SUM RULES and ENERGY SCALES in BiSrCaCuO

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Abstract From very high accuracy reflectivity spectra, we have derived the optical conductivity and estimated the spectral weight up to various cut-off frequencies in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212). We show that, when evaluating the optical spectral weight over the full conduction band (1 eV), the kinetic energy decreases in the superconducting state, unlike in conventional BCS superconductors. As a consequence, the Ferrell-Glover-Tinkham sum rule is not satisfied up to this energy scale. This stands as a very unconventional behavior, contrasted with the overdoped Bi-2212 sample.

Keywords: High Tc superconductors, infrared conductivity, spectral weight

Introduction

A long lasting debate about the cuprate superconductors, stems from the differences and similarities with BCS behavior. The actual pairing mechanism, which eventually results in lowering the free energy of the system, is not yet known. In BCS superconductors, the free energy gain results from a competition of electronic kinetic energy increase and an eventually larger potential energy decrease. If the free energy in cuprates can be (somewhat arbitrarily) separated between potential and kinetic energy [1, 2], then the latter is measured by optics [3, 4]. The kinetic energy can be inferred from the partial sum rule or spectral weight W, defined as:

$$W(\Omega) = \int_0^{\Omega} \sigma_1(\omega, T) d\omega \qquad (1)$$

where $\sigma_1(\omega, T)$ is the frequency (ω) and temperature (T) dependent conductivity, and Ω is a cut-off frequency. Setting $\Omega = \Omega_B$, where Ω_B is the conduction band widht, one can get the kinetic energy E_k per copper site [4], through:

$$W(\Omega_B) = \frac{\pi}{2} \frac{e^2}{\hbar^2} \frac{a^2}{V_u} [-E_k] \qquad (2)$$

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where a is the lattice parameter, and V_u the volume per Cu site.

In the superconducting state, the integral in eq.1 or 2 includes the contribution of the superfluid, i.e. the weight of the $\delta(\omega)$ function centered at zero frequency. The Ferrell-Glover-Tinkham (FGT) sum rule [5, 6] requires that the spectral weight lost at finite frequency in the superconducting state must be retrieved in the spectral weight W_s of the δ function. In conventional superconductors, it was found to be fulfilled if integrating up to $\hbar\Omega_0 \sim 4\Delta$ (Δ is the superconducting gap). $\hbar\Omega_0$ is a characteristic energy of the boson spectrum responsible for the pairing mechanism. The FGT sum rule would then be exhausted for cuprates, if conventional, for $\hbar\Omega_0 \sim 0.1$ eV (assuming $\Delta \sim 25$ meV) [7, 8].

The studies of the FGT sum rule, first performed from c-axis (interlayer) optical conductivity data, showed a strong violation, interpreted as a change of interlayer kinetic energy [3, 9]. However, the amount of kinetic energy saving which was found is too small to account for the condensation energy. Although *in-plane* data appeared firstly to yield a conventional behavior [9], these early results were subsequently contradicted by ellipsometric and infrared data [10]. Our own infrared-visible reflectivity experiments, in Bi-2212, showed that in-plane spectral weight lost from the visible range is transferred into the δ function [11]. These two independent sets of data yielded a *decrease* of kinetic energy below T_c of the order of 1 meV.

The present paper implements our previous report [11] by using the *partial sum rule* in Eq.2. Although this is in principle equivalent to the FGT sum rule, we found this method to be more robust, because we can trace the entire temperature evolution of the spectral weight. We focus here on the underdoped thin film from the Bi-2212 family. We pin down the raw reflectivity data which allows to establish small changes at high energy (up to 10000 cm^{-1}) in the conductivity, hence in the spectral weight, thus illustrating why our unprecedented resolution is a necessary condition to trace this phenomenon. Our present, more elaborate analysis confirms that within error bars, the in-plane kinetic energy, calculated from Eq.2, decreases in the superconducting state. Using the partial sum rule in Eq.2 (and not only the FGT sum rule, as in [11]) shows qualitatively the opposite, conventional behavior in the case of the overdoped sample [12].

1. Experimental results

Reflectivity spectra, recorded in the range 30-25000 cm^{-1} for 15 temperatures between 300 K and 10 K, are reported elsewhere [13]. An example is recalled in Fig.1, for selected temperatures, up to 3000 cm^{-1}, for the underdoped sample (T_c=70 K). Most of the change with temperature occurs below 1000 cm^{-1}. However, the difference between the spectra at 10 K and 100 K extends up to 5000 cm^{-1}, displaying a reflectivity increase in the superconducting state of 0.3%, in this range, to be compared to the 4% increase below 1000 cm^{-1} (see inset of fig.1).

