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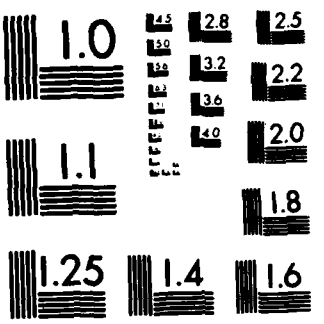

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Our recent development of self-consistent local (spin) density energy band approaches have provided a powerful theoretical/computational tool for determining the electronic structure and properties of complex materials. The importance of charge transfer between constituent atom species has been demonstrated as has its inclusion by means of accurate self-consistent solutions. Some examples of areas in which progress has been achieved include: the high field superconductivity in Chevrel phase compounds; the unusual →

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magnetic and superconducting properties of some C15 compounds including a better understanding of their relationship between the electronic, lattice and superconducting properties of these materials; self-consistent energy band calculations including all electrons and all atoms in the 16 atoms per unit cell of the linear chain transition metal trichalcogenide, $TaSe_3$; and a detailed assessment of theoretical determinations of the electron phonon coupling parameter in metals and intermetallic compounds.

PROGRESS REPORT

Our recent development of self-consistent local (spin) density energy band approaches to determining the electronic structure and properties of complex materials have provided a powerful theoretical/computational tool. The importance of charge transfer between constituent atom species has been demonstrated as has its inclusion by means of accurate self-consistent solutions. Some examples of the progress achieved in the last year include the following:

A. High Field Superconductivity and Magnetism of Chevrel Phase Compounds

One of the unusual characteristics of the Chevrel phase compounds is that the compounds Mo_6S_8 and Mo_6Se_8 are stable and in the case of Mo_6Se_8 also superconducting with a relatively high transition temperature of 6 K. This transition temperature is comparable to that of $PbMo_6Se_8$ and $LaMo_6S_8$ (7K) despite the fact that these compounds have an additional metal atom (Pb, Sn or La).

As proposed, we have studied the electronic structure and properties of Mo_6S_8 and Mo_6Se_8 and, for comparison, $LaMo_6S_8$ also. As before for the other Chevrels, we have calculated self-consistently the electronic band structure, density of states, electron-phonon coupling parameter and transition temperature for these materials. The results provide us with a first principles understanding of the behavior of these materials and the origin of their superconducting properties. In addition to several publications in international journals, the work has been reported in a book chapter in "Superconductivity in Ternary Compounds". Our work on the Chevrel compounds was presented as an invited paper at the International

Conference on "Superconductivity in d and f Band Metals" in Karlsruhe, Germany, in June, 1982.

B. Superconductivity and Magnetism in C15 Compounds

Our successful studies of the C15 compounds $ZrZn_2$ and $TiBe_2$, discussed in last year's report, led us to undertake an in-depth study of the electronic structure and properties of $CeAl_2$ and $LaAl_2$. These studies were undertaken in order to understand the role played by the 4f electrons - a role obviously essential for explaining the valence fluctuation properties of $CeAl_2$. Both paramagnetic and spin-polarized antiferromagnetic state calculations were carried out in order to elucidate the magnetic properties of the system. In the case of $CeAl_2$, considerable f electron charge was found on the Ce sites and so justified the use of an extended basis set in the self-consistent procedure to account correctly for the f charge density during the iterative process.

The Ce and La atoms are found to be the dominant factor in determining the electronic structure near the Fermi energy and this is enhanced by the presence of f-bands close to ($LaAl_2$) or at ($CeAl_2$) the Fermi energy. In paramagnetic $CeAl_2$, the f-bands are about 1 eV wide and, although principally above the Fermi energy, extend down so that the additional electron compared to $LaAl_2$ is accommodated in the f-density. The ferromagnetic state is found not to be stable but the antiferromagnetic state is found to be stable with the experimentally observed moment. A significant narrowing of the f-bandwidth is observed in the antiferromagnetic state.

C. Soft Phonon Modes and Superconductivity in C15 Compounds

The relationship between electronic, lattice and superconducting properties of materials has attracted considerable theoretical and experimental attention. Recent interest has centered on the C15 super-

conducting compounds HfV_2 and TaV_2 as ideal candidates for these studies. Experimentally, it is known that HfV_2 is a high T_c superconductor ($T_c \sim 9K$); it has a high critical field, a lattice phase transition at 113 K, a large electronic specific heat coefficient, and a large temperature (T) dependent susceptibility. In contrast, TaV_2 has a low T_c (3.6 K), an average electronic specific heat, an almost T independent susceptibility, and no lattice anomalies. In addition, much is known about certain average properties from a series of heat capacity measurements of $Hf_{1-x}TaV_2$ compounds. These revealed a remarkably strong correlation between the electronic and lattice properties, and showed that the Fermi surface electrons were largely responsible for the soft mode behavior of the phase transition and the high T_c of HfV_2.

