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Umklapp scattering in cuprates

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Abstract
The insulating Mott state is formed due to repulsive electron–electron Coulomb interaction in the presence of the lattice (or in other words, due to electron–electron Umklapp processes). The second moment conductivity sum rule is derived, and it allows us to evaluate the strength of Umklapp scattering and the static electron–lattice energy. Mid-infrared spectra of current–current and density–density correlation functions in cuprates are suggested as evidence of strong Umklapp scattering.

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1. Introduction

High-temperature superconductivity in cuprates appears in the proximity of the insulating Mott state. Strong electron–electron (el–el) correlations, present in the Mott state, remain upon doping away from half-filling and, these correlations are revealed by anomalous normal state and pseudogap state properties. Since Mott–Hubbard insulator (distinct from Wigner insulator) is formed due to el–el Umklapp scattering, Umklapp processes are, presumably, relevant for the understanding of normal state phases as well as superconductivity. Umklapp scattering is principally different from other types of el–el scattering, because it breaks the conservation law of momentum and may lead to new liquid states distinct from Landau–Fermi liquid [1].

Interactions in many-body systems lead to various phases, distinguished by broken symmetries and order parameters. Conventionally, the presence of periodic lattice potential is accounted for by the Bloch theory (for non-interacting particles). But the Bloch band theory can fail in the presence of strong interactions[1], and not only new phases with broken

[1] Mott–Hubbard insulator is conventionally considered in the framework of Hubbard model on a lattice, and for this reason Hubbard model necessarily, although implicitly, includes periodic lattice potential. Therefore, a consistent treatment of correlated electron gas in proximity of Mott state has to include static electron–lattice coupling (last term of Hamiltonian (equation (1)). In the language of reduced Bloch band scheme, el–el scattering between Bloch states can be distinguished as momentum-conserving and momentum-non-conserving (Umklapp). In this language, Umklapp el–el scattering due to the presence of static electron–lattice potential causes the formation of Mott–Hubbard state (the commensuration—one electron per unit cell—leads to the increase of the effective strength of el–el Umklapp processes).

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symmetries (e.g., antiferromagnetic Mott insulator) but also new electron liquids (distinct in their phenomenology from Landau–Fermi liquid) may form. Namely, the ground state and excitations of many-body electron system (at strong coupling) may be different in the presence of the lattice (not protected by momentum conservation law) and without the lattice (leading to Landau–Fermi liquid). Therefore, it is important to have direct unambiguous measurements of the strength of Umklapp scattering for a many-body interacting system. In this paper, I show that the strength of Umklapp scattering can be evaluated from the second moment conductivity sum rule. The electromagnetic spectra of cuprates are discussed demonstrating the relevance of Umklapp scattering in the doping range where superconductivity exists. Coulomb energy (short- and long-range parts) of an inhomogeneous electron system depends on the lattice potential [2] especially in the proximity of Mott state, and therefore the understanding of Umklapp scattering is relevant for many theories of cuprates. Strong Umklapp scattering is particularly relevant in the context of 'mid-infrared scenario' [3, 4], which suggests a possibility of saving the long-range part of Coulomb energy at the superconducting transition.

The system of interest is the many-body system of electrons in the periodic lattice potential. If the dynamic lattice effects (e.g., phonons) are neglected (by assumption), the Hamiltonian can be written as follows:

$$\hat{H} = \sum_p \frac{p^2}{2m} c_p^+ c_p + \frac{1}{2} \sum_{q \neq 0} V_q \hat{\rho}_q \hat{\rho}_{-q} + \sum_\kappa U_{-\kappa} \hat{\rho}_\kappa \tag{1}$$

where $\hat{\rho}_q = \sum_k c_{k-q}^+ c_k$ is the density operator. The first and second terms are kinetic and Coulomb interaction energy of electrons. The third term describes the periodic potential of the lattice on the electrons which can be represented by the Umklapp pseudopotential $U_{-\kappa}$ with the sum over wavevectors of the reciprocal lattice $\kappa = l2\pi/a$, where $l = \{\pm 1, \pm 2, \ldots\}$. Various factors (the symmetry and profiles of electron–ion potential and relevant electronic valence orbitals, etc) are encrypted in the periodic pseudopotential. The interaction term with positive homogeneous background ($q = 0$) is omitted. The Coulomb energy and static electron–lattice (el–lat) energies are by definition

