Final Report for Period: 08/1996 - 07/2000 **Submitted on:** 08/15/2000

Principal Investigator: Freericks, James K. **Award ID:** 9627778

Organization: Georgetown University

An Integrated First-Principles and Many-Body Theory Description of Electron-Phonon Superconductors

Project Participants

Senior Personnel

Name: Freericks, James
Worked for more than 160 Hours: Yes
Contribution to Project:

Name: Liu, Amy
Worked for more than 160 Hours: Yes
Contribution to Project:

Post-doc

Name: Porezag, Dirk
Worked for more than 160 Hours: Yes
Contribution to Project:
Dirk Porezag was a post-doctoral fellow at Georgetown from 1997 to 2000. During this period he received support from this NSF award, a fellowship from the Humboldt Foundation, and a grant from the Naval Research Laboratory. He carried out a systematic study of the importance of nonlinear core corrections in pseudopotential calculations. He also studied the adsorption of nitrogen-containing molecules on aluminum surfaces.

Name: Rudin, Sven
Worked for more than 160 Hours: Yes
Contribution to Project:
Sven Rudin was at Georgetown as a post-doctoral fellow from Fall 1997 to Fall 1999. He investigated the electron-phonon interaction in Indium using both first-principles and many-body methods. He also studied structural phase transitions and superconductivity in highly compressed Sulfur.

Graduate Student

Name: Bauer, Rudolf
Worked for more than 160 Hours: Yes
Contribution to Project:
Rudolf Bauer, then a graduate student from the University of Regensburg in Germany, spent six months visiting Georgetown in 1997 and returned for a one-month stay in 1998. His stay in 1997 was funded by the DAAD. His primary efforts were devoted to improving the density-functional linear-response code by testing and comparing different algorithms and implementations.

Undergraduate Student

Name: Troyer, Stephanie
Worked for more than 160 Hours: No
Contribution to Project:
Stephanie Troyer was a teaching assistant for the course 'The Quantum World Around Us.'

Name: Lupas, Maria
Worked for more than 160 Hours: No
Contribution to Project:
Lupas served as an teaching assistant for the course 'The Quantum World Around Us.'

Name: Deese, Heather
Worked for more than 160 Hours: No
Contribution to Project:
Deese was a teaching assistant for the tutorials used in the introductory physics course. Georgetown serves as a test site for the tutorials being developed by Lillian McDermott's Physics Education Research Group at the University of Washington.

Name: Fromm, Allison
Worked for more than 160 Hours: No
Contribution to Project:
Allison Fromm was a teaching assistant for the 'Quantum World Around Us' course.

Name: Jones, Emily
Worked for more than 160 Hours: Yes
Contribution to Project:
During the 1997-97 academic year, Emily Jones was an undergratudate research assistant on the project investigating the phase diagram of Lithium.

Name: Benson, Daniel
Worked for more than 160 Hours: No
Contribution to Project:
Benson was a teaching assistant for the tutorials in the introductory physics course. Georgetown served as a test site for the tutorials being developed by Lillian McDermott's Physics Education Research group at the University of Washington.

Organizational Partners

Other Collaborators or Contacts

Andrew Quong of Lawrence Livermore National Laboratory.
Woonki Chung of Georgetown University.
Mark Jarrell of University of Cincinnati.
Elisabeth Nicol of the University of Guelph.

Activities and Findings

Project Activities and Findings:
Work was completed on an inversion of the experimental tunneling data for Lead, including the effects of vertex corrections. We have also investigated the accuracies of perturbation theories for treating the electron-phonon problem off of half-filling. When vertex corrections are included, there are a number of approximation schemes that a priori seem equally reasonable, and we have compared the accuracy of these various approximations.

We have improved our density-functional linear-response codes, and are porting these codes to parallel platforms. These codes have been used to investigate the electron-phonon interaction in Lithium, Indium, NbC, and some high-pressure phases of S and Se that have been found to be superconducting in recent diamond-anvil-cell experiments. These codes have also been used to investigate the thermal properties of metals within the quasiharmonic approximation.

We have presented our work in about 20 invited talks at conferences and institutions in the US and Europe, including Old Dominion University, Virginia Commonwealth University, George Mason University, the University of California at Berkeley, the University of California at Santa Barabara, the University of Cincinnati, Kansas State University, ETH (Zurich), the Institute of Low Temperature and Structure Research, Wroclaw, and the Institute of Physics in Zagreb Croatia.

