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Energy Scales in the High-T_c Superconductor $YBa_2Cu_3O_{6+x}$

C. C. Homes,[1] S. V. Dordevic,[1] D. A. Bonn,[2] R. Liang,[2] and W. N. Hardy[2]

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The optical conductivity sum rule is used to examine the evolution of the spectral weight $N(\omega)$ in both the normal and superconducting states of optimally and underdoped $YBa_2Cu_3O_{6+x}$ along the a axis. Differences in $N(\omega)$ above and below T_c allow the strength of the superconducting condensate ρ_s to be determined. In the optimally-doped material, ρ_s is fully formed at energies comparable to the full superconducting gap maximum ($\simeq$0.1 eV), while in the underdoped material the energy scale for convergence is considerably higher ($\simeq$0.6 eV). This difference is discussed in terms of normal-state properties.

KEY WORDS: superconductivity; infrared; spectral weight; $YBa_2Cu_3O_{6+x}$.

Sum rules and conservation laws play an important role in physics. In spectroscopy, the conductivity sum rule is particularly useful and is an expression of the conservation of charge [1]. In BCS superconductors, below the critical temperature T_c some fraction of the carriers collapse into the δ-function at zero frequency, with a commensurate loss of spectral weight from low frequencies (below twice the superconducting energy gap). This missing spectral weight may be quantified by another sum rule as discussed by Ferrell, Glover, and Tinkham (the FGT sum rule) [2,3] to determine the strength of the superfluid ρ_s (or superfluid stiffness), where $\rho_s = c^2/\lambda_L^2$, and λ_L is the London penetration depth. The BCS theory holds that while the kinetic energy of the superconducting state is greater than that of the normal state [4], this increase is compensated by the reduction in potential energy which drives the transition [5]. However, it has been proposed that in certain hole–doped materials the superconductivity could arise from a lowering of the kinetic rather than the potential energy [6]. In such a case, the FGT sum rule might appear to be violated and ρ_s would be too small (λ_L would be too large). Similar models in the cuprate materials presume either strong coupling [7,8], or that the normal state is not a Fermi liquid and that superconductivity is driven either by the recovery of frustrated kinetic energy when pairs are formed [9,10], by lowering the in-plane zero-point kinetic energy [11], or by the condensation of preformed pairs [12].

Here we examine the evolution of the spectral weight along the a axis of $YBa_2Cu_3O_{6+x}$ for two different oxygen dopings; optimally doped ($x = 0.95$) with $T_c \simeq 91$ K, and underdoped ($x = 0.60$) with $T_c \simeq 57$ K. The presence of copper–oxygen chains along the b axis is a complicating factor in these materials. However, by examining the a-axis direction, it is thought that only the dynamics of the copper–oxygen planes are probed. Details of the crystal growth and the reflectance technique, from which the optical properties are calculated from a Kramers–Kronig analysis, have been discussed in previous report [13]. While the experimental error in the optical conductivity precludes definitive statements based on sum rules about possible kinetic energy changes (the accuracy of the sum rule is no better than 5%, while the maximum kinetic energy contribution is less than 0.3% of the total weight of the condensate in the optimally-doped material) (D. van der Marel, private communication), the spectral weight shows distinctly different behavior in the optimally doped and underdoped materials, establishing different energy scales for the superconductivity in these two systems.

Optical sum rules comprise a useful set of tools to study and characterize the lattice vibrations and

[1]Department of Physics, Brookhaven National Laboratory, Upton, New York 11973.
[2]Department of Physics and Astronomy, University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1.

