Elementary Excitations in the Metallic CuO_2 Planes of High-T_c Systems

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We determine from a two-band Hubbard Hamiltonian the spin susceptibility and the self-energy for the holes in the CuO_2 planes. From this, the quasiparticle lifetime τ, the spectral density, the electrical resistivity ρ resulting from spin scattering, and the NMR relaxation rate $1/T_1$ at Cu sites are calculated. Our results, which are expected to be valid for larger hole doping, are in reasonable agreement with experiment and indicate that an interplay of narrow-band phenomena and spin fluctuations leads to the observed anomalous normal-state properties. It is shown how to extend the theory to smaller hole doping when local magnetic moment behavior should be more pronounced.

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The normal-state properties of high-T_c superconductors show puzzling deviations from conventional Fermi-liquid behavior. The resistivity increases linearly with temperature and is 2 orders of magnitude larger than in conventional metals [1,2]. The quasiparticle lifetime, as extracted from photoemission data [3], appears to be proportional to ω^{-1}. Furthermore, the NMR spin-lattice relaxation rate $1/T_1$ is enhanced by an order of magnitude compared with Korringa behavior [4,5]. However, photoemission experiments [6] also indicate the existence of a well-defined Fermi surface, which may justify a Fermi-liquid-like description.

In this paper we develop a model for strongly correlated itinerant holes using the two-band Hubbard Hamiltonian and a slave-boson-type theory to determine various magnetic, spectroscopic, and transport data. In particular, we study elementary excitations of the spin system in the strongly correlated CuO_2 planes, by calculating the Cu spin susceptibility and the hole self-energy due to spin scattering. It is discussed how to extend our model to determine effects due to more pronounced local magnetic moments.

We start from the two-band Hubbard Hamiltonian for the Cu $3d_{x^2-y^2}$ and O $2p_{x,y}$ orbitals in the CuO_2 planes:

$$H=\sum_{j,\sigma}\epsilon_d^o d_{j\sigma}^\dagger d_{j\sigma}+\sum_{i,\sigma}\epsilon_p p_{i\sigma}^\dagger p_{i\sigma}+2t\sum_{k,\sigma}\gamma_k(p_{k\sigma}^\dagger d_{k\sigma}+\text{H.c.})+U\sum_j d_{j\uparrow}^\dagger d_{j\uparrow}d_{j\downarrow}^\dagger d_{j\downarrow}. \quad (1)$$

Here, $d_{l\sigma}^\dagger$ ($p_{l\sigma}^\dagger$) creates a hole at a Cu (O) site l with spin σ, and ϵ_d^o (ϵ_p) are the corresponding on-site energies. $d_{k\sigma}^\dagger$ and $p_{k\sigma}^\dagger$ are the Fourier-transformed operators. The coherence factor $\gamma_k=[\sin^2(k_x/2)+\sin^2(k_y/2)]^{1/2}$ describes nearest-neighbor Cu-O hopping processes for orthogonal oxygen orbitals, with k_x and k_y given in units of the Cu-Cu bond length. In the following we use the parameters $\Delta=\epsilon_p-\epsilon_d^o=2$ eV, $U=7.6$ eV, and $t=1.6$ eV which appear to be reasonable for high-T_c systems.

Using the spin-rotation-invariant form [7] of the slave-boson method proposed by Kotliar and Ruckenstein [8], we introduce boson operators e, s_o [$\mathbf{s}=(s_x,s_y,s_z)$], and δ which describe empty, singly occupied, and doubly occupied copper sites. In order to obtain the correct matrix elements of the resulting fermion-boson Hamiltonian one has to impose constraints on the bosonic and fermionic operators. The corresponding Lagrange multipliers $\lambda^{(1)}$, $\lambda_o^{(2)}$, and $\boldsymbol{\lambda}^{(2)}$ act as internal molecular fields and have a bosonic kinematic arising from a local gauge transformation [9]. In the saddle-point approximation, the bosons are supposed to be real numbers, which are obtained by a minimization of the free energy. This leads to an effective tight-binding Hamiltonian,

$$H_{\text{eff}}=\sum_{j,\sigma\sigma'}d_{j\sigma}^\dagger\{(\epsilon_d^o+\lambda_{oj}^{(2)})\tau_{o\sigma\sigma'}+\boldsymbol{\lambda}_j^{(2)}\boldsymbol{\tau}_{\sigma\sigma'}\}d_{j\sigma'}+\sum_{i,\sigma}\epsilon_p p_{i\sigma}^\dagger p_{i\sigma}+2t\sum_{kk',\sigma\sigma'}\gamma_k(p_{k\sigma}^\dagger z_{k-k',\sigma\sigma'}d_{k'\sigma'}+\text{H.c.}). \quad (2)$$