Project Training and Development:
We discovered that the effects of vertex corrections on the superconducting tunneling data in Pb may have an observable effect at the 1% level. Our work motivates further experimental study on low-temperature superconductors to see whether effects of vertex corrections can be definitively seen.

Our approximate calculations have focused on four different perturbative methods that all produce the same perturbation theory in the large phonon-frequency limit, which is known to be accurate for the Hubbard model. We find that when the phonon frequency is small to moderate, none of these approximation methods are adequate over a wide range of interaction strength, but we hope to improve the accuracy by examining how to properly dress the phonons.

We have compared the electron-phonon spectral function calculated from first-principles with functions extracted from tunneling data. Indium was used as a test case because (i)clean tunneling data are available and (ii)it is a relatively simple sp metal. We found that although the electron-phonon coupling parameter is quoted to three significant digits in the literature, the uncertainties in the data at low and high voltage can result in large uncertainties in the extracted parameters. Based on our first-principles calculations as well as our 'most-likely' fit to the tunneling data, we estimate lambda = 0.9 +/- 0.1 for indium, as compared to the value of 0.804 in the literature.

The pressure-temperature structural phase diagram for Lithium was calculated within the quasiharmonic approximation. Low-lying phonon modes are found to stabilize the bcc phase with increasing temperature. These low-energy modes also enhance the electron-phonon coupling in bcc Li. However, the electron-phonon interaction in the low-temperature close-packed 9R phase is calculated to be significantly weaker, and is consistent with the observed lack of a superconducting transition in Li.

Pressure-induced structure transitions in Sulfur have been investigated. Contrary to expectations based on the high-pressure behavior of the heavier chalcogens, Se and Te, we find the open simple cubic structure to be favored by S over a wide range of pressures (280-540 GPa). Furthermore, the calculated phonon spectrum and electron-phonon coupling strength in the lower-pressure beta-Po phase are consistent with the large superconducting transition temperature of 17 K that has been measured experimentally at 160 GPa.

Our systematic comparison of results from all-electron and pseudopotential calculations demonstrates the importance of including a core correction to account for the nonlinearity of the exchange-correlation potential between core and valence charge densities when pseudopotentials are used. We proposed a new form for the nonlinear core corrections and introduced a scheme for choosing the core cutoff radius.

Research Training:
Emily Jones spent a year as an undergraduate research assistant working on the project involving the phase diagram of Lithium. In the process, she improved her programming skills and developed proficiency with the unix operating system. Upon graduating, she went to TRW to work as a computer programmer. She currently works at the US Patent and Trademark Office.

During his six-month visit at Georgetown, Rudolf Bauer spent significant time learning how to optimize our codes for different computational platforms.

Sven Rudin came to Georgetown with computational experience in quantum chemistry. He has since learned about density-functional calculations as well as the Eliashberg theory of superconductivity.

Dirk Porezag, an expert in all-electron ab initio calculations for clusters, learned about pseudopotential theory and periodic planewave based methodologies.

Outreach Activities:
Seven seminars were given about the course the Quantum World Around Us, which is a course in quantum mechanics and materials science that we have been developing for nonscience students. These talks were presented at Institute of Physics, Zagreb, Croatia (2), the University of Split, Croatia (2), the Ecole Polytechnique Federale de Lausanne, University of Maryland at College Park, and University of Cincinnati.
A presentation about this course was also given at the 1999 Project Kaleidoscope meeting in a session devoted to 'Preparing Science-Savvy Citizens.'

