Theory of photoemission from copper oxide materials

Shiping Feng

CCAST (World Laboratory), P. O. Box 8730, Beijing 100080, China;
Department of Physics, Beijing Normal University, Beijing 100875, China;*
and National Laboratory of Superconductivity, Academia Sinica, Beijing 100080, China

Yun Song

CCAST (World Laboratory), P. O. Box 8730, Beijing 100080, China
and Department of Physics, Beijing Normal University, Beijing 100875, China

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A mean-field theory which satisfies the electron on-site local constraint in the relevant regime of density for the high-temperature superconductors is developed. Within this approach, the electron spectral function, the electron dispersion, and the electron density of states of copper oxide materials are discussed, and the results are qualitatively consistent with the numerical simulations. [S0163-1829(97)01001-1]

The discovery of the high-temperature superconductivity in copper oxide materials has initiated an enormous theoretical effort on quantum antiferromagnets in two-dimensions (2D).[1–3] Although experiments have not yet isolated the key elements of the electronic structure necessary for a global understanding of the physical properties of copper oxide materials, a significant body of reliable and reproducible data has been amassed by using many probes.[2,3] The normal-state properties of these materials exhibit a number of anomalous properties in the sense that they do not fit in the conventional Fermi-liquid theory.[4] When undoped, with one hole per copper site, these materials are antiferromagnetic Mott insulators.[2] When doped with sufficient carriers, they are superconductors with no antiferromagnetic long-range order.[2] There is strong evidence from transport and neutron scattering that no sharp phase transition occurs, and the strong correlations are very important to the electronic structure.[5] The angle-resolved photoemission spectroscopy is the definitive way to determine the electron dispersion relation for any materials. In copper oxide materials, the angle-resolved photoemission experiments[6] have produced interesting data that introduce important constraints on theories. It has been shown[2,3] that the hole-doped copper oxide materials exhibit universal properties likely induced by the behavior of carriers in their common copper oxide sheets. It is believed[1] that the physics of these materials may be effectively described by a 2D, large-U, single-band Hubbard model. In the large-U limit, the Hubbard model is transferred into the t-J model acting on the space with no doubly occupied sites. Furthermore, Zhang and Rice[1] derived the t-J model from a multiband Hubbard model described with 2D copper oxide planes.

It is clearly very important to establish appropriate formalism for the problem and to show that this leads to behavior similar to that seen in experiments. In order to account for real experiments under the t-J model, the crucial requirement is to impose the electron local constraint.[7] The local nature of the constraint is of prime importance, and its violation may lead to some unphysical results.[8] Recently a fermion-spin theory based on the charge-spin separation is proposed[9] to incorporate this constraint. In this approach, the

electron on-site local constraint for single occupancy is satisfied even in the mean-field approximation (MFA). In the framework of the fermion-spin theory, the ground-state properties, such as the ground-state energy, ground-state kinetic energy, phase separation, specific-heat data, and doping dependence of the antiferromagnetic long-range order, are discussed[9–11] and the results are in qualitative agreement with experiments and numerical simulations. In this paper, we develop a mean-field theory in the optimal doping regime within the fermion-spin theory to study the photoemission spectrum, the electron dispersion, and the electron density of states, which is useful for understanding of the electronic structure of the copper oxide materials.

We start from the t-J model which describes the electrons moving on a planar square lattice,

$$H = -t \sum_{\langle ij \rangle \sigma} C_{i\sigma}^{\dagger} C_{j\sigma} + \text{H.c.} - \mu \sum_{i\sigma} C_{i\sigma}^{\dagger} C_{i\sigma} + J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j,$$
(1)

where $C_{i\sigma}^{\dagger}(C_{i\sigma})$ are the electron creation (annihilation) operators, $\mathbf{S}_i = C_i^{\dagger} \boldsymbol{\sigma} C_i / 2$ are spin operators with $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ as Pauli matrices, and μ is the chemical potential. The summation $\langle ij \rangle$ is carried over nearest nonrepeated bonds. The t-J Hamiltonian (1) is supplemented by the on-site local constraint, $\Sigma_\sigma C_{i\sigma}^{\dagger} C_{i\sigma} \leq 1$, i.e., there be no doubly occupied sites. With the help of the fermion-spin transformation[9]

