Scaling Laws in High-temperature Superconductors as Revealed through Infrared Spectroscopy

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Superconductivity refers to a fascinating state of matter where the electrical resistivity is precisely zero. Originally discovered in elemental metals such as mercury and tin in the early part of the last century, the mechanism of superconductivity was elusive and nearly 50 years passed before a comprehensive theory for superconductivity in metals was proposed by Bardeen, Cooper and Schrieffer [1] (the "BCS" theory). In a normal metal, the resistivity is determined by the elastic scattering of carriers. However, when a metal becomes a superconductor, the charge carriers are no longer single electrons, but rather pairs of electrons ("Cooper pairs"), which are bound together by a phonon interaction (phonons are the vibrations of the atomic lattice), and flow without resistance. The BCS theory of superconductivity has been tremendously successful at describing this phenomenon in metals and alloys. However, an aspect of the phonon-mediated pairing mechanism is that the superconducting critical temperature (T_c) will be restricted to values below ~30 K [2]. This prediction, as well as the observation for a number of years of a record $T_c = 23$ K in Nb₃Ge, implied that the superconductivity would remain a curiosity restricted to low-temperature physics labs, removed from mainstream applications.

This field was revolutionized by the discovery of superconductivity at elevated temperatures ("high-T_c") in 1986 in a class of copper-oxide materials [3]. The critical temperatures of these materials increased rapidly, with the current record of T_c ~ 140 K in a class of mercury-based copper-oxide materials. One of the best known (and most studied) of the high-T_c superconductors is the bilayer material YBa₂Cu₃O_{7- δ} (YBCO), shown in Figure 1; this was the first material with a T_c above liquid-

Figure 1: The unit cell of $YBa_2Cu_3O_7$, with Y (yellow), Ba (green), Cu (blue), and O (red, orange) denoting the different atoms. This high-temperature superconductor has two copper-oxygen sheets that form the a-b planes in the unit cell. The pyramids illustrate the in-plane oxygen coordination and the apical oxygen along the c-axis direction. This material also has copper-oxygen chains that run along the b axis. The copper-oxygen planes are the common building blocks of the high-temperature superconductors.



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nitrogen temperatures (77 K). The fact that the T_c 's of the copper-oxide materials are much higher than those predicted by the BCS theory indicates that the superconductivity in these materials is probably not phonon-mediated, at least in the usual sense. Understanding high- T_c materials is currently one of the great challenges in condensed matter physics. Despite an unprecedented effort and over 60,000 published papers in the field, the mechanism of superconductivity in these materials is sill not understood.

Spectroscopy of superconductors

Infrared reflectance spectroscopy is a powerful technique for determining the optical properties of solids. The reflectance is a complex quantity, consisting of an amplitude and a phase. Typically, in an experiment only the amplitude is measured. However, if the reflectance is measured over a very wide frequency range (in this case from a few meV to several eV), then the Kramers-Kronig relation may be used to calculate the phase. Once the amplitude and the phase are known, then the frequency dependence of all the other complex optical properties, such as the dielectric function and the conductivity, may be determined [4]. While the range of measurements spans the microwave (below 1 meV) through to the visible and ultraviolet (typically above 4 eV), the infrared and far-infrared regions (1-125 meV) are of particular interest because they correspond to the energy scales that are important for superconductivity in both the elemental and high-T_c superconductors. This allows the optical signatures of superconductivity to be investigated in a wide variety of materials. Standard Michelson-type Fourier transform spectrometers are used for these measurements, and different combinations of sources, beam splitters and detectors are used to sample different frequency regions. The tremendous advantage of the synchrotron is that it is a highly collimated "white-light" source that may be used across the entire spectral region. Many of these experiments were performed at the infrared beamline U10A at the National Synchrotron Light Source at Brookhaven National Laboratory.

Two very useful properties of metals and superconductors may be determined using optical methods: the dc conductivity σ_{dc} and the superfluid density ρ_s (a measure of the number of carriers in the superconducting state). Very often these values are obtained using different techniques on different samples. The advantage of optical techniques is that in a single experiment the real and imaginary parts of the dielectric function may be determined, and through this the values for σ_{dc} and ρ_s . In the region of interest, the depth to which light penetrates is typically a micron or greater, making infrared a bulk probe.

