High Temperature Superconductivity and Ultrasound

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Resonant ultrasound reveals a thermodynamic phase that may be important in understanding high temperature superconductors.

The phenomenon of superconductivity is one of the many great surprises of science. Building on the liquefaction of helium on 10 July 1908, on 28 April 2011 H. Kamerlingh Onnes (Onnes, 1911) cooled mercury to a low enough temperature that its resistance was "…near enough null". By 1986, the highest temperature T_c at which superconductivity had been observed was just above 20K. Work over the seven decades after the discovery of superconductivity revealed many unusual properties of superconductors. Of those properties, the most obvious is the vanishing of electrical resistance. The resistance was so low that no simple resistance measurement was found to be good enough to confirm this. Instead, a persistent current was induced in a closed loop of superconductor and the decay of the current was measured to a part in 10^5 over year time scales to put bounds on the resistance of the loop of less than 10^{-21} ohms, with million-year decay times. However the defining characteristic of superconductivity is the Meissner effect (Meissner and Ochsenfeld, 1933), which is the property that when a metal in a magnetic field is cooled to below its superconducting transition temperature, it expels the magnetic field from the interior of the superconductor (we'll come back to this later). This is not what a simple perfect electrical conductor would do--it has no problem with the magnetic field threading through it. The expulsion of the magnetic field by a superconductor requires energy and that can only come from a thermodynamic phase transition like that of water to ice. The difference in energy between the normal and superconducting state is the energy of the magnetic field in the volume of the material when it is in the normal state, just like the latent heat that must be removed from water at 273.15K to make ice.

To do this, persistent currents form on the surface of a superconductor in just the right way to cancel the interior magnetic field. The currents penetrate a small distance, the London penetration depth, λ_L, and for magnetic fields above some critical magnetic field H_c, superconductivity is destroyed. It is a curiosity that the Meissner effect is intimately related to "gauge symmetry" breaking. The phenomenon of gauge symmetry breaking, which we will not explain here, originally introduced for superconductivity in metals and reviewed by Anderson (Anderson, 1966), is now an important part of the "standard model" of particle physics and is the basis for the "Higgs mechanism" which gives mass to all elementary particles.

In 1986 scientists were stunned by the discovery of a superconductor with twice the transition temperature of the best superconductors found in the preceding seven decades. Adding to this baffling discovery was that this sudden jump in the usable temperature of superconductors occurred not in a conventional metal but in a copper oxide compound (a cuprate). The phenomenon was so outside the current thinking about superconductivity that it was distinguished with the name "high temperature superconductivity", or HTS. It is, today, still not understood.

Understanding high temperature superconductivity will require new theoretical insight outside the scope of the existing theory of metals. The success of the 1957 Bardeen, Cooper, and Schreiffer (BCS) theory of superconductivity (Bardeen et al., 1957) was a direct result of a deep understanding of the conventional metallic state. In cuprate HTS, the normal metallic state out of which HTS emerges is today not understood and this is the main scientific attraction. Even though theory is in the dark, HTS has motivated enormous advances in many measurement techniques that have also proved fruitful in understanding other condensed-matter systems. We review here some aspects of superconductivity, lay out incompletely and with bias some of the problems before us in the grand challenge to understand high temperature superconductivity, and briefly describe our recent insights (Shekhter et al., 2013) using Resonant Ultrasound Spectroscopy (RUS) that reveal a "pseudogap" and hint at the origin of the unusual metallic state in cuprates.

Important Ingredients for a Theory of Superconductivity

The microscopic theory of superconductivity is extensively reviewed and beyond our intended scope here. Here we only provide a broad sketch of the theory of conventional superconductivity that indicates how scientists direct work toward understanding HTS, and why acoustic measurements are relevant. This will also provide the context for our recent measurements using RUS. Let's begin with the theory of metals (Ashcroft, and Mermin, 1976).

In empty space an electron has well-defined momentum, "theory jargon" for an electron moving at constant speed in a fixed direction. It also has spin. Surprisingly, when an electron moves in a periodic array of atoms (crystal) that make up a metal, its quantum wavelike nature enables it to do this without losing energy just like sound waves diffracting in a phononic crystal or light through a diffraction grating.