$$\langle V_c \rangle \equiv \frac{1}{2\Omega} \sum_q V_q [\langle \hat{\rho}_q \hat{\rho}_{-q} \rangle - N] \tag{2}$$

$$E_{\text{el–lat}} = \sum_\kappa U_\kappa \langle \rho_{-\kappa} \rangle. \tag{3}$$

Coulomb energy in the small-q limit ($q \ll q_{TF}$) for Hamiltonian (1) was analyzed in the publication [2]. It was shown that Umklapp processes are important (especially for two-dimensional Coulomb systems) in determining density–density correlation function. Sum rule for current–current correlation function, derived in this paper, can directly give valuable information about the static el–lat energy. This sum rule allows analysis of changes of static el–lat energy in the superconducting (or any other) transition.

2. Sum rule derivation

The standard way to derive various sum rules is to calculate commutators of an appropriate operator with the Hamiltonian. The first moment of the current–current correlation function is given by the expectation value of the following commutators:

$$\frac{2}{\pi} \int_0^{+\infty} \mathrm{d}\omega\, \omega \,\mathrm{Im}\, \Pi(q, \omega) = \langle [[j_q, H], j_{-q}] \rangle. \tag{4}$$

The current–current correlation function is defined in the standard way:

$$\Pi(q,\omega) \equiv \frac{\mathrm{i}}{\hbar}\int_0^{+\infty} \mathrm{d}t\, \mathrm{e}^{\mathrm{i}(\omega+\mathrm{i}\delta)t}\langle[\hat{j}(q,t),\hat{j}^{+}(q,0)]\rangle \tag{5}$$

where the current is $j_q = \frac{\mathrm{e}}{2m}\sum_p (2\vec{p}-\vec{q})c^{+}_{p-q}c_p$. This current can be split into the longitudinal and transverse components with respect to the direction of the perturbation $\vec{q}$:

$$j_q^{\parallel} = \frac{\mathrm{e}}{2m}\sum_p \left(\frac{2\vec{p}\vec{q}}{q^2}-1\right)\vec{q}c^{+}_{p-q}c_p \qquad j_q^{\perp} = \frac{\mathrm{e}}{2m}\sum_p \left(2\vec{p}-\frac{2\vec{p}\vec{q}}{q^2}\vec{q}\right)c^{+}_{p-q}c_p. \tag{6}$$

Using standard relations between the conductivity and the current–current correlation function (e.g., [5])

$$\frac{\omega^2}{4\pi}\,\mathrm{Im}\,\epsilon_{L,T}(q,\omega) = \mathrm{Im}\,\Pi_{L,T}(q,\omega) = \omega\,\mathrm{Re}\,\sigma_{L,T}(\omega) \tag{7}$$

the sum rule can be written equivalently

$$\frac{2}{\pi}\int_0^{+\infty}\mathrm{d}\omega\,\omega^3\,\mathrm{Im}\,\epsilon(q,\omega) = 8\int_0^{+\infty}\mathrm{d}\omega\,\omega^2\,\sigma(q,\omega) = \langle[[j_q,H],j_{-q}]\rangle. \tag{8}$$

The explicit calculation gives the following results for the sum rules of the longitudinal current correlations

$$\left[\left[j_q^{\parallel},\hat{H}\right],j_{-q}^{\parallel}\right] = \frac{\mathrm{e}^2}{(2m)^3}\left[2q^4\hat{N}+24\sum_p(\vec{p}\vec{q})^2c_p^{+}c_p\right]+\left(\frac{\mathrm{e}}{2m}\right)^2\sum_t 2(\vec{t}\vec{q})V_t[(2\vec{t}\vec{q})\rho_{t+q}\rho_{-(t+q)}$$
$$-(2\vec{t}\vec{q}+2q^2)\rho_{-t}\rho_t]-4\left(\frac{\mathrm{e}}{2m}\right)^2\sum_\kappa(\vec{\kappa}\hat{\vec{q}})[(\vec{\kappa}\hat{\vec{q}})-q^2]U_\kappa\rho_{-\kappa} \tag{9}$$