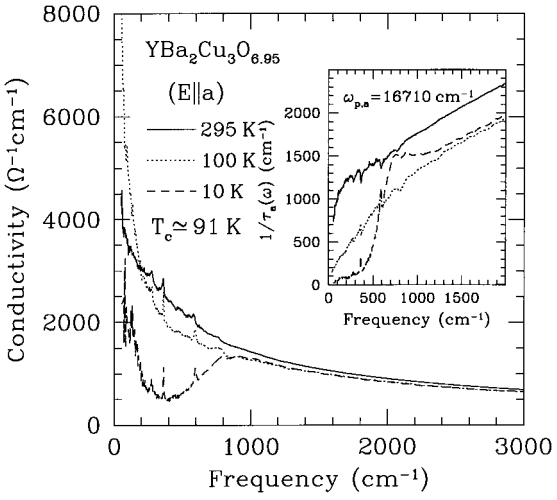

Fig. 1. The temperature dependence of the optical conductivity of $YBa_2Cu_3O_{6.95}$ for light polarized along the *a* axis. There is considerable narrowing of the Drude-like response as the temperature decreases in the normal state, followed by a loss of low-frequency spectral weight below T_c. Inset: The frequency-dependent scattering rate, which shows no evidence for a pseudogap in the normal state; $\omega_{p,a}$ is the estimated value of the plasma frequency used to scale $1/\tau_a(\omega)$.

electronic properties of solids [1]. The spectral weight may be estimated from a partial sum rule of the conductivity [14][3]

$$N(\omega_c) = \frac{120}{\pi}\int_0^{\omega_c} \sigma_1(\omega)\, d\omega \overset{\omega_c\to\infty}{\longrightarrow} \omega_p^2, \qquad (1)$$

where $\omega_p^2 = 4\pi ne^2/m$ is the classical plasma frequency, n is the carrier concentration, and m is the mass. In the absence of bound excitations, this expression is exact in the limit of $\omega_c \to \infty$, thus $N(\omega_c \to \infty) \equiv \omega_p^2$ and the spectral weight is $\propto n/m$. A variation of the conductivity sum rule is used to study the amount of spectral weight that collapses into the superconducting δ function at the origin for $T \ll T_c$.

This is illustrated by the optical conductivity for $YBa_2Cu_3O_{6.95}$ for light polarized along the *a* axis, shown in Fig. 1. The optical conductivity can be described simply by using a "two-component" picture, which consists of a series of Lorentzian oscillators including a Drude term at zero frequency and several bound excitations to describe the broad mid-infrared response [15]. The Drude-like response narrows considerably at low temperature. However, for $T \ll T_c$ there is a considerable loss of low frequency spectral weight. An alternative description is the "single-component" approach in which the scattering rate is assumed to have a frequency dependence [16]. The frequency-dependent scattering rate, shown in the inset, does not display a pseudogap [17] and may obey its own sum rule [18].

The missing spectral weight represents the strength of the condensate ω_{pS}^2 (expressed as a plasma frequency in units of cm^{-2}). This area may be estimated by the sum rule

$$\omega_{pS}^2 = \frac{120}{\pi}\int_{0+}^{\omega_c} [\sigma_{1,n}(\omega) - \sigma_{1,s}(\omega)]\, d\omega, \qquad (2)$$

where $\sigma_{1,n}(\omega) \equiv \sigma_1(\omega, T \gtrsim T_c)$, and $\sigma_{1,s}(\omega) \equiv \sigma_1(\omega, T \ll T_c)$. This is the FGT sum rule [2,3]. An alternative method for extracting the superfluid density relies on only the real part of the dielectric function (the imaginary part of the conductivity). A simple demonstration of this is to consider a simple Drude metal with a dielectric function $\tilde{\epsilon}(\omega) = \epsilon_\infty - \omega_p^2/p[\omega(\omega + i\Gamma)]$, where $\Gamma = 1/\tau$ is the scattering rate. If upon entering the superconducting state for $T \ll T_c$ it is assumed that all of the carriers collapse into the condensate, then $\omega_{ps} \equiv \omega_p$ and $\Gamma \to 0$, so that the dielectric function becomes $\tilde{\epsilon}(\omega) \to \epsilon_1(\omega) = \epsilon_\infty - \omega_{pS}^2/\omega^2$; in the limit of $\omega \to 0$, $\rho_s \propto \omega_{pS}^2 = -\omega^2\epsilon_1(\omega)$. It should be noted that this is a general result and the response to the formation of a δ function is not model dependent. The determination of ρ_s from $-\omega^2\epsilon_1(\omega)$ has two main advantages: (i) it relies only on the value of $\epsilon_1(\omega)$ for $T \ll T_c$ and thus probes just the superfluid response, and (ii) ρ_s is determined in a low-frequency limit, which removes the uncertainty of the high-frequency cut-off frequency ω_c in the FGT sum rule estimates of the condensate. We will distinguish between values of the condensate determined from $-\omega^2\epsilon_1(\omega)$ as ρ_s, and the FGT sum rule as ω_{pS}^2. The two techniques should in fact yield the same result, and it is indeed useful to compare the high-frequency estimates of ω_{pS}^2 with ρ_s.