We can imagine constructing a metal by adding electrons one by one to a fixed array of atoms (ions) whose position is determined by the crystal lattice structure. The first electron goes to the lowest energy state permitted by the lattice, almost a state of rest (~zero momentum). You can think of this like sound waves in a room. The lowest resonance of the room is a sound wave where a half wavelength fits in the longest dimension of the room. But we're dealing with electrons. No two electrons (fermions) can be in the same momentum (and spin) state. The second electron can also be in this low est energy state (but with opposite spin), however the third

"...Similar to magnetization, heat capacity, electric polarization, the elastic stiffness is fundamentally connected to thermodynamics and the free energy."

electron is out of luck. It must be in a state of higher energy. The acoustics analogy is a half wavelength now fits in the second-longest dimension of the room. By the time we throw in 10^{23} electrons to make the metal electrically neutral, the last electrons have the highest energy in the metal (the Fermi energy) and are moving at about 300 times the speed of sound or about 1% of the speed of light.

Because the atoms in the crystal are equally spaced (periodic) there are only a finite number of meaningful solutions to the electron wave equation, and therefore only a finite number of allowed energy states for an electron in a solid. The acoustics and digital electronics analogy here has to do with aliasing and the Nyquist limit. Consider a digitizer acting on a sinusoidal signal. Let's say the digitization rate is exactly twice the sine wave frequency, and the digitization starts at the first zero crossing of the sine wave. The next data point will be at the next zero crossing and so on. The end result is that the digitizer output is all zeros. This is the Nyquist limit. For a sine wave of lower frequency, the digitizer more or less captures the sine wave. The curious effect occurs if the sine wave frequency is slightly higher than that half that of the digitizer. The digitizer captures a sine wave, but it is at a very low frequency—namely one that is at the difference between sine wave and half the digitizer frequencies. This is called aliasing, and is used in such things as cell phones so that the very high radio frequencies can be reduced to a lower value for processing (called an undersampling mixer). If we consider the ions as the digitizer for electron wave functions, then just as in the electronics case, any electron wave function with wavelength shorter than twice the lattice spacing is the same physically as a much shorter wavelength electron. This forces the system to have only a finite number of physically-meaningful allowed electron wavelengths, and therefore a finite number of allowed energies.

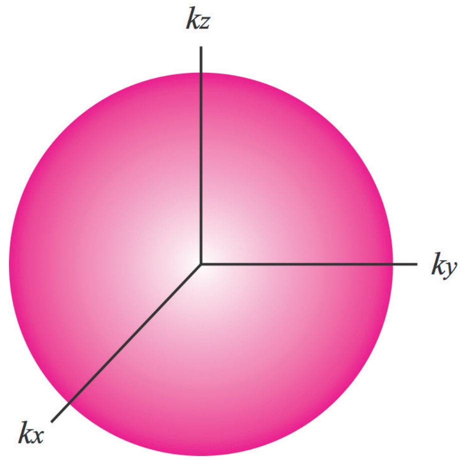

Figure 1. *The theory of metals takes the crystallographic array of nuclei and pours electrons in, each going into a different quantum state until enough go in to cancel the positive charge of the nuclei. When a plot is made of energy versus momentum, in the very simplest case, the electrons fill a sphere whose surface is at a single energy, the Fermi energy— ~30,000K, so that the electrons at the Fermi surface are moving on order 300 times the speed of sound.*

Electrons move in 3-dimensional space, their momenta are vectors. This is the starting point for any modern discussion of a metal. An important concept is the Fermi surface. It is a surface in a plot of electron energy versus momentum inside of which all electron energy states are occupied, and all states above it are empty (Figure 1). The difference between a metal and an insulator is that in a metal, not all the allowed energy levels are occupied, while in an insulator, every one is occupied, and, not surprisingly, in any particular direction for metal or insulator, there are an equal number of electrons moving one way as another, so there is zero net electrical current. The empty energy states in a metal are very close to the occupied ones, of order the Fermi energy divided by Avogadro's number. The tiny energy needed to shift an electron to an empty state makes it easy for an electric field to induce an electron to change its momentum, unbalancing the number of electrons moving in a particular direction so that an electric current is the result.

