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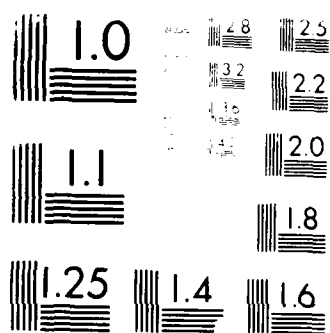
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Arthur J. Freeman

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A new technique for determining the electronic structure of bulk solids has been developed by solving Poisson's equation for a general charge density and potential. All contributions to the potential are completely taken into account in the Hamiltonian matrix elements. The high accuracy of this approach allows us to go beyond traditional treatments and to consider the total energy which allows the accurate determination of structural information (lattice parameters, force constants, etc.) and total energy determinations of the lowest energy phase. The problems treated during this period include: (i) excited state properties of rare-earths and their compounds, (ii) room temperature superconductivity in metallic hydrogen under high pressure and the nature of the molecular to metal phase transition in hydrogen under high pressure, (iii) total energy theory of mixed valence, (iv) electronic structure and superconductivity in the high T_c AlB compounds, and (v) superconductivity and magnetism of C15 intermetallic compounds.

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22a. NAME OF RESPONSIBLE INDIVIDUAL

Karl L. S. J. J. J. J.

22b. TELEPHONE NUMBER
(Include Area Code)

(202) 767-2011

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NP

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The aims and purposes of this research were to (i) develop sophisticated theoretical and computational methods which will (ii) permit the accurate theoretical determination of the electronic structure of complex systems and of the collective phenomena which are the origin of their electronically driven phase transitions. It is widely recognized that the many important and increasingly complex properties of such materials require the most developed form of the quantum theory of matter. Unique information derived from such studies is increasingly attracting the interest of materials scientists seeking solutions and/or understanding of phenomena (including some of the more classical metallurgical domain) thought previously to be too complex for first principles theoretical treatment. The work carried out under AFOSR support, has become an active and prolific research program.

II. METHODS AND APPROACH TO THE PROBLEM

The newly discovered exotic phenomena in complex materials, such as superconductivity in organic salts, have challenged the traditional understanding acquired from approximate theoretical treatments within the quantum theory of matter. This challenge has been met by recent developments of the energy band method which have made it an increasingly powerful, sophisticated, and successful tool of the solid state physicist. The success of the energy band approach is based on the work of Hohenberg, Kohn, and Sham in developing the local density functional formalism for investigating various ground state properties of solids. Aside from providing a rigorous basis and justification of the single particle energy band description, it has led to accurate tractable computational schemes for describing them from first principles.

Because of the importance of charge transfer between different atom

species, meaningful results are possible only if carried out self-consistently. For magnetic systems, local spin density functional theory carried out as self-consistent spin-polarized energy band calculations (different potentials for different spins) are obviously necessary. Both self-consistency (which requires iterative procedures to obtain convergence) and spin polarization make these calculations enormously more complicated and time consuming and so have been carried out, using traditional methods, only on simple systems.

Our work has been at the forefront of these developments in the local density theory energy band approach. Several novel schemes, such as the self-consistent linear combination of atomic orbitals (LCAO) method and the self-consistent linearized augmented plane wave (LAPW) and the self-consistent spin-polarized linearized muffin-tin orbital (LMTO) methods are now among the basic methods we have developed and are using in our fundamental theoretical studies of both bulk solids and surfaces.

In a major advance, we have completed development of the full-potential linearized augmented plane wave (FLAPW) method for carrying out accurate calculations using the local (spin) density approach without any shape approximation to the potential. This capability, which we exploited in the work completed, permits highly accurate determinations of the total energy of bulk solids and therefore of structural properties.

The examples described later under progress indicate some of the richness and variety of properties and phenomena which are being and can be studied today. Comparisons with experiment provide such a demanding test of the predictions of one-electron theory that our work has the additional virtue in permitting, by the comparison with experiment, accurate determinations of the relative magnitude and importance of many-body effects in real solids.



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III. PROGRESS

A. Excited State Properties of Rare-earths and Their Compounds .

As is apparent from the list of publications attached for the final year 1984-85 of this AFOSR grant, there was considerable progress reported indicating a very high level of productivity and accomplishment.

Some of the challenges involved in several of the problems in treating mixed valence in rare earth compounds were addressed. Studies initiated earlier on Ce and its compounds (such as CeN and CeSb) were completed. A major aim was to understand the information obtained with photoemission experiments. This was done with molecular cluster calculations for Ce as well as with supercell energy band calculations for Ce, CeN, and CeSb. We also extended our cluster and supercell calculations to yield ratios of the intensities of the photoemission peaks which can be a sensitive test when compared with experiment.