On the theoretical side, our recent detailed self-consistent energy band studies of some C15's have elucidated their electronic structure and have yielded some surprising results. For ZrV_2, the calculations predict a very large electron-phonon coupling $\lambda(=2.4)$ and a very high T_c of 32 K — assuming that the C15 structure is retained to low T with unchanged lattice constant and geometric phonon frequencies. These provocative results have raised some questions concerning the role of lattice properties, which are not known but crudely approximated.

Our work has successfully related, for the first time, the microscopic lattice parameters (obtained experimentally) and electronic parameters (obtained theoretically) to the superconductivity of HfV_2 and TaV_2. First, results of EXAFS measurements taken by Knapp, Georgopoulos and Pan of the mean square relative displacements (MSRD) in HfV_2 and TaV_2 were utilized to calculate some important lattice properties, notably the effective near-neighbor force constants and the effective Debye temperatures of all

pairs of near neighbors. When combined with the results of our self-consistent energy band calculations for HfV_2 and TaV_2, these constants allowed us to calculate the λ and T_c values of these materials. For TaV_2, we find $\lambda = 0.4$, consistent with the low T_c, whereas $\lambda = 2.6$ for cubic HfV_2. Using strong coupling theory, such a large λ would give a T_c of 28 K. This large a value of T_c calculated for HfV_2 is presumably not found experimentally because of the phase transition below which the DOS (and presumably the η values) decrease markedly.

In conclusion, we have shown that the EXAFS technique yields important new information concerning the phonon parameters in a relatively complex system. For the first time all the relevant force constants necessary to calculate T_c have been determined and are found to be much smaller in HfV_2 than in TaV_2. The increase in the DOS and η values for HfV_2 relative to TaV_2 are responsible for the unusually large decrease in the force constants that drives the lattice phase transition at 113 K.

D. Electronic Structure of $TaSe_3$, A Linear Trichalcogenide Compound

Last year we proposed to complete our investigation of the electronic structure and properties of $TaSe_3$, a "linear" chain transition metal trichalcogenide compound. The self-consistent electronic energy band structure, density of states, Fermi surface and charge densities have now been accurately determined. These results are now to be compared with experiments being undertaken both at Northwestern University and elsewhere. The calculations represent a forefront state of the art approach to a very complex system of low symmetry (monoclinic structure) which has a total of 16 atoms per unit cell. From the electronic structure results we also hope to understand the origin of the superconducting properties of this system.

E. Assessment of Theoretical Determinations of the Electron-Phonon Coupling Parameter, λ, in Metals and Intermetallic Compounds

The electron-phonon coupling parameter, λ_{el-p}, while playing a centrally important role in superconductivity and other phenomena, is still a difficult quantity to determine from first principles theory. The availability of accurate ab initio self-consistent energy band calculations of partial and total density of states (DOS) allow λ to be determined from the electronic specific heat or from simple theoretical treatments such as the rigid ion approximation. We have assessed the accuracy of these determinations (and the band structure results) in a number of transition metals and A15 and C15 intermetallic compounds. We have included comparisons with results obtained using the McMillan equation parameterization of T_c along with experimental results from tunneling measurements, NMR, and comparisons of high and low T specific heat data. As a result of these comparisons, we have been able to show that for many of the high DOS materials serious descrepancies exist between the theoretical and experimental determinations. As a result, the role of spin fluctuations and lattice transformations in connection with these findings has been clarified.

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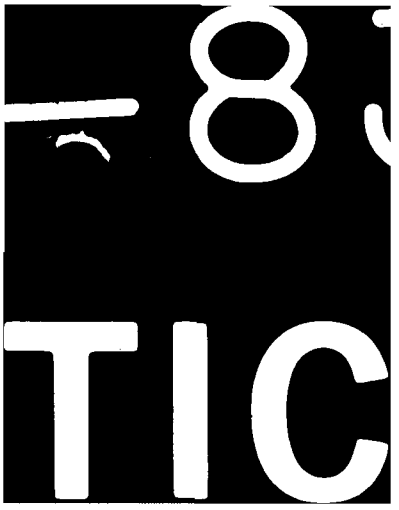
1. "Soft Phonon Modes and Superconductivity in C15 Compounds" (G.S. Knapp, H.-K. Pan, A.J. Freeman and T. Jarlborg).

2. "Magnetism versus Superconductivity in $HfZn_2$" (Jian-hua Zu, T. Jarlborg, and A.J. Freeman).

3. "Self-consistent Relativistic Electronic Band Structure and Properties of RuO_2 and NbO_2", Shi-min Chen, A.J. Freeman, and D. D. Koelling).

4. "Self consistent Relativistic Electronic Band Structures and Properties of A15 Compounds", Shi-min Chen, A.J. Freeman, and D. D. Koelling).

5. "Self-consistent LMTO Determination of Electronic Structure of Rutile-dioxides" (Jian-hua Xu, T. Jarlborg and A.J. Freeman).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