

# 2016 Superconductivity Program Review

Drs., Weinstock, Pomrenke, Nahcman; Marshall | November 10, 2016 | Arlington, VA

Abstracts

ORAL PRESENTATIONS

## **The Search and Theoretical Guidance for Higher T<sub>c</sub> Superconducting Materials: an Update**

(Ian R. Fisher, Stanford)

We present a summary of recent advances, both experimental and theoretical, affecting understanding of several anomalous superconductors. Following advances in recent years, we have continued to focus our attention mostly in two areas; the roles played by valence skipping elements, and the effect of charge density wave correlations. Materials studied during the last year include Na- and TI-doped PbTe, Pd-intercalated RTe<sub>3</sub> (R = rare earth), Li-intercalated InF<sub>3</sub>, and (La,Sr)<sub>2</sub>CuO<sub>4</sub>.

## **Discovery of New Superconductors By: Deliberate, Combinatorial or Natural Synthesis**

(Ivan K. Schuller, UC San Diego)

We have used combinatorics with a broad range of synthesis methods combined with high sensitivity Magnetic Field Microwave Spectroscopy (MFMMS) to search for new superconductors. Once an interesting signal is uncovered in a particular system, this is subject to a battery of conventional tests including Energy Dispersive X-ray, X-ray Diffractions, magnetotransport and magnetic measurements. This was shown earlier to be a very promising strategy in the search for new superconductors.

We have finished and published a detailed study of the electromagnetic signal observed at 117K in the Tlacotepec meteorite, found new magnetic phases in CuCl, and investigated in detail a variety of Graphene based materials. The new materials systems started during the past period include: ferromagnetic oxchlorides (chemical synthesis), bismuthates, irridates and nitrides (metallurgical synthesis), and BaFe<sub>2</sub>S<sub>3</sub> (high pressure synthesis). Additionally, we have investigated naturally and artificially synthesized minerals (Parkinsonite, Asisite, Hematophinite) and studied inclusions from meteorites (Mundrabilla and GRA95205). Extensive doping experiments are underway. In some of these materials, we have found evidence for new, unknown magnetic phases and also signatures of not yet identified (low temperature) superconductors.

We have done service measurements to search for superconductivity in samples from a number of groups in the US and elsewhere, which claimed unusual signals in a variety of compounds.

Heavily boron-doped diamond structures have been theoretically predicted to be candidates for high temperature superconductivity and indeed signatures of high  $T_c$  values of about 25K have recently been reported in high quality B doped (1.8 at %) diamond layers grown homoepitaxially on single crystal substrates using chemical vapour deposition. According to some predictions, if the degree of ordering could be increased, a  $T_c$  of 100K may be possible.

In the present work, we report the results of very high dose B implantation into single crystal diamond at high temperatures. We employ MeV ion energies in order to create a buried B doped layer far from potentially compensating defects near the surface. The use of high temperatures (600C) during the implantation allows for dynamic annealing of defects, which in turn inhibits the transformation of diamond into graphite, which would normally occur for implantation at such high fluences. In this way highly doped layers about 100nm thick, buried about 1.5  $\mu\text{m}$  below the surface and containing more than 6 at % of B have been fabricated.

### **Investigation of New Iron-Chalcogenide Superconductors**

(JP Paglione, Maryland)

The Maryland team's efforts to synthesize new advanced superconductors will be reviewed, including traditional materials exploration of known and new superconducting compounds and variants using bulk crystal growth techniques, high pressure experiments and exploration of natural mineral compounds. We will focus on recent progress in hydrothermal synthesis of new metastable iron chalcogenide phases as well as intercalation techniques to substantially increase  $T_c$  values, reviewing growth techniques as well as physical properties and anisotropies that review some new facets of iron-based superconductivity.

### **AFRL Search for New Superconductors, and Air Force Goals for Applications and Basic Research**

(Timothy J. Haugan, AFRL/RQQM)

The potential significant impact of superconductivity to the AF is increasingly being realized in many applications, such as high power devices, photonic sensors, microelectronics, antennas, and directed energy. The benefits and new capabilities enabled by superconductivity arise from unique scientific properties that cannot be achieved by other technologies. The lab at RQQM is focused on developing components and ancillary technologies required for megawatt-class power electric drivetrains, which are needed for applications such as hybrid-electric propulsion or directed energy. As a result of working on device development for many years, new basic research and materials needs are discovered, that are

critical to enable development or improve device or system performance. Examples of AF applications and basic research needs for those applications will be provided.

