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THEORETICAL STUDIES ON THE ELECTRONIC STRUCTURE AND PROPERTIES OF COMPLEX CERAMIC CRYSTALS AND GLASSES

Annual Progress Report
for Period July 1, 1990 - June 30, 1991

Wai-Yim Ching

University of Missouri-Kansas City
Kansas City, Missouri 64110

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Summary

This progress report summarizes the research activities for the period 7/1/90 - 6/30/91, the first year of a three-year renewal. The major accomplishments for the current year are: (a) Study of electronic structures and optical properties of several important ceramic crystals such as Y_2O_3, Al_2O_3, MgO, $MgAl_2O_4$, ZrO_2, V_2O_3, V_2O_5 and all polycrystalline forms of SiO_2. (b) Theoretical study on superconducting oxides. (c) First-principles calculation of total energies and structural phase transitions in oxides and nitrides. (d) Basic study on metallic glasses including calculation of transport properties. (e) Theory of rare-earth and transition metal compounds. The major thrust for the next year's effort will be concentrating on the following: (1) Continuation of electronic and optical studies of important ceramic systems. Particular emphasis will be on the total energy calculations on complex crystals such as various polymorphic forms of SiO_2, ZrO_2, and Si_3N_4. (2) Fundamental studies on the structure and properties of different phases of Boron and B-related compounds. (3) Further development of the OLCAO method such as application of self-interaction correction to wide gap insulators, spin-polarized calculations on the magnetic glasses and extraction of effective interatomic pair potentials for simulational studies.

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I. Progress Report (7-1-90 to 6-30-91)

This section succinctly summarizes our major accomplishments for the one year period from July 1, 1990 to June 30, 1991 which include some of the work currently still in progress. They are loosely divided into five categories not necessarily in the order of importance. More detailed description can be found from the published papers which are listed in the following section.

(a) Electronic and optical properties of ceramic crystals.

Detailed studies on the electronic structures and optical properties on the following important ceramic crystals have been carried out: Y_2O_3, α-Al_2O_3, MgO, $MgAl_2O_4$ (spinel), $SrTiO_3$, $BaTiO_3$, V_2O_3, V_2O_5. ZrO_2 (3-phases), BN (3-phases), polycrystalline forms of SiO_2. These are all selfconsistent calculations using the state-of-the-art OLCAO (orthogonalized linear combination of atomic orbitals) method. Accurate band structures, density of states, charge distributions and effective ionic charges are obtained using the first-principles approach. For Y_2O_3 and $MgAl_2O_4$ crystals, these represent the first-time studies. The calculated optical properties are generally found to be in good agreement with recent measurements, indicating the accuracy of the calculated electronic structures on these crystals.

(b) Theory of high Tc superconductors.

We have continued to study the electronic properties states of high temperature superconductors that was initiated few years ago shortly after their discoveries. The present study includes the the analysis of orbital-resolved partial density of states in $YBa_2Cu_3O_7$ and using them to

interpret several key experiments. The effects of metal substitution such as Ga and Zn on Cu and/or Y sites on the electronic structures in $YBa_2Cu_3O_7$ have also been studied. In collaboration with an experimental group in the Chemistry Department, we have successfully used the crystal potentials and charge distributions obtained from our selfconsistent band calculations to explain the positron annihilation data on several different high Tc superconductors.

(c) Total energy and structural phase transitions.

After the successful total energy calculation in ten different phases of Silicon a year ago, we have extended our bulk structural studies to more complicate crystals. We have initiated studies on α-quartz, cosite, β-Si_3N_4, cubic and hexagonal phases of BN and three different phases of ZrO_2. The results obtained so far are very encouraging. In case of BN, excellent values for the equilibrium volume, bulk modulus and cohesive energy have bee obtained. However, for the structurally more complicated crystals, the required accuracy for the total energy calculation is much more stringent than in the simple crystals such as Si or BN. We are currently working towards significant improvement in this area.

(d) Basic study on metallic glasses.