The spectral weight in the normal $[N_n(\omega) \equiv N_n(\omega, T \gtrsim T_c)]$ and superconducting state $[N_s(\omega) \equiv N_s(\omega, T \ll T_c)]$, as well as difference (the FGT sum rule), is shown for $YBa_2Cu_3O_{6.95}$ in Fig. 2; $N_n(\omega)$ increases rapidly with frequency, but does not display any unusual structure. On the other hand, $N_s(\omega)$ evolves more slowly, and has several inflection points at low frequency which are thought to be related to the

[3]The term $120/\pi$ before the integral assumes that the units of conductivity are in $\Omega^{-1}cm^{-1}$, so that the integral yields cm^{-2}. The factor in front of the integral is sometimes expressed as $2m^*V_c/\pi e^2$, where V_c is the volume of the unit cell, in which case the integral yields the effective number of carriers.

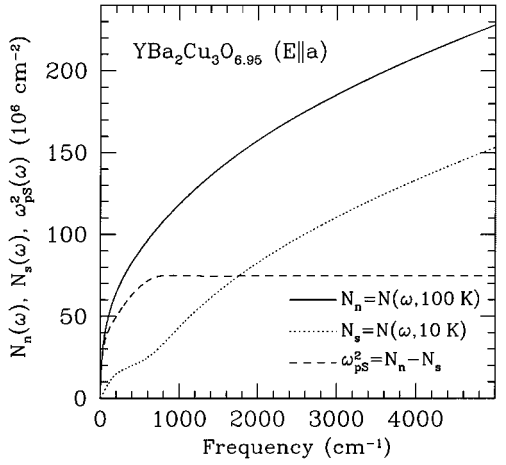

Fig. 2. The temperature dependence of the spectral weight of $YBa_2Cu_3O_{6.95}$ for light polarized along the a axis at $T \simeq T_c$ (solid line), and $T \ll T_c$ (dotted line). The difference between the two is ω_{pS}^2 (dashed line). Note that ω_{pS}^2 is essentially fully-formed by 800 cm^{-1}.

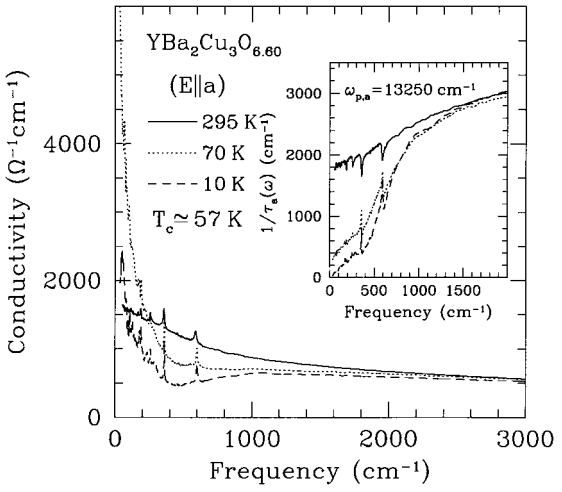

Fig. 3. The temperature dependence of the optical conductivity of $YBa_2Cu_3O_{6.60}$ for light polarized along the a axis. The normal-state conductivity changes dramatically with decreasing temperature, but the Drude-like response is quite narrow at $T \simeq T_c$. The loss of spectral weight for $T \ll T_c$ is not as pronounced at low frequency, and it is evident that more spectral weight has been shifted to higher frequencies. Inset: The frequency-dependent scattering rate, which shows a pseudogap in the normal state; $\omega_{p,a}$ is the estimated value of the plasma frequency used to scale $1/\tau_a(\omega)$.

spectral function [19]. The difference between the two curves $\omega_{pS}^2 = N_n(\omega) - N_s(\omega)$ is shown by the dashed line in Fig. 2. This quantity increases quickly and then saturates above $\approx$800 cm^{-1} to a constant value. This is of the order of twice the gap maximum, $2\Delta_0$, which is the relevant energy scale observed in a BCS system.

