I. INTRODUCTION.

Theoretical progress in the understanding of microscopic origin of high-temperature superconductivity appears to be ambiguous. The novelty and difficulty is to describe the strong effects of the electron-electron interactions which determine the strongly correlated phases of cuprates at various dopings: the Mott antiferromagnetic insulator, the “anomalous” metallic state and the superconductor. In fact, the fundamentally new microscopic origin of superconductivity (SC) in these materials is, perhaps, due to electron-electron interactions unlike standard phonon-mediated superconductivity [1,2]. We explore the general aspects of such a scenario with the help of sum rules for the density-density correlation function. The particular question of interest is the origin of the condensation energy, the difference in the energy between the “normal” state extrapolated to the superconducting ground state. The famous BCS theory of superconductivity [3] is based on the assumption that the attractive interaction between electrons arises from the lattice vibrations. Here we would like to investigate a general alternative to the BCS phonon-mediated superconductivity, namely, that the superconducting state (e.g. the condensation energy) is promoted either by the long-range part of electron-electron Coulomb interaction or by the short-range part of electron-electron and static electron-ion interactions. The identification of the part of the full electron-ion Hamiltonian responsible for the condensation energy would be an important step towards a complete and consistent theory of high-temperature superconductivity.

Many proposals for the condensation energy have been suggested [4], most of them (except Ref. [3]) being based on phenomenological Hamiltonians. Experimental confirmation of the origin of the condensation energy based on a phenomenological Hamiltonian would still require the validation of the phenomenological Hamiltonian (by various other experiments) in order to construct a self-consistent theory. Alternatively, the essential terms of the full [3] original electron-ion Hamiltonian (and changes of the expectation values thereof upon transitions) can be established consistently from experiments with the use of sum rules at the outset. Such an approach, taken in this paper, can potentially identify the origin of the condensation energy and the phenomenological Hamiltonian sufficiently to describe superconductivity and other strongly correlated phases.

If the ion kinetic and ion-ion Coulomb energies are assumed irrelevant (or in other words, these terms do not change upon the phase transitions and do not determine the important correlated phases), the full [3] electron-ion Hamiltonian can be reduced to the following form

\[ H = \sum_{p,\sigma} \frac{p^2}{2m} c_{p,\sigma}^{+} c_{p,\sigma} + \frac{1}{2\Omega} \sum_{q \neq 0} V_q [\hat{\rho}_q \hat{\rho}_{-q} - N] + \sum_{\kappa \neq 0} U_{-\kappa} \hat{\rho}_{\kappa}, \]  

where \( \hat{\rho}_q = \sum_{k,\sigma} \hat{c}_{k-q,\sigma}^{+} \hat{c}_{k,\sigma} \) is the total density operator. \( N \) is the number of electrons, and \( \Omega \) is a total volume. The first and second terms are the kinetic and Coulomb interaction energy of the electrons. The third term describes the interaction of the electrons with the periodic potential of the lattice, which can be represented by the Umklapp pseudopotential \( U_{-\kappa} \) with the sum over corresponding wavevectors \( \kappa \) of the reciprocal lattice [3]. The term of interaction between electrons and positive homogeneous ion background is omitted. In spite of making the “static lattice” assumption (the dynamic lattice effects (e.g. phonons) are neglected), the Hamiltonian [3] is quite general. For instance, the Hubbard model (and multi-band versions of it) is only a reduced version of the Hamiltonian [3], which neglects the long-range part of the Coulomb interaction. Presumably, the Hamiltonian [3] is sufficient not only to describe the Mott insulating state and the metallic state at high den-
ties (far away from the half-filled band) but also all other important phases of the cuprates.

We can analyse the electron Coulomb energy in situations of different dimensionality. In an isotropic medium the three-dimensional (3D) Coulomb potential is \( V_q = \frac{e^2}{\epsilon_0 \epsilon_\infty q} \), where \( \epsilon_\infty \) is the high-frequency dielectric constant due to the screening by the core electrons \([11,12]\). In a two-dimensional (2D) plane, the Coulomb potential is \( V_q = \frac{e^2}{2\epsilon_0 \epsilon_\infty} \). For a layered electron gas with interplane distance \( d \) (relevant for the discussion of single-layer cuprates) the Coulomb interaction is

\[
V_{q,q'} = \frac{e^2 d}{2\epsilon_0 \epsilon_\infty q} \sinh(qd) - \cos(qd)
\]

(2)

with different dependencies on the wavenumbers \( q \) parallel and \( q_z \) perpendicular to the planes \([13]\). The Coulomb potential of a layered gas becomes three-dimensional \( V_q = \frac{e^2}{\epsilon_0 \epsilon_\infty} \) in the long-wavelength limit \((q_d \ll 1 \text{ and } qd \ll 1)\) and two-dimensional, \( V_q = \frac{e^2}{2\epsilon_0 \epsilon_\infty} \), for short wavelengths \((qd \gg 1, \text{ independently of } q_z \) momentum).