The shape of the Fermi surface and the properties of electrons very near this surface determine most metallic properties including electrical and thermal conductivity, heat capacity, magnetization, and more. The electrons with energy close to the Fermi surface are the only electrons that participate in superconductivity (note that we will, throughout, use temperature as the unit of energy where $E=k_BT=\hbar\omega$ and where E is energy, T is absolute temperature, $\hbar$ is Planck's reduced constant, and k_B is Boltzmann's constant).

There are several energy scales at play when we discuss superconductivity. The first energy scale is the one just discussed, the Fermi energy T_F, about 30,000K, the same order of magnitude as chemical binding energies. We see, then, that metals are very "cold", that is, the primary energy scale for electrons in a metal is 100 times room temperature.

The second energy scale is the Debye energy. Although this, as we will show, is connected to sound and ion motion, it is intimately also connected to electron motion. We've ignored electron and ion charge up to this point. Electrons and ions are charged and we would expect them to interact over long distances via the ordinary coulomb electrostatic force. However electrons near the Fermi surface in a metal can adjust their motion to screen all long range electrical forces. That is, if, say, a positive charge were placed inside a metal then a cloud of electrons would form around it so that the positive ion and its negatively charge electron cloud would appear electrically neutral some short distance away. This ensures that in the metal the "coulomb forces" act only over very, very short distances-less than a unit cell. Such screening makes the forces between ions short range so that a useful model of ions in a solid is an array of masses and springs, as diagrammed in Figure 2. This model of a huge array of masses and springs is of order Avogadro's number of quantum harmonic oscillators, and is responsible for all the acoustic properties of a metal. Each oscillator contains quanta of vibrational energy or phonons. The spectrum of phonons form the dispersion curve in a solid, Figure 3.

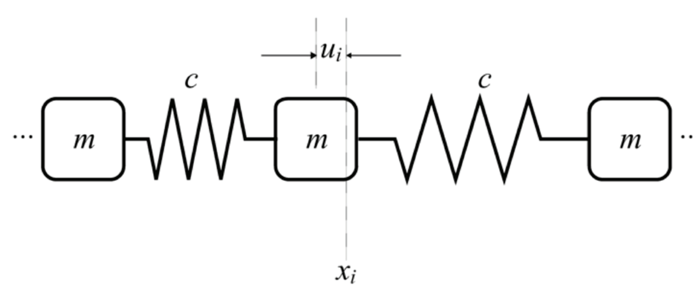

Figure 2. *Screening of charges from each other by weak dynamic motion of electrons shields the the positive ion motion u_i at x_i and makes a useful model of vibrations in solids to be that of masses m more or less connected within a few nearest neighbors by springs c.*

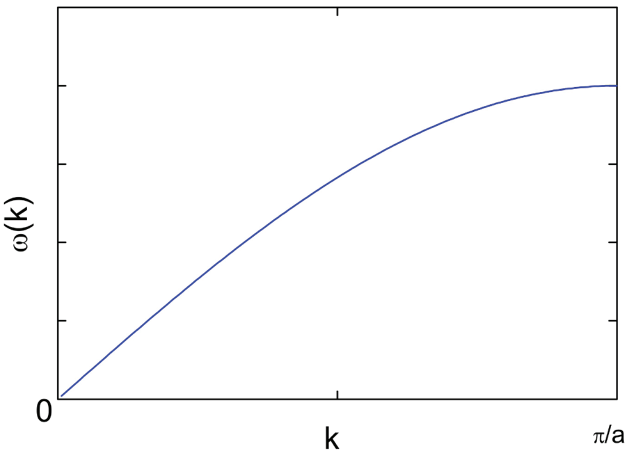

Figure 3. *The solution to an array of order Avogadro's number of simple quantum harmonic oscillators, called the phonon dispersion curve. The horizontal axis is the momentum in units of ħ. The maximum momentum is that of a vibration whose wavelength is twice the unit cell distance. The vertical axis is in arbitrary units of angular frequency. In a typical solid, the maximum frequency of a phonon is 300K or so and the solid line is really of order 10^7 discrete points.*

The allowed phonon wavelengths are also subject to a Nyquist-like argument but, because phonons are bosons, a solid can have as many phonons as you like. The hotter the solid, the more phonons there are. The maximum frequency of vibration in a solid (maximum energy for a single phonon) is of order 300K, or the Debye energy. Notable, the linear slope of the phonon dispersion curve at low momentum is the speed of sound.