Our group has a long history of making accurate calculations on noncrystalline materials in both metallic glasses and insulating glasses based on realistic structural models. Recent progress includes calculation on the electronic, optical and transport properties of several different types of metallic glasses. We intend to continue this

tradition and will study other important metallic glass systems such as $Fe_{1-x}B_x$, a soft magnet important to electric power industry. Spin-polarized calculation on pure amorphous Fe glass shows, for the first time, the distribution of Fe moments in an amorphous environment and its relation to the short range order. The distribution of the Fe moment and its dependence on the B concentration in amorphous $Fe_{1-x}B_x$ is a subject of great interest.

(e) Spin-polarized calculation on Fe-B compounds.

We have extended the OLCAO code to include the spin-polarization and applied it to study the electronic and magnetic properties of FeB, Fe_2B and Fe_3B intermetallic compounds. The results obtained are in good agreement with experiment. This gives us great confidence in future studies on the magnetic properties of some selective transition metal alloys, rare-earth compounds and magnetic glasses which are of crucial importance to the energy science.

II. List of scientific papers published, accepted and submitted.

(since last report and for the period 7/1/1990 - 6/30/1991)

(A) Work fully or partially supported by the project

(1) Y.-N. Xu, **W.Y. Ching** and K.W.Wong, "Electronic Structures of Ga- and Zn- Substituted $YBa_2Cu_3O_7$ Superconductors", MRS Symposium M, High Temperature Superconductors: Fundamental Properties and Novel Materials Processing. Edited by J. Narayan, C.W. Chu and L.F.Schneermyer, MRS, Pittsburg, PA (1990), p41-46.

(2) F. Zandiehnadem and **W.Y. Ching**, "Total Energy,Lattice Dynamics and Structural Phase Transition of Si by the Orthogonalized Linear Combination of Atomic Orbitals Method", Phys. Rev. B**41**, 12162 (1990).

(3) **W.Y. Ching** and Y.-N Xu, "Electronic and Optical Properties of Yttria", Phys. Rev. Lett., **65**, 895 (1990).

(4) P.C.W. Fung, Z.C. Lin, Z.M. Liu, X.Ying, Z.Z. Sheng, F.T. Chan, K.W. Wong, Y.-N. Xu and **W.Y. Ching**, "Vanadium Substituted 2212 and 2223 Superconducting Ceramics", Solid State Commun. **75**(3), 211 (1990).

(5) **W.Y. Ching**, Y.-N Xu, B.N. Harmon and J. Ye and C.T. Leung,"Electronic Strutures of FeB, Fe_2B and Fe_3B Compounds by First-Principles Spin-Polarized Calculations", Phys. Rev. B**42**, 4460, (1990).

(6) J. C. Parker, D. J. Lam, Y.-N. Xu and **W.Y. Ching**, "The Optical Properties of Vanadium Pentoxide Determined From Ellipsometry Measurements and Band Structure Calculations." Phys. Rev. B**42**, 5289, (1990).

(7) Y.-N. Xu, **W.Y. Ching** and R.H. French, "Selfconsistent Band Structures and Optical calculations in Cubic Ferroelectric Perovskites." FERROELECTRICS, Vol. **111**, 23-32 (1990).

(8) **W.Y. Ching**, "Theroretical Studies of Electronic Properties of Ceramic Materials", Journal of American Ceramic Society, **71** (11), 3135 (1990).

(9) J.C. Parker, U.W. Gelser, D.J. Lam, Y.-N. Xu and **W.Y. Ching**, "The Optical Properties of the Vanadium Oxides: VO_2 and V_2O_5", J. Amer. Ceram. Soc. **71** (11), 3206 (1990).

(10) A. Bharathi, C.S. Sundar, **W.Y. Ching**, Y.C. Jean, P.H. Hor and C.W. Chu "Positron Distribution in Understanding Annihilation Characteristics Across Tc in Oxide Superconductors", Phys. Rev. B**42**, 10199 (1990).

(11) **W.Y. Ching**, G.-L. Zhao and Yi He, "Theory of Metallic Glasses, I: Electronic Properties", Phys. Rev. B**42**, 10878 (1990).

(12) G.-L. Zhao, Yi He and **W.Y. Ching**, "Theory of Metallic Glasses, II: Transport and Optical Properties", Phys. Rev. B.**42**, 10887 (1990).