The frequency-dependence of the real part of the conductivity of $Bi_2Sr_2CaCu_2O_8$ (BSCCO), another bilayer material with a T_c similar to that of YBCO, is shown at close to optimal doping (where T_c is a maximum) in Figure 2 in the infrared region for a variety of temperatures [5]. The conductivity is a measure of the imaginary part of the dielectric function, $\sigma_1(\omega) = \omega \epsilon_2(\omega)/60$; the frequency is expressed in units of cm⁻¹, also referred to as wavenumbers (1 eV = 8,065 cm⁻¹), so that the con-

ductivity has units of Ω^{-1} cm⁻¹. In the normal state (T > T_c) the low-frequency conductivity is representative of many copper-oxide materials and may be described by $\sigma_1(\omega) = \sigma_{dc}/(1 + \omega^2 \tau^2)$, a Lorentzian centered at zero frequency with width $1/\tau$ (the carrier scattering rate), which narrows with decreasing temperature. The dc conductivity is simply σ_{dc} = $\sigma_1(\omega \to 0)$, which makes a connection with transport measurements. Below T_c, large changes in the conductivity have taken place that signal the onset of superconductivity and the formation of a condensate; below ~100 meV there is a strong suppression of the conductivity, although there is still a substantial amount of residual conductivity at low frequency. This behavior is typical of most, if not all, of the copper-oxide superconductors. If the spectral weight is defined as simply the area under the conductivity, then the ρ_s may be estimated by tracking the changes in the spectral weight above and below T_c--the so-called "missing area" in Figure 2. This is also known as the Ferrell-Glover-Tinkham sum rule [6]. While techniques based on sum rules often yield accurate values for ρ_s , this may not always be the case [7].

A more direct estimate comes from an examination of the real part of the dielectric function for temperatures well below T_c where the condensate is fully formed. In a material where all of the charge carriers collapse into the condensate, the dielectric function consists only of a real part that can be expressed as $\varepsilon_1(\omega) = \varepsilon_{\infty} - \omega_{ps}^2/\omega^2$, where $\omega_{ps}^2 =$



Figure 2: The real part of the optical conductivity for Bi₂Sr₂CaCu₂O₈ for light polarized along the a-axis direction (there is a weak in-plane anisotropy in these materials) at several temperatures above and below T_c . Large changes in the normal state at low frequency reflect a decrease in the scattering rate (1/ τ), while the loss of weight at low frequencies below T_c is indicative of the formation of a superconducting condensate. Optical values for the dc conductivity are determined by taking $\sigma_{dc} = \sigma_1(\omega \rightarrow 0)$.

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 $4\pi ne^2/m^*$ is the strength of the condensate expressed as the square of a plasma frequency (i.e., $\rho_s = \omega_{ps}^2$), and ε_{∞} is a high-frequency contribution. The strength of the condensate may simply be written as $\rho_s = -\omega^2 \varepsilon_1(\omega)$ in the $\omega \rightarrow 0$ limit. This is illustrated for BSCCO in Figure 3; at 100 K the material is in the normal state and the extrapolated value for $\rho_s = 0$, while below T_c extrapolated value is $\rho_s \sim 95 \times 10^6$ cm⁻², which may also be expressed as a penetration depth of about 0.16 microns.

Scaling laws

Scaling laws express a systematic and universal simplicity among complex systems in nature. For example, such laws are of enormous significance in biology, where the scaling relation between body mass and metabolic rate spans 21 orders of magnitude [8]! Scaling relations are equally important in the physical sciences. Since the discovery of the high-T_c superconductors, there has been considerable effort to find trends and correlations between the physical quantities as a clue to the origin of the superconductivity. One of the earliest patterns that emerged was the linear scaling of the superfluid density $\rho_s \propto 1/\lambda^2$ (where λ is the superconducting penetration depth) with T_c in the copper-oxygen planes

of the hole-doped copper-oxide superconductors. This is referred to as the Uemura relation [9] and it works reasonably well for the underdoped materials. However, it does not describe very underdoped, optimally doped, overdoped [10], or electron-doped materials [11]. A similar attempt to scale ρ_s with σ_{dc} was only partially successful [12].

We have recently demonstrated that the scaling relation $\rho_s \propto \sigma_{dc}T_c$ may be applied to a large number of high-temperature superconductors, regardless of doping level or type, nature of disorder, crystal structure, or direction (parallel or perpendicular to the copper-oxygen planes) [13]. The optical values of ρ_s (T \ll T_c) and σ_{dc} (T \cong T_c) within the copper-oxygen (*a-b*) planes have been determined for a large number of copper-oxide superconductors using the optical methods previously outlined and are shown as a log–log plot in Figure 4. Within error, all the data points fall on the line $\rho_s \cong 35 \sigma_{dc} T_c$ (in this instance, both sides of the equation possess the same units, so that the constant is dimensionless). In addition, the elemental BCS superconductors Nb and Pb (without any special regards to preparation) are also observed to follow this scaling relation reasonably well. The fact that the Nb and Pb follow this scaling relation is curious given the strong evidence that the superconductivity in the copper-oxide materials is thought to be of a fundamen-

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Figure 3: The frequency dependence of $-\omega^2 \varepsilon_1$ in $Bi_2 Sr_2 CaCu_2 O_8$ for light polarized along the *a* axis just above T_c in the normal state, and well below T_c . The strength of the condensate is taken as $\rho_s = -\omega^2 \varepsilon_1(\omega)$ in the $\omega \to 0$ limit. In the normal state, this extrapolates to zero, while in the superconducting state, $\rho_s \sim 95 \times 10^6$ cm⁻².