II. SUM RULES.

The electron-electron Coulomb energy can be related to the density response function \( \chi(q,\omega) \), and thus it is instructive to analyse various sum rules for this response function \([14]\). In particular, the expectation value \(<V_c>\) of the Coulomb energy can be written in the form

\[
<V_c> = \frac{1}{2\Omega} \sum_q V_q <\hat{\rho}_q \hat{\rho}_{-q}> - N = \\
\sum_q <V_{c,q}> - \frac{1}{2} <V_q> \text{,}
\]

\[
<V_{c,q}> = \frac{1}{2} V_q \int \frac{h \omega}{2\pi} Im \chi(q,\omega) \coth \left( \frac{h \omega}{2kT} \right)
\]

(3)

where \( n = N/\Omega \) is the density of the electron system, and the density-density correlation function \( \chi(q,\omega) \) is defined in the standard way \([11,12]\).

\[
\chi(q,\omega) = \frac{i}{\hbar \Omega} \int_0^{+\infty} dt e^{i(\omega + i\delta)t} <[\hat{\rho}(q,t),\hat{\rho}^+(q,0)]> .
\]

(4)

\(<V_{c,q}>\) is the expectation value of the partial Coulomb energy corresponding to a particular value of momentum \( q \). The structure factor \( S_q \)

\[
S_q = \frac{1}{N} <\hat{\rho}_q \hat{\rho}_{-q}> - N \delta_{q,0},
\]

(5)

can be expressed again through the imaginary part \( Im \chi(q,\omega) \) using the fluctuation-dissipation theorem

\[
nS_q = \int_{-\infty}^{+\infty} Im \chi(q,\omega) \coth \left( \frac{h \omega}{2kT} \right) \frac{h \omega}{2\pi} \text{,}
\]

\[
<V_{c,q}> = \frac{1}{2} V_q nS_q.
\]

(6)

Various sum rules for the imaginary part of the susceptibility \( Im \chi(q,\omega) \) (valid for arbitrary dimensional system with appropriate form of Coulomb potential \( V_q \)) can be derived by calculating the commutators of the density operator and the Hamiltonian. To calculate the well-known \( f \)-sum rule (or the first moment sum rule) it is necessary to calculate the expectation value of the operator \([\hat{\rho}_q, \hat{H}, \hat{\rho}_{-q}]\). Another additional sum rule is the causality (Kramers-Kronig) relation. These two well-known sum rules are

\[
J_{-1} = \frac{2}{\pi} \int_0^{+\infty} \frac{Im \chi(q,\omega)}{\omega} d\omega = \chi(q,0),
\]

(7)

\[
J_1 = \frac{2}{\pi} \int_0^{+\infty} \omega Im \chi(q,\omega) d\omega = \frac{nq^2}{m},
\]

(8)

To calculate the \( \omega^3 \)-moment sum rule \([13]\) the expectation value of the operator \([[[\hat{\rho}_q, \hat{H}, \hat{H}], \hat{\rho}_{-q}]\) must be calculated. The result of a long calculation is

\[
J_3 = \frac{2}{\pi} \int_0^{+\infty} \omega^3 Im \chi(q,\omega) d\omega = \\
= \frac{1}{m^2} \frac{1}{\Omega} \sum_k (\vec{k}q)^4 (\omega - \epsilon_k) > + \\
+ q^4 \frac{n^2}{m^2} V_q q^4 \frac{3}{m^2} T_{pr} > + \frac{2n^2 h^2}{(2m)^3} + \\
+ \frac{n}{m^2} \Omega \sum_{p \neq \pm q} [V_{p+q}\vec{q} + q^2]^2 - V_p\vec{q}^2] S_p,
\]

(9)

where \( T_{pr} = \frac{1}{\Omega} \sum_p \frac{(\vec{k}p)^2}{2m} c_p^+ c_p = \int \frac{d^2 p}{2\pi^2} \frac{\vec{k}p^2}{2m} c_p^+ c_p \) is the projected kinetic energy operator, and \( \vec{q} = \vec{q}/|q| \) is a unit vector along the direction of \( \vec{q} \). This higher order sum rule (Eqn. \(5\)) is convenient for analysis of high frequency transitions in the density response, because it weights higher frequencies by a factor \( \omega^3 \). The existence (or convergence) of the third moment sum rule can be demonstrated by showing that all terms on the right-hand side of Eqn. \(5\) are finite. The first and the third terms are expected to be finite, because they are essentially related to the finite expectation values of the electron-lattice and the kinetic energy in the ground state. The convergence and \( q \)-dependence of the last term will be discussed in detail separately for 2D and 3D cases.

The three sum rules (Eqn. \(6\)) allow us to derive upper and lower bounds \([13]\) on the electron Coulomb energy, and hence to discuss the possible changes of Coulomb energy due to the phase transition. Since the imaginary part of the susceptibility \( Im \chi(q,\omega) \) is a real positive definite function, the two Cauchy-Schwartz inequalities can be written for the partial Coulomb energy \(<V_c> \) at \( T = 0 \) K.
\[ \frac{1}{2}(V_{q}^{2}J_{-1}J_{1})^{1/2} \geq < V_{c,q} > \geq \frac{1}{2}(V_{q}^{2}J_{3}^{3})^{1/2}. \] (10)

It is convenient to introduce the notional “plasma frequency” \( \omega_{p}(q) \) defined by \( \omega_{p}(q) = \left( \frac{q^{2}V_{q}^{2}}{m} \right)^{1/2} \). In fact, defined as above the “plasma frequency” has the right asymptotics for the corresponding plasma waves in the three-dimensional case \( (\omega_{3D}^{2}(q) = e^{2}n_{3D}q/(2\epsilon_{0}m)) \) and the two-dimensional case \( (\omega_{2D}^{2}(q) = e^{2}n_{2D}q/(2\epsilon_{0}m)) \).

Another relation, which is useful in order to rewrite the \( J_{-1} \) sum rule for \( q \to 0 \), can be derived in the long-wavelength limit \( (q \ll q_{TF}, \text{ where } q_{TF} \text{ is the inverse of Thomas-Fermi screening length}) \) for the full susceptibility \( \chi(q,0) \), if we express \( \chi(q,\omega) \) through the “bare” (or local) susceptibility \( \chi_{0}(q,\omega) \):

\[ \chi(q,\omega) = \frac{\chi_{0}(q,\omega)}{1 + V_{q}\chi_{0}(q,\omega)} \approx \frac{1}{V_{q}}. \] (11)

The discussion up to this point is valid for any dimensionality of the system (with the corresponding form of Coulomb potential \( V_{q} \)). In what follows we analyse the two-dimensional and three-dimensional cases separately and find important differences.