Journal Publications

J. K. Freericks, E.J. Nicol, A. Y. Liu, and A. A. Quong, "Vertex-Corrected Tunneling Inversion in Electron-Phonon Mediated Superconductors: Pb.", *Physical Review B*, p. 11651, vol. 55, (1997).) Published
A. A. Quong and A. Y. Liu, "First-Principles Calculations of the Thermal Expansion of Metals", *Physical Review B*, p. 7767, vol. 56, (1997).) Published
S. P. Rudin, R. Bauer, A. Y. Liu, J.K. Freericks, "Reevaluating Electron-Phonon Coupling Strengths: Indium as a Test Case for Ab Initio and Manybody Theory Methods", *Physical Review B*, p. 14511, vol. 58, (1998).) Published
J. K. Freericks, V. Zlatic, W. Chung, M. Jarrell, "Vertex-corrected perturbation theory for the electron-phonon problem with nonconstant density of states", *Physical Review B*, p. 11613, vol. 58, (1998).) Published
A. Y. Liu, A. A. Quong, J. K. Freericks, E. J. Nicol, E. C. Jones, "Structural phase stability and electron-phonon coupling in lithium", *Physical Review B*, p. 4028, vol. 59, (1999).) Published
J. K. Freericks, V. Zlatic, M. Jarrell, "Approximate scaling relation for the anharmonic electron-phonon problem", *Physical Review B (Rapid Communication)*, p. R838, vol. 61, (2000).) Published
J. K. Freericks, S. P. Rudin, and A. Y. Liu, "First-principles determination of superconducting properties of metals", *Physica B*, p. 425, vol. 284, (2000).) Published
D. Porezag, M. R. Pederson, A. Y. Liu, "Adsorption and dissociation of hydrazoic acid on Al(111)", *Physical Review B*, p. 13230, vol. 61, (2000).) Published
A. Y. Liu
, "Linear-response studies of the electron-phonon interaction in metals", *Physica Status Solidi B*, p. 419, vol. 217, (2000).) Published
D. Porezag, M. R. Pederson, A. Y. Liu, "Importance of nonlinear core corrections for density-functional based pseudopotential calculations", *Physical Review B*, p. 14132, vol. 60, (1999).) Published
S. P. Rudin and A. Y. Liu, "Predicted simple-cubic phase and superconducting properties for compressed sulfur", *Physical Review Letters*, p. 3049, vol. 83, (1999).) Published
R. Bauer, A. Y. Liu, and D. Strauch, "Phonon linewidths due to electron-phonon coupling in NbC", *Physica B*, p. 452, vol. 263-263, (1999).) Published

Books or Other One-time Publications

J. K. Freericks and A. Y. Liu, "The Quantum World Around Us: Teaching Q", (1997). *Contributed Poster to Conference Pr*, Published
Editor(s): Edward F. Redish and John S. Rigden
Collection: The Changing Role of Physics Undergradu
Bibliography: AIP Conference Proceedings 399

Web/Internet Sites

URL(s):

Description:

Other Specific Products

Contributions

Contributions within Discipline:
We hope our work on Indium and Lead will motivate further experimental investigations of electron-phonon superconductors. In particular, if error bars were reported in measurments of the tunneling conductance and superconducting gap, we would be able to use a maximum entropy technique to determine the best fit spectral function. This would allow more accurate determinations of coupling parameters, and in addition, in materials where vertex corrections may be important, the improved accuracy of the extracted spectral function would also allow the effects of vertex corrections to be oberved in the multiphonon region.

Our study of the thermal properties of metals in the quasiharmonic approximation shows that it is reasonable approach for calculating thermodynamic properties of materials from first-principles.

Contrary to earlier suggestions that the large superconducting Tc observed in the beta-Po phase of compressed S could indicate a novel mechanism for pairing, we find that this material can be understood within the standard theory of electron-phonon mediated superconductivity. In addition, our prediction of a simple cubic structure for compressed S has motivated new experiments to search for this unusual open phase. The predicted transition pressure of about 280 GPa is just within reach of current diamond-anvil cell techniques.

While non-linear core corrections for pseudopotential calculations were introduced 15 years ago, the importance of including them for elements throughout the periodic table has not been fully appreciated. Our systematic study emphasizes that these corrections should be used for many different types of elements to improve the accuracy of pseudopotential calculations, and our proposed scheme for choosing the parameters that characterize the correction is straightforward to apply.

Contributions to Other Disciplines:

Contributions to Human Resource Development:

We have continued our development of the course 'The Quantum World Around Us'. This course introduces quantum mechanics, solid-state physics, and materials science to nonscientists. Much of this course is based on the methods and materials developed by Richard Feynman, Daniel Styer, and Edwin Taylor. This course has been successfully introduced at Georgetown and we are disseminating information about it to other universities in the hope that it will be adopted elsewhere.

Contributions to Science and Technology Infrastructure:

Beyond Science and Engineering:

<u>Categories for which nothing is reported:</u>

Organizational Partners
Any Product
Contributions: To Any Other Disciplines
Contributions: To Any Science or Technology Infrastructure
Contributions: Beyond Science or Engineering