The optical conductivity for oxygen–underdoped $YBa_2Cu_3O_{6.60}$ for light polarized along the a axis, is shown in Fig. 3. While there is a dramatic change in the nature of the normal state with decreasing temperature, for $T \gtrsim T_c$ the Drude-like conductivity has narrowed considerably, and the scattering rate is in fact smaller in the underdoped sample ($\Gamma \simeq 100$ cm^{-1}) than in the optimally-doped case ($\Gamma \simeq 140$ cm^{-1}); it is always the case that $\Gamma < 2\Delta_0$. For $T \ll T_c$ the loss of low-frequency spectral weight is not as pronounced, and it is evident that spectral weight has been shifted to higher frequencies. The frequency-dependent scattering rate is shown in the inset, and clearly indicates the formation of a pseudogap in the frequency-dependent in-plane scattering rate. This has been interpreted as a change in the density of states near the Fermi surface at temperatures above T_c.

The development of the spectral weight above and below T_c of $YBa_2Cu_3O_{6.60}$ for light polarized along the a axis is shown in Fig. 4. While much of the overall behavior of $N_n(\omega)$ and $N_s(\omega)$ is similar to the optimally-doped material, ω_{pS}^2 does not converge to a value nearly as quickly. This point may be illustrated more clearly by plotting the normalized value of the spectral weight of the condensate for the optimally doped and underdoped materials, shown in Fig. 5. The values of $\rho_{s,a}$ have been obtained from an analysis of $-\omega^2\epsilon_1(\omega)$. For optimally doped $YBa_2Cu_3O_{6.95}$ the strength of the condensate, expressed in terms of a

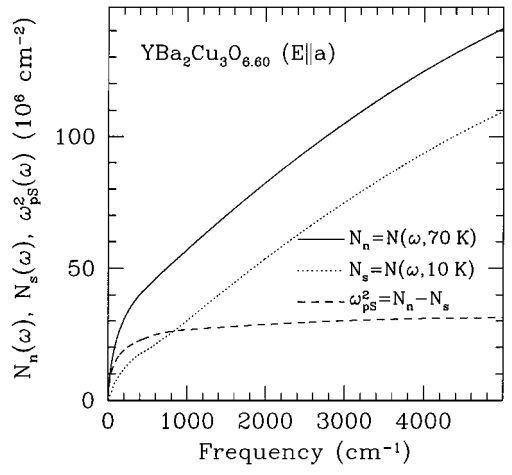

Fig. 4. The temperature dependence of the spectral weight of $YBa_2Cu_3O_{6.60}$ for light polarized along the a axis at $T \simeq T_c$ (solid line), and $T \ll T_c$ (dotted line). The difference between the two is ω_{pS}^2 (dashed line). The spectral weight is not recovered until $\simeq$5000 cm^{-1}, or about 0.6 eV.

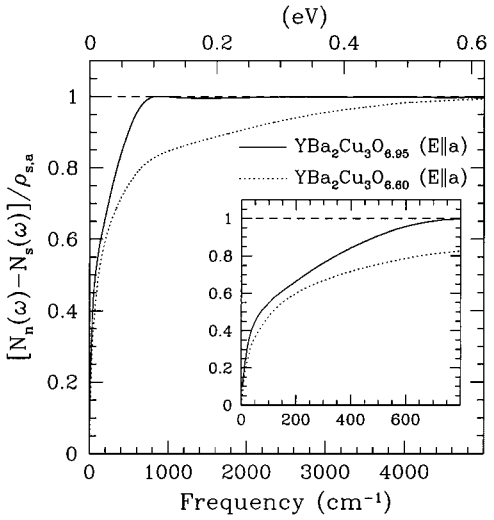

Fig. 5. The normalized weight of the condensate $[N_n(\omega) - N_s(\omega)]/\rho_{p,a}$ for optimally-doped $YBa_2Cu_3O_{6.95}$ (solid line) and underdoped $YBa_2Cu_3O_{6.60}$ (dotted line) along the a-axis direction. The curves describing the condensate have been normalized. The condensate for the optimally-doped material has saturated by $\simeq$800 cm^{-1}, while in the underdoped material the condensate is roughly 80% formed by this frequency, but the other 20% is not recovered until much higher frequencies. Inset: The low-frequency region.

plasma frequency is $\sqrt{\rho_{s,a}} = 8670\ cm^{-1}$, while for the underdoped material it is $\sqrt{\rho_{s,a}} = 5620\ cm^{-1}$, in good agreement with previous estimates [13].