$$C_{i\uparrow} = h_i^{\dagger} S_i^{-}, \quad C_{i\downarrow} = h_i^{\dagger} S_i^{+},$$
(2)

where the spinless fermion operator h_i keeps track of the charge (holon), while the pseudospin operator S_i keeps track of the spin (spinon), the t-J model (1) can be rewritten as[9]

$$H = -t \sum_{\langle ij \rangle} h_i h_j^{\dagger} (S_i^{+} S_j^{-} + S_i^{-} S_j^{+}) + \text{H.c.} - \mu \sum_i h_i^{\dagger} h_i$$

$$+ J \sum_{\langle ij \rangle} (h_i h_i^{\dagger})(\mathbf{S}_i \cdot \mathbf{S}_j)(h_j h_j^{\dagger}),$$
(3)

where S_i^{+} and S_i^{-} are pseudospin raising and lowering operators, respectively. It is shown[9] that the constrained elec-

tron operator can be mapped exactly onto the fermion-spin transformation defined with an additional projection operator. However, this projection operator is cumbersome to handle for the actual calculation possible in 2D, we have dropped it in Eq. (3). It has been shown in Ref. 9 that such treatment leads to errors of the order δ in counting the number of spin states, which is negligible for small doping δ. Within the MFA, the t-J model (3) can be decoupled as

$$H = H_t + H_J - 8Nt\chi\phi, \tag{4a}$$

$$H_t = 2\chi t \sum_{i,\eta} h_{i+\eta}^\dagger h_i - \mu \sum_i h_i^\dagger h_i, \tag{4b}$$

$$H_J = \frac{1}{2}J_{\text{eff}}\epsilon \sum_{i,\eta}(S_i^+ S_{i+\eta}^- + S_i^- S_{i+\eta}^+) + J_{\text{eff}} \sum_{i,\eta} S_i^z S_{i+\eta}^z, \tag{4c}$$

with $\eta = \pm\hat{x}, \pm\hat{y}$, N is the number of sites, and $J_{\text{eff}} = J[(1-\delta)^2 - \phi^2]$. The nearest-neighbor spin bond-order amplitude χ and holon particle-hole parameter ϕ are defined as $\chi = \langle S_i^+ S_{i+\eta}^- \rangle$ and $\phi = \langle h_i^\dagger h_{i+\eta} \rangle$, respectively, where the site subscripts of the order parameters χ and ϕ have been dropped since the system is translation invariant. In this mean-field level, the spinon part is described by an anisotropic Heisenberg model with the anisotropic parameter given by

$$\epsilon = \frac{J_{\text{eff}} + 2t\phi}{J_{\text{eff}}}. \tag{5}$$

The quantum spin operators obey the Pauli spin algebra, and this problem can be discussed in terms of the two-time spin Green's function within the Tyablikov scheme.[12] In this case, the one-particle spinon and holon two-time Green's functions are defined as

$$D(i-j, t-t') = -i\theta(t-t')\langle[S_i^+(t), S_j^-(t')]\rangle$$
$$= \langle\langle S_i^+(t); S_j^-(t')\rangle\rangle, \tag{6a}$$

$$D_z(i-j, t-t') = -i\theta(t-t')\langle[S_i^z(t), S_j^z(t')]\rangle$$
$$= \langle\langle S_i^z(t); S_j^z(t')\rangle\rangle, \tag{6b}$$

and

$$g(i-j, \tau-\tau') = -i\theta(t-t')\langle[h_i(t), h_j^\dagger(t')]\rangle$$
$$= \langle\langle h_i(t); h_j^\dagger(t')\rangle\rangle, \tag{7}$$

respectively, where $\langle \dots \rangle$ is an average over the ensemble. Because the spinon system is an anisotropic, we have defined the two spinon Green's function $D(i-j, t-t')$ and $D_z(i-j, t-t')$ to describe the spinon propagations. The time-Fourier transform of the two-time Green's function satisfies the equation

$$\omega\langle\langle A; B\rangle\rangle = \frac{1}{2\pi}\langle[A, B]\rangle + \langle\langle[A, H]; B\rangle\rangle, \tag{8}$$

therefore the Green's functions can be obtained by applying the Tyablikov decoupling technique, and the correlation functions can be obtained by the spectral representations as

$$\langle B(t')A(t)\rangle$$
$$= i\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{\langle\langle A; B\rangle\rangle_{\omega+i0^+} - \langle\langle A; B\rangle\rangle_{\omega-i0^+}}{e^{\beta\omega} - 1} e^{-i\omega(t-t')}. \tag{9}$$