In real metals, the coulomb interactions are not weak. However the short-range (screened) interactions enable a very successful description of metallic and elastic properties, the "Fermi liquid" theory of metals in which the ensemble of electrons is treated as liquid. In so-called "correlated" metals such as the HTS cuprates, the effect of coulomb interactions is not captured by Fermi liquid theory—electrons are no longer independent as they are in our simple description of the effects of screening. The breakdown of the Fermi liquid description is why we do not understand HTS and what makes the physics of cuprates a grand challenge of condensed matter.

There is another way to break the Fermi liquid ground state. Fifty years after the discovery of superconductivity, Cooper (Cooper, 1956) had the essential insight that the Fermi surface is unstable if electrons attract each other, no matter how weakly. He showed that two electrons above the Fermi surface form a bound state for any weak attraction, the famous "Cooper pair". That Cooper pair is no longer a part of the Fermi surface. Bardeen, Cooper, and Schreiffer (BCS) showed that in a real metal with an attractive inter-electron potential, then all electrons (of order Avogadro's number) will form pairs, and the liquid of these pairs forms a single quantum state, the so-called superconducting condensate, lowering the system energy. The properties of this condensate explain all superconducting properties. All Cooper pairs have zero momentum. To break up a pair takes a lot of energy, of order T_c, the so-called "superconducting gap". What this means is that for a single electron to change its state, it has to have not the tiny amount of energy needed in an ordinary metal to begin electrical conduction, but now a large amount of energy to jump across the superconducting gap. But the electrons in the condensate are a different story. Because the condensate is a single quantum state with a large number of electrons in it, and because it takes a lot of energy, of order T_c to break it up, it can move without resistance. That is, an electric field applied to it accelerates the entire quantum state as if it were a large chunk of electric charge in a vacuum. The explanation of this led to the second superconductivity-related Nobel Prize, the one for BCS. One important aspect of this condensate is that the number of electrons that participate in it is not well defined, or equivalently, for this state charge is not conserved (it is of course conserved for the totality of electrons). This breaks gauge symmetry, something we won't explain (Anderson, 1966). Though very controversial at the time, this idea led to the Higgs mechanism, at the basis of the standard model of particle physics.

What did BCS realize? If like charges repel, how can electrons attract each other? Bardeen, convinced by an ion-mass isotope effect, connected the missing "glue" to ion motion. The isotope effect for most superconductors known at the time is not the one that gave Bardeen this idea, nevertheless in the end his theory was proved correct and predictive for a restricted (BCS) class of superconductors. The pairing "glue" in BCS superconductors is phonons. How does this attraction work?

It was known before 1957 that electrons couple to phonons. What does this coupling look like? Electrons move at 300 times the speed of sound. The electrons tweak the lattice for short times (a non-resonant drive) creating a distortion in the crystal lattice that takes a long time to recover and so is "retarded" because ions are heavy. Other electrons will see that distortion as a brief increase in a local positive charge background, creating a weak interaction between electrons. To keep close to reality we must discuss the quantum mechanical form of this interaction (Abrikosov, 1965), with $\omega(k)$ the frequency for a given energy of an object with momentum k, e the electron energy, p the phonon momentum, $u(p)$ the phonon dispersion curve. Figure 4 shows the energy

$$\frac{\omega_0^2\left(\vec{k}\right)}{\omega^2\left(\vec{k}\right)-\omega_0^2\left(\vec{k}\right)}=\frac{u^2\left(\vec{p}_3-\vec{p}_1\right)^2}{\left(e_3-e_1\right)^2-u^2\left(\vec{p}_3-\vec{p}_1\right)^2}$$

"landscape" for this expression and the "Feynman diagram" used to represent it. The second term in the denominator on the right of Equation 1 is the energy of the phonon that is exchanged between two interacting electrons, always less than the Debye energy ω_0 (300K). If the change in energy (e.g. $e3$-$e1$) of each electron is much less than the Debye energy, the phonons "mediate" an attractive interaction (the sign of the right side is negative). Figure 5 shows how the energy landscape is modified to form the superconducting gap in the energy spectrum of a superconductor.

