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Polarized angle resolved infrared spectroscopy of high temperature superconductors.

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Summary

Spectroscopic Probes

In order to investigate the properties of a material one must use an experimental probe, which will interact with the material. It is by examining the effect of this interaction that one is able to deduce or calculate a physical property of the material. There are two types of probe commonly used to investigate a material, transport and spectroscopic. A transport probe such as resistivity gives information about the density of electrons and the average time between electronic collisions as a function of temperature. A spectroscopic probe excites the material, and by varying the temperature one can investigate the superconducting and the normal state. Each different type of spectroscopic probe, probes a different aspect of the material, so that all of them together provide a complete picture. Some examples of spectroscopic probes are FIR (Far Infrared), Raman, Angle Resolved Photo Electron Spectroscopy (ARPES), Inelastic Neutron Scattering (INS), Muon Spin Resonance (μ SR) and Nuclear Magnetic Resonance (NMR).

A spectroscopic probe can produce an electronic excitation, a collective excitation or a nuclear excitation. One can either measure the incoming and outgoing beam or the excitation produced. In Nuclear Magnetic Resonance (NMR) one excites the oxygen and copper nuclei, which before relaxing interact with the valence electrons. Inelastic Neutron Scattering (INS) provides information about the magnetic excitations such as magnons. In Inelastic Neutron Scattering a beam of monochromatic neutrons impinges upon the material and one measures the scattered beam. Having measured the energy and momentum (E, k) of the incoming and outgoing neutrons one can calculate the dispersion relationship for the excitations. These may be phonons or magnons. In Photo Electron Spectroscopy (PES) the material is excited by a beam of photons but one measures the outgoing excited electrons. Here the exciting particle (photons) are not the same as the measured particle (electrons). Very often a model will be used to predict the outcome of an experiment. By comparing theory with experiment a better understanding of the material is obtained.

In Far Infrared Spectroscopy (FIR) we excite the material with photons (electromagnetic waves) of different energy and measure the rate of absorption. In this thesis we use reflectivity as a measure of the absorption. In order to obtain the absolute reflectivity, a perfectly reflecting mirror is used as a reference. Wherever the absolute reflectivity deviates from one, the material has absorbed

or transmitted photons at this frequency. The conservation of energy holds so that $R+T+A=1$, where R is the absolute reflectivity, T is the transmission and A is the absorption.

The normal approach to determining whether a material is a metal, insulator or a semiconductor is to examine the frequency dependence of the reflectivity. A metal has a high reflectivity upto the plasma frequency, a semiconductor has a small and an insulator has a large energy gap with clear phonons.

For simplicity one often uses the independent particle approximation for the theoretical description of solids, even though electrons interact both directly (via Coulomb interaction) and indirectly (via electron-phonon interaction) with each other. This approach breaks down when the interaction energy of the particles is larger than the kinetic energy. This situation occurs in strongly correlated electron systems (SCES) and the HTSC's are considered to fall into this category. Single electron band calculations predict the parent compound La_2CuO_4 to be a metal whereas it is an insulator. Not only are the HTSC's strongly correlated, but also anisotropic which means their optical and transport properties depend upon which optical axis is being excited or along which axis the transport takes place. The cuprates are therefore complex materials for which the current solid state theory is inadequate.

Far infrared spectroscopy is the probe used in this thesis to investigate the HTSC's. The measurements were performed over a temperature range of 4K to 300K in a 113v Bruker Fourier Transform Infrared (FTIR) spectrometer. This spectrometer allows the material to be probed over a range of frequencies in one or two seconds. Repeating this measurement improves the noise statistics of the spectrum. For weak signals or small samples several hundred scans are normally performed. FIR spectroscopy measures the dressed electronic excitations (usually called quasiparticles), both in-phase (real part of σ) and out of phase (imaginary part of σ) values of the conductivity are obtained. The energy range of the spectrometer was $10 - 8000 \text{ cm}^{-1}$. The superconducting energy gap for all HTSC's falls well within this range. If one uses the BCS weak coupling relationship $2\Delta/kT_c=3.5$, then for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ ($T_c \approx 32\text{K}$) the gap energy 2Δ is $\approx 82 \text{ cm}^{-1}$ (10 meV). For HgBaCaO ($T_c \approx 132\text{K}$) the gap is $\approx 340 \text{ cm}^{-1}$ (41 meV). Even if we assume the case of extremely strong coupling eg. $2\Delta/kT_c=7$, then the superconducting gap still falls within the measurement range of the spectrometer.

Symmetry of the Order Parameter

Near normal incidence, when the angle of incidence θ of the incoming beam is $\approx 5^\circ - 7^\circ$, FIR spectroscopy has a limitation when measuring the spectrum of materials which are highly reflecting i.e. $R \approx 1$. The resulting optical conductivity then depends upon the noise level in the reflectivity measurements. Certainly

5.5. Conclusion

if small changes in the noise level they will be reflected in the reflectivity is one then the analysis of such a measurement at normal incidence θ is now complicated by $1/\cos\theta$. If θ is small. Therefore small changes in the noise level. This novel technique is called PARIS (Polarized Anisotropic Reflectivity Infrared Spectroscopy). The shape of $\sigma_1(\omega)$ is a function of the order parameter. By measuring the order parameter, one can determine the symmetry of the order parameter upon the symmetry of the order parameter. Measurements of L (at 29K) show that no order parameter implies that the order parameter is a d-wave order parameter. The validity of the Ginzburg-Landau theory for the effective number of charge carriers in the overdoped case to the overdoped case. High accuracy measurements of the order parameter number of charge carriers.