and of the transverse current correlations

$$\left[\left[j_q^{\perp},\hat{H}\right],j_{-q}^{\perp}\right] = \frac{\mathrm{e}^2}{(2m)^3}4\sum_p[p^2q^2-(\vec{p}\vec{q})^2]c_p^{+}c_p+\left(\frac{\mathrm{e}}{2m}\right)^2 4\sum_t(t^2-(\vec{t}\hat{\vec{q}})^2)V_t[\rho_{t+q}\rho_{-(t+q)}$$
$$-\rho_t\rho_{-t}]-\left(\frac{\mathrm{e}}{2m}\right)^2 4\sum_\kappa(\kappa^2-(\vec{\kappa}\hat{\vec{q}})^2)U_\kappa\rho_{-\kappa} \tag{10}$$

where $\hat{\vec{q}} = \frac{\vec{q}}{q}$ is a unit vector along the direction of $\vec{q}$, and $\hat{N} = \sum_p c_p^{+}c_p$ is the number operator. In the long wavelength limit ($q \to 0$), the sum rules can be written in a simple form

$$\int_0^{+\infty}\mathrm{d}\omega\,\omega^2\,\sigma_T(\omega) \simeq \frac{\mathrm{e}^2}{8m^2}\sum_\kappa(\kappa^2-(\vec{\kappa}\hat{\vec{q}})^2)(-1)\langle U_\kappa\rho_{-\kappa}\rangle \tag{11}$$

$$\int_0^{+\infty}\mathrm{d}\omega\,\omega^2\,\sigma_L(\omega) \simeq \frac{\mathrm{e}^2}{8m^2}\sum_\kappa(\vec{\kappa}\hat{\vec{q}})^2(-1)\langle U_\kappa\rho_{-\kappa}\rangle+\left[\frac{4\pi\,\mathrm{e}^2n^2}{m^2}\right] \tag{12}$$

and define $\hat{A} \equiv \sum_\kappa(\vec{\kappa}\hat{\vec{q}})^2(-1)U_{-\kappa}\hat{\rho}_\kappa$. The sum rule for the transverse conductivity (equation (11)) is determined by Bragg scattering from the periodic potential such that $\vec{\kappa}\perp\vec{q}$. The sum rule for the longitudinal conductivity (equation (12)) is given by two terms: one due to Bragg scattering from the lattice distortion with $\vec{\kappa}\|\vec{q}$, and another one, due to the long-range part of el–el Coulomb interaction (only in three-dimensional case). This last term of equation (12) can be omitted under many circumstances if sum rule is written for *local* longitudinal conductivity. The conductivity defined in equation (7) is nonlocal, and difficulties associated with such a conventional definition were discussed in detail [6]. Under many circumstances [6] the long-range part of Coulomb potential can be considered self-consistently,

so that only the short-range part of Coulomb potential is included into effective Hamiltonian for calculation of local conductivity. In the absence of long-range order current fluctuations have finite correlation length, and therefore local transverse and longitudinal conductivities has to be equal in the limit $q \to 0$. Consequently, the right-hand sides of equations (11) and (12) have to be equal (for square(or cubic) lattice).

Sum rules (equations (11) and (12)) can be used to determine (e.g., from optical reflectivity experiments) the expectation value of $\langle \hat{A} \rangle$ and closely related $E_{\text{el–lat}}$ (or rather changes thereof upon phase transition). The analysis of these sum rules would allow us to measure the changes of static el–lat energy (usually included into 'effective kinetic energy', see discussion below). In the Mott state, conductivity $\sigma_T(\omega)$ is suppressed up to high frequencies (of interband transitions, charge-transfer transitions, etc). Since most of the spectral weight of $\sigma_T(\omega)$ is transferred to high frequencies, the right-hand side of equation (11) increases. This is consistent with the decrease of $E_{\text{el–lat}}$ (increase of $\langle \hat{A} \rangle$) in the Mott state.