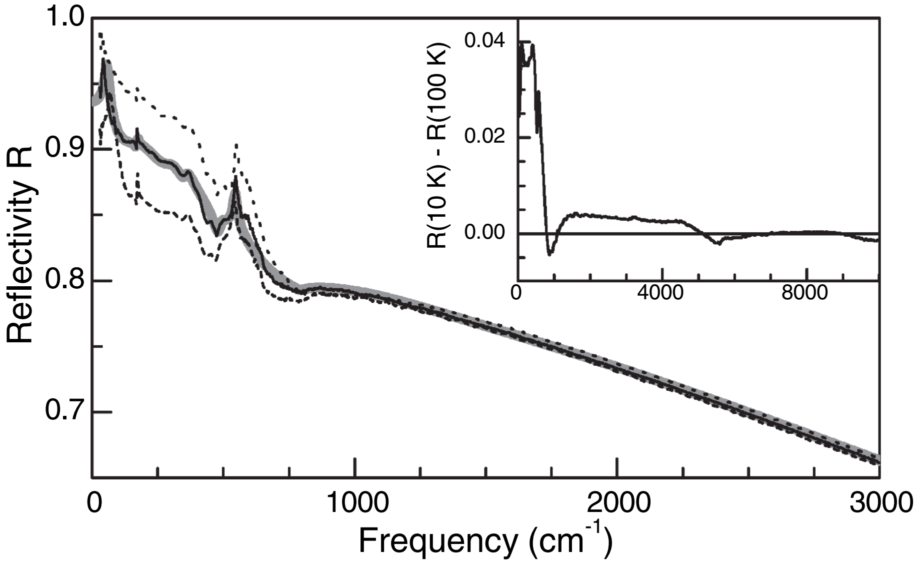

Figure 1. Reflectivity spectra in the 0,3000 cm^{-1} range at three temperatures: 200 K: dashed, 100 K: solid, 10 K: dotted line. The thick gray line is the fitted spectrum at 100 K. Inset: difference R(10 K)- R(100 K) between the reflectivity at 10 K and 100 K.

A standard thin film fitting procedure was applied in order to derive the optical conductivity [13, 14]. The fit is accurate enough so as to reproduce within less than 0.1 %, the small reflectivity difference between 100 and 10 K. This illustrates the accuracy of our data *and* analysis. A Kramers-Kronig based argument shows that such a small difference in reflectivity over a given frequency range $\Delta\omega$ results into changes in conductivity extending over a frequency range 2-3 $\Delta\omega$ [14]. The changes of spectral weights over an unusually large frequency range (possibly up to 16000 cm^{-1} as discussed further), arises from this general argument. Without the ability of resolving 0.1 % in the reflectivity, the results to be described further could not possibly be firmly established. The optical conductivity is shown in Fig.2, in the same frequency range and the same temperatures as in Fig.1.

2. Kinetic energy

From the optical conductivity, we compute the spectral weight or partial sum rule defined in Eq.1. We show in Fig3-a and -b the temperature variation

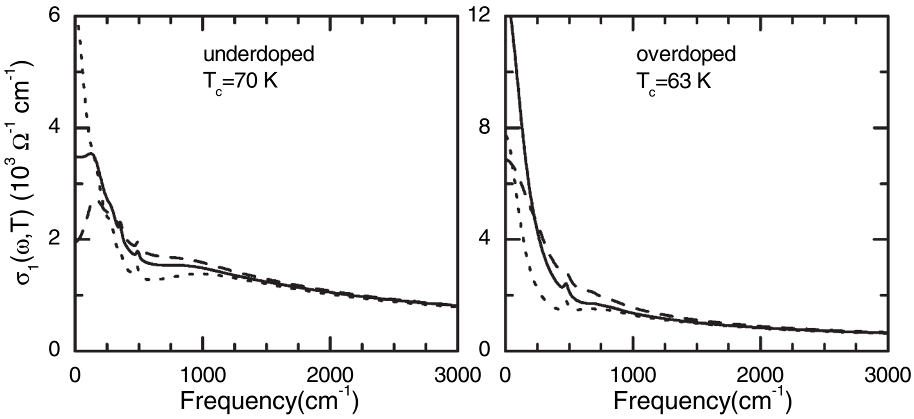

Figure 2. Real part $\sigma_1(\omega, T)$ of the optical conductivity, for the underdoped and overdoped (for comparison) samples. Temperatures are the same as in fig.1. Note that, in the former, a very large Drude-like contribution persists in the superconducting state.

of the spectral weight for selected cut-off frequencies Ω ranging from 500 to 8000 cm^{-1}, both for the underdoped sample (500, 1000, 8000 cm^{-1}), and for the overdoped sample (8000 cm^{-1}). For sake of clarity, we have normalized each spectral weight to its value at 300K. The integration starting from 0^+, in the superconducting state, does not include the spectral weight W_s of the δ function. We have added below T_c the weight of the superfluid, using the input parameter determined by the best fit [15]. We thus obtain, below T_c, the data represented by the open symbols in Fig.3, hereafter referred to as the *total* spectral weight. We have carefully worked out the accuracy of the data by using various sets of fitting parameters (mainly the superfluid weight and the Drude contribution) and by estimating the uncertainty on each experimental point accordingly. The size of symbols in Fig.3-b has been adjusted so as to represent the uncertainty.