The search for new superconductors is being pursued, to achieve improved materials properties. Several areas are being investigated. 1. RE-Ti-C-O rare-earth titanates. Several interesting microwave (MFMMS) transitions are observed at 16K and 25K, that can't be assigned to known compounds in this system, and are believed to be associated with a phase with small < 1 Vol% that is difficult to isolate thus far. 2. The (FeSe)-M, (M = Sn, Ag, Cu) chalcogenide system. This is a 2D van der Waals gapped material being investigated for: i) the possibility of intercalation for increasing the superconducting T<sub>c</sub>, and ii) new compounds with correlated properties such as ferromagnetic (FM) or ferroelectric. Initial compounds discovered show promise as a possible 2D FM material with Curie temperature T<sub>C</sub> ~ 400K, and new magnetic/structural phase transitions at 5K, 40K and 130K and 180K, the exact nature of which have yet to be determined. 3. Doping of carbon allotropes with S, P, O elements and C<sub>8</sub>H<sub>18</sub> octane. For O and octane doping there are interesting anomalies being discovered, such as positive-to-negative-resistivity vs temperature transition at 100-150K for low current values (low ampacity), sharp magnetic transitions at 200-250 K, resistivity that varies with current values, creation of a lightweight ferromagnetic material at 300 K, and others.

### **Investigation of Superconductivity in Boron-Doped Diamond**

(Steven Praver, Melbourne)

Heavily boron-doped diamond structures have been theoretically predicted to be candidates for high temperature superconductivity and indeed signatures of high T<sub>c</sub> values of about 25K have recently been reported in high quality B doped (1.8 at %) diamond layers grown homoepitaxially on single crystal substrates using chemical vapour deposition. According to some predictions, if the degree of ordering could be increased, a T<sub>c</sub> of 100K may be possible.

In the present work, we report the results of very high dose B implantation into single crystal diamond at high temperatures. We employ MeV ion energies in order to create a buried B doped layer far from potentially compensating defects near the surface. The use of high temperatures (600C) during the implantation allows for dynamic annealing of defects, which in turn inhibits the transformation of diamond into graphite, which would normally occur for implantation at such high fluences. In this way highly doped layers about 100nm thick, buried about 1.5 μm below the surface and containing more than 6 at % of B have been fabricated.

Raman and photoluminescence measurements reveal that the single crystal diamond lattice has been preserved and that following high temperature treatment, most defects have been annealed. SIMS measurements showed that all of the implanted B has been retained in the lattice, and that no observable diffusion has occurred. However, despite the high B concentration and excellent crystal quality, electrical measurements over the range 35mK – 300K failed to reveal a superconducting transition. Measurements of magnetoresistance over the range -6 to 6T suggest evidence of weak

localization and the presence of residual compensating defects (electrons), thus pointing to the need to further optimize the annealing techniques.

### **Electric-Field-Driven Insulator-Metal transition and Superconducting Correlations in CuCl**

(Yakov Kopelevich, Campinas)

We present original experimental results in the context of a long-standing problem of possible high-temperature superconductivity in Copper Chloride (CuCl). In particular, we observed the electric-field-driven insulator-metal transition (IMT) in CuCl, related to the formation of Cu channels (filaments). Close to the IMT and at the “metallic” side of the transition, electrical transport properties are similar to those of Kondo systems, demonstrating negative magneto-resistance,  $[R(H)-R(0)] < 0$ , minimum in  $R(T)$  at a temperature  $T_m \sim 20$ -25 K, and a logarithmic rise of the resistance for  $T < T_m$ . We attribute this Kondo-like behavior to the spin-flip scattering of conduction electrons by magnetic moments localized at the Cu/CuCl interfaces. Besides, for “metallic” samples with low enough resistance, the superconducting (SC) - type transition reveals itself. The transition temperature  $T_c(0)$  for localized SC increases with the electric field application time:  $T_c = 9$  K ( $t = 3$  h),  $T_c = 16$  K ( $t = 10$  h),  $T_c = 35$  K ( $t = 15$  h). Hence, the filamentary SC in CuCl can be engineered and enhanced by means of applied electric field. We discuss a possible origin of SC in the context of the interplay between Kondo- and SC-like behavior in CuCl.

