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The Search and Theoretical Guidance for Higher Tc Superconducting Materials

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14. ABSTRACT

The overarching goal of the research was to identify mechanisms for high temperature s-wave superconductivity based on a combination of theory and experimental measurements of paradigmatic materials, and then to perform targeted searches for materials exhibiting these effects based on theoretical insights. Our work primarily focused on the inter-relation of specific types of charge ordered states and superconductivity, but we also took the opportunity to explore a variety of other promising directions based on opportunities that arose. Principle research directions included (1) the possibility of enhanced pairing interactions associated with materials that support a charge disproportionated charge density wave (CD-CDW) in part of their phase diagram, as distinct from CDWs arising from Fermi surface nesting, and/or that comprise valence skipping elements; (2) the relation between superconductivity, charge density wave correlations, and disorder; and (3) the search for and growth of higher Tc superconducting materials using molecular-beam epitaxy. Material systems that were studied included BaBiO3 (BBO) and the related solid solutions Ba1-xKxBiO3 (BKBO) and Ba(Bi1-xPbx)O3 (BPBO); analogs of BBO including CsTlCl3, CsTlFl3 and Cs1.17In0.81Cl3; Tl-doped PbTe; Pd-intercalated RTe3 (R = rare earth); Sr1−xLaxCuO2; La2-xBaxCuO4; Ca2RuO4; WO3; and Cu-intercalated TiSe2. This work resulted in 24 articles published in peer reviewed journals.

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Final Report for Project FA9550-14-1-0331
The Search and Theoretical Guidance for Higher Tc Superconducting Materials

M. R. Beasley, I. R. Fisher (Department of Applied Physics, Stanford University)
& G. Kotliar (Department of Physics and Astronomy, Rutgers University)

24/09/2019

Overview

The overarching goal of the research was to identify mechanisms for high temperature s-wave superconductivity based on a combination of theory and experimental measurements of paradigmatic materials, and then to perform targeted searches for materials exhibiting these effects based on theoretical insights. Our work primarily focused on the inter-relation of specific types of charge ordered states and superconductivity, but we also took the opportunity to explore a variety of other promising directions based on opportunities that arose.

Our program comprised three components: bulk synthesis and transport/thermodynamic measurements (Fisher + half a post-doc); band structure / theory (Kotliar + half a post-doc); and MBE thin film deposition (Beasley + staff scientist, C. Adamo).

Principle research directions included (1) the possibility of enhanced pairing interactions associated with materials that support a charge disproportionated charge density wave (CD-CDW) in part of their phase diagram, as distinct from CDWs arising from Fermi surface (FS) nesting, and/or that comprise valence skipping elements; (2) the relation between superconductivity, charge density wave correlations, and disorder; and (3) the search for and growth of higher T_c superconducting materials using molecular-beam epitaxy.

This program built upon ideas that originated during an earlier AFOSR-supported MURI award, while including some new directions enabled by the acquisition of the new Veeco oxide MBE system purchased on an AFOSR DURIP award.

The period of the award was from Sept 15th 2014 to Sept 14th 2019.

We outline in bullet form some of our main program accomplishments below. References are to the attached publication list. A more detailed summary of each of these accomplishments can be found in the earlier annual reports. A list of students and post-docs supported by the award is provided at the end of the report.

Accomplishments

(a) Theory:

The overarching goal of the research was to identify mechanisms for high temperature s-wave superconductivity in mixed valent compounds close to valence and charge instabilities, as in the paradigmatic compound $BaBiO_3$.

For disordered charge density wave materials (CDW) we proposed a Tc enhancement by elimination of the competing order via the introduction of disorder, namely superconducting order-by-disorder, and demonstrated its applicability in the $TaSe_{2-x}S_x$ alloy [10].

For $Ba_{1-x}K_xBiO_3$ (BKBO) system, we showed that static correlations near a metal to insulator transition (captured by GW and Hybrid Functionals) increase the electronic bandwidth and enhance the electron phonon interaction to the point that it can produce substantially higher Tc's than the corresponding BCS estimates (Phys. Rev. X **3**, 021011 (2013)). This proposal was confirmed experimentally [15] by angle-resolved photoemission spectroscopy studies on $Ba_{0.51}K_{0.49}BiO_3$. These measurements revealed that the effective mass was enhanced by a factor close to two together with the electron-phonon interactions which had a coupling constant $\lambda \sim 1.3 \pm 0.2$. See below for further discussion of the role of disorder in the T_c in the bismuthates.

Having established the mechanism for enhanced pairing interaction in BKBO, we focused on finding realizations of other "parent" compounds similar to the $BaBiO_3$ system which had not been synthesized previously and therefore were not listed in the ICSD database of known compounds. In the previous funding period of this grant, we designed the new compounds $CsTlCl_3$ and $CsTlFl_3$ ("Rational material design of mixed-valent high-Tc superconductors", Z. P. Yin and G. Kotliar EPL **101**, 2, 27002, (2013)). It was later synthesized and its mixed valent nature proved experimentally [2]. Given the toxicity of Tl, we used this award to address the design problem of new mixed valent systems based on In, and indeed were successful in achieving this goal [23]. Achieving efficient ways of doping these compounds remains a challenge, but the research has definitely established the theoretical capabilities to predict new mixed valent compounds which are thermodynamically stable, as one of these compounds were later synthesized [24].

