particles. For the ground-state properties, formalisms have been developed to reduce the many-electron problem into an effective single particle self-consistent-field problem. The electron-ion part of the interaction energy is often simplified by the use of pseudopotentials which removes the inert core electrons from the problem and thus allows a more precise determination of small energy differences resulting from changes in valence electron configurations. Further, by focusing only on the valence electrons, it is no more difficult to treat the very heavy elements than the light ones. Another new important development is a theory which deals successfully with excited-state (quasiparticle) properties.

In the past several years, calculations have been carried out to explain and predict with high accuracy the properties of a whole host of materials with only input being the atomic number and atomic mass of the constituent elements. Examples of these include calculation of the static structural properties, vibrational properties, electron-phonon and phonon-phonon interactions, optical spectra, solid-solid structural phase transitions, superconducting transition temperatures, surface electronic and geometric structures, and so forth. The only major limitation on these studies is that present computer resources restrict the calculations to relatively simple systems with simple chemical composition and in highly ordered phases (see next section).

The successes thus far have been extremely encouraging. With improved computing facilities, we believe many exciting objectives are realizable.
One could predict structures. One could analyze new proposed materials theoretically and search for those with special properties. Studies of this kind would be particularly useful in connection with modern materials fabrication experiments, e.g., those using molecular beam epitaxy techniques. Many materials have desirable properties only at high pressures. Theory could help in understanding the high pressure phases and in finding ways to bring back the material to zero pressure while retaining the desirable properties.

Another example is the search for a high transition temperature superconductor. Theoretical calculations provide information on the electron-phonon interactions. The object is then to find materials with strong electron-phonon interactions which would give a high transition temperature. Other areas of interest in which the ab initio studies would have a major impact include clusters, surfaces and interfaces, and impurities and defects in solids. These are all areas of fundamental physics interest and of technological importance.

MATHEMATICAL MODELS AND COMPUTER REQUIREMENTS

(1) Mathematical Models--The properties of solids are determined by the valence electrons which hold the atoms together. Thus, the most basic quantities required in our studies are the wavefunctions and energies of the valence electrons. They are obtained by solving a second order differential equation (a Schrödinger-like equation) with the potential seen by the electron self-consistently dependent on the charge density of all the electrons. Iterative self-consistent field (SCF) solution techniques are used. An important mathematical simplification results from the lattice periodicity of a crystal. The states are described by the crystal momentum quantum number k so that at each iterative step, the problem subdivides into $N_k$ differential equations which need only be solved within one unit cell as opposed to the whole crystal. For convergent results, $N_k$ ranges from about 10 to a few hundred for typical systems.

The basic procedure is as follows: (a) set up and solve the SCF equations as a matrix eigenvalue problem $N_k$ times; (b) accumulate solutions and modify original equations; and (c) iterate. There are, of course, many further manipulations based on the solution of the SCF problem to obtain the physical quantities of interest.

(2) Algorithms--Present program codes make use of efficient fast Fourier transform routines and matrix operations such as diagonalization and inversion of non-sparse matrices. Vectorization capability is essential in the construction and manipulation of large matrices. However, the matrix element evaluation part is generally less vectorizable. There is at present a strong need for very fast and reliable I/O routines capable of handling very large files for repeated use in the iteration process.

(3) Parallelism--Our problems can be readily partitioned in step (a) described in (1) since all $N_k$ solutions may be obtained in parallel. Thus, parallelism will be useful. However, the accumulation and update step will eventually be the bottleneck.
(4) **Speed and Memory**—In this area, both very high computational rates and large central memory are needed. The calculational effort in general scales at least as $N^2$ in computation memory and $N^3$ in computation time where $N$ is the number of atoms in the unit cell. For the next generation problems, matrix sizes of the order of several thousands are envisioned.

(5) **Productivity**—It is desirable to have interactive editing, debugging, compilation, etc., on the supercomputer. However, they should not be implemented at the expense of "number crunching" performance. Having compiler emulators on frontend/local machines such as a VAX 11/780 or workstations would be very beneficial.

(6) **Access**—Presently, we have access to the supercomputers at the Lawrence Livermore National Laboratory through MFENET and to the supercomputer at Purdue University through ARPANET. We find the LBL-LLNL MFENET quite satisfactory since it offers high transmission speed and is capable of transferring large files in reasonable time (i.e., 10-30 minutes).

(7) **Ranking of Items**—To our research activity, we find essential: high compiler technology, high I/O performance, large storing capacity on rotating disks or solid state disks, access to scientific libraries, debugging tools and workstations for pre- and postprocessing. We find useful: performance evaluation tools and comfortable graphics library. We find not useful at present: languages other than FORTRAN, high-speed image generation, or real-time image rotation, zoom, and translation.

**FUTURE**

In my opinion, all signs indicate a very bright future for this field. In this country, over 20 groups in universities, industrial laboratories, and national laboratories are actively applying similar theoretical techniques in calculating materials properties from first principles. Researchers in Europe and Japan are pushing toward the same goals. With the proper computing facilities, I believe many exciting objectives and possibilities are realizable since one is now in a position to predict properties of systems which formerly were not accessible to theory or experiment.

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