We consider first the 3D case. The leading terms at small \( q \) for the third moment sum rule are

\[ J_{3} \approx \frac{q^{2}}{m^{2}} < A > + q^{4} \frac{q^{2}}{m^{2}}V_{q}, \] (13)

where \( \hat{A} = \frac{1}{\pi} \sum_{\kappa}(\hat{q}\hat{\kappa})^{2}(-1)U_{-\kappa}\hat{\rho}_{\kappa} \). The third and fourth terms in Eqn. (13), being proportional to \( q^{4} \) and \( q^{6} \) powers, are subdominant. The last term in (13)

\[ \frac{1}{m^{2}} \int d^{D}p_{\hat{q}}V_{[\hat{p}+\hat{q}]^{2}[(\hat{p}\hat{q})^{2} + q^{2}]^{2} - V_{p}(\hat{p}\hat{q})^{2}]}n_{S_{p}} \] (14)

requires careful analysis of its convergence and \( q \)-dependence. The \( q \)-dependence (and convergence), due to the part of the integral over small momenta \( p \), is evident, since the upper bound on the pair-correlation function \( S_{p} \) (see Eqn. (13)) is \( S_{p} \leq Bq^{(D-1)/2} \) (where \( B \) is a constant, \( D \) is dimensionality) (17):

\[ \int d^{3}pV_{p}[(\hat{p}\hat{q})^{2}S_{p+q} - S_{p}] \sim \int d^{3}pV_{p}[(\hat{p}\hat{q})^{2}]S_{q} \] (15)

for small momenta \( p < q \). Since in 3D case the upper bound \( S_{q} \leq Bq^{2} \), the contribution from small momenta \( p \) integration is at least of order \( q^{4} \) (or higher power of \( q \) for small \( q \)). To analyse the \( q \)-dependence of the part of the integral (Eqn. (14)) from the integration over large momenta \( p \), we use the “cusp theorem” (18), which gives the asymptotic behaviour at large momenta \( p \) (3D case)

\[ S_{p} = 1 - \frac{C_{3D}}{p^{4}} + o\left( \frac{1}{p^{4}} \right), \] (16)

where \( C_{3D} \) is some constant. The integral of Eqn. (14) with \( S_{p} = 1 \) is identically zero (this is why \( S_{p} \) can be substituted by \( S_{p} - 1 \) if convenient). It is easy to see that the integral over \( p \) with the second term of expansion \( C_{3D}/p^{4} \) is convergent. By expanding in powers of \( (q/p) \) for large \( p \), the leading \( q \)-dependence of the considered term

\[ \int d^{3}pV_{p}[(\hat{p}\hat{q})^{2}]S_{p+q} - S_{p} \] (17)

is found to be proportional to \( q^{4} \). The terms, proportional to \( q^{3} \) and other odd powers of \( q \), are equal zero after the integration over angle \( \theta \) ( \( \cos \theta = (\hat{p}\hat{q})/pq \) is the angle between \( \hat{p} \) and \( \hat{q} \)). It can be seen that for large \( q \) the last term (Eqn. (14)) grows no faster than \( q^{3} \) as well. What is important for the ensuing discussion is that, both for small and large \( q \), the last term of Eqn. (14) has subleading \( q^{4} \) dependence on the wavevector \( q \).

The upper and lower limits on the partial Coulomb energy can be conveniently written in terms of the 3D “plasma frequency”

\[ \frac{\hbar}{2} \omega_{p,3D}(q) + o(q^{2}) \geq \geq < V_{c,q} >, \]

\[ \leq \frac{\hbar}{2} < q_{TF,3D} > + o(q^{2}). \] (18)

The inequalities (18) have an interesting significance for the possibility of gaining energy, in the SC condensation, from small-\( q \) modes. We first notice that in 3D case if \( < A > \geq 0 \) in both normal and superconducting states, which is certainly the case if there is no crystalline potential, the terms proportional to \( q^{4} \) (the third and last terms of the right-hand side of Eqn. (4)) determine the difference between the upper and lower bounds. Therefore if \( < A > = 0 \), then for given \( q \) the maximum possible saving is proportional to \( (\hbar/2)\omega_{p,3D}(q)/q_{0} \) (where \( q_{0} \approx q_{TF,3D} \)). \( q_{TF,3D} = \sqrt{\frac{e^{2}k_{F}m_{s}}{2\epsilon_{0}m_{s}}} \) in 3D, \( k_{F} \) is the Fermi wavevector, which is a negligible portion of the partial Coulomb energy \( \frac{\hbar}{2} \omega_{p,3D} \) at long wavelengths. In fact, in the absence of the Umklapp processes the sum rules (Eqn. (8) and (10)) essentially fix the density spectrum at long wavelengths \( q < q_{TF} \) to the plasma pole contribution: \( Im\chi(q,\omega) \sim \frac{\hbar}{2} \omega_{p,3D}(q) \delta(\omega - \omega_{p,3D}(q)) \), which satisfies completely all three sum rules at \( q \to 0 \) . Other
terms in the $J_3$ sum rule become comparable with the dominant term $q^4 V_q \frac{\omega_m}{m^2}$ only at $q \geq q_{TF,3D}$. Therefore, a large saving of Coulomb energy in 3D case in the absence of the lattice due to the phase transition is possible only for short wavelengths $q \geq q_{TF,3D}$.

