PHYSICAL REVIEW B **66**, 100506(R) (2002)

Quasiparticle formation and optical sum rule violation in cuprate superconductors

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(Received 5 August 2002; published 12 September 2002)

Using a simple model for the frequency dependent scattering rate, we evaluate the in-plane optical integral for cuprate superconductors in the normal and superconducting states. In the overdoped region, this integral is conserved. In the optimal and underdoped region, though, the optical integrals differ, implying a lowering of the in-plane kinetic energy in the superconducting state. This sum rule violation, due to the difference of the non-Fermi-liquid normal state and the superconducting Fermi-liquid state, has a magnitude comparable to recent experimental results.

DOI: 10.1103/PhysRevB.66.100506 PACS number(s): 74.25.Gz, 74.72.−h

In superconductors, there is a dramatic change in the conductivity due to opening of an excitation gap in the finite frequency response, and the formation of a zero frequency δ function peak representing the dissipationless response of the condensate. The change is such as to preserve the optical sum rule, in that the finite frequency weight removed by the opening of the excitation gap is recovered by the condensate peak.[1]

In cuprate superconductors, though, there is experimental evidence that the sum rule is violated for *c*-axis conductivity.[2] Over the measured frequency range, more weight is present in the condensate peak than can be accounted for by the loss of finite frequency weight. Since the total optical sum rule must be preserved, the extra weight in the condensate peak is coming from outside this frequency range. This is unusual, since, in classic superconductors, the change in the optical integral is exhausted over a frequency range of order 4Δ, where Δ is the superconducting gap. Anderson[3] stressed that such sum rule violations are connected to the lack of quasiparticle poles in the normal state, and their emergence in the superconducting state. This is unlike the premise of BCS theory, where quasiparticles are assumed to exist in the normal state.

Although the sum rule violation for the *c*-axis response is profound, its contribution to the condensation energy is small due to the smallness of the *c*-axis kinetic energy in the cuprates. If kinetic energy effects are to play a role in the condensation energy, then they must be coming from the in-plane response, since the in-plane kinetic energy is quite large, of order an eV.[4] This largeness, though, means that the violation is difficult to see. That is, a 1 meV change in the kinetic energy would represent $\sim 1\%$ change in the optical integral. Recently, though, two groups claimed to observe such a change. Ellipsometry data on optimal and underdoped Bi2212 (Ref. 5) have been quantified as corresponding to a change in the kinetic energy of 1 meV. The same kinetic energy change has been inferred from reflectance data[6] on an underdoped Bi2212 film, though no such change could be resolved in an overdoped film. These results are intriguing, since a 1-meV kinetic energy savings per plane is in excess of the condensation energy inferred from specific heat data.[7]

In this paper, using a simple model for the frequency dependent scattering rate based on angle resolved photoemission (ARPES) and infrared data, we calculate the change in the optical integral from the normal to the superconducting state, and find its sign and magnitude to be comparable to these recent findings.

The full optical integral, integrating over all energy bands, is proportional to the bare carrier density over the bare electron mass, and thus must be conserved. Of greater interest here is the optical response of the band around the Fermi energy, correlating with the experimental data which are typically integrated out to an energy of order the plasma frequency (1 eV). This leads to a consideration of the single band sum rule[8]

$$\int_0^\infty Re\,\sigma_{xx}(\omega)d\omega=\frac{\pi e^2a^2}{2\hbar^2V}E_K, \tag{1}$$

where the restriction of σ to the single band response is implicit, and where a is the in-plane lattice constant, V the unit cell volume, and

$$E_K=\frac{2}{a^2N}\sum_k\frac{\partial^2\epsilon_k}{\partial k_x^2}n_k, \tag{2}$$

with N the number of k vectors; ϵ_k is the bare dispersion as defined by the effective single band Hamiltonian,[9] and n_k is the momentum distribution function. For a Hamiltonian with near neighbor hopping,[4] E_K is equivalent to minus the kinetic energy [$E_{kin}\equiv(2/N)\Sigma_k\epsilon_k n_k$], but in general these two quantities differ.

For free electrons, the inverse mass tensor is a constant in momentum, and thus this integral is conserved due to charge conservation. This is not generally the case, since the sum of the inverse mass tensor over the Brillouin zone vanishes.[10] When considering the change in this integral between different electronic states, the emphasis in the past has been on a possible change in the inverse mass tensor.[4,11] In general, though, we expect ϵ_k to be invariant, and therefore the change should instead be due to changes in n_k. A simple case is BCS theory,[1] where the kinetic energy increases in the superconducting state due to particle-hole mixing. If a near neighbor tight binding model is applied, the BCS opti-

cal integral would be smaller in the superconducting state than in the normal state, opposite to the recent experimental results.