Recently, we[9-11] have employed the fermion-spin theory to study the ground-state properties of the 2D t-J model and obtained some interesting results. Within the random-phase approximation, we[11] have shown that the antiferromagnetic long-range order is destroyed by hole doping of the order $\sim 5\%$ for the reasonable value of the parameters $t/J = 5$. Then in the following discussions, we only study the systems in the optimal doping regime ($20\% > \delta > 5\%$), where there is no antiferromagnetic long-range order, i.e., $\langle S_i^z \rangle = 0$. In this case, the basic equations for the mean-field spinon two-time Green's function in one-dimension have been discussed in detail by Kondo and Yamaji.[13] Following their discussions, we can obtain the mean-field spinon Green's functions of H_J in 2D,

$$D(\mathbf{k}, \omega) = \frac{\Delta[(2\epsilon\chi_z + \chi)\gamma_k - (\epsilon\chi + 2\chi_z)]}{2\omega(k)}$$
$$\times \left(\frac{1}{\omega - \omega(k)} - \frac{1}{\omega + \omega(k)}\right), \tag{10a}$$

$$D_z(\mathbf{k}, \omega) = \frac{\Delta\epsilon\chi(\gamma_k - 1)}{2\omega_z(k)}\left(\frac{1}{\omega - \omega_z(k)} - \frac{1}{\omega + \omega_z(k)}\right), \tag{10b}$$

where $\gamma_{\mathbf{k}} = (1/Z)\sum_\eta e^{i\mathbf{k}\cdot\hat{\eta}}$, and

$$\omega^2(k) = \Delta^2 \left\{ \alpha\epsilon\left(\chi_z\gamma_k - \frac{1}{Z}\chi\right)(\epsilon\gamma_k - 1) \right.$$
$$\left. + \left[\alpha C_z + \frac{1}{4Z}(1-\alpha)\right](1 - \epsilon\gamma_k) \right\}$$
$$+ \Delta^2\left(\frac{1}{2}\alpha\epsilon\chi\gamma_k(\gamma_k - \epsilon)\right.$$
$$\left. + \frac{1}{2}\epsilon\left[\alpha C + \frac{1}{2Z}(1-\alpha)\right](\epsilon - \gamma_k)\right), \tag{11a}$$

$$\omega_z^2(k) = \Delta^2\left(\epsilon^2\left[\alpha C + \frac{1}{2Z}(1-\alpha)\right]\right.$$
$$\left. - \alpha\epsilon\chi\gamma_k - \frac{1}{Z}\alpha\epsilon\chi\right)(1 - \gamma_k), \tag{11b}$$

with $\Delta = 2ZJ_{\text{eff}}$, Z is the number of the nearest-neighbor sites, and the order parameters

$$\chi_z = \langle S_i^z S_{i+\eta}^z \rangle, \quad C = (1/Z^2)\sum_{\eta,\eta'}\langle S_{i+\eta}^+ S_{i+\eta'}^- \rangle,$$

and

$$C_z = (1/Z^2)\sum_{\eta,\eta'}\langle S_{i+\eta}^z S_{i+\eta'}^z \rangle.$$

In order not to violate the sum rule of the correlation function $\langle S_i^+ S_i^- \rangle = 1/2$ in the case $\langle S_i^z \rangle = 0$, the important decou-

pling parameter α has been introduced as discussed by Kondo and Yamaji,[13] which can be regarded as the vertex corrections. At half-filling, the t-J model is reduced as the isotropic antiferromagnetic Heisenberg model, where $\epsilon = 1$, $\chi_z = (1/2)\chi$, and $C_z = (1/2)C$ in the rotational symmetrical case, and we obtain $D_z(\mathbf{k},\omega) = (1/2)D(\mathbf{k},\omega)$, which is just those discussed by Shimahara and Takada.[14]