(13) X.L. Yang, S.-H. Guo, F.T. Chan, K.W. Wong and **W.Y. Ching**, "Analytic Solution of Two-dimensional Hydrogen Atom. I. Non-relativistic Solution", Phys. Rev. A, (in press) Feb. 1, 1991.

(14) F.T. Chan, S.-H. Guo, X.L. Yang, K.W. Wong and **W.Y. Ching**, "Analytic Solution of Two-dimensional Hydrogen Atom. II. Relativistic Solution with Chern-Simon Action", Phys. Rev. A, (in press) Feb. 1, 1991.

(15) **W.Y. Ching**, G.-L. Zhao, Y.-N. Xu and K.W. Wong, "Orbital-resolved Partial Density of States in $YBa_2Cu_3O_7$", Phys. Rev. B. (accepted).

(16) Y.-N. Xu and **W.Y. Ching**, "Selfconsistent Band Structures, Charge Distributions and Optical Properties of MgO, α-Al_2O_3 and $MgAl_2O_4$", Phys. Rev. B. (accepted).

(17) Y.-N. Xu and **W.Y. Ching**, "Distribution of Iron Moments in a-Fe and a-$Fe_{1-x}B_x$ Glasses by First Principles Spin-Polarized Calculations", to appear in J. Appl. Phys.

(18) Y.-N. Xu and **W.Y. Ching**, "Electronic Structure, Total Energy and Optical Constant Calculations in Cubic and Hexagonal Phases of Boron Nitride", submitted to Physical Rev. B.

(B) Scientific Work not directly related to the grant but representing scholarly work in the general area of condensed matter theory.

(Supercomputer time allocated by Office of Basic Energy Research may have been used in some cases)

(1) **W.Y. Ching** and D.L. Huber, "Zero Temperature Statics and Dynamics of a Random Exchange Model for the Magnetic Properties of $La_{2-x}Sr_xCuO_4$ in the Insulating Phase", Phys. Rev. B**42**, 493 (1990).

(2) D.L. Huber and **W.Y. Ching**, "Studies of the Trapping of the Frenkel Excitons in One Dimensional Systems", Chem. Physics, **146**, 409 (1990).

(3) D.L. Huber and **W.Y. Ching**, "Time-Domain Analysis of the Dynamics of Frenkel Excitation in Disordered Systems", Phys. Rev. B**42**, 7718 (1990).

(4) C.S. Sundar, A.Bharathi, **W.Y. Ching**, Y.C. Jean, P.H. Hor, R.L. Meng, Z.J. Huang and C.W. Chu, "Positron Annihilation Studies on the Tl-Ba-Ca-Cu-O Superconductors", Phys. Rev. B**42**, 2193, (1990).

(5) C.S. Sundar, A.Bharathi, **W.Y. Ching**, Y.C. Jean, P.H. Hor, R.L. Meng, Z.J. Huang and C.W. Chu, "Positron Annihilation Studies on the Bi-Sr-Ca-Cu-O Superconductor", Phys. Rev. B**43**, (accepted).

III. **Efforts of scientific personnel involved.**

(1)	W.Y. Ching, Principal Investigator	
	15% academic year	not paid by the grant
	2 summer months	paid by the grant
(2)	Yong-Nian Xu, Research Associate	paid by the grant
	Xue-Fu Zhong*, Visiting Scientist	partly paid by the grant
(3)	Graduate Research Assistants:	
	Farzien Zandiehnadem+	partly paid by the grant
	F. Gan&	Paid by the Grant
	Dung Li%	Paid by the grant

* Prof. Xue-Fu Zhong of the Institute of Semiconductor, Beijing, China had participated in the DOE project from January 1, 1988 to December 20, 1989. After returning to China for a year, he is currently a visiting scientist in the Department of Physics and will be partly involved in the DOE project.

\+ Mr. Zandiehnadem completed his Ph. D. degree in May, 1990. He was paid 2 months last summer to finish some of the research work on ZrO2. He is now a post-doctoral fellow in the Chemistry Department of the University of Missouri-Kansas City.

& Mr. Gan expects to finish his MS degree in the summer of 1991.

% Mr. Dung Li is a new research assistant replacing Mr. Zandiehnadem who graduated with a Ph.D. last summer.

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