Final Report for Period: 08/1999 - 07/2003 **Submitted on:** 10/02/2003
Principal Investigator: Freericks, James K.
Award ID: 9973225
Organization: Georgetown University
Title:
Combining ab initio Methods and many-Body Theory to Describe the Electron-Phonon Interaction in Real Materials

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:

Name: Zlatic, Veljko
Worked for more than 160 Hours: Yes
Contribution to Project:
Zlatic is collaborating on two main fields of study---the first is an examination of the Falicov-Kimball model and how the properties of the exact solution relate to the experimental behavior in YbInCu4 and SmB6. The second is a broad study of transport properties ranging from examining models that can be solved exactly to analyzing how one applies density functional theory to describe transport in metals with strong electron-phonon interaction. He visits the US for one month each year, and people from the US team visit Croatia for one month each year, receiving support from Zlatic's grant from the Croatian Ministry of Science.

Post-doc

Name: Quandt, Alexander
Worked for more than 160 Hours: Yes
Contribution to Project:
Alex Quandt has been a post-doctoral fellow at Georgetown since January 2000. He has been working a the A15 project.

Name: Demchenko, Denis
Worked for more than 160 Hours: Yes
Contribution to Project:
Denis worked full time on research on density functional theory calculations for magnetic systems and on many body physics for transport of charge and heat in correlated materials.

Graduate Student

Undergraduate Student

Name: King, Peter
Worked for more than 160 Hours: No
Contribution to Project:
Peter King was an undergraduate teaching assistant for the introductory physics course.

Name: Grimes, Ross
Worked for more than 160 Hours: No
Contribution to Project:
Ross helped with teaching tutorials in relativity and quantum mechanics. Georgetown University is a test site for developing new tutorials in collaboration with the physics education group at the University of Washington. Ross served as a peer instructor and

interacted directly with the students in the class.

Name: Peterson, Courtney

Worked for more than 160 Hours: Yes

Contribution to Project:

For her senior thesis, Courtney Peterson constructed a tight-binding model for the electronic bands in MgB2. She also analyzed a simple BCS-like two-gap model for describing superconductivity in MgB2 to see where it succeeds and where it fails.

Technician, Programmer

Other Participant

Research Experience for Undergraduates

Organizational Partners

Institute of Physics, Zagreb Croatia

We engage in regular exchanges with the Institute. Zlatic will typically visit the US for one month a year and people from the US team will visit the Institute for one month per year. Zlatic supports his travel to the US and the local costs of the US team in Croatia through his grant from the Croatian Ministry of Science. The Institute provides office space, computers, etc. for the US team when in Croatia.
One visit in 2001 involved introducing a junior researcher, Alex Quandt, to the lab in Croatia during a visit with Freericks. A joint project with Zlatic was initiated, but did not produce publishable results.

Other Collaborators or Contacts

Sven Rudin of Los Alamos National Laboratory.
Thomas Devereaux of the University of Waterloo.
Mark Jarrell of the University of Cincinnati.
Igor Mazin of NRL.
Romuald Lemanski of the Institute for Low Temperature and Structure Research.
Ralf Bulla of Augsburg.
Andrij Shvaika of Lviv, Ukraine.
Barri Letfulov of Ekaterinberg (deceased, 2002).

Activities and Findings

Research and Education Activities:

We have implemented a tunneling inversion code that takes into account non-constant density of states needed to properly evaluate the tunneling data for the A15 superconductors. The electronic density of states from DFT calculations are used as input to analyze the high-quality tunneling data generated by Geerk and his collaborators.

We have used density functional calculations to investigate structural instabilities in the A15 compounds, structural transitions and superconductivity in compressed chalcogens, superconductivity in
diborides and related ternary compounds, and stability of diboride-inspired metal-boron nanotubes.

We have developed an exact theory for nonresonant Raman scattering that carries one through a metal-insulator transition. Comparison of the results from the exact solution with experiment on nontrivial materials like the cuprates or SmB6 show remarkable agreement in the temperature dependence of the low-frequency spectral weight, the appearance of an isosbestic point, and in the appearance of charge-transfer peaks.The theory was also extended to include an examination of nonresonant inelastic light scattering by x-rays. Here we could examine transfering both momementum and energy with the correlated electron system. We found the charge transfer peaks dispersed little, while the low energy peaks had substantial dispersion throughout the Brillouin zone. We also identified that nonresonant scattering does not depend on the polarization channel when one is at the zone corner, and hence if there is polarization dependence in experiments, it is a direct measure of the effects of nonlocal charge fluctuations.