The *c* Axis Electronic Excitations

The HTSC's are characterized by their anisotropic nature like *c*-axis conductivity. In two dimensional metals the incidence FIR measurement shows that the conductivity exceeds the Mott-Ioffe limit which normally denotes the Mott-Ioffe limit. Below T_c a situation arises where the collective excitation frequency is below T_c and it shows a large increase in conductivity. In the overdoped metallic, T_c decreases and the frequency doubles i.e.

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of the incoming beam is measuring the spectrum of resulting optical conductivity measurements. Certainly

if small changes induced during the phase transition are of the order of the noise level they will be lost. Considering a BCS superconductor, if $\sigma_1(\omega) = 0$ then the reflectivity is one. However, if due to noise the measured value is $R = 0.995$, then the analysis will produce an artificially large value of $\sigma_1(\omega)$. The error of such a measurement would make the interpretation of σ_1 dubious. If the angle of incidence θ is now increased, the absorption and the sensitivity will be increased by $1/\cos\theta$. If θ is increased to 80° the sensitivity is increased by a factor of 6. Therefore small superconductivity induced changes are amplified by a factor of 6. This novel technique is the basis of this thesis and is referred to by the acronym PARIS (Polarized Angle Resolved Infrared Spectroscopy).

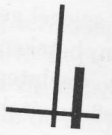
The shape of $\sigma_1(\omega)$ contains information about the symmetry of the order parameter. By modelling the conductivity based upon a s and a d-wave order parameter, one can compare the calculations with measurements and comment upon the symmetry of the order parameter for that particular material. PARIS measurements of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ ($T_c = 32\text{K}$) and for $\text{La}_{1.80}\text{Sr}_{0.20}\text{CuO}_4$ ($T_c = 29\text{K}$) show that no clear gap develops in the conductivity spectrum at 4K. This implies that the order parameter cannot be s-wave. Model calculations using a d-wave order parameter show a reasonable agreement. This does not affect the validity of the Ginzburg-Landau theory. It had long been assumed that the effective number of charges continued to increase in going from the optimally doped case to the overdoped situation. But it was never understood why T_c decreased. High accuracy measurements of $\text{La}_{1.80}\text{Sr}_{0.20}\text{CuO}_4$ show in fact that the effective number of charge carriers decreases and so tracks T_c (see chapter 3).

The c Axis Electrodynamics

The HTSC's are characterized by a metallic ab-plane conductivity and a insulator like c-axis conductivity. Within a simple picture they may be viewed as two dimensional metallic sheets separated by insulating material. Near normal incidence FIR measurements of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ (optimally doped crystal) shows that the conductivity in the normal state is $8 (\Omega \text{ cm})^{-1}$ at 40K which is below the Mott-Ioffe limit of $100 (\Omega \text{ cm})^{-1}$ for a metal. There is no zero crossing of ϵ_1 which normally denotes the plasma frequency. Furthermore the scattering rate exceeds the theoretically calculated plasma frequency which is a very unusual situation. Below T_c a plasma edge appears at 50 cm^{-1} . This is unusual in that it's frequency is below the BCS gap value of 82 cm^{-1} , therefore it is a sub-gap collective excitation. Measurements on the overdoped crystal $\text{La}_{1.80}\text{Sr}_{0.20}\text{CuO}_4$ show a large increase in the conductivity σ_1 at 40K but the value is still below the Mott-Ioffe limit of $100 (\Omega \text{ cm})^{-1}$. As the c-axis conductivity becomes more metallic, T_c decreases. For a change of 3K in the critical temperature, the plasma frequency doubles i.e. it becomes 100 cm^{-1} which is unexpected (see chapter 4).

Experimental Test of the Anderson Interlayer Tunnelling Model

P. W. Anderson and coworkers proposed that the frequency of the *c*-axis plasma edge was the Josephson plasma frequency ω_J . Furthermore that the critical temperature T_c was directly proportional to ω_J . This was a consequence of the Inter Layer Tunnelling (ILT) model. Due to the complicated coupling structure in a material with more than one copper-oxygen plane per unit cell, this model was applicable only to single layer compounds. The Josephson plasma frequency can be calculated, as it is based upon physical parameters. The calculated and measured value of ω_J for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ are in reasonable agreement with each other. The question remained as to whether it was valid for other single layer compounds. The concept had not been tested due to the lack of availability of crystals with sufficient *c*-axis dimensions to perform optical measurements. Crystals of $\text{Tl}_2\text{Ba}_2\text{CuO}_6$, $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ were available but only with *c*-axis dimensions of 50 - 100 μm . However as the plasma edge is a longitudinal excitation it may be detected using the PARIS technique. The assumption was tested and verified on a crystal of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ where the plasma edge was correctly detected. Reflectivity measurements on $\text{Tl}_2\text{Ba}_2\text{CuO}_6$, $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ did not show a plasma edge near the predicted value. In fact no plasma edge was measured within the measurement range. The ILT model prediction for ω_J appears to be incorrect (see chapter 5).



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