### **Interface-Induced 25 K Superconductivity in Ca122 at Ambient**

(C. W. Chu, Houston)

Superconductivity has been reversibly induced/suppressed in undoped  $\text{CaFe}_2\text{As}_2$  (Ca122) single crystals through proper thermal treatments, with  $T_c$  at  $\sim 25$  K at ambient pressure and up to 30 K at 1.7 GPa. We found that Ca122 can be stabilized in two distinct tetragonal phases (T) at room temperature and ambient pressure: PI with a non-magnetic collapsed tetragonal (cT) phase at low temperature and PII with an antiferromagnetic orthorhombic phase (O) at low temperature, depending on the low temperature annealing condition. Neither phase at ambient pressure is superconducting down to 2 K. However, systematic annealing for different time-periods at 350 °C on the as-synthesized crystals, which were obtained by quenching the crystal ingot from 850 °C, reveals the emergence of superconductivity over a narrow time-window. While the onset  $T_c$  is insensitive to the anneal time, the superconductive volume fraction evolves with the time in a dome-shape fashion. Detailed X-ray diffraction profile analyses further reveal mesoscopically stacked layers of the PI and the PII phases. The deduced interface density correlates well with the superconducting volume measured. The transport anomalies of the T-cT transition, which is sensitive to lattice strain, and the T-O transition, which is associated with the spin-density-wave transition (SDW), are gradually suppressed over the superconductive region, presumably due to the interface interactions between the non-magnetic metallic cT phase and the antiferromagnetic O phase. The results provide the most direct evidence to date for interface-induced superconductivity in undoped Ca122, consistent with the recent theoretical prediction.

### **Can Stoichiometric La<sub>2</sub>CuO<sub>4</sub> Be an Undoped High Temperature Superconductor?**

(Kyle Shen, Cornell)

In the cuprates, carrier doping of the Mott insulating parent state is necessary to realize superconductivity as well as a number of other exotic states involving charge or spin density waves. Cation substitution is the primary method for doping carriers into these compounds, and is the only known method for electron doping in these materials. Here, we report electron doping without cation substitution in epitaxially stabilized thin films of La<sub>2</sub>CuO<sub>4</sub> grown in the T' structure via molecular-beam epitaxy. We use angle-resolved photoemission spectroscopy to directly measure their electronic structure and conclusively determine that these compounds are electron doped with a carrier concentration of  $x = 0.09 \pm 0.02$  electrons per Cu. We propose that intrinsic defects, most likely oxygen vacancies, are the sources of doped electrons in these materials. Our results suggest a new approach to electron doping in the cuprates, one which could lead to a more detailed experimental understanding of their properties.

### **Examination of the Evidence of Superconductivity in Doped Graphite and Graphene**

(Grover Larkins, FIU)

We have observed evidence at temperatures in the vicinity of 260K of superconductivity in phosphorous- doped graphite and graphene. This includes transport current, magnetic susceptibility, Hall Effect and (pancake) vortex state measurements. All of these measurements indicate a transition that is that of a type II superconductor with no type I phase until below the limits of our measurement capabilities. These results will be discussed in detail. In addition we have also examined a number of other dopants, including boron, aluminum, arsenic, fluorine, lithium, nitrogen, sodium and sulfur in graphene and Highly Oriented Pyrolytic Graphite. The results of these studies will be presented and discussed in the context of the more widely published results using phosphorous as a dopant in graphene and Highly Oriented Pyrolytic Graphite.