We are examining transport properties (electrical conductivity, thermopower, and thermal conductivity) in both many-body systems that can be solved exactly and in real materials that can be analyzed within DFT, but which have strong electron-phonon interactions.We discovered a few scenarios for a large figure of merit ZT for thermal transport. We found that close to half filling, a correlated insulator could show a sharp low temperature peak to ZT, but the peak might be wiped out by lattice thermal transport effects. We saw a much wider regime of large ZT at high temperature, appropriate for power generation applications. At moderate T, we found some instances where the Lorentz number could dip well below 3, which could be useful for low heat load conducting wires in cryogenic electronics packaging.

We studied transport properties of YbInCu4, which is an interesting valence change material, undergoing a first-order transition from a high temperature local moment phase to a low temperature intermediate valence state. The high temperature phase appears to be well described by the Falicov-Kimball model, which can be generalized to include all of the local atomic physics of the local moments. The model can also be applied to EuNi2(SiGe) alloys.

We examined an exact solution for the two-dimensional Falicov-Kimball model and showed how it could describe some of the physics present in stripe formation in the cuprates. We found diagonal stripes prevalent near half filling, giving way to axial stripes away from half filling, but we also saw the stability of a number of more two-dimensional structures at intermediate fillings, and we saw a wide range where phase separation prevailed. The results support the Kivelson-Emery scenario for stripe formation at large U, and the Scalapino-White scenario at small U. What remains unknown is how relevant the Falicov-Kimball model results are to the Hubbard model, but they certainly indicate a need to examine two dimensional structures in addition to stripe like structures in the Hubbard model.

We have also completed a Reviews of Modern Physics article on the Falicov-Kimball model, to appear this October.

Approximately 25 invited talks have been given on this research, at, for example, Univ. of Kansas, Kansas State, NRL, Virginia Commonwealth, Univ. of Illinois, Ohio State, IBM Almaden, Lawrence Livermore National Lab, Univ. of Fribourg, the Ecole Polytechnique Federale de Lausanne, ETH, an International Conference on Soft Condensed Matter Physics in Lviv, Ukraine, March meeting of APS in 2002, SCES conference in Poland, NIST, and the Workshop on Concepts in Electron Correlations in Hvar, Croatia.

In educational activities, we have been a test site for tutorials in relativity and quantum mechanics with the University of Washington Physics Education group, working to revise a set of tutorials for a sophmore-level class. We also have worked on training undergraduates to be peer instructors in a class on materials physics and quantum mechanics given to approximately 40 nonscientists each year.

Findings:

The A15 compounds have sharp structure in the electronic density of states near the Fermi level. Our analysis of the tunneling data for Nb3Sn indicates that taking into account this structure in the density of states (as opposed to using the standard procedure of assuming a constant density of states near the Fermi level) can significantly affect the extracted electron-phonon spectral function. The extracted Eliashberg function displays a number of features that are expected to be present for high-quality samples. However, we find that inclusion of the energy dependence in the density of states alone is not enough to resolve all the puzzles in the superconducting tunneling characteristics of this material.

Many of the A15 compounds undergo a tetragonal to cubic structural transformation as the temperature is raised. We have investigated this transformation in the Nb3Sn. As found in previous DFT studies, at T=0, the cubic structure is unstable against a sublattice distortion corresponding to dimerization along Nb chains. We carried out linear response calculations of the phonon spectra to determine if vibrational contributions to the free energy could account for the stabilization of the cubic phase at high temperatures. Because of the sharp structure in the electronic density of states near the Fermi level, we found that the phonon calculations were extremely sensitive to details in how the Brillouin zone was sampled, and we were unable to reliably determine the phonon contributions to the free energy.