In the presence of the periodic lattice potential ($\hat{A} \neq 0$), it can be seen from the inequalities [21] that for $q < q_{TF,3D}$ the maximum theoretical saving is a finite fraction of the zero-point plasma energy $\frac{\hbar}{2} \omega_{p,3D}(q)$. Therefore, substantial saving of the Coulomb energy in the long-wavelength limit is possible only in the presence of strong crystalline potential. In 3D (where the Coulomb interaction $V_q = e^2/\epsilon_0 q^2$), the Umklapp term $\frac{q^4}{m^2} \hat{A} < \hat{A}$ contributes in the same leading order of powers of $q$ into the $J_3$ sum rule as the Coulomb term $q^4 \frac{n^2}{m^2} V_q$ (see Eqn. [13]). The Umklapp term is then responsible for the finite width (or “lifetime”) of the plasmon peak.

In the 2D case, the leading terms are again

$$ J_3 = \frac{q^2}{m^2} < \hat{A} > + q^4 \frac{n^2}{m^2} V_q,_{2D}. \quad (19) $$

Other terms are subleading and proportional to $q^4$ and $q^6$ powers. The last term can be analysed similarly to 3D case and be shown proportional to $q^4$ at small $q$. Using the “cusp theorem” for 2D case [5],

$$ S_p = 1 - \frac{C_{2D}}{p^3} + o(\frac{1}{p^4}), \quad (20) $$

we can expand in powers of $q/p$ for large momenta $p$ and keep the leading term as a function of $q/p$:

$$ \int d^2 p V_p,_{2D}(\vec{p} \hat{q})^2 [S_{p+q} - S_p] \approx \int d^2 p V_p,_{2D}(\vec{p} \hat{q})^2 C_{2D} * \left[ \frac{1}{(1 + (q/p)^2 + 2(q/p)\cos \theta)^{1/2} - 1} \right]. \quad (21) $$

Due to the angle $\theta$ integration, terms proportional to $q^3$ and other odd powers vanish. For large $q$, the last term grows no faster than $q^3$ also (due to the first term in the bracket of Eqn. [24]). In the absence of Umklapp scattering ($\langle \hat{A} > = 0$), the density spectrum is given by the expression $Im \chi(q, \omega) \sim \frac{\hbar}{2} \omega_{p,2D}(q) \delta(\omega - \omega_{p,2D}(q))$, which satisfies all three sum rules at small $q$. The maximum possible saving of partial Coulomb energy at long wavelengths is of order $\frac{\hbar}{2} \omega_{p,2D}(q) q^{1/2}$ (where $q_{TF,2D} = \frac{e^2 m}{2\pi\epsilon_0 \hbar^2}$ is a 2D Thomas-Fermi screening wavevector).

The presence of the Umklapp term $q^2 < \hat{A}$ has a much more dramatic effect on the density spectrum in 2D, because this term, proportional to $q^2$, has a leading power of $q$ in the third moment sum rule at small $q$ dominating over the Coulomb term (unlike 3D case, where the Umklapp term has the same power-$q$ dependence as the Coulomb term). The density spectrum cannot be even approximated by a plasma pole expression, and the plasmon is never really a well-defined excitation in 2D in the presence of Umklapp scattering. It means that the Umklapp scattering modifies strongly (or “nonperturbatively”) the spectrum of the density fluctuations, and the spectrum is dominated by multi-pair and pair excitations rather than by plasmon. Of course, the mere existence of a large value of $< \hat{A} >$ is in itself perfectly compatible with a traditional textbook picture, in which the sum rules are satisfied by taking proper account of interband transitions; in such a case there is no a priori reason why a plasmon associated with the excitations of the conduction band must automatically be ill-defined. However, in a system where Umklapp (quasi-momentum-nonconserving) interactions between the Bloch quasiparticles are strong it seems natural that these alone could give rise to a substantial value of $Im \chi$ even below the first band gap; particularly in view of the above remark about the enhanced effect in 2D, it is tempting to view the so-called midinfrared peak in the cuprates in this light.

The upper bound on the partial Coulomb energy is still the half of the plasmon energy ($\omega_{p,2D}(q) \sim \sqrt{\pi}$), but the lower bound at small $q$ if $< \hat{A} > \neq 0$ is essentially given by

$$ < V_{c,q} > \geq \frac{\hbar}{2} \omega_{p,2D}(q) \left( \frac{nm}{< \hat{A} >} \right)^{1/2} \sim q. \quad (22) $$

and so in the limit $q \rightarrow 0$,

$$ < V_{c,q} > \geq \frac{\hbar}{2} \omega_{p,2D}(q) \left( \frac{nm}{< \hat{A} >} \right)^{1/2} \sim q. \quad (23) $$

Therefore, bounds, based on sum rules, are compatible with saving of almost all Coulomb energy $\frac{\hbar}{2} \omega_{p,2D}(q)$ in 2D when $< \hat{A} > \neq 0$.

It is necessary to mention extensive literature (for instance, Ref. [14]) using sum rules (in particular, third moment sum rule) in order to analyze and derive various local-field corrections and approximations of the density response, whereas our goal in this paper is to analyze general constraints on the electron Coulomb energy at small $q$ without relying on any approximation.

It is also interesting to discuss briefly for comparison the interaction energy of a many-particle system interacting via a short-range potential ($V_q \rightarrow V_0 = const$, for $q \rightarrow 0$). The upper bound on the partial interaction energy $< V_{int,q} >$ is given by the “acoustic mode” $\frac{\hbar}{2} (\frac{V_0 n}{m s})^{1/2} \sim q$.

$$ \frac{\hbar}{2} (\frac{V_0 n}{m s}) q \geq < V_{int,q} >, \quad (24) $$

where $\chi_0(q,0)$ is given by the compressibility sum rule [13]: $\chi_0(q,0) = \frac{\hbar}{ms}$, where $s$ is the velocity of
sound. Therefore, the maximum available interaction energy at long wavelength is insignificant (especially when weighted by the phase volume). For instance, it implies that in most phase transitions in neutral systems (i.e., many-particle systems interacting via a short-range potential) the interaction energy is saved predominantly at short distances.