The mean-field Green's function of H_t for holons is very simple, and can be written as

$$g(\mathbf{k},\omega) = \frac{1}{\omega - (\varepsilon_k - \mu)}, \qquad (12)$$

where $\varepsilon_k = 2Z\chi t\gamma_k$. With help of the spinon and holon Green's functions and the spectral representations of the correlation functions, the order parameters $\chi, C, \chi_z, C_z, \phi$, and chemical potential μ can be obtained by the self-consistent equations,

$$\chi = \frac{1}{N}\sum_k \gamma_k \frac{\Delta[(2\epsilon\chi_z + \chi)\gamma_k - (\epsilon\chi + 2\chi_z)]}{2\omega(k)}\coth\left(\frac{\beta\omega(k)}{2}\right), \qquad (13a)$$

$$C = \frac{1}{N}\sum_k \gamma_k^2 \frac{\Delta[(2\epsilon\chi_z + \chi)\gamma_k - (\epsilon\chi + 2\chi_z)]}{2\omega(k)}\coth\left(\frac{\beta\omega(k)}{2}\right), \qquad (13b)$$

$$\frac{1}{2} = \frac{1}{N}\sum_k \frac{\Delta[(2\epsilon\chi_z + \chi)\gamma_k - (\epsilon\chi + 2\chi_z)]}{2\omega(k)}\coth\left(\frac{\beta\omega(k)}{2}\right), \qquad (13c)$$

$$\chi_z = \frac{1}{N}\sum_k \gamma_k \frac{\Delta\epsilon\chi(\gamma_k - 1)}{2\omega_z(k)}\coth\left(\frac{\beta\omega_z(k)}{2}\right), \qquad (13d)$$

$$C_z = \frac{1}{N}\sum_k \gamma_k^2 \frac{\Delta\epsilon\chi(\gamma_k - 1)}{2\omega_z(k)}\coth\left(\frac{\beta\omega_z(k)}{2}\right), \qquad (13e)$$

$$\phi = \frac{1}{2N}\sum_k \gamma_k\left(1 - \mathrm{th}\frac{\beta(\varepsilon_k - \mu)}{2}\right), \qquad (13f)$$

$$\delta = \frac{1}{2N}\sum_k \left(1 - \mathrm{th}\frac{\beta(\varepsilon_k - \mu)}{2}\right). \qquad (13g)$$

As we have shown in previous works,[11] the present MFA self-consistent calculation is just the usual self-consistent Hartree-Fock approximation.

We[15] have performed a numerical calculation for these mean-field self-consistent equations. The results for the order parameters at optimal doping regime are very close to our previous works[9,10] based on the 2D Jordan-Wigner approach, and detailed discussions will be given elsewhere.[15] In this paper, we hope to discuss the electronic structure of copper oxide materials, and therefore it is necessary to calculate the electron Green's function $G(i-j, t-t')$ $= \langle\langle C_{i\sigma}(t); C_{j\sigma}^\dagger(t')\rangle\rangle$. According the fermion-spin transformation (2), the electron Green's function is a convolution of the spinon Green's function $D(i-j, t-t')$ and holon Green's function $g(i-j, t-t')$, and can be obtained at the mean-field level as
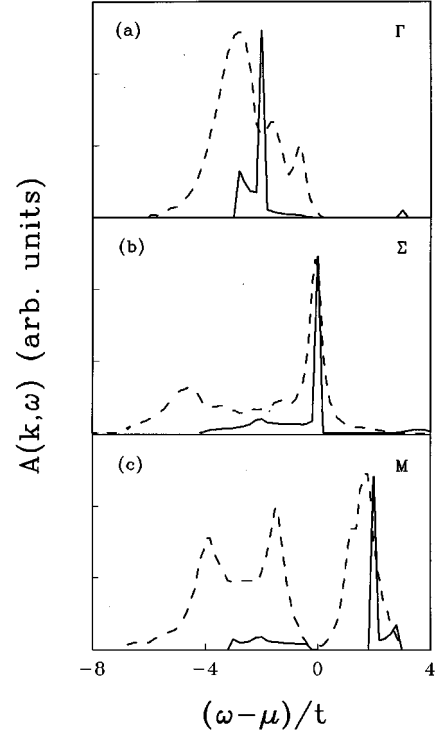