We have found that despite their chemical similarity, S and Se
differ significantly in their behavior under pressure. Our calculations find that with increasing pressure, Se adopts a sequence
of ever more closely packed structures while S favors some more open structures. These differences are attributed to differences in the S and Se core states. All the compressed phases of S and Se we considered are calculated to have weak to moderate electron-phonon coupling strengths. Of particular interest, we have found that the surprisingly high superconducting Tc of 17 K measured in S at 160 GPa arises from a combination of only moderate electron-phonon coupling strength and a high phonon energy scale. Our work shows that despite suggestions otherwise, the conventional theory of electron-phonon superconductivity is able to explain the observed behavior of high-pressure phases of S. Recent diamond-anvil-cell experiments on S above 160 GPa have confirmed our predictions about the pressure dependence of the superconducting Tc.

The discovery of superconductivity near 40 K in MgB2 prompted a lot of speculation about the nature of the pairing in this material. Our DFT calculations indicated that the isotropic electron-phonon coupling parameter lambda is about 0.8. While this value is consistent with thermodynamic data, it is too small to explain the observed Tc, even taking into account the high phonon energies arising from the light B

mass. However, we have found that allowing for different order parameters on different sheets of the Fermi surface leads to a clean-limit superconducting lambda that is significantly larger. This interband anisotropy is important in MgB2 because there are two very different types of bands crossing the Fermi surface. Tunneling data on single crystals now support this multiple-gap model. Our calculations also suggested the importance of anharmonicity and nonlinear electron-phonon coupling in this material. We are now extending this work to investigate the ternary compounds AAlSi (A=Ca, Sr), which are isostructural with the diborides. While the electronic structure of CaAlSi and SrAlSi are similar, some superconducting properties such as gap-to-Tc ratio and pressure dependence of Tc have been measured to be very different in the two materials.

We have investigated the stability of hypothetical metal-B nanotubes inspired by the layered diborides (like MgB2 and AlB2). The prototype tubular systems studied were found to be competitive in energy with their bulk counterparts. Given the interesting physical and chemical properties of the bulk systems (e.g., superconductivity, and ability to accomodate wide range of metal atoms), these boron-based tubes could form an interesting new class of materials.

We have presented the first exact solution for nonresonant Raman scattering in any system where the electrons are interacting, solving a long-standing problem in electronic Raman scattering. We find on the insulating side of the metal-insulator transition that Raman response is 'universal' or model-independent, and displays the three features seen in experiments on a wide range of different materials: (i) there is a large increase in low-energy spectral weight and loss of higher-energy spectral weight as the temperature is raised: (ii) there is an isosbestic point, where the Raman response is independent of temperature separating the low-energy region from the high-energy one; and (iii) the ratio of the onset temperature, where low-energy spectral weight begins to appear, to the range in frequency over which the spectral weight increases as T increases, is much smaller than 1/3.52. We have been able to examine nonresonant Raman scattering in all symmetry sectors for the Falicov-Kimball model (which is not a Fermi liquid) and in the B1g sector for the Hubbard model (which is a Fermi liquid in the metallic phase). We find an essential model independence of the results in the correlated insulator in the sense that the results are essentially the same in both models, but we found interesting behavior in the Hubbard model when we looked at the Fermi liquid off of half filling. These results could be relevant to experiments that will hopefully be done on heavy fermion compounds in the future.

We also generalized our results to examine inelastic x-ray scattering, to see what happens when momentum can be transferred in addition to energy. We found that generically, the fermi-liquid like peak broadens and gets pushed higher in energy as the momentum transferred increases. We found little dispersion of the charge transfer peak, which is to be expected since the insulator is driven by essentially local physics. We also found that at the zone corner, there is a polarization independence of the results, indicating that any polarization dependence found there indicates the influence of nonlocal charge fluctuations, which may be one of the cleanest experimental probes to measure those effects in solid state systems.

In our thermal transport calculations, we were able to prove the Jonson-Mahan theorem by a direct evaluation of the operator averages in the Falicov-Kimball model, which provided an interesting counterpart to their derivation of those results. We also qualitatively examined thermal transport in a wide array of different correlated insulators and metals described by the Falicov-Kimball model and found that on the hypercubic lattice, near half filling, one could get a sharp peak in ZT at low temperature (but it might be washed out by lattice effects) and that over a wide range of parameters, one could get large ZT values at high temperature for power generation applications. We also found a curious region where the Lorenz number dipped well below 3.2, indicating the possibility of creating low heat load current leads for cryogenic applications.