The BCS model, though, assumes the existence of quasiparticle poles in the normal state. It is straightforward to demonstrate that the kinetic energy can indeed be lowered in the superconducting state if the normal state is a non-Fermi-liquid, and the superconducting state a Fermi liquid.[9] This occurs if the effect of quasiparticle formation on sharpening n_k is larger than the smearing due to particle-hole mixing. This effect is anisotropic in momentum, due to anisotropies in the scattering rate and the *d*-wave order parameter. Given these anisotropies and the anisotropy of the mass tensor, it is not obvious what the effect of the kinetic energy lowering will be on the optical integral, since a near neighbor tight binding model is inadequate to describe $\epsilon_{\mathbf{k}}$. In addition, ARPES measurements indicate a substantial doping dependence of the scattering rate, which implies that the sum rule violation will also be doping dependent.

We start by considering a simple model for the frequency dependent scattering rate, based on fits to ARPES data at the $(\pi,0)$ point.[12] This was used in work on the condensation energy[9] and the *c*-axis sum rule.[13] The model assumes a large frequency independent scattering rate in the normal state, consistent with the broad Lorentzian line shapes. In the superconducting state, the broad peak is replaced by a sharp peak at the superconducting gap energy, followed at higher binding energy by a spectral dip, then a broad maximum (the ''hump''). This change is modeled by cutting off $\mathrm{Im}\Sigma$ at the energy of the spectral dip. The resulting Σ is

$$\Sigma_\Gamma = \frac{\Gamma}{\pi}\ln\left|\frac{\omega-\omega_0}{\omega+\omega_0}\right| - i\Gamma\Theta(|\omega|-\omega_0), \tag{3}$$

where ω_0 is the spectral dip energy. This self-energy is then used in the spectral function[14]

$$A = \frac{1}{\pi}\mathrm{Im}\frac{Z\omega+\epsilon}{Z^2(\omega^2-\Delta^2)-\epsilon^2}, \tag{4}$$

where $Z=1-\Sigma/\omega$. For this form of Σ, the spectral function has two δ functions located at $\pm E$, where E satisfies the pole condition [denominator of Eq. (4) vanishes]. Such poles always exist for $E<\omega_0$ because of the log divergence of $\mathrm{Re}\Sigma$ at $\pm\omega_0$. The weight of the poles are determined as[15] $|dA^{-1}(\pm E)/d\omega|$. In addition, there are incoherent pieces for $|\omega|>\omega_0$.

For now, we assume Γ is k independent. ω_0 is also assumed to be k independent, as implied by ARPES experiments.[16] ϵ_k is taken from a six parameter tight binding fit to normal state ARPES data.[17] For the order parameter, the *d*-wave form $\cos(k_x a)-\cos(k_y a)$ is assumed. The k sum is done using a 100×100 grid in the irreducible quadrant of the zone. The quasiparticle pole weight contribution to n_k is analytic.[18] The incoherent contribution is evaluated by trapezoidal integration. We consider the $T=0$ limit, and thus $n_k=\int_{-\infty}^{0}A(\omega)d\omega$. In practice, the lower cutoff is taken to be -10 eV. In the normal state with no lower cutoff, $n_k=1/2-\tan^{-1}(\epsilon/\Gamma)/\pi$.

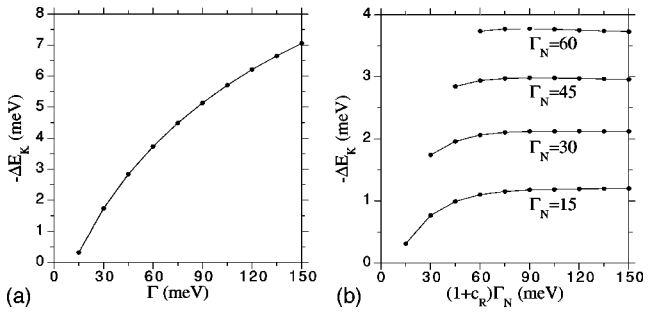

FIG. 1. $-\Delta E_K$ versus (a) isotropic scattering rate and (b) antinodal scattering rate (for various nodal scattering rates). $\omega_0=71$ meV and $\Delta_{max}=32$ meV.