FIG. 1. Spectral function $A(\mathbf{k},\omega)$ of the 2D t-J model within the fermion-spin mean-field theory (solid line) and the exact diagonalization and quantum Monte Carlo methods (dashed line) for the parameter $t/J = 2.5$.

$$G(\mathbf{k},\omega) = \frac{1}{N}\sum_p \frac{\Delta[(2\epsilon\chi_z + \chi)\gamma_p - (\epsilon\chi + 2\chi_z)]}{2\omega(p)}$$
$$\times\left(\frac{F_1(\mathbf{k},\mathbf{p})}{\omega - \omega(p) + \varepsilon_{p-k}} + \frac{F_2(\mathbf{k},\mathbf{p})}{\omega + \omega(p) + \varepsilon_{p-k}}\right), \qquad (14)$$

where $F_1(\mathbf{k},\mathbf{p}) = n_B(\omega_p) + n_F(\varepsilon_{p-k} - \mu)$, $F_2(\mathbf{k},\mathbf{p}) = 1 + n_B(\omega_p) - n_F(\varepsilon_{p-k} - \mu)$, with $n_B(\omega_p)$ and $n_F(\varepsilon_{p-k} - \mu)$ are the boson and fermion distribution functions for spinons and holons, respectively. From the electron Green's function (14), we obtain the electron spectrum function,

$$A(\mathbf{k},\omega) = -2\,\mathrm{Im}\,G(\mathbf{k},\omega)$$
$$= 2\pi\frac{1}{N}\sum_p \frac{\Delta[(2\epsilon\chi_z + \chi)\gamma_p - (\epsilon\chi + 2\chi_z)]}{2\omega(p)}$$
$$\times (F_1(\mathbf{k},\mathbf{p})\delta[\omega - \omega(p) + \varepsilon_{p-k}]$$
$$+ F_2(\mathbf{k},\mathbf{p})\delta[\omega + \omega(p) + \varepsilon_{p-k}]). \qquad (15)$$

In the t-J model, the doubly occupied Hilbert space has been pushed to infinity as Hubbard $U \to \infty$ and therefore the spectrum function only describes the lower Hubbard band. Our mean-field result of the spectral functions at the doping $\delta = 0.12$ for the parameter $t/J = 2.5$ is shown in Fig. 1 (solid line). For comparison, the exact diagonalization and quantum Monte Carlo result at $\delta = 0.12$ for $t/J = 2.5$ obtained by Moreo et al.[16] is also shown in Fig. 1 (dashed line). Although the particular details of the spectral function and dispersion may differ from compound to compound, some qualitative features seem common to all copper oxide mate-
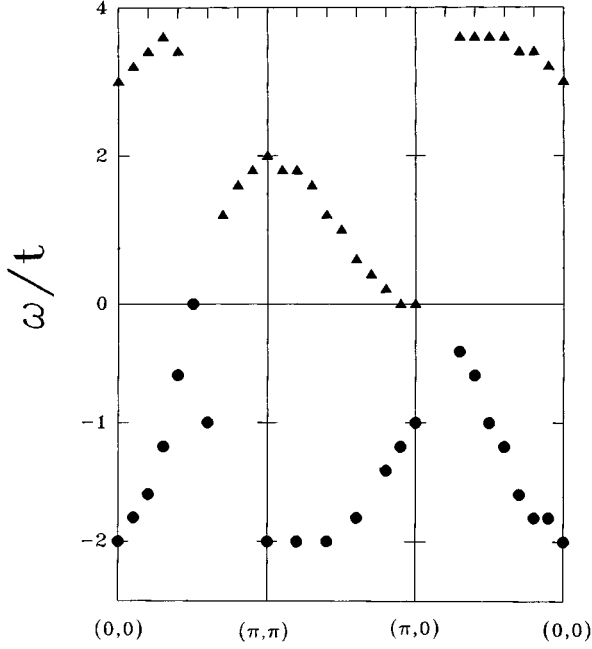
FIG. 2. Position of the dominant peaks in $A(\mathbf{k}, \omega)$ as a function of momentum.