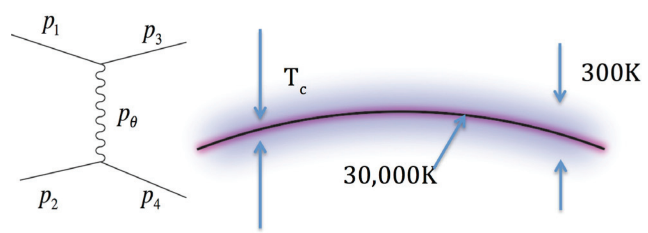

Figure 4. *The Feynman diagram used to describe the electron-phonon interaction, and the energy landscape for the process. For electrons within T_C of the Fermi energy, which is much less than the Debye energy of 300K, the phase shift from driving a harmonic oscillator (phonon) below resonance produces an attractive interaction.*

Superconductivity and Ultrasound

How does ultrasound connect to superconductivity? Similar to magnetization, heat capacity, electric polarization, the elastic stiffness is a fundamental thermodynamic susceptibility of the free energy (Migliori, 2008). The free energy difference ΔF between the superconducting and normal states is the energy of the (maximum possible) magnetic field expelled from the volume of the superconductor, proportional

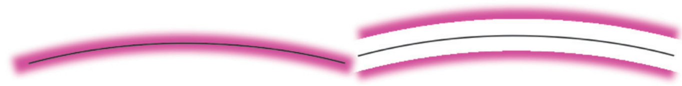

Figure 5. *A cartoon of the occupied energies of electrons above T_C (left) and below T_C (right). The bound pairs sweep out an energy gap. For electrons to show electrical resistance, they must dissipate energy, only possible if they are driven hard enough to cross the energy gap. Driving them gently produces motion of the entire superfluid without dissipation, resulting in zero resistance.*

to magnetic field squared. Because the superconducting phase transition is second-order or continuous, absolutely nothing seems to happen right at the phase transition so that

$$\Delta F\big|_{Tc} = H_c^2\left(T_c, B, P\right) = 0$$

where T is temperature, B is magnetic field, P is pressure (we have set all constants equal to unity). Equation 3 tells us the volume change across a superconducting phase transition ΔV is the derivative of Equation 1which is (pressure change times volume change is energy change)

$$\left.\frac{\partial \Delta F}{\partial P}\right|_{Tc} = \Delta V\big|_{Tc} = H_c\left.\frac{\partial H_c}{\partial P}\right|_{Tc} = 0$$

because at T_c the critical field H_c is zero. The change in volume with pressure is directly related to the elastic stiffness, Equation 4, and is at T_c.

$$-\left.\frac{\partial^2 \Delta F}{\partial P^2}\right|_{Tc} = -\left.\frac{\partial \Delta V}{\partial P}\right|_{Tc} = \Delta\frac{1}{c_{ij}} = H_c\left.\frac{\partial^2 H_c}{\partial P^2}\right|_{Tc} + \left.\left(\frac{\partial H_c}{\partial P}\right)^2\right|_{Tc}.$$

The first term on the far right of Equation 4 is again zero, but the last term is positive definite. For a continuous (or second-order) phase transition such as superconductivity (or the pseudogap), the elastic moduli c_{ij} are *discontinuous*--there is a step change downward upon entering the ordered (low temperature) phase. There is also a break in slope of moduli versus temperature. *Thus elastic moduli, and the sound speeds that they determine, are direct probes of a second order (or any) phase transition and reveal it with the very strong response of a discontinuous jump even though the phase transition is continuous.*

The method we used to study high temperature superconductivity, called Resonant Ultrasound Spectroscopy or RUS is reviewed extensively (Migliori and Maynard, 2005). In brief the mechanical resonances of a specimen of regular shape (easy to measure) are analyzed (difficult computational problem) to obtain the full elastic tensor. With good control over vibration and temperature, we can detect 10^{-7} changes in elastic moduli. We developed new and powerful techniques to determine accurately the frequency and width of resonances. Figure 6 illustrates part of this approach where the in-phase and quadrature response across the two resonances is shown. Balsa wood (we cannot seem to find anything less acoustically dead at 4K) for the cell improves vibrational isolation down to 4K. Even with these improvements, the measurements we were after required perfect detwinned single crystals (Shekhter et al. 2013), only recently available.