In Fig. 1(a), $\Delta E_K \equiv E_K^N - E_K^S$ (where N denotes normal state and S superconducting state) is plotted as a function of Γ. The strong dependence on Γ is expected, since as Γ increases, the change in n_k ($\Delta n_k \equiv n_k^N - n_k^S$) becomes increasingly pronounced, leading to a larger sum rule violation. This implies that the sum rule violation becomes larger as the doping decreases, since Γ from ARPES measurements increases with underdoping.

One issue with Fig. 1(a) is the rather large value of the sum rule violation for realistic values of Γ (the antinodal scattering rate from ARPES is $\sim$100 meV for optimal doping). It is known, though, that the scattering rate from ARPES is a strong function of momentum.[19] We consider a simple model for the anisotropy with $\Gamma_k=\Gamma_N[1+c_R(\cos(k_x a)-\cos(k_y a))^2/4]$, where Γ_N is the nodal scattering rate and $\Gamma_N(1+c_R)$ the antinodal one. In Fig. 1(b), we plot ΔE_K versus c_R for various Γ_N, and find that it rapidly saturates with c_R, and thus with the antinodal scattering rate. We have also considered the influence of the anisotropic pseudogap on the normal state,[20] and found this had little effect on ΔE_K.

To gain further insight, we plot in Fig. 2(a) the integrand of ΔE_K as a function of momentum. Note that the overall integral is negative, with negative regions corresponding to unoccupied states near the *d*-wave node [$(0,0)-(\pi,\pi)$ Fermi crossing] and occupied states near the $(\pi/2,0)$ points, and positive regions to occupied states near the node and unoccupied states near the antinode [$(\pi,0)-(\pi,\pi)$ Fermi crossing]. To understand this, we plot two curves in Fig. 2(a), one the Fermi surface, the other the zero of the inverse mass tensor. In our model n_k is equal to 1/2 on the Fermi surface, and thus Δn_k changes sign there. Therefore, the optical integrand, which is the product of the inverse mass tensor times Δn_k, changes sign each time one of these two curves is crossed. From this, one can easily understand the various sign regions in the plot.

Perhaps more instructive is to convert the optical integral to the equivalent one involving $-\nabla n_k \cdot \nabla \epsilon_k$ (using Greens' theorem for periodic functions[10]). The resulting integrand is plotted in Fig. 2(b), and as expected, is localized about the Fermi surface. The important point is that the integrand peaks at the node. This can be easily understood. In the superconducting state there are quasiparticle poles, but at the node, $\Delta_k=0$, so there is a true step discontinuity in n_k there.

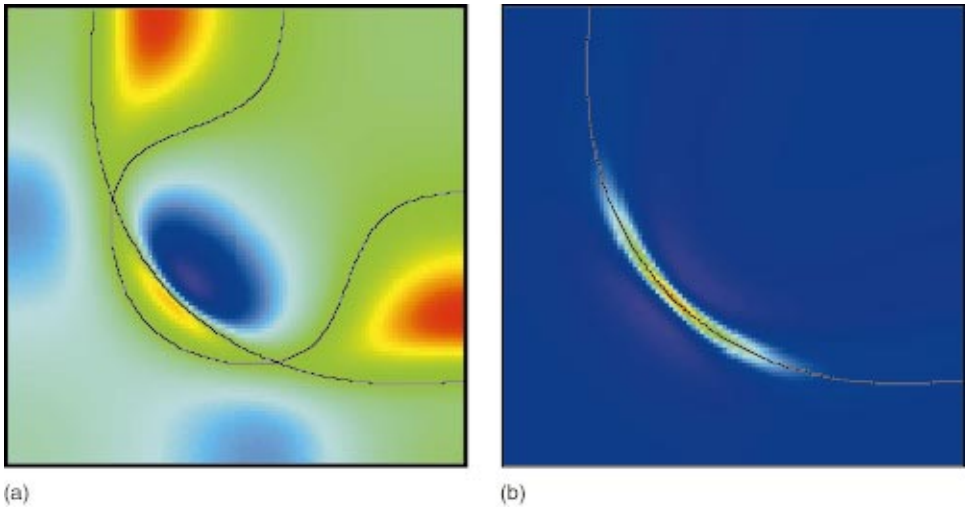

FIG. 2. (Color) (a) ΔE_K versus k (red is positive, green near zero, blue negative). The curves are the Fermi surface and zero of the inverse mass tensor. (b) $\Delta(\nabla n_k \cdot \nabla \epsilon_k)$ versus k (red is positive, blue near zero). $\Gamma = 150$ meV, $\omega_0 = 71$ meV, and $\Delta_{max} = 32$ meV.