Here, τ_o and $\boldsymbol{\tau}$ are the unit matrix and the vector of the Pauli matrices, respectively. $z_{q,\sigma\sigma'}$ takes the bosonic hopping processes into account that accompany any fermionic hopping [7,8]. In the case of a translationally invariant paramagnetic system, which is expected to describe the high-T_c systems for larger doping, H_{eff} is a tight-binding Hamiltonian with a renormalized on-site energy ($\epsilon_d=\epsilon_d^o+\lambda_o^{(2)}$) and hopping element ($t\to zt$, where $z\le 1$), and $\boldsymbol{\lambda}_j^{(2)}=0$. On the other hand, for small hole doping the presence of pronounced local moments requires us to take into account the local exchange field $\boldsymbol{\lambda}_j^{(2)}$. This may be done approximately by assuming $\boldsymbol{\lambda}_j^{(2)}=\pm(0,0,\Lambda)$ and using an alloy-analogy description.

In order to study the dynamical properties of the highly doped system we consider Gaussian fluctuations around the saddle point of the translationally invariant Hamiltonian. This corresponds to the well-known $1/N$ expansion (Ref. [10] and references therein), where N is the spin degeneracy of a copper electron, and leads to a Dyson equation [$D^{-1}=(D^o)^{-1}-\Pi$] for the fluctuating bosons as shown in Fig. 1(a). This is a matrix equation

involving (11×11) matrices because of the large numbers of bosons occurring in the Hamiltonian. In particular the matrix element D_{s_z,s_z} leads to the copper spin susceptibility [11],

$$\chi_q^d(\omega)=4s_o^2 D_{s_z,s_z}(q,\omega)=\frac{\chi_{oq}^{dddd}(\omega)}{1+J_d\chi_{oq}^{dddd}(\omega)+\frac{1}{2}J_{pd}[\chi_{oq}^{pddd}(\omega)+\chi_{oq}^{ddpd}(\omega)]+J_{pd}^2\Gamma_q(\omega)}\,, \tag{3}$$

where the Lindhard susceptibilities $\chi_{oq}^{\alpha\beta\gamma\delta}(i\omega_n)$ are calculated from the hole Green's functions $G_{\alpha\beta}^o(k,i\omega_m)$ $(\alpha,\beta\in\{p,d\})$ of the saddle-point Hamiltonian. $\Gamma_q(\omega)$ is found to be small and will be neglected in the following. The spin fluctuation energies J_d and J_{pd} are given by

$$J_d=\frac{1}{2s_o^2}\left\{\lambda^{(1)}-\lambda_o^{(2)}+\frac{\partial\langle H_{\rm eff}\rangle}{\partial z}\frac{\partial^2 z}{\partial s_z^2}\bigg|_{\rm SP}\right\} \tag{4}$$