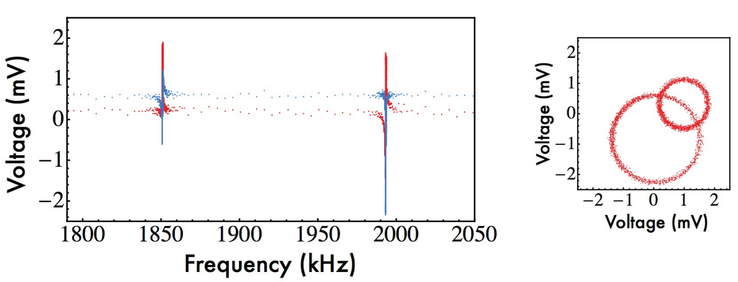

Figure 6. *Shown here are raw resonance in-phase and quadrature data for two YBCO resonances (left) and the plot in in-phase, quadrature space (ReV-ImV) of the same peaks (right). Note how by choosing the data point interval to be uniform in ReV-ImV space, the inset easily reveals that two peaks are present, while greatly reducing the time for a measurement with no compromise in signal to noise ratio.*

How big are the effects? At the superconducting transition, a fraction T_c / T_F of electrons change energy by the superconducting gap, proportional to T_c so it is expected that the fractional step discontinuity in moduli is of order $(T_c / T_F)2$ and this is about what we observed (10^{-4}) in $YBa_2Cu_3O_{6+\delta}$ (YBCO), Figure 7. This result is a first in that it suggests a rather conventional thermodynamic signature of superconductivity in cuprates. This means that this elastic properties of high temperature superconductivity in cuprates are not anomalous, an important result made possible by acoustics. Note that the transition we observed ultrasonically is very sharp for a HTS superconductor, indicating that the specimens we measured are nearly perfect. RUS is exceptionally intolerant of flaws of any sort, but produces exceptional results when flaws are absent.

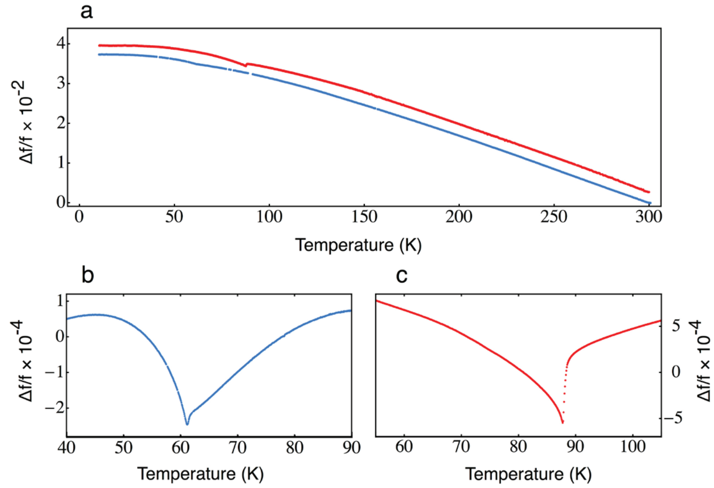

Figure 7. *The overall resonant frequency response (proportional to elastic modulus) of a compressional modulus of YBCO for both near-optimally doped (red) and underdoped (blue), left, and the superconducting transitions in expanded plots, right, measured with RUS.*

The Pseudogap and Ultrasound

The pairing glue in cuprates is not phonons. Instead, the pairing glue is mediated by electronic excitations, yet-unknown. It has long been recognized that the physics of superconducting pairing in HTS is related to the physics responsible for the anomalous metallic behavior in the normal state. Electrical conductivity and other measurements indicate a change in the metallic behavior in cuprates across a boundary in the temperature-oxygen-doping phase diagram. This boundary defines the so called "pseudogap" phase (Figure 8) (Shekhter et al., 2013). The conjecture is that the physics of the