(b) <u>Experiment (synthesis and measurement)</u>:

Our work focused on several paradigmatic materials exhibiting superconductivity and one or more of CD-CDW order, valence disproportionation, and/or short-range CDW correlations, including $Ba(Bi_{1-x}Pb_x)O_3$ (BPBO), Tl-doped PbTe and Pd-intercalated RTe_3. We also explored a wide range of other novel materials using the new oxide MBE system that we acquired, this latter work being accomplished by the staff scientist Carolina Adamo working on this project. Some specific achievements are listed below.

For BPBO, we discovered a previously unknown stripe-like nano-scale phase separation of two distinct polymorphs for superconducting compositions, and tracked its evolution across the phase diagram [5]. Combined with our tunneling measurements [1], these results point towards a clear role for disorder in reducing the maximum possible critical temperature in this system relative to values predicted by theory, possibly accounting for the startling difference in maximum Tc between BPBO and the closely related case of BKBO. Collaborative work based on our samples also revealed evidence for highly anomalous relaxation kinetics, possibly related to the interplay of disorder and the incipient CD-CDW state [9]. A unified theory including disorder and the GW/hybrid functionals is a challenging problem for the future.

For Tl-doped PbTe, we performed a series of experiments that definitively established (a) the shape of the Fermi surface [8, 20]; (b) that the Fermi level is approximately pinned by Tl-impurity states and that these states contribute additional incoherent carriers [18]; (c) that the effective mass increases in this regime [21]; (d) that the Tl impurities exist as distinct valence states, Tl^{1+} and Tl^{3+} based on spectroscopic measurements [19]; (e) that there is a strong spatial dependence in the DOS close to Tl impurities, and that this has a temperature dependence characteristic of the Kondo effect, suggesting the presence of dynamic valence fluctuations [12]; and (f) that the pairing interaction is enhanced relative to expectations based solely upon variations in the DOS (i.e. that the pairing interaction must be stronger when the Fermi level is pinned by the Tl impurities) [18]. The sum of these observations define a possible avenue for designing enhanced pairing interactions in other related materials, and possibly connect to the theory ideas introduced above.

For Pd-intercalated RTe_3 (R = rare earth), we demonstrated that disorder rapidly suppresses unidirectional CDW order, eventually yielding a superconducting state, but that short range CDW correlations persist over the entire phase diagram [22]. Moreover, these correlations are bi-directional, reflecting the nominally 4-fold-symmetric electronic susceptibility. Our work establishes the material as a potential model system to explore the inter-relation of CDW order, nematic order (which in principle can persist in the presence of disorder, even when long range CDW order is suppressed) and superconductivity.

Undoped $TiSe_2$ is another charge density wave material that we investigated. Doping with Cu lowers the CDW transition and a superconducting dome appears. We applied our point contact tunneling spectroscopy to this material and observed a sizable correlation gap, indicating the presence of Coulomb effects in the doped materials. However, the degree of correlation associated with this disorder was small [3] at optimal doping. Thus, Cu-doped $TiSe_2$ appears to be more like K-doped $BaPbO_3$ than Bi-doped $BaPbO_3$.

c) Novel applications MBE synthesis:

Carolina Adamo of our group is an outstanding MBE oxide thin film synthesizer. As this program wound down, she moved to TRW Research Labs in Los Angeles. While she was with us, she was in high demand as a film grower, and we tried to accommodate those groups (principally the Cornell Group) when their interests and ours permitted seminal work. These include the ability to control the topology of the Fermi surface in the ruthenates by means of epitaxial growth on suitable substrates (notably providing lattice expansion) [6, 7], and, again using epitaxy, to stabilize the T'phase of La2CuO4 over a wide range of background oxygen pressures during growth. ARPES data was carried out on these samples. The results provides definitive evidence that the electron doping in this materials is due to oxygen vacancies [11]. The origin of the doping in this material is a matter of considerable debate. Other examples of work made possible by the film growth by Adamo can be found in the references [16, 17].

Finally, we note that mixtures of gold and silver-oxide particles were recently reported to exhibit very high temperature superconductivity. Using MBE we grew silver oxide thin films upon which gold was deposited. Various deposition conditions were explored. No interface superconductivity was observed, consistent with tests by others of the reported high temperature superconductivity.

Publications

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Katherine Luna, Paula Giraldo-Gallo, Theodore Geballe, Ian Fisher, and Malcolm Beasley
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(Acknowledgment of this award was inadvertently omitted from this publication, but C. Adamo was supported 100% by this program for this study.)

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Students, post-docs and staff scientists supported during this award

(a) Students: P. Giraldo-Gallo (Stanford), J. Straquadine (Stanford)
(b) Post-docs: P. Walmsley (Stanford), M. Ikeda (Stanford), Zhiping Yin (Rutgers), C.J. Kang (Rutgers)
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