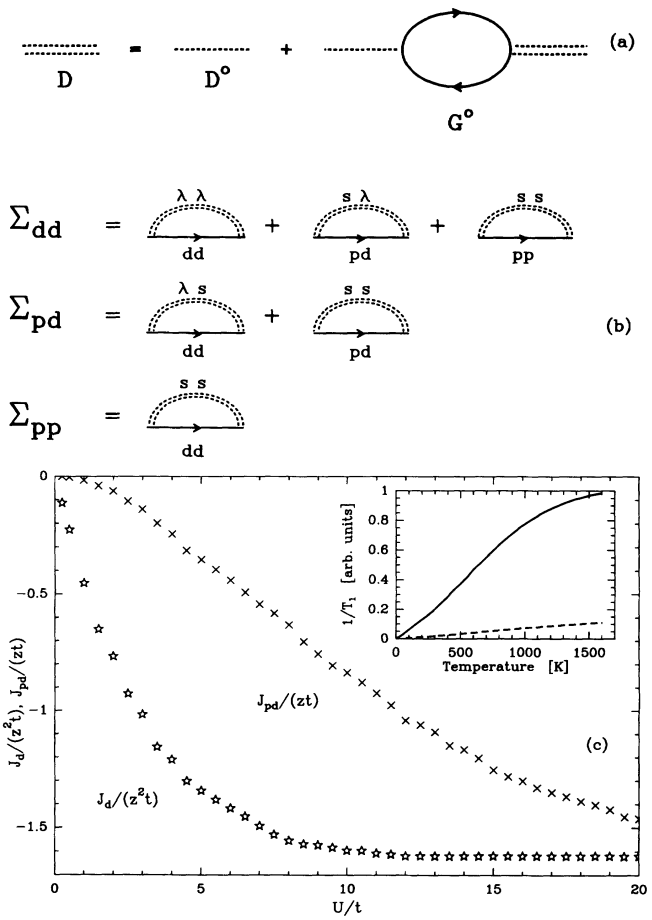

FIG. 1. (a) Dyson equation for the slave bosons described by the propagator $D_{\mu\nu}=-\langle T(\tau)a_\mu(q,\tau)a_\nu^\dagger(q,0)\rangle$, where $a_\mu,a_\nu\in\{\boldsymbol{\lambda}^{(2)},\mathbf{s}\}$, and (b) the contributions to the total self-energy Σ arising from the summation of all possible spin-scattering processes caused by $D_{\mu\nu}$. The Green's function G^o refers to the paramagnetic saddle-point Hamiltonian. (c) The U dependence of the effective spin fluctuation energies J_d and J_{pd} at the doping value $x=0.2$. z is the renormalization of t. Inset: The temperature dependence of the NMR relaxation rate $1/T_1$ at the Cu sites according to Eq. (3) in arbitrary units. For comparison we show the behavior of the noninteracting system (dashed line).

and

$$J_{pd}=-\frac{4t}{s_o}\frac{\partial z}{\partial s_z}\bigg|_{\rm SP}\propto(z^2-1)t\,, \tag{5}$$

where the derivatives are taken at the saddle point. In the limit $U/W<1$, where W is the bandwidth of the noninteracting system, we find $J_d\simeq -U$ and $J_{pd}=O((U/W)^2)$, see Fig. 1(c). Thus, we recover the random-phase approximation (RPA). For strong coupling $(U/W>1)$, J_d and J_{pd}, scaled by the corresponding band renormalizations, saturate and are of the same order of magnitude. J_d is obviously an effective, screened U, which has already been introduced phenomenologically by Bulut *et al.* [12] in their description of the magnetic properties of the high-T_c materials. In Eq. (3), the J_{pd} term leads essentially to a renormalization of J_d. Thus, J_{pd} represents a dressing of the Cu spin fluctuations by the neighboring oxygen holes and becomes important if the system tends to localize. Note, if one maps the Hamiltonian of Eq. (2) onto a one-band model by introducing only an effective nearest-neighbor hopping parameter $\tilde{t}$ $[\tilde{t}=t^2/(\epsilon_p-\epsilon_d)]$, then the Cu-O-Cu hopping processes where the holes return to the initial Cu site are not included, and J_{pd} is found to be small.

Furthermore, we calculate the hole self-energy arising from the spin excitations which are represented by the internal magnetic molecular field $\boldsymbol{\lambda}^{(2)}$ and the on-site magnetic moment $2s_o\mathbf{s}$. The total self-energy which results from these interactions with spin excitations is given by

$$\Sigma(k,\omega)=u_k^2\Sigma_{dd}(k,\omega)-2u_kv_k\Sigma_{pd}(k,\omega)+v_k^2\Sigma_{pp}(k,\omega)\,. \tag{6}$$

The various contributions to Σ are shown in Fig. 1(b) and are calculated from $\Sigma_{\alpha\beta;\mu\nu}=G_{\alpha\beta}D_{\mu\nu}$, with $\mu,\nu\in\{\boldsymbol{\lambda}^{(2)},\mathbf{s}\}$. The coherence factors u_k^2 and $v_k^2=1-u_k^2$ ensure that we consider the excitations at the Fermi surface in the lower (bonding) band. Furthermore we replace G by the Green's function G^o of the saddle-point Hamiltonian (Born approximation) and approximate D by using the local approximation [13]. For small U, where $J_{pd}\ll1$ and $J_d\approx -U$, only $\Sigma_{dd,\lambda\lambda}$ contributes to Σ and we recover the well-known RPA self-energy.