The continuity equation $\vec{q}\,\vec{j}_{q,\omega} = -\omega\rho_{q,\omega}$, due to the charge conservation, allows us to relate the longitudinal current–current correlation function and the density–density correlation function

$$q^2 \operatorname{Im} \Pi_{j^{\parallel},j^{\parallel}}(q,\omega) = \omega^2 \operatorname{Im} \chi_{\rho,\rho}(q,\omega). \tag{13}$$

Thus one way to derive the f-sum rule for the longitudinal conductivity is to use the first moment sum rule for the density–density correlation function. Since [5]

$$\frac{2}{\pi}\int_0^{+\infty} \omega \operatorname{Im} \chi_{\rho,\rho}(q,\omega)\,\mathrm{d}\omega = \frac{ne^2q^2}{m} \tag{14}$$

then the f-sum rule for the longitudinal conductivity follows:

$$\frac{2}{\pi}\int_0^{+\infty} \mathrm{d}\omega\, \frac{\operatorname{Im} \Pi_{j^{\parallel},j^{\parallel}}(q,\omega)}{\omega} = \frac{2}{\pi}\int_0^{+\infty} \mathrm{d}\omega\, \sigma_L(\omega) = \frac{n\,\mathrm{e}^2}{m}. \tag{15}$$

One can also check directly the equivalence between the third moment sum rule for the density–density correlation function (given in [2]) and the first moment sum rule for the longitudinal current–current correlation function (equation (9)).

For $q = 0$ limit, the sum rule (equation (11)) was derived by Hopfield [7]. Namely, in real space the sum rule can be written

$$\frac{2}{\pi}\int_0^{+\infty} \mathrm{d}\omega\, \omega^3 \operatorname{Im} \epsilon(q,\omega) = -\frac{\omega_0^2\,\mathrm{e}}{3mn}\sum_b \int_{\text{all space}} \mathrm{d}^3 r\, \delta\rho(r)\nabla^2 V_b(r) \tag{16}$$

where the sum is over different atoms in the unit cell (numbered by b). Thus, in real space the sum rule (equation (11)) is given by the integral of the product of the Laplacian of the crystal potential $V_b(r)$ and the distortion of the electron density $\delta\rho(r)$.

3. Discussion

In the recent literature, the extensive use of the tight-binding model expression for the kinetic energy was used unlike as defined in the Hamiltonian (equation (1)). Such an approach has several obvious limitations: (a) the tight-binding kinetic energy is only 'effective' kinetic energy, which is the arbitrary sum of the kinetic and partial electron–lattice energy, (b) the upper cutoff on the bandwidth of the tight-binding band is ambiguous (especially because this cutoff cannot be determined unambiguously in strongly correlated materials), (c) finally, the 'truncated' f-sum rule (equation (15)) is not constant, but rather the 'effective' kinetic energy (the validity is limited to only the closest-neighbour hopping (for instance, neglecting

the next-nearest neighbour hopping)). The Hamiltonian used in this paper has much wider applicability than the tight-binding model (perhaps, the only not well-controlled assumption is the assumption of the 'pseudopotential', representing the 'effective' periodic potential from ions and core electrons).

Electron-energy loss spectroscopy (EELS) [8] and optical reflectometry (OR) [9] provide direct evidence that the Umklapp scattering is very strong in cuprates (in wide doping range including superconductivity). Indeed, the peak for the loss function $\mathrm{Im}\left[\frac{-1}{\epsilon(q,\omega)}\right]$ (for more detailed discussion of density–density correlation function of Hamiltonian (1), see [2]) is so wide that it can barely be associated with narrow plasmon peak as observed in simple metals. Another widely known fact from optical reflectivity measurements [9] is that the optical conductivity $\sigma_{ab}(\omega)$, in addition to the Drude contribution at low frequencies, contains a so-called 'mid-infrared band' stretching from 0.2 eV to 2 eV. This can be contrasted with the situation for simple metals, where at high frequencies the conductivity is small and given by the tail of the Drude peak. The vanishing conductivity $\sigma(\omega \sim \omega_p)$ at the plasma frequency implies the narrowness of the plasma peak for the loss function $\mathrm{Im}(-1/\epsilon(q = 0, \omega)) = \mathrm{Im}(-1/(1 + 4\mathrm{i}\pi\sigma(\omega)/\omega))$. Correspondingly, for cuprates the mid-infrared contribution to the conductivity $\mathrm{Re}\,\sigma(\omega) \sim \omega\,\mathrm{Im}\,\epsilon(\omega)$ and the wide plasmon peak for $\mathrm{Im}(-1/\epsilon(q = 0, \omega))$ are closely related facts (both due to Umklapp scattering).