As one moves away from the node, Δ_k increases from zero, and so $|\nabla n_k|$ decreases in magnitude. From Fig. 2(b), it is easy to appreciate the result of Fig. 1 that the optical integral is sensitive to the nodal scattering rate and not so sensitive to the antinodal one. As our model was motivated by fitting ARPES data in the antinodal region of the zone, this indicates that a model based directly on the nodal region should be considered.

Normal state ARPES data[19] are consistent with a scattering rate of the form $-\mathrm{Im}\Sigma = \Gamma_k + \alpha|\omega|$, where Γ_k has the anisotropy described above and α is momentum independent.[21] For simplicity, we will assume that both of these terms have an infrared cutoff at ω_0 as we did for the Γ model. When determining $\mathrm{Re}\Sigma$, it is important to provide an ultraviolet cut-off to $\mathrm{Im}\Sigma$. A hard cut-off at ω_c leads to a log singularity in $\mathrm{Re}\Sigma$ at ω_c. Rather, we take $\mathrm{Im}\Sigma$ to saturate at ω_c. This gives

$$\mathrm{Re}\Sigma_\alpha = \frac{\alpha}{\pi}\left(\omega \ln\left|\frac{\omega^2-\omega_0^2}{\omega^2-\omega_c^2}\right| + \omega_c \ln\left|\frac{\omega-\omega_c}{\omega+\omega_c}\right|\right), \qquad (5)$$

where $\Sigma = \Sigma_\alpha + \Sigma_\Gamma$. The normal state Σ is obtained by setting $\omega_0 = 0$. ω_0 is the energy of the dispersion kink along the zone diagonal, which is the same energy as the spectral dip at $(\pi,0)$,[16] and thus ω_0 is k independent. For ω_c, fits to ARPES are consistent with a value of 500 meV.[22]

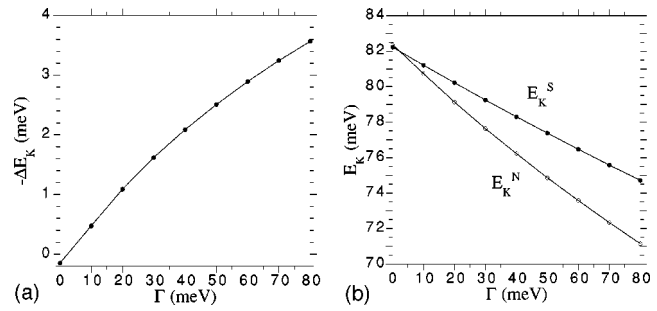

FIG. 3. (a) $-\Delta E_K$ and (b) E_K versus Γ. $\alpha = 0.75$, $\omega_0 = 71$ meV, $\Delta_{max} = 32$ meV, and $\omega_c = 500$ meV.

For illustrative purposes, we consider first the case with no anisotropy in Γ. In Fig. 3, we plot the sum rule violation versus Γ for a typical value of α. The variation with Γ is similar to Fig. 1(a) despite the presence of a substantial α term. Thus for a "pure" marginal Fermi liquid ($\Gamma = 0$), the sum rule violation is essentially zero. The reason is that with $\Gamma = 0$, the normal state posseses quite sharp spectral peaks, and thus the change in n_k when going into the superconducting state is reduced.

To make quantitative comparisons to experiment, realistic values of Γ_k and α as a function of doping are needed. We can obtain them from the optics data. In Fig. 4(a), we plot $1/\tau(\omega)$ for four Bi2212 samples in the superconducting state extracted from reflectivity data.[23] The linear high frequency behavior is of the form $a + b\omega$. Let us relate these parameters to Γ_k and α. The α term is easy to obtain, since it is k independent. At T=0, $1/\tau$ is an average of $-2\mathrm{Im}\Sigma$ over a frequency range of 0 to ω.[23] Since the α term is linear in ω, then $\alpha = b$.