Our results for the examination of the valence change transition in YbInCu4 and related compounds indicated that the results of the experiments are likely tied to the opening of a correlated Falicov-Kimball like gap at high temperature, although we are unable to describe the low temperature state within the FK model.

In our examination of charge stripes, we found that both the Kivelson-Emery scenario, coupling stripes to a proximity to phase separation, and the Scalapino-White scenario, which says stripes form naturally as a competition between kinetic and potential energy effects in the presence of correlations and fluctuations, occur in the FK model. In addition, we found a wide range of two-dimensional phases that also are stabilized, and we conjecture that some of these phases are likely to be important in the Hubbard model as well, although little work to identify such states has taken place.

Training and Development:

Alex Quandt came to Georgetown with a strong background in the area of quasicrystals. Working on this project has given him the opportunity to learn more about lattice dynamics, superconductivity, and transport in solids. Alex also participated in a trip to Croatia to work with Veljko Zlatic on thermal transport and density functional theory. Unfortunately no publishable results came out of that collaboration.

Denis Demchenko came to Georgetown with a background in magnetic materials and spin-based devices. At Georgetown, he has been working on electronic-structure calculations for magnetic interfaces and molecular magnets. He has also taken the opportunity to learn many-body physics techniques, and has written two papers on transport in correlated materials. He was an enthusiastic participant in the 2003 Boulder Summer School, which added both breadth and depth to his perspective on condensed-matter physics.

Courtney Peterson was an undergraduate physics major at Georgetown. In the course of doing her senior thesis on applying simple models to describe the electronic structure and superconductivity in MgB2, she learned basic solid-state physics and significantly strengthened her computational skills.

Ross Grimes and Peter King, both undergraduates at Georgetown, spent time learning how to be peer instructors for tutorial-based instruction in both a relativity and quantum mechanics course for majors and in a quantum mechanics course for nonscientists.

Outreach Activities:

Journal Publications

V. Zlatic and J.K. Freericks, "Theory of valence transitions in Ytterbium-based compounds", Proceedings of the NATO ARW conference, Bled, Slovenia
in Open Problems in Strongly Correlated Electron Systems, edited by J. Bonca, P. Prelovsek, A. Ramsak, and S. Sarkar, (Kluwer, Dordrecht, 2001), p. 371, vol. , (2001). Published

J.K. Freericks and T.P. Devereaux, "Non-resonant Raman scattering through a metal-insulator transition: an exact analysis of the Falicov-Kimball model", Condensed Matter Physics (Ukraine), p. 149, vol. 4, (2001). Published

J. K. Freericks and R. Lemanski, "Segregation and charge-density-wave order in the spinless Falicov-Kimball model", Phys. Rev. B, p. 13438, vol. 61, (2000). Published

V. Zlatic, J. K. Freericks, R. Lemanski, and G. Czycholl, "Exact solution of the multi-component Falicov-Kimball model in infinite dimensions", Phil. Mag.B, p. 1443, vol. 81, (2001). Published

J. K. Freericks and V. Zlatic, "Gap ratio in anharmonic charge-density-wave systems", Phys. Rev. B, p. 07310, vol. 64, (2001). Published

B. M. Letfulov and J. K. Freericks, "Dynamical mean-field theory of a double-exchange model with diagonal disorder", Phys. Rev. B, p. 174409, vol. 64, (2001). Published

S. P. Rudin, A. Y. Liu, J. K. Freericks, and A. Quandt, "Comparison of structural transformations and superconductivity in compressed Sulfur and Selenium", Phys. Rev. B, p. 224107, vol. 63, (2001). Published

A. Y. Liu, I. I. Mazin, and J. Kortus, "Beyond Eliashberg superconductivity in MgB2: anharmonicity, two-phonon scattering, and multiple gaps", Phys. Rev. Lett., p. 087005, vol. 87, (2001). Published

J. K. Freericks, T. P. Devereaux, and R. Bulla, " An exact theory for Raman scattering in correlated metals and insulators", Phys. Rev. B, p. 233114, vol. 64, (2001). Published