The origin of the mid-infrared band can be understood from the following relation, derived by Hopfield [10],

$$\mathrm{Re}\,\sigma_t(\omega) \sim \frac{1}{4\pi m^2\omega^3}\sum_{\kappa} U_\kappa^2\kappa^2\kappa_\mu^2\,\mathrm{Im}\left[-\frac{1}{\epsilon_l(\kappa,\omega)}\right] \tag{17}$$

that is the real part of transverse conductivity (at $q \to 0$) has a contribution proportional to the imaginary part of the inverse longitudinal dielectric function at the zone boundary (or the reciprocal lattice vectors). The loss function $\mathrm{Im}\left[-\frac{1}{\epsilon_l(\kappa,\omega)}\right]$ far from the centre of the Brillouin zone (up to a third of the lattice vector) was measured by EELS [8] to be a wide band of density excitations (mixed collective and el–hole excitations), and it can be approximated to be of similar form up to the zone boundary. In the presence of strong Umklapp potential U_κ, *this band should be associated by virtue of equation (17) with the mid-infrared band observed for the transverse conductivity*. The essence of equation (17) is that in the presence of the lattice potential (or any potential breaking translational invariance) the longitudinal excitations become partially transverse (and thus can be observed in the transverse response). In spite of the limited validity of equation (17) (valid only to second order in U_κ), the suggested origin of the mid-infrared band in the transverse conductivity deserves further experimental analysis. Inter alia, the fact that high-frequency modes (e.g., plasmons and interband transitions) are affected by the appearance of SC is contrary to the simple RPA treatment [11]. Thus a band theory (even with strong short-range Hubbard repulsion) is not a valid description essentially at all dopings including the SC phase. This is not surprising, since the band theory fails utterly in the Mott insulating state (at half-filled band). Moreover, since OR observes changes due to the SC transition at high frequencies [12, 13] of collective modes and interband transitions (0.2–4 eV), the complete understanding of high-T_c superconductivity cannot be limited to effective low-energy phenomenological theories.

Dynamical mean field theory (DMFT) [14] successfully describes Mott transition and related consequences of this transition (for instance, 'mid-infrared band' in the optical conductivity). DMFT, not limited to low energies, can correctly explain the local physics of Hubbard repulsion, while the description of superconductivity and the long-range part of Coulomb interaction remains a difficult challenge. Sum rules (equations (11) and (12)) can be used to check the self-consistency of DMFT calculations of optical conductivity.

One of the important questions about high-temperature superconductivity is the origin of the condensation energy (the energy difference between superconducting state and normal state). Hamiltonian (equation (1)) is sufficiently close to the original truly first-principle Hamiltonian so that the terms of Hamiltonian (equation (1)) can be clearly identified with terms of the original Hamiltonian. Similar identification is quite problematic for many phenomenological Hamiltonians suggested in the literature (making the origin of condensation energy ambiguous for such models). The change of el–lat energy (static part of electron–ion energy) can be analyzed from experimental optical data with the use of sum rule (equation (11)) in order to test whether $E_{\text{el–lat}}$ contributes to the condensation energy. It should be noted that although BCS Hamiltonian is a reduced (phenomenological) Hamiltonian, it correctly ascribes condensation energy to the reduction of the dynamic part of electron–ion interaction energy (rewritten as attractive el–el term in BCS Hamiltonian).

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