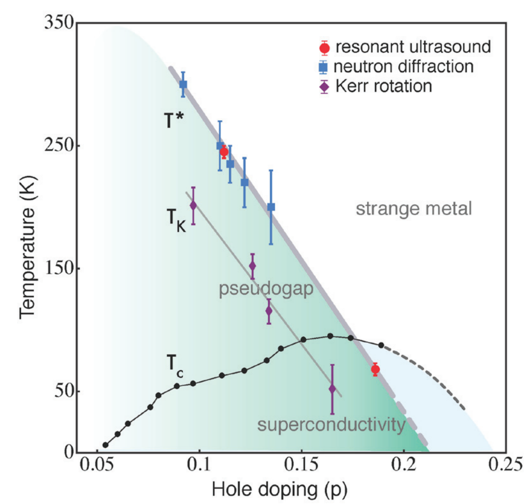

Figure 8. (Shekhter et al., 2013) *The various measurements indicating the presence of the pseudogap, with RUS results in red. The transition temperatures for superconductivity are shown by the solid black circles (left).*

"pseudogap" region of cuprates phase might provide insights into the physics of the anomalous metallic state in cuprates. Until it was not certain that the pseudogap was a thermodynamic phase. A few years ago polarized neutron scattering (Figure 8) measurements identified the onset of magnetic order at the pseudogap boundary. Recent resonant ultrasound measurements (Shekhter et al. 2013) reveal a thermodynamic signature at the pseudogap boundary that extends to where the superconductivity is strongest. Because fluctuations of the order parameter (basically the transition temperature) associated with the pseuodogap have a similar energy scale to that of phonons, those critical fluctuations just might act as a glue for pairing. The jury is still out.

Summary

Understanding the anomalous metallic state in cuprates and the high temperature superconductivity that emerges from it are grand challenges of condensed-matter physics today. Acoustics and RUS have proven to be revealing in studies of HTS. The acoustic observation of the pseudogap thermodynamic phase in HTS, though far from understood, and certainly not established as a mechanism for the glue of high temperature superconductivity, has the ingredients needed to replace phonons in assembling the pairs that upon formation become the charged superfluid of superconductivity.

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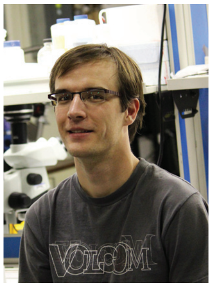
Biosketches

Albert Migliori received his B. S. in physics in 1968 from Carnegie Mellon University, his M. S. and Ph.D. in physics from the University of Illinois in 1970 and 1973. He is co-discoverer of acoustic heat engines, Co-Chair of the Science Advisory Council for the National High Magnetic Field Laboratory (UF, FSU, LANL), Director of the Seaborg Institute for Actinide Science at Los Alamos National Laboratory, and is a leading expert in the use of resonant ultrasound spectroscopy as a solid-state physics tool for which he has won RD100 awards in 1991 and 1994. He is a fellow of the Los Alamos National Laboratory, the American Physical Society, the American Association for the Advancement of Science, and the Acoustical Society of America. He is Chair, Physical Acoustics Technical Committee, Acoustical Society of America, and Chair, General Instrumentation and Measurement Topical Group, American Physical Society. He holds 25 patents, is the author of about 200 publications, six book chapters, and one book. Recent interests include elasticity of PU, and state-of-the-art research and development of new measurement techniques.

Arkady Shekhter received his PhD in theoretical condensed matter at Weizmann Institute in Israel in 2006 for his work on correlation effects in two-dimensional electron liquids. He continued his research of thermodynamic and transport phenomena in metallic cuprates as a postdoctoral research associate at UC Riverside. Challenging the boundary between theory and experiment he proceeded with experimental work as a Dirac Postdoctoral Fellow at the NHMFL-PFF. His work has been reported in peer-reviewed journals including Nature, Phys. Rev. Lett.



Brad Ramshaw got his PhD in experimental condensed matter physics from the University of British Columbia in 2012 working on quantum oscillations in the high temperature superconductor YBa2Cu3O6+delta. Brad came to LANL in 2012 and has developed new resonant ultrasound spectroscopy techniques for looking at the symmetry breaking of phase transitions, and extended his previous work on quantum oscillations to 100 Tesla using the unique capabilities at the NHMFL. His publications have appeared in journals such as Nature, Nature physics, Nature Communications and Physical Review Letters.

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