Figure 4: The log–log plot of the superfluid density ρ_s vs. $\sigma_{dc} T_c$ for the *a*-b planes of a variety of single and double-layer electron- and hole-doped high- T_c superconductors. Within error, all the points may be described by a single (dashed) line, $\rho_s \approx 35 \sigma_{dc} T_c$. The upper and lower dotted lines, $\rho_s \approx 44 \sigma_{dc} T_c$ and $\rho_s \approx 28 \sigma_{dc} T_c$ respectively, represent approximately the spread of the data. The points for Nb and Pb, indicated by the atomic symbols, also fall close to the dotted line.

tally different nature. In general, the BCS formalism can be considered for two limiting cases; the "clean-limit" case where there is little normal-state carrier scattering and essentially all of the normal-state carriers collapse into the superconducting state, and the "dirty-limit" case where the scattering rate is considerably larger and only some small fraction of the carriers collapses into the condensate. In the extreme dirty limit the scaling relation $\rho_s \cong 65 \sigma_{dc} T_c$ is recovered [14], which is similar to the empirical result obtained for the high-T_c superconductors. The fact that the points for Nb fall close to the scaling line indicates that they are in the dirty limit. This might further suggest that within a BCS framework that the high-T_c materials may also be thought of as being in the dirty limit.

The copper-oxide materials are highly anisotropic in transport, with highly conducting copper-oxygen (a-b) planes, while the c axis is poorly conducting with the transport often described as activated and governed by site-to-site hopping. However, the superconductivity is a bulk transition, and values for ρ_s and σ_{dc} may also be determined optically along the c axis (although the values are considerably smaller than those observed in the *a-b* planes). Because the transport is remarkably different in these two directions, it was therefore very surprising that the *ab*-plane and *c*-axis data are all described by the same scaling relation, as shown in Figure 5, which spans nearly five orders of magnitude. The activated nature of the c-axis transport precludes any description in terms of scattering rates. However, the superconductivity along the c axis is described very well by considering a series of coupled superconducting planes; this is referred to as the Josephson effect. Interestingly, the Josephson-coupling model yields the same scaling relation as the dirty limit; $\rho_s \approx 65 \sigma_{dc} T_c$ [15]. This indicates that there is some crossover from a Josephson coupling to dirty limit behavior, and provides a basis understanding why the *ab*-plane and *c*-axis data all follow the same general scaling relation. The difference in the numerical constant (i.e., $\cong 65$ and $\cong 35$) is most likely due to the simple way that the superconductivity has been modeled in these materials.

In summary, optical techniques are essential for determining the values of ρ_s and σ_{dc} in the high- T_c materials, and in some cases may only be determined by optical methods. A scaling relation $\rho_s \cong 35 \sigma_{dc}$ T_c has been observed in a wide variety of copper-oxide superconductors, regardless of the doping level or type, nature of the disorder, crystal structure, or direction (parallel or perpendicular to the copper-oxygen planes). While precisely what the scaling relation tells us is currently a source of considerable debate, this result should provide new insights into the origins of superconductivity in these materials [16].

Acknowledgements

It is a pleasure to acknowledge many useful discussions with Y. Ando, D. N. Basov, D. A. Bonn, I. Bozovic, A. V. Chubukov, M. Greven, W. N. Hardy, P. D. Johnson, S. A. Kivelson, P. A. Lee, M. Strongin, T.



Figure 5: The log–log plot of the superfluid density expressed as the square of a plasma frequency $\rho_s \equiv \omega_{ps}^2 vs. \sigma_{dc} T_c$ for the a-b planes and the c axis for a variety of cuprates. Within error, all of the points fall on the same universal (dashed) line defined by $\rho_s \cong 35 \sigma_{dc} T_c$; the dotted line is the dirty limit result $\rho_s \cong 65 \sigma_{dc} T_c$ for the BCS weak-coupling case, and also represents the Josephson result for the BCS weak-coupling case, used to describe the scaling along the c-axis. (The values for T_c , σ_{dc} and ρ_s for the a-b planes and the c-axis are listed in tables provided in the on-line supplemental information of Ref. 13.)

M. Rice, D. B. Tanner, T. Timusk, J. J. Tu, and T. Valla. Work at Brookhaven was supported by the DOE under contract number DE-AC02-98CH10886.

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