J. K. Freericks and V. Zlatic', "Thermal transport in the Falicov-Kimball model", Phys. Rev. B, p. 245118, vol. 64, (2001). Published

J. K. Freericks, Amy Y. Liu, A. Quandt, and J. Geerk, "Nonconstant electronic density of states tunneling inversion for A15 superconductors: Nb3Sn", Phys.Rev. B, p. 224510, vol. 65, (2002). Published

B.M. Letfulov and J. K. Freericks, "Phase separation in the combined Falicov-Kimball and static Holstein model", Phys. Rev. B, p. 033102, vol. 66, (2002). Published

R. Lemanski, J.K. Freericks and G. Banach, " Stripe phases in the two-dimensional Falicov-Kimball model", Phys. Rev. Lett., p. 196403, vol. 89, (2002). Published

F. Venturini, M. Opel, T. P. Devereaux, J. K. Freericks, I. Tutto, B. Revaz, E. Walker, H. Berger, L. Forro, and R. Hackl, "Observation of an unconventional metal-insulator transition in overdoped CuO2 compounds", Phys. Rev. Lett., p. 107003, vol. 89, (2002). Published

T. P. Devereaux, G. E. D. McCormack and J. K. Freericks, " Inelastic x-ray scattering in correlated (Mott) insulators", Phys. Rev. Lett., p. 067402, vol. 90, (2003). Published

J. K. Freericks, T. P. Devereaux, R. Bulla, and Th. Pruschke, "Nonresonant inelastic light scattering in the Hubbard model", Phys. Rev. B, p. 155102, vol. 67, (2003). Published

J. K. Freericks, D. Demchenko, A. Joura, and V. Zlatic', "Optimizing thermal transport in the Falicov-Kimball model: binary-alloy picture", Phys. Rev. B, p. 1551XX, vol. 68, (2003). Accepted

J. K. Freericks and V. Zlatic', "Exact dynamical mean field theory of the Falicov-Kimball model", Rev. Mod. Phys., p. , vol. , (2003). Accepted

A. M. Shvaika and J. K. Freericks, "Equivalence of the Falicov-Kimball and Brandt-Mielsch forms for the free energy of the infinite-dimensional Falicov-Kimball model", Phys. Rev. B, p. 153103, vol. 67, (2003). Published

T. P. Devereaux, G. E. D. McCormack and J. K. Freericks, " Inelastic x-ray scattering as a probe of electronic correlations", Phys. Rev. B, p. 075105, vol. 68, (2003). Published

J. K. Freericks, T. P. Devereaux, and R. Bulla, "Inelastic Light Scattering and the Correlated metal-Insulator Transition", Acta Physica Polonica B, p. 737, vol. 34, (2003). Published

V. Zlatic' and J. K. Freericks, "DMFT solution of the Falicov-Kimball model with an internal structure", Acta Physica Polonica B, p. 931, vol. 34, (2003). Published

J. K. Freericks, T. P. Devereaux, and R. Bulla, "Inelastic light scattering and the correlated metal-insulator transition", (Proceedings of the NATO ARW on strongly correlated electrons, Hvar, Croatia) Nato Science series II: Mathematics Physics and Chemistry: Vol. 110 (Kluwer, Dordrecht, 2003), p. 115, vol. , (2003). Published

V. Zlatic' and J. K. Freericks, "Describing the valence-change transition by the DMFT solution of the Falicov-Kimball model", (Proceedings of the NATO ARW on strongly correlated electrons, Hvar, Croatia) Nato Science series II: Mathematics Physics and Chemistry: Vol. 110 (Kluwer, Dordrecht, 2003, p. XXX, vol. , (2003). Published

A. Quandt, A. Y. Liu, and I. Boustani, "Density-functional calculations for metal-boron nanotubes", Phys. Rev. B, p. 125422, vol. 64, (2001). Published

Books or Other One-time Publications

Web/Internet Site

URL(s):

http://www.physics.georgetown.edu/~jkf/talk_list.html
http://www.physics.georgetown.edu/~jkf/pub_list.html
http://www.physics.georgetown.edu/~jkf/res_int.html
http://www.physics.georgetown.edu/~liu/pubs/list.html

Description:

These web sites includes some of the invited talks in powerpoint format that have been presented during the period of the grant, include publications from the grant, and include descriptions of the work for nonspecialists.