The Γ_k term is a different story. If it were isotropic, then $\Gamma = a/2$. For the anisotropic case, these two quantities are

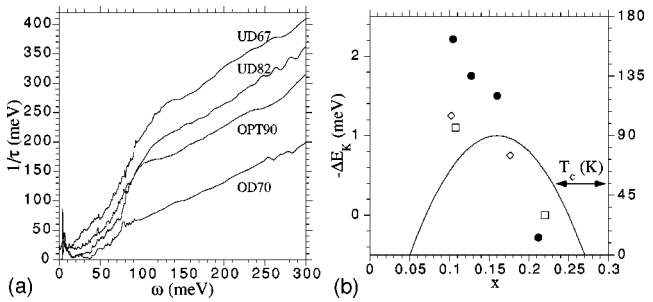

FIG. 4. (a) $1/\tau(\omega)$ versus ω for various Bi2212 samples from Ref. 23 (OD overdoped, OPT optimal doped, UD underdoped). (b) Calculated sum rule violation ($-\Delta E_K$) versus doping, x. The curve is T_c. The parameters (meV) extracted from (a) are Γ_N (1, 22, 27, 37), α (.65, .75, .88, .98), ω_0 (54, 71, 76, 83), and Δ_{max} (24, 32, 41, 54) for OD70, OPT90, UD82, and UD67, respectively. Also shown in (b) are the experimental results (open squares from Ref. 6, open diamonds from Ref. 5). The theoretical doping trend in (b) is due to the increasing offset in $1/\tau$ seen in (a).

related by a Fermi surface integral. We can do this analytically by replacing the anisotropy term by $\cos^2(2\phi)$, where ϕ is the Fermi surface angle (the node is at $\phi=\pi/4$). We find that the Fermi velocity along the Fermi surface can also be fit to the same anisotropic form. The resulting transport integral is[15] $2\tau(0)=[\int d\phi v(\phi)/\Gamma(\phi)]/\int d\phi v(\phi)$, where $v(\phi)=v_N[1+v_R\cos^2(2\phi)]$ (v is the modulus of the velocity) and $\Gamma(\phi)=\Gamma_N[1+c_R\cos^2(2\phi)]$. From ARPES[19] $c_R=3$, and from the tight binding fit, $v_R=-0.72$. Solving, we find that $\Gamma_N^{-1}=3.4\tau(0)$.

For the other parameters, we note that the deviation of $1/\tau(\omega)$ from linearity sets in at an energy $\Delta_{max}+\omega_0$, where ω_0 is the single particle scattering rate gap. This is easily shown from the Kubo bubble by dressing one of the two lines. From ARPES and tunneling, $\omega_0=\Delta_{max}+\omega_{res}$, where ω_{res} is the energy separation of the peak and the dip.[12,24,25] This is found to vary with doping as $5T_c$,[25] and we use this to extract Δ_{max} from the optics scattering rate gap. The resulting Δ_{max} values are consistent with ARPES (Ref. 24) and tunneling[25] measurements.

These values are used to determine the sum rule violation versus doping, shown in Fig. 4(b). We find no sum rule violation for the overdoped sample. This is the expected BCS like behavior, and is consistent with the experimental result on an overdoped film.[6] For the other samples, we find a sum rule violation which increases from 1.5 to 2.2 meV as the doping decreases. The doping trend is consistent with the reported experimental results [also shown in Fig. 4(b), although the values are perhaps too large by a factor of 2. This may be due to the approximation of using a hard infrared cutoff on the scattering rate in the superconducting state. Still, given the simplicity of our model, and the substantial experimental error bars, the agreement with experiment is surprisingly good. The doping trend in our model is due to the increase in Γ_N with underdoping. We also note that the kinetic energy change is about twice $-\Delta E_K$.

As for where the extra condensate weight is coming from, we note that the the experimental optical integrals balance at an energy cutoff of about 2 eV.[5,6] This value is comparable to the Mott gap of the insulator, so we speculate that the extra weight comes from the upper and lower Hubbard bands. This would be in accord with the more delocalized nature of the electrons in the superconducting state.

In conclusion, using a simple model for the frequency dependent scattering rate, we can understand recently reported results for the sum rule violation for the in-plane conductivity. The effect is due to the formation of quasiparticles in the superconducting state, and confirms earlier speculations by Anderson.[3] As the doping increases into the overdoped region, we find the sum rule violation goes away, consistent with the more Fermi-liquid-like nature of the normal state.

We acknowledge discussions with N. Bontemps, A. Santander-Syro, D. van der Marel, D. Basov, T. Timusk, and A. Georges. This work was supported by the U.S. Dept. of Energy, Office of Science, under Contract No. W-31-109-ENG-38, the Aspen Center for Physics, and the CNRS (France). We would like to thank Professor Bontemps and the ESPCI for their hospitality while this work was in progress, Professor Timusk for providing his data, and Professor. Bontemps and van der Marel for use of their unpublished work.

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