Results for the temperature and frequency dependence of the imaginary part of the self-energy for a doping concentration $x=0.2$ are given in Fig. 2 and the inset of Fig. 3, respectively. Below 300 K we find the canonical T^2 dependence and above 500 K a linear temperature dependence for $\mathrm{Im}\Sigma(\omega=0,T)$. A Drude analysis [2,14] of the

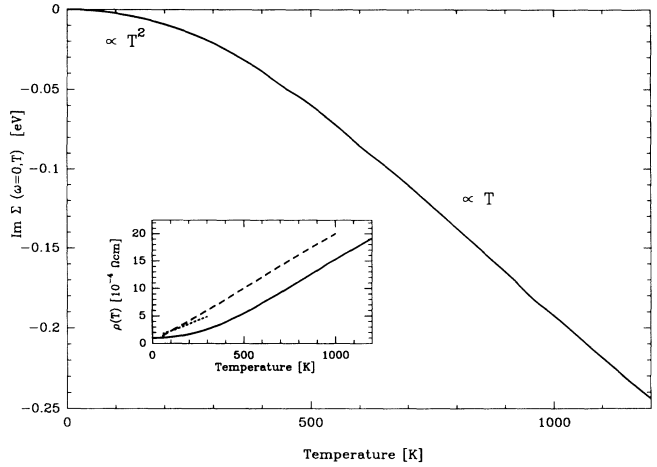

FIG. 2. Temperature dependence of the imaginary part of the total self-energy Σ at the excitation energy $\omega=0$ due to all spin-scattering diagrams shown in Fig. 1. Notice the change from $\mathrm{Im}\Sigma \propto T^2$ for smaller temperatures to $\mathrm{Im}\Sigma \propto T$ at larger T. Inset: A comparison of our estimated results for the electrical resistivity $\rho(T)$, using an extrapolated experimental residual resistivity, with experiments for $La_{2-x}Sr_xCuO_4$ by Gurvitch and Fiory [1] ($x=0.15$, dashed line) and by Suzuki [2] ($x=0.2$, dotted line). As the former experiments refer to a polycrystalline sample they may overestimate [1] the in-plane resistivity by a factor of 2.

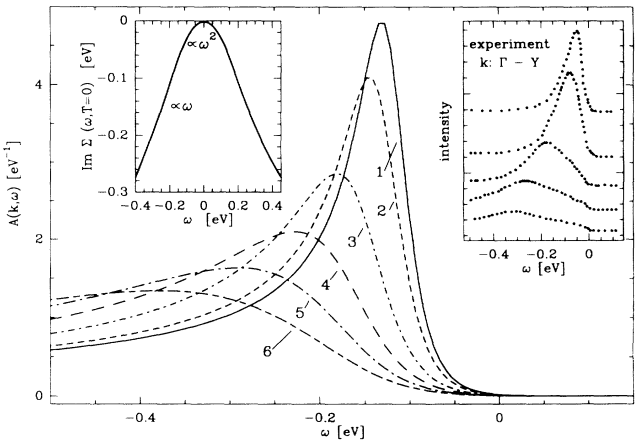

FIG. 3. Results for the spectral density $A(k,\omega)$. The curves 1-6 refer to $k_y=0.51$, 0.50, 0.47, 0.44, 0.42, and 0.39, respectively ($k_x=0$, throughout). For comparison photoemission results are shown in the right-hand inset. The left-hand inset shows the imaginary part of the low-temperature total self-energy Σ due to spin scattering as a function of the excitation energy ω. Notice that $\mathrm{Im}\Sigma \propto \omega^2$ for $\omega < 0.1$ eV, and $\mathrm{Im}\Sigma \propto \omega$ otherwise.

zero-frequency lifetime of the ac conductivity at 100 K of the La-Cu-O and Y-Ba-Cu-O compounds yields about 0.015–0.03 eV for τ^{-1}. This is in agreement with our results shown in the inset of Fig. 3 if we take $1/\tau = -\mathrm{Im}\Sigma$. Using the sample Drude relation $\rho=(m^*/ne^2)/\tau$ and $\omega_p=1$ eV for the plasma frequency [2] at $x=0.2$ we obtain the electrical resistivity of the holes. The results are shown in the inset of Fig. 2 and are compared with experiment. Note that we obtain in particular the change in the temperature dependence from $\rho \propto T^2$ to $\rho \propto T$ which was previously derived by Millis, Monien, and Pines [15], and Ihle, Kassner, and Plakida [16] from a model susceptibility and which has already been observed for $La_{1.8}Sr_{0.2}CuO_4$ [2] and $Tl_2Ba_2CuO_{6-y}$ [17] at $T^* \approx 180$ K. Moreover, we determine $d\rho/dT=1.95\ \mu\Omega$ cm/K which also agrees well with the experimental value [1,2] $(d\rho/dT)_{\mathrm{expt}}=1.77\mu\Omega$ cm/K.

It is also of interest to study the frequency dependence of the self-energy, because photoemission experiments [3] on the Bi-Sr-Ca-Cu-O compound demonstrate that the reciprocal quasiparticle lifetime varies linearly with ω. As can be seen from the inset of Fig. 3, we obtain a linear dependence of the inverse lifetime on the excitation energy for $\omega > 0.125$ eV and quadratic behavior for $\omega < 0.1$ eV which guarantees the existence of a Fermi surface. From the photoemission experiments it was deduced that $1/\tau \approx 0.6\omega$ which is comparable with our results ($1/\tau \approx 0.8\omega$).