III. EXPERIMENTAL PROBES.

The remainder of this paper is devoted to a brief discussion of the experimental spectroscopies which should, at least in principle, be able to shed light on the origin of the condensation energy in the SC transition, and the inferences which we may currently draw from them (cf. also Ref. [3], section 4.2). For simplicity we will consider explicitly a single-plane cuprate such as $Tl-2201$, so that in the normal phase there is only one characteristic length (other than, possibly, the electron mean free path) large compared to the quantity $q_{TF}^{-1}$, namely the interplane spacing $d$ (typically $\sim 10\,\AA$, i.e., $\sim 10-20q_{TF}^{-1}$) (note notable differences from Ref. [3]). The case of a multi-layer cuprate such as $Bi-2212$ is more complicated, as there is now a second "large" characteristic length, namely the intra-bilayer spacing ($\sim 3 - 5\,\AA$); however, the general pattern of the results is unchanged. In addition, we will assume tetragonal symmetry.

The two spectroscopies which most directly probe the Coulomb energy, or something closely related to it, are electron energy loss spectroscopy (EELS) [14] and optical reflectivity [20]; in the latter case we shall assume that ellipsometric measurements are possible in the interesting frequency regime [21,22] so that we may deduce the relevant complex dielectric constant without the use of Kramers-Kronig relations. In a bulk isotropic 3D metal the situation is very simple: to the approximation that we neglect multiple scattering and the effect of the ionic cores, the transmission EELS cross-section $\sigma(q, \omega)$ is a direct measure of the quantity $V_0^2 Im\chi(q, \omega)$, where $\chi(q, \omega)$ is the “true” density susceptibility as defined in Eqn. 4. Since in 3D the longitudinal dielectric constant $\epsilon_{\parallel}(q, \omega)$ is identically equal to $1 + \frac{e^2}{\pi\epsilon_0 q^2} \chi_0(q, \omega)$ with the help of Eqn. 4, we find the simple result

$$\sigma(q, \omega) = const \frac{1}{q} Im \left[ -\frac{1}{\epsilon_{\parallel}(q, \omega)} \right],$$

(25)

where the constant is of purely geometrical origin and can be calculated, and the quantity $Im \left[ -\frac{1}{\epsilon_{\parallel}(q, \omega)} \right]$ is usually known as the loss function. This formula is valid for arbitrary $q$, including values larger the inverse lattice spacing in the approximation of neglect of the direct scattering effect of the ionic cores. If the latter is taken into account, the effect is to multiply the formula (25) by a factor which is in general a function of $q$ but not of $\omega$ provided that the latter is small compared to typical core excitation energies ($\sim 20\,eV$), and expected to be unaffected by the superconducting transition. Writing out the integrand of Eqn. 4 explicitly in terms of $\chi_0(q, \omega)$ (see Eqn. 11) and using the 3D relation between the latter and $\epsilon_{\parallel}(q, \omega)$, we see that apart from a function of $q$ the transmission EELS cross-section is a direct measure of the Coulomb energy locked up in $dq\tilde{q}\omega$.

It is well known that the condensation energy due to the SC transition is extremely small (of order $10^{-4}\,eV$ per electron (or per unit cell)) in comparison with the atomic energies ($10\,eV$). It implies stringent requirements on experimental techniques, nevertheless, changes associated with the SC transition were observed by optics at mid-infrared frequencies [21,22] of magnitude sufficient to provide the condensation energy (measured directly by specific heat measurements). It is worth noting that we do not discuss the changes at frequencies comparable or lower than a superconducting gap (although these changes of course are most remarkable consequences of superconductivity!), because the change of Coulomb energy associated with this region of frequencies is negligible (if limited to small momenta $q \ll q_{TF}$). The optical reflectivity measurements [21,22] have enough precision to explore the type of questions discussed in this paper, while it is hoped that the transmission EELS can achieve required accuracy in the near future.

In a 3D bulk metal ellipsometric optical measurements can measure the complete transverse dielectric constant $\epsilon_{\perp}(q, \omega)$, in the limit $\tilde{q} \rightarrow 0$, and hence the corresponding “transverse” loss function $Im \left[ -\frac{1}{\epsilon_{\perp}(q, \omega)} \right]$. Since in the normal phase, at least, there should be no distinction, in the limit $\tilde{q} \rightarrow 0$, between $\epsilon_{\parallel}(q, \omega)$ and $\epsilon_{\perp}(q, \omega)$, it follows that in this phase the $\tilde{q} \rightarrow 0$ limit of “loss functions” measured by EELS and by optics should coincide. It is a somewhat delicate question, once one renounces reliance on some specific model such as the Fermi liquid one, what is to count as “the $\tilde{q} \rightarrow 0$ limit”; in addition to the obvious scale $q_{TF}$ or $q_F$, it is not immediately clear that the inverse electron mean free path $1/l$ might not be a relevant quantity. However, it is plausible that this quantity should not play a major role for $\omega$ in the mid-infrared region, so that we shall tentatively take “$\tilde{q} \rightarrow 0$” to mean in the 3D case $q \ll q_{TF}, q_F$.