FIG. 3. Electron spectral density of the 2D t-J model at the parameter $t/J = 2.5$ for (a) doping $\delta = 0.12$ and (b) $\delta = 0.06$.

rials. Hence a quantitative comparison between theory and experiment is still early, but the qualitative tendency of the spectral function and dispersion in an adequate theoretical description should be consistent with experiments and numerical simulations. In the present mean-field theory, the most important feature is that the intensity peaks are qualitatively consistent with the numerical simulation.[16] The low-energy peak is well defined at all momenta, and the positions of the dominant peaks in $A(\mathbf{k}, \omega)$ as a function of momentum are shown in Fig. 2, which is also in qualitative agreement with the numerical simulation[16] and the experimental result.[6]

Now we consider the electron density of states, which is defined as

$$\rho(\omega) = \frac{1}{N} \sum_k A(\mathbf{k}, \omega). \tag{16}$$

The numerical analysis of the electron density of states in the t-J model as a function of doping has been done by many authors.[17] On the other hand, oxygen x-ray-absorption spectra measured[18] on $La_{2-x}Sr_xCuO_4$ may be interpreted in terms of a picture in which hole doping introduces carriers into the lower hand. Our mean-field result at doping $\delta = 0.12$ and $\delta = 0.06$ for the parameter $t/J = 2.5$ is shown in Fig. 3. We find that the chemical potential μ moves from nearly zero at small doping $\delta = 0.06$ to the top edge of the lower Hubbard band, which also is qualitatively consistent with the numerical simulation[17] and experimental result.[18]

In summary, we have developed a mean-field theory which satisfies the electron on-site local constraint in the relevant regime of density for the high-temperature superconductors, namely in the vicinity of optimal doping within the fermion-spin theory. Within this mean-field theory, we have studied the electron spectral function, the electron dispersion, and the electron density of states of copper oxide
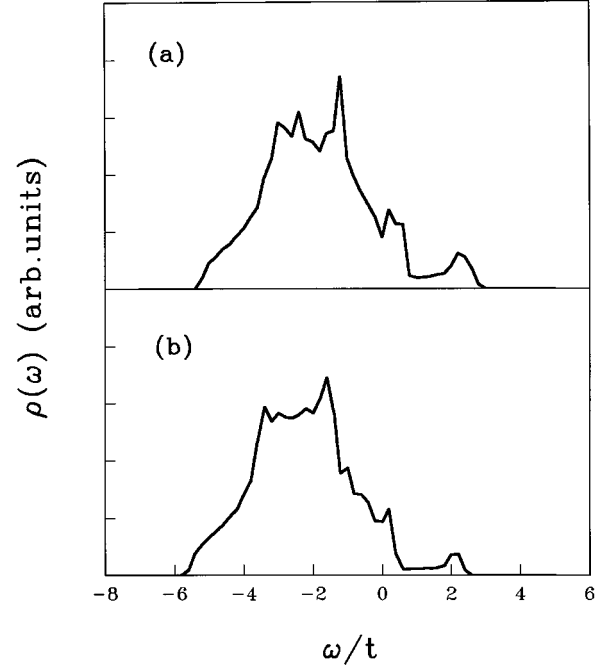
materials, the results are qualitatively consistent with the numerical simulations and experiments.

At zero doping, the t-J model reduces to the antiferromagnetic Heisenberg model, which has a local SU(2) symmetry in the fermion representation.[19] This symmetry does imply that the spinon particle (hole) state with spin up is the same state as a spinon hole (particle) with spin down. In the conventional slave-boson theory,[20] the SU(2) is broken to U(1) upon doping, and this U(1) gauge degree of freedom is introduced to incorporate with the single occupancy local constraint. However, in the fermion-spin theory, the SU(2) symmetry is also broken upon doping. Moreover, since the local constraint is satisfied exactly even in the MFA, the extra gauge degree of freedom occurring in the slave-particle approach does not appear here, which is consistent with the discussions in Ref. 21.

Finally we also note that recently Wen and Lee[22] have developed a slave-boson theory for the t-J model at underdoped regime which preserves SU(2) symmetry, they argued that spin-gap phase at the underdoped regime can be understood as the staggered flux phase. However, the area of the Fermi surface produced by this SU(2) mean-field theory is larger than that predicted by the Luttinger theorem which reveals a drawback of the SU(2) mean-field theory at optimal doping, and therefore the U(1) mean-field theory is better at the optimal doping.

*Mailing address.

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