The energy scale required to recover the full strength of the condensate $\rho_{s,a}$ in the optimally-doped material is $\omega_c \simeq 800\ cm^{-1}$ ($\simeq 2\Delta_0$), which is consistent with BCS model. However, in the underdoped materials the energy scale is much higher, $\omega_c \gtrsim 5000\ cm^{-1}$. This effect cannot be attributed to dirty limit effects as a result of increased scattering, as $\Gamma < 2\Delta_0$ in both materials. A similar effect has also been observed in the optical properties of optimally doped and underdoped $Bi_2Sr_2CaCuO_{8+\delta}$ [20]. The two very different types of behavior observed in the optimal and underdoped materials, in the normal and superconducting states, suggests that the behavior of the electronic correlations in the normal state determines the nature of the superconductivity and the energy scales associated with it in these materials [21].

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REFERENCES

1. D. Y. Smith, in *Handbook of Optical Constants of Solids*, E. D. Palik, ed. (Academic Press, New York, 1985), pp. 35–68.
2. R. A. Ferrell and R. E. Glover III, *Phys. Rev.* **109,** 1398 (1958).
3. M. Tinkham and R. A. Ferrell, *Phys. Rev. Lett.* **2,** 331 (1959).
4. M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1966).
5. S. Charkravarty, *Eur. Phys. J. B* **5,** 337 (1998).
6. J. Hirsch, *Physica C* **199,** 305 (1992).
7. M. R. Norman, M. Randeria, B. Jankó, and J. C. Campuzano, *Phys. Rev. B* **61,** 14742 (2000).
8. R. Haslinger and A. V. Chubukov, *Phys. Rev. B* **67,** 140504 (2003).
9. P. A. Lee, *Physica C* **317,** 194 (1999).
10. P. W. Anderson, *Physica C* **341–348,** 9 (2000).
11. V. J. Emery and S. A. Kivelson, *J. Phys. Chem. Solids* **61,** 467 (2000).
12. S. Alexandrov and N. F. Mott, *High Temperature Superconductors and Other Superfluids* (Taylor and Francis, London, 1994).
13. C. C. Homes, D. A. Bonn, R. Liang, W. N. Hardy, D. N. Basov, T. Timusk, and B. P. Clayman, *Phys. Rev. B* **60,** 9782 (1999).
14. D. B. Tanner, H. L. Liu, M. A. Quijada, A. M. Zibold, H. Berger, R. J. Kelley, M. Onellion, F. C. Chou, D. C. Johnston, J. P. Rice, *et al.*, *Physica B* **244,** 1 (1998).
15. D. B. Tanner and T. Timusk, in *Physical Properties of High Temperature Superconductors III*, D. M. Ginsberg, ed. (World Scientific, Singapore, 1992), p. 408.
16. A. V. Puchkov, D. N. Basov, and T. Timusk, *J. Phys: Cond. Matt.* **8,** 10049 (1996).
17. T. Timusk and B. Statt, Rep. *Prog. Phys.* **62,** 61 (1999).
18. D. N. Basov, E. J. Singley, and S. V. Dordevic, *Phys. Rev. B* **65,** 054516 (2002).
19. J. J. Tu, C. C. Homes, G. D. Gu, D. N. Basov, and M. Strongin, *Phys. Rev. B* **66,** 144514 (2002).
20. A. F. Santander-Syro, R. P. S. M. Lobo, N. Bontemps, Z. Konstantinovic, Z. Z. Li, and H. Raffy, *Europhys. Lett.* **62,** 568 (2003).
21. M. R. Norman and C. Pépin, *Phys. Rev. B* **66,** 100506 (2002).