In addition, we calculate the spectral density of the *electrons*,

$$A_k(\omega) = -\frac{1}{\pi}\frac{\Sigma''(-\omega)[1-f(-\omega)]}{[-\omega-\epsilon_k-\Sigma'(-\omega)]^2+\Sigma''(-\omega)^2}.$$

Here, $\Sigma(\omega)=\Sigma'(\omega)+i\Sigma''(\omega)$ is the self-energy due to spin *and* charge fluctuations averaged over the Brillouin zone (local approximation), $f(\omega)$ represents the Fermi distribution function, and $\epsilon(k)$ describes the dispersion of the bonding Cu-O band within the saddle-point approximation. Our results [18] for the angular-resolved spectral distribution $A(k,\omega)$ are shown in Fig. 3 and are in good agreement with experimental results [3]. This is an important clue for the correctness of our treatment of the electronic structure.

In the inset of Fig. 1(c) we show, for $x=0.2$, numerical results for the temperature dependence of the NMR relaxation rate at Cu sites,

$$T_1^{-1} = A_{dd}^2 T \lim_{\omega \to 0} \mathrm{Im}[\chi_{\mathrm{loc}}^d(\omega)/\omega],$$

obtained by using the local approximation [13] for the susceptibility and assuming an isotropic on-site hyperfine coupling constant A_{dd}. We find a linear increase of $1/T_1$ up to $T^*=650$ K and deviations from this behavior for higher temperatures. For $T=300$ K the relaxation rate is enhanced by a factor of 10 compared with the noninteracting case. The spin contribution to the Cu Knight shift $K_s^d \propto \chi_d(q \to 0, \omega=0)$, which is found to be temperature independent in our model, is enhanced by a factor of 2.3. An enhancement of these quantities was observed in NMR and nuclear quadrupole resonance experiments. Experimental data [19,20] for $Tl_2Ba_2CuO_{6-y}$ and overdoped $La_{2-x}Sr_xCuO_4$ ($x > 0.2$), which clearly *indicate*

that $(T_1T)^{-1}$=const for a wide temperature range above T_c, support our results for $1/T_1$ at larger hole doping.

The crossover temperatures T^* calculated for $x=0.2$ appear to be larger than the experimentally determined ones by a factor of 2. Note, if one includes the oxygen-oxygen hopping t_{pp} then the Van Hove singularity is shifted towards the Fermi energy causing a decrease of T^*.

The very low crossover temperatures observed for $x<0.2$ may result from the localized behavior of the copper spins which can be included in our model by assuming $\lambda_j^{(2)}=\pm(0,0,\Lambda)$ and using an alloy-analogy approximation. Within a single-site coherent potential approximation description of the fictitious alloy a pseudogap in the density of states occurs [21]. As a result of the temperature and doping dependence of the local exchange field Λ, the pseudogap decreases with increasing temperature and hole doping, causing an increase of the Knight shift with T and x and of $(T_1T)^{-1}$ with T, all observed experimentally. This behavior may have interesting consequences regarding the observation [22] of an inflection point in $\mathrm{Im}\chi(q,\omega)$ in recent neutron-scattering experiments which is interpreted as a consequence of a gap in the spin excitation spectrum. Such a behavior results mainly from the relation $\gamma_{k+Q_{af}}^2=2-\gamma_k^2$ which is related to the perfect nesting of the Fermi surface for $x=0$ and which is expected to be still fulfilled when local moments are present.

In conclusion, we derived from an electronic theory involving hybridized Cu-O states at the Fermi surface fulfilling the Luttinger theorem, the spin susceptibility $\chi^d(q,\omega)$, the electrical resistivity $\rho(T)$, and the self-energy $\Sigma(\omega,T)$. The theory presents a useful extension of interesting previous studies of Bulut *et al.* [12] and Lu and co-workers [23,24] and is expected to be valid for larger hole doping where local spin effects are not dominant. Otherwise a better treatment of the interplay of itinerant hole behavior and localized Cu spins is necessary by including Λ.

Via $\lambda_j^{(2)}=\pm(0,0,\Lambda)$ we obtain another temperature scale and a general theory permitting a uniform treatment of the T and x dependence of T_1, K, and the dynamical structure factor $S(q,\omega)\propto\mathrm{Im}\chi(q,\omega)$.

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