Some care is needed in adapting the above results to the case of a layered material such as cuprates, even if we specialize (as we shall) to the limit $q_{\parallel} \ll 1/d$, which is automatically fulfilled in optical experiments and may be satisfied in transmission EELS by a suitable choice of geometry. In the context of EELS experiments, we now have to distinguish the cases $qd \ll 1$ and $qd \gg 1$ (where $q$ is the ab-plane component of the momentum loss). In the former case the 3D bulk formula (Eqn. 25) applies unchanged, provided that $\epsilon_{\parallel}(q, \omega)$ is defined to be
the tensor component of the longitudinal dielectric constant corresponding to current flow in the ab-plane; note in particular that, at least in the normal phase, we expect that \( Im \left( \frac{1}{\epsilon_{\|}(q, \omega)} \right) \) is nearly independent of \(|\vec{q}|\) in the limit \( \vec{q} \to 0 \). In the opposite limit \( qd \gg 1 \), we could choose to continue to use the 3D formula (Eqn. 25), but would then find that the \( \epsilon_{\|}(q, \omega) \) so defined has a strong explicit dependence on \( q \). A much more natural convention in this limit is to treat the scattering as occurring independently from the different CuO\(_2\) planes, and to define a two-dimensional (“per-plane”) version \( \chi^{(2)}(q, \omega) \) of \( \chi_0(q, \omega) \), or equivalently a quantity (cf. Ref. [3], section 4.1)

\[
K(q, \omega) \equiv \frac{1}{2qd^2} \chi^{(2)}_0(q, \omega) = \frac{d}{2} (\epsilon_{\|}(q, \omega) - \epsilon_b),
\]

(26)

where \( \epsilon_{\|}(q, \omega) \) is the “natural” definition of the 3D bulk ab-plane dielectric constant, i.e. the quantity which relates the local polarization to the local field, and is expected to be nearly constant over a range \( q \gg 1/d \), and \( \epsilon_b \) is the “background” (off-plane) contribution to it (cf. Ref. [3]). With this definition we find that, apart from factors depending only on \( q \), both the transmission EELS cross-section and the (single-plane) Coulomb energy locked up in the range \( d\vec{q}\vec{d}\omega \) are proportional to the quantity

\[
-Im \left( \frac{1}{1 + qK(q, \omega)/\epsilon_{sc}} \right),
\]

(27)

where \( \epsilon_{sc} \) is the dielectric constant which screens the Coulomb interactions of the in-plane electrons (note that in general \( \epsilon_{sc} \) is not equal to \( \epsilon_b \)). Thus, just as in the bulk 3D case, the transmission EELS cross-section is a direct measure of the Coulomb energy locked up in the relevant region of \((q, \omega)\)-space; note however that Eqn. 27 introduces an extra explicit \( q \)-dependence which absent in the bulk case. This subtlety seems to have been overlooked in the analysis of existing normal state EELS data [14] on the cuprates, where it seems to be assumed that even in the regime \( qd \gg 1 \) EELS experiments measure the “bulk” \( \epsilon_{\|}(q, \omega) \). In the intermediate case \((qd \sim 1)\) a similar analysis using Eqn. 3 is possible, but will not be given here.

In the case of optical experiments on the cuprates (with the sample surface assumed to lie in the ab-plane) we always have \( qd \ll 1 \), \( \vec{q}d \ll 1 \), and thus at first sight we would expect the complex dielectric constant inferred from reflectivity measurements to be identical to the \( \epsilon_{\|}(q, \omega) \) inferred, via Eqn. 25-27, from EELS experiments in the limit \( \vec{q} \to 0 \). Existing measurements in the normal phase do appear to be consistent with this prediction. In the superconducting state, however, there are three complications: first, it is not clear that even in the mid-infrared regime the Cooper-pair radius \( \xi_0 \) is not a relevant length scale, so that it may be illegitimate to use the “true” \( \vec{q} \to 0 \) \((q\xi_0 \ll 0)\) behaviour observed in the optics to infer the behaviour in the regime 1/\(\xi_0 \ll q \ll qTF \). Secondly, it is not completely obvious, particularly in the former limit \( q\xi_0 \ll 1 \), that the finite-frequency longitudinal and transverse dielectric constants must be equal in the superconducting state. This latter complication may be somewhat mitigated by a third consideration, namely that at the non-normal angles of incidence necessary in the ellipsometric technique what is measured, in a layered material, is not simply \( \epsilon_{\|} \) but a combination of \( \epsilon_{\perp} \) and \( \epsilon_{\perp} \).

We will not attempt to develop those points further here, but will rather use them to draw the conclusion that, while the spectacular changes observed [21,22] in the optically measured dielectric constants of the cuprates at and below the superconducting transition are strongly suggestive, a quantitative test of any scenario (such as the mid-infrared one of Ref. [3]) which attributes the energy saving largely to a regime of \( q \) small compared to \( qTF \) but large compared to \( \xi_0 \) (and 1/d) will require accurate transmission EELS data taken across the transition, something which (as regards the mid-infrared regime of frequencies) does not to our knowledge at present exist.

We finally address head-on the question: which of the three terms in Eqn. 3 is (are) reduced in the superconducting transition? If it is the second (Coulomb) one, in what regime(s) of \( q \) and \( \omega \) does the saving predominantly occur? Part of the interest of this question is that as we have seen above, a conjectured answer can be tested directly in transmission EELS experiments.

We start by recalling a well-known result: since the original (“true”) Hamiltonian of the N-body system (the nonrelativistic limit of the Dirac Hamiltonian) is composed exclusively of kinetic energy terms and (unscreened) Coulomb interactions, the virial theorem immediately tells us that the change in total kinetic energy (of electrons and ions) must be exactly minus half that of the total Coulomb energy (electron-electron, electron-ion and ion-ion), and thus Coulomb energy must be saved in the superconducting transition (and indeed in any other phase transition into a lower-energy state). While this conclusion is very generic and rigorous, it is not usually regarded as giving much insight into the “mechanism” of superconductivity in the cuprates (or for that matter in the classic superconductors) since the term “mechanism” is often held to refer to a low-energy effective Hamiltonian in which the separation of the original kinetic and potential energies may no longer be explicit. However, “intermediate-level” effective Hamiltonian [1] is sufficiently close to the original truly first-principles one that the virial-theorem result for the latter might at least suggest that it is one or both of the last two terms of [1] which are saved [23].