### **Chemical Doping and High Pressure Studies of Mineral related $\beta$ -PdBi<sub>2</sub> Single Crystals**

(Bing Lv, UT-Dallas)

In conjunction with AFOSR's mission of searching for superconductivity in the naturally occurring minerals, we have adopted a parallel approach to determine the advantages and potential challenges of searching for novel superconductors in minerals: to look for superconductivity in minerals acquired and to synthesize the superconducting components followed by comparing them. We have purchased and

tested three minerals (after failing to get permission from Houston Museum of Natural Science for us to test their collection to low temperature, one of the best in the country), namely, Froodite, Polarite, and Sobolevskite, all consist of Pd-Bi-Pb compounds (although superconductivity associated with CuS<sub>3</sub> was detected in Covellite in 2006). We did make single crystals of PdBi<sub>2</sub>-xPbx for x = 0 – 0.5 and found T<sub>c</sub> = 5.4 K for x = 0 and decreases to below 2 K for x > 0.35. Unfortunately, no superconducting signal has been detected in the above minerals down to 2 K in the presence of a very strong magnetic background. A rapid decrease of T<sub>c</sub> with Na intercalation is also observed in the synthesized electron-doped Na<sub>x</sub>PdBi<sub>2</sub>. High-pressure study of β-PdBi<sub>2</sub> shows that the T<sub>c</sub> is linearly suppressed under pressure. The possibility of topological superconductivity in the undoped β-PdBi<sub>2</sub> and associated implications will also be discussed.

### **Determining Latitude and Longitude for Quantum Spins**

(Irfan Siddiqi, UC-Berkeley)

In quantum mechanics, measurements cause wavefunction collapse that yields precise outcomes, whereas for non-commuting observables such as position and momentum Heisenberg's uncertainty principle limits the intrinsic precision of a state. We simultaneously apply two continuous quantum non-demolition probes of non-commuting observables to a superconducting circuit. We implement multiple readout channels by coupling the quantum oscillator to multiple modes of a cavity. To control the measurement observables, we implement a 'single quadrature' measurement by driving the oscillator and applying cavity sidebands with a relative phase that sets the observable. Here, we use this approach to show that the uncertainty principle governs the dynamics of the wavefunction by enforcing a lower bound on the measurement-induced disturbance. Consequently, as we transition from measuring identical to measuring non-commuting observables, the dynamics make a smooth transition from standard wavefunction collapse to localized persistent diffusion and then to isotropic persistent diffusion. Although the evolution of the state differs markedly from that of a conventional measurement, information about both non-commuting observables is extracted by keeping track of the time ordering of the measurement record, enabling quantum state tomography without alternating measurements. Our work creates novel capabilities for quantum control, including rapid state purification, adaptive measurement, and measurement-based state.

### **Graphene-Superconductor Weak Links**

(Xu Du, Stony Brook)

I will discuss our recent work on probing the intrinsic Dirac electron Josephson effect and Andreev reflection in suspended graphene Josephson weak links. With ballistic charge transport, the Josephson current shows a linear dependence on the Fermi energy in graphene, as a result of its Dirac energy spectrum. At extreme close vicinity to the Dirac point, we confirm the long-standing theoretical proposal

of the Pseudo-diffusive charge transport through its signature in multiple Andreev reflections. I will also discuss our preliminary work on understanding the impact of magnetic field on Andreev reflection in graphene-superconductor weak links, where a strong suppression of Andreev reflection by magnetic field as well as its dynamics was observed. All our work paves the way towards a systematic study of the interplay between Andreev reflection and quantum Hall effect.

### **Direct-Write Nano Josephson Superconducting Tunnel Junctions**

(Shane Cybart, UC Riverside)

High-TC materials are very anisotropic and the superconducting properties vary along the different crystallographic directions. Our group has developed a direct-write Josephson junction fabrication technique that allows us to probe this anisotropy along these directions using electrical transport and confined nanowire geometries. From our results we find very large variations in the Josephson binding energy in the a-b plane which we attribute to the d-wave order parameter symmetry. I will present our recent results in these investigations and demonstrate some novel aspects of this technology.

### **SFAs: Superconducting Fraunhofer Amplifiers**

(Kevin Pratt, Tristan)

We present a novel sensor design based on linear arrays of long Josephson junctions. These long junctions, fabricated in YBCO by a direct-write approach using a helium ion microscope, possess inherent linearity over a wide dynamic range. The sensitivity of these devices scales linearly with the number of junctions in the array, enabling the development of wide bandwidth receivers with minimal intermodulation distortion.