We see in Fig.3-a that, when integrating up to 1000 cm^{-1}, the normal state spectral weight above T_c follows a T^2 increase, as indicated by a dotted line. The total spectral weight exceeds significantly this normal state spectral weight. Considering 1000 cm^{-1} as a conventional energy scale as discussed in the introduction, we are led to conclude that the energy scale over which one must extend the integration, in order to retrieve the spectral weight of the δ function, exceeds significantly any conventional scale. The excess in total spectral weight in the superconducting state, with respect to the normal state, is present throughout the conduction band, i.e. up to typically 1 eV=8000 cm^{-1} (fig.3-b). In this range, the spectral weight follows a T^2 behavior down to 130 K, then levels off. Between the normal (horizontal dotted line) and su-

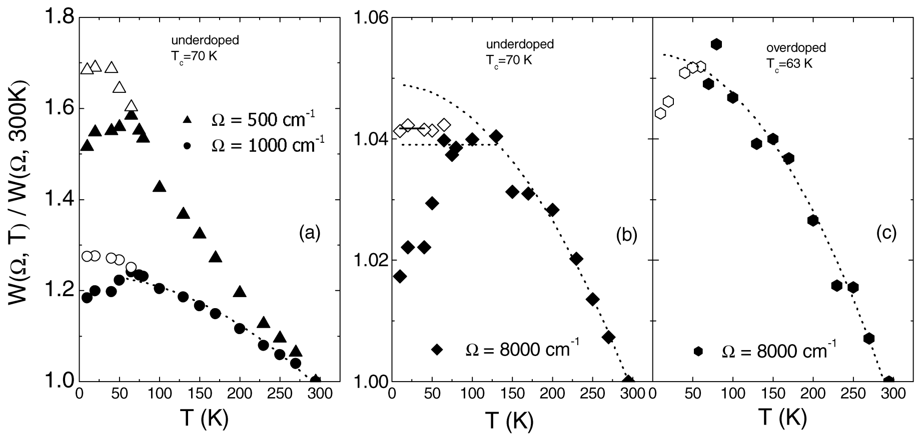

Figure 3. Partial sum rule shown for underdoped (a) and (b), and overdoped (c) samples, for selected cutoff frequencies. Full symbols represent the spectral weight, integrated from 0^+, hence without the superfluid contribution. Open symbols include (below T_c) the superfluid weight. Fig3-b and -c represent the intraband spectral weight, hence $-E_k$, as a function of temperature. The dotted lines are T^2 best fits to the normal state data.

perconducting state (horizontal solid line), we observe a small but definite increase. Equation 2 yields then the change of kinetic energy associated with this change of spectral weight. We find $\Delta E_k = -(0.5 \pm 0.3)$ meV per copper site, thus consistent with our early data [11] and confirming that the onset of superconductivity in the underdoped Bi-2212 sample is associated with a decrease of kinetic energy. In contrast, the overdoped sample displays a very clear decrease of spectral weight when performing the integration up to 8000 cm^{-1} (fig.3-c), i.e. an *increase* of kinetic energy. This is the conventional BCS behavior [17]. In order to make the connection with our earlier work, we now translate briefly these results in terms of the FGT sum rule. The FGT sum rule is fulfilled for the overdoped sample over a conventional range (1000 cm^{-1}) [11]. Conversely, it is *not satisfied* up to 8000 cm^{-1} in the underdoped sample. With our refined analysis, we can set a lower limit of 10000 cm^{-1} and an upper limit of 16000 cm^{-1} for the energy scale required to satisfy the FGT sum rule. Recent work also shows a much larger energy scale for the exhaustion of the FGT sum rule in underdoped YBCO, as compared to the optimal doping [18]. Such large energy scale is difficult to reconcile with a simple phonon mechanism, and seems therefore to call for a different excitation spectrum or more elaborate mechanisms.

Condensation by kinetic energy saving [19, 20] has been considered by several groups, sometimes with reasonable quantitative agreement [19]. Other models have been discussed in order to account for these data [21].

3. Summary

In conclusion, we have shown that according to the doping level in Bi-2212, the onset of superconductivity is associated with a decrease of kinetic energy in the underdoped material, whereas the overdoped material exhibits an increase of kinetic energy. Such a drastic difference confirms that the superconducting state in the underdoped regime of cuprates is unconventional.

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