If we assume for the sake of argument that Coulomb energy is indeed saved, then where in the space of \( q \) and \( \omega \) is it saved? It is at this point that the sum rules argu-
ments of section 2 come into their own. For convenience we reproduce here the three relevant sum rules with terms of relative order $q^4$ and higher omitted on the right-hand sides: with notation as in Eqs. 28-30

$$J_{-1} = \frac{1}{V_q}, \quad (28)$$

$$J_{1} = \frac{m a^2}{4}, \quad (29)$$

$$J_{3} = \frac{n^2 q^4 V_q + q^2}{m^2} < \hat{A} >, \quad (30)$$

where in the case of a layered system $V_q$ is given by Eqn. 3 and tends to $e^2/(\epsilon_0 \epsilon_{\infty} q^2)$ for $q d \ll 1$ (and $q_{\perp} d \ll 1$, see above) and to $e^2/(2\epsilon_0 \epsilon_{\infty} q)$ for $q d \gg 1$, and where $\hat{A}$ is defined below Eqn. 3. The $(T = 0 K)$ contribution $< V_{e,q} >$ to the expectation value of the Coulomb energy from wave vector $q$ is, up to a factor, just $J_0$ (see Eqn. 3).

The arguments of section 2 now show that the maximum change of $< V_{e,q} >$ in the “essentially 3D” regime $qd \ll 1$ is proportional to a finite fraction of the “3D plasmon energy” but weighted by $q^2$ of the phase space ($d^q q$), and hence the maximum total saving possible from this regime is very small. On the other hand, the contribution from the regime $1/d \ll q \ll q_{TF}$ (where the truncated forms Eqs. 28-30 are still a good approximation) can be of order $q^{-7/2}$ (cf. the conclusion after Eqn. 23), provided the quantity $< \hat{A} >$ is substantial, while the phase space allows significant saving of Coulomb energy since $d q d^q q \sim (2\pi/d^2)\pi q dq d$ and $1/d \ll q \ll q_{TF}$ and $d^q q$. Thus, in a quasi-2D system with a large value of $< \hat{A} >$ substantial energy is in principle available for saving in this small-$q$ regime. To estimate the value of $< \hat{A} >$ we return for a moment to the limit $qd \ll 1$ and refer to the normal state optical loss function data: using Eqs. 23-24 with the appropriate form $(e^2/(\epsilon_0 \epsilon_{\infty} q^2))$ of $V_q$, we see that the quantity $< \hat{A} >_n$ is given, in the natural units of $n^2 e^2/(\epsilon_0 \epsilon_{\infty})$ by the expression (“n” is the normal-state value)

$$< \hat{A} >_n = \frac{J_3 - J_1^2}{J_{-1}}. \quad (31)$$

Although a strict evaluation of the right-hand side of Eqn. 31 from the optical loss-function requires us to know about the effective frequency cutoff (since at high frequencies there will be contributions to $\epsilon(q, \omega)$ from “core” processes not described by Eqn. 3), it is clear that the mere existence of mid-infrared(MIR) peak extending over an order of magnitude in frequency already implies that it at least of order of 1. Thus, a very appreciable fraction of the Coulomb energy locked up, in the normal state, in the low-$q$, MIR-frequency regime is in principle available for saving in the SC transition(or indeed in other possible phase transitions). Whether it is in fact saved and to what extent, as in fact postulated in the “MIR scenario” of Ref. 3, depends of course on the cost of the formation of the Cooper pairs in kinetic and/or static lattice energy. Actually, rather than asking as above for the fraction of the Coulomb energy which is in principle available for saving (something which is not that significant if the original value is itself small), it may be more informative to estimate the relative contribution of the small-$q$ regime in 2D and 3D for a given change in $\chi_0$ due to the phase transition. Taking into account both the phase space factor and the extra factor of $q$ in the denominator of the expression (27) in the 2D case, we find that in the regime where the $(qK)$ dominates the contribution of small $q$ is proportional to $q^2$ in 3D but to a constant in 2D, so that the relative importance of the long-wavelength regime is vastly enhanced in the 2D case.

On the experimental front, it has to be said that as noted in Ref. 21, the optical data, if extrapolated into the relevant $(q_{TF} \gg 1)$ regime with several other assumptions, indicate rather the opposite, i.e. that the Coulomb energy associated with the MIR regime actually increases in the SC state. However, because of the various considerations noted above, this extrapolation may be problematic, and a definitive test of the MIR hypothesis must await quantitative transmission EELS measurements across the superconducting transition (or a better theoretical understanding of the generic $q$-dependence of $\epsilon(q, \omega)$ in the SC state).

In sum, we analyzed the electron-electron Coulomb energy in the presence of the periodic lattice potential using various sum rules for the density-density response function. We believe that in this paper we have made it plausible that two specific properties of cuprates, namely, (a) the layered (two-dimensional) structure of the $CuO_2$ planes and (b) the occurrence of a broad and strong peak in the optical loss function, can be essential ingredients in the occurrence of high-temperature superconductivity in these materials by conspiring to the saving of small-$q$ ($q < q_{TF}$) part of electron-electron Coulomb energy.

We are grateful for discussions to Dirk van der Marel and Neil Ashcroft. A.J.L. acknowledges the support of grant NSF-DMR-99-86199. M.T. was supported by EP-SRC.