The origin of the two peak photoemission and inversion photoemission spectra in Ce and Ce compounds was determined by means of total energy supercell calculations. This work was further developed and several publications appeared which applied the theory to the case of the cerium pnictides and later extended to include a detailed analysis and interpretation of the experiments in PrSb and NdSb. These results demonstrated the validity of the local density functional total energy supercell description of excited state properties of solids.

B. Room Temperature Superconductivity in Metallic Hydrogen Under High Pressure and the Nature of the Molecular to Metal Phase Transition in Hydrogen Under High Pressure .

The structural properties, superconductivity, and magnetism of metallic hydrogen was studied using highly precise total energy local density methods. This study further gave support for the possibility of high temperature superconductivity in metallic hydrogen under pressure. Local spin

density calculations were also performed for metallic hydrogen at low density in the antiferromagnetic phase. This was part of a detailed study of the structure and properties of hydrogen at ambient and high pressures.

As a next step in our investigations we evaluated the electronic structure and total energy of molecular hydrogen using our FLAPW approach. This was done for the P_3 structure (which consists of 4 molecules of hydrogen per unit cell). In this way, we were able to treat, for the first time, both the molecular and the metallic phases in the same way so that total energy results are really comparable. The results yielded a detailed understanding of the nature of the phase transition which occurs under pressure between molecular and metallic hydrogen. It should be emphasized that these very challenging calculations for an extremely anisotropic and open structure with 8 atoms per unit cell truly demanded the high numerical precision of our total energy FLAPW approach. These unique calculations showed that the FLAPW method which we developed had the precision and stability to study the properties of systems as complex as hydrogen in both its molecular and metallic phases.

C. Total energy theory of mixed valence .

We completed an investigation of TmSe which is one of the most interesting of the intermediate valence compounds. This is in part due to the fact that it exhibits antiferromagnetic ordering at low temperatures. TmSe is also a metamagnet: applying a field of 5 kOe changes the antiferromagnetic structure into a ferromagnetic structure. In the similar compound TmTe there is no mixed valence behavior and as a result, the alloy $\text{TmSe}_{1-x}\text{Te}_x$ reveals some interesting and unexpected phase transitions. In order to explain these experimental results we performed electronic structure calculations for the compounds TmSe and TmTe. This provided insight as to why the comparable compounds TmSe and TmTe show such a different behavior; further, it formed the

basis for understanding the alloy system $\text{TmSe}_{1-x}\text{Te}_x$. As a result of these investigations, we demonstrated that the local density total energy approach could give important information about the electronic structure and properties of a well known and well studied mixed valence system.

D. Electronic Structure and Superconductivity in the High T_c Al5 Compounds .

The second stage of the calculations using our FLAPW approach were carried out on the high T_c compounds Nb_3Ge and Nb_3Sn as well as the very low T_c in Nb_3Sb . All electrons of the 8 atoms per unit cell were included in a fully self-consistent fashion; we also included all relativistic effects including spin orbit coupling. These calculations provided a severe test of the validity of local density theory for these complex systems and revealed some of the interesting properties of these high T_c superconductors.

E. Superconductivity and Magnetism of C15 Intermetallic Compounds .

Our self-consistent full potential LAPW (FLAPW) electronic structure calculations were completed for the compounds ZrZn_2 (a ferromagnet) and ZrV_2 (a superconductor). This allowed us to determine the effect of non-spherical terms in the potential and also to assess the importance of spin orbit coupling on the properties.

A.J. Freeman - Publications Supported by The Air Force Office of Scientific Research (Grant No. 81-0024) 1984-1986

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"Magnetism and Superconductivity in C15 Compounds: $ZrZn_2$ and ZrV_2 ", (with M.C. Huang and H.J.F. Jansen) to be submitted to J. Mag. Magn. Matls.

"LAPW Approach to the Electronic Structure of NbO_2 and RuO_2 ", (with S.M. Chen and D.D. Koelling) to be submitted to Phys. Rev.

"Electronic Structure and Superconductivity of Nb_3Ge and Nb_3Sb ", (with S.M. Chen, H.J.F. Jansen, and D.D. Koelling) to be submitted to Phys. Rev.

"Electronic Structure and Properties of Rutile Dioxides", (with J.H. Xu and T. Jarlborg) in final preparation.

"Self-consistent FLAPW local density electronic structure of magnetism and superconductivity in C15 compounds: $ZrZn_2$ and ZrV_2 ", (with M.-C. Huang and H.J.F. Jansen) in final preparation.

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