Other Specific Products

Contributions

Contributions within Discipline:

While the conventional theory of superconductivity, in which the interaction between electrons and phonons gives rise to the superconducting state, is well understood, properties of some specific electron-phonon superconductors remain puzzling. We have used a combination of theoretical and computational methods to examine some of these puzzles. For example, for the A15 compounds, the standard method of experimentally probing the electron-phonon interaction through tunneling yields strange and sometimes unphysical results (such as negative regions of the spectral function.) Our results indicate that taking into account the actual shape of the electronic density of states, which is sharply peaked in the A15 materials, rather than assuming a constant density of states, as is usually done in interpreting the tunneling data, can signficantly change the deduced spectral function. This helps explain some of the unphysical results obtained in the past and provides a better understanding of the superconducting properties of the A15 materials. Another example is MgB2, a material that chemists have long known about and studied, but which was only recently discovered to be a superconductor with a relatively high transition temperature. Different types of experimental probes have yielded different estimates for the strength of the electron-phonon interaction in MgB2. Calculations based on standard approximations for the electron-phonon interaction yield values that are consistent with thermodynamic data, but inconsistent with the high transition temperature. We have proposed one solution to this puzzle based on avoiding the usual approximation of a single isotropic gap, and there is now significant experimental evidence supporting this multiple-gap model.

We have also carried out a theoretical study of what happens when laser light is shone on a wide variety of materials that lie close to a metal-insulator transition. The scattering of light by the electrons has strong temperature and energy dependence and reveals interesting results about how the electrons in the material interact with each other. This contributes to our basic understanding of the phenomenon of the metal-insulator transition. Materials exhibiting this phenomenon are of interest for a variety of applications including detectors and nonlinear devices. Our work also examined what happens when x-rays scatter inelastically with correlated electrons. These experiments give more information about the dispersion relations of charge fluctuations in correlated systems, and our work has been able to make a number of predictions of what might be able to be observed if careful polarized measurements are made at different temperatures. Such experiments are currently being planned in next-generation RIXS studies.

Our work on charge stripes brings up the neglected possibility of two-dimensional structures being present in the Hubbard model phase diagram. We see these structures compete with stripes in the Falicov-Kimball model, so it is possible they also appear in the Hubbard model, at least for some range of parameters.
Our work on thermal transport shows what happens to thermal transport near a metal-insulator transition. This has been particularly difficult to model, since the most popular models of the correlated metal-insulator transition show the insulating phase only at the particle-hole symmetric point, where the thermopower must vanish. But we can investigate these properties in the Falicov-Kimball model since the MIT occurs away from the particle-hole symmetric point as well.

Contributions to Other Disciplines:

Ever since their discovery, carbon nanotubes have been the focus of intense multi-disciplinary research aimed at elucidating their basic properties and their potential for nanotechnological applications. We have proposed that it may be possible to synthesize other types of nanotubes such as boron-based systems as well. Such materials would differ from the carbon tubes electronically and could serve complementary purposes. Boron-based nano structures (tubes and needles) have been synthesized by at least two experimental groups, though further characterization of these materials is needed.

Our work on inelastic light scattering, especially that of scattering by x-rays, could have influence outside of materials theory as it can help guide new experiments and interpretation of data that is generated from the next generation of advanced light sources.

Our work on thermal transport could have applications outside of physics into engineering if some of the ideas for low temperature thermoelectrics can be realized in real systems, they could lead to low temperature thermocoolers. We also have some interesting ideas for low thermal loss electrical leads that could be used in the packaging of cryo-electronics.

Contributions to Human Resource Development:

We are a test site for the Tutorials in Introductory Physics, focusing on helping develop the tutorials in relativity and quantum mechanics. We have been working with the physics education group at U. Washington for eight years now, and our work has helped train other tutorial instructors and has helped improve the quality of the tutorials themselves before they have been disseminated publically nationwide.

Contributions to Resources for Research and Education:

Contributions Beyond Science and Engineering:

Categories for which nothing is reported:

Activities and Findings: Any Outreach Activities

Any Book

Any Product

Contributions: To Any Resources for Research and Education

Contributions: To Any Beyond Science and Engineering