[1] J. R. Schrieffer, *Theory of Superconductivity*, Benjamin, Reading, Mass., 1964.
[2] Although the phonon mechanism for superconductivity is not ruled out irrevocably for the cuprate superconductors, we explore the general scenario of an electronic mechanism, at least, as a matter of principle.
[3] A. J. Leggett, Proc. Natl. Acad. Sci. USA vol. 96, 8365-8372 (1999).
We take here a definition of the density correlation function such that for positive frequencies ($\omega > 0$) the imaginary part of the susceptibility is positive $\text{Im} \chi(q, \omega) > 0$, and also $\text{Re} \chi(q, \omega) > 0$. This definition is different by a minus sign from the definition in Ref. [12] (section 4.1).

See also below, that $\chi_0$, defined in Eqn. 11, is different only by minus sign from $\chi_{xc}$ defined in Ref. [12] (section 4.1).

[4] P. W. Anderson, *The Theory of Superconductivity in the High $T_c$ Cuprates* (Princeton Univ. Press, Princeton, 1997), D. J. Scalapino, S. R. White, Phys. Rev. B 58, 8222 (1998), E. Demler, S.-C. Zhang, Nature 396, 733 (1998), V. Emery, S. Kivelson, O. Zachar, Physica C 282-287, 174 (1997), M. R. Norman et al, Phys. Rev. B 61, 14742 (2000).

[5] The separation into conduction and core electrons is assumed, where the core electrons contribute to the screening constant $\epsilon_\infty$ and to the effective periodic potential. For simplicity, we do not consider here possible effects of anisotropic (or $q$-dependent) screening by the core electrons. We also neglect the effects of disorder.

[6] Throughout this paper we do not use the reduced zone scheme (in order not to be based on this one-electron approximation in the presence of strong el-el interaction), and all momenta are real (not quasimomenta).

[7] Here the terms, Umklapp and lattice scattering, are used interchangeably to denote the static effects of the lattice potential. In the reduced zone scheme, one can distinguish between “lattice” and “Umklapp” effects, the former being the real momentum-non-conserving effects of the static lattice, while the latter due to effects of el-el Coulomb interaction (rigorously conserving real momentum, but in general not conserving quasimomentum).

[8] A. L. Fetter, Annals of Physics 88, 1-25 (1974).

[9] The sum rule analysis is applicable only to homogeneous phases of cuprates.

[10] Of course, in a system with crystalline periodicity the coordinate-space form of $\chi$ is a function of the center-of-mass coordinate as well as the relative one, and the momentum-space form is correspondingly a matrix quantity $\chi(q + \kappa_i, q + \kappa_j)$, where $\kappa_i, j$ are reciprocal lattice vectors. The quantity discussed everywhere in the text is in this notation $\chi(q, q)$.

[11] We take here a definition of the density correlation function such that for positive frequencies ($\omega > 0$) the imaginary part of the susceptibility is positive $\text{Im} \chi(q, \omega) > 0$, and also $\text{Re} \chi(q, \omega) > 0$. This definition is different by a minus sign from the definition in Ref. [12] (section 4.1).

See also below, that $\chi_0$, defined in Eqn. 11, is different only by minus sign from $\chi_{xc}$ defined in Ref. [12] (section 4.1).

[12] P. Nozieres, D. Pines, *The Theory of Quantum Liquids*, Perseus Books, Cambridge, Mass., 1999.

[13] N. Mihara, R. D. Puff, Phys. Rev. 174, 221 (1968).

[14] N. Iwamoto, Phys. Rev. A 30, 3289 (1984), K. Sturm, Phys. Rev. B 52, 8028 (1995).

[15] In different contexts, upper and lower bounds on the structure factor for classical and quantum liquids were discussed in the literature, for instance, Ref. [13], N.D. Mermin, Phys. Rev. 171, 272 (1968) and references therein.

[16] Here we give arguments that the expectation value of $< \hat{A} >$ is positive. Namely, if $U(x) \sim U_0 \cos(\kappa x)$, the electron density minimises the interaction energy by taking the form $\rho(x) \sim -\rho_0 \cos(\kappa x)$. In other words, the electron-lattice energy $< \sum_\kappa U_{\kappa} \rho_0 >$ (without the zero-momentum “background”) harmonic $\kappa = 0$ is always negative, because the harmonics of the density $\rho_0$ are shifted by the phase $\pi$ to the harmonics of the lattice potential $U_{\kappa}$. In addition, from inequalities Eqn. [14], it follows that the expectation value $< \hat{A} >$ is positive.

[17] It can be convenient for the analysis to rewrite Eqn. 14 as $(m/E)^2 \int d^3 p \rho(p) \{ S_{p+q} - S_p \}$ by making a change of variable $q = p + q$ in the first term in the bracket of Eqn. 14.

[18] T. Kato, Commun. Pure Appl. Math. 10, 151 (1957), J.C. Kimball, Phys. Rev. A 7, 1648 (1973), A.K. Rajagopal and J.C. Kimball, Phys. Rev. B 15, 2819 (1977).

[19] N. Nucker et al, Phys. Rev. B 39, 12379 (1989), Yun-Yu Wang et al, Phys. Rev. B 42, 420 (1990).

[20] Raman scattering is also relevant, but is a less direct probe and will not be discussed here.

[21] H. J. A. Molegraaf et al, Science 295, 2239 (2002).

[22] M. Rubhausen et al, Phys. Rev. B 63, 224514 (2001).

[23] Of course we cannot apply the virial theorem directly to Hamiltonian (Eqn. [1]), because the lattice potential $U_\kappa$ is not in general a sum of simple Coulomb terms.