Eur. Phys. J. B **3**, 149–154 (1998)

THE EUROPEAN PHYSICAL JOURNAL B

Optical conductivity of the Bechgaard salts: the sum rules revisited

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Received: 19 December 1997 / Received in final form: 9 March 1998 / Accepted: 23 March 1998

Abstract. The validity of the optical sum rules has been addressed eversince and was always matter of debate. Particularly controversial is the proof that the *partial* sum rules can be extended to both optical conductivity and energy loss function. We show in this paper that for both transverse (optical conductivity) and longitudinal (energy loss function) absorption processes the corresponding sum rule can be theoretically established and through appropriate conditions for the integration limits exactly verified. We also focus our attention on the one-dimensional case within the microscopic Hubbard model. An application of these concepts to the quasi one-dimensional systems, for which we have chosen the organic (TMTSF)$_2$PF$_6$ material, will also be presented.

PACS. 78.20.-e Optical properties of bulk materials and thin films – 75.30.Fv Spin-density waves

1 Introduction

1.1 Partial sum rule for the loss function

The global sum rule on the real part of the optical conductivity

$$I_\sigma = 8\int_0^\infty \sigma_1(\omega)d\omega = -\frac{4\pi e^2}{\Omega}\langle\Psi_0|H_{kin}|\Psi_0\rangle, \qquad (1)$$

where Ω is the volume and H_{kin} is the kinetic energy Hamiltonian, has been shown to remain valid for a single tight-binding band by Maldague in 1977 [1], in which case one talks of a *partial* sum rule. The proof that the similar global sum rule for the loss function

$$I_p = \frac{2}{\pi}\int_0^\infty \omega Im\left(\frac{-1}{\varepsilon^\perp(\omega)}\right)d\omega = -\frac{4\pi e^2}{\Omega}\langle\Psi_0|H_{kin}|\Psi_0\rangle, \qquad (2)$$

where $\varepsilon^\perp(\omega)$ is the transverse complex dielectric function, can also be extended to a single tight-binding band has not been given so far.

On the basis of experimental data, Jacobsen was led to the conclusion that there is no such *partial* sum rule for the loss function [2]. In fact, Jacobsen presented reflectivity data and the analysis of the optical sum rules for the quasi-one-dimensional organic conductor DBTTF-TCNQCl$_2$ at room temperature [2]. The main conclusion of his analysis is that since the sum rule of the real part of the optical conductivity (I_σ) is related to the kinetic energy [3], short-range electron-electron interactions increasing the kinetic energy, lead to a reduction in the oscillator strength and thus the saturation value of the sum rule I_σ is smaller than the saturation limit of the sum rule of the loss function (I_p). The sum rule of the loss function is supposed to be unaffected by short range correlations and I_p should saturate close to ω_p^2, where ω_p is the plasma frequency. From this apparent discrepancy between the saturation limit of I_p and I_σ, Jacobsen estimated values of the correlation parameters U, the on-site repulsion, and V, the nearest-neighbour repulsion, for DBTTF-TCNQCl$_2$ [2].

Here, we want first of all to demonstrate that the argument by Jacobsen is theoretically not strictly correct since in fact the global sum rule for the loss function can be extended to a single band. The response to a longitudinal probe is given by the causal response function $\frac{1}{\varepsilon^\parallel} - 1$ which, under very general conditions, satisfies:

$$2\int_0^\infty \omega\left(\frac{1}{\varepsilon^\parallel(\mathbf{q},\omega)} - 1\right)d\omega = \frac{-i\pi e}{\Omega}V(\mathbf{q}) \times \langle\Psi_0\big|\big[[H,\rho_{-\mathbf{q}}],\rho_{\mathbf{q}}\big]\big|\Psi_0\rangle. \qquad (3)$$

Here $V(\mathbf{q})$ is the Fourier transform of the interaction between electrons, $\varepsilon^\parallel(\mathbf{q},\omega)$ is the longitudinal complex dielectric function and $|\Psi_0\rangle$ is the ground state of H. Let us now consider the Hamiltonian:

$$H = H_{kin} + \sum_{\mathbf{r},\mathbf{r}'} V_{\mathbf{r},\mathbf{r}'}n_{\mathbf{r}}n_{\mathbf{r}'}. \qquad (4)$$

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To describe a single band model, we choose a tight binding Hamiltonian for the kinetic part:

$$H_{kin} = -\sum_{\mathbf{r},\sigma,\alpha\in\{a,b,c\}} t_\alpha (c^\dagger{}_{\mathbf{r}+\boldsymbol{\delta}_\alpha,\sigma} c_{\mathbf{r},\sigma} + h.c.) \tag{5}$$

where $\boldsymbol{\delta}_a$, $\boldsymbol{\delta}_b$, $\boldsymbol{\delta}_c$ are the basic lattice vectors. The interaction term does not contribute to the double commutator, which can be easily shown to be equal to:

$$[[H,\rho_{-\mathbf{q}}],\rho_{\mathbf{q}}] = -\sum_\alpha 4\sin^2\left(\frac{q_\alpha}{2}\right) H_{kin,\alpha} \tag{6}$$

with $H_{kin,\alpha} = -\sum_{\mathbf{r},\sigma} t_\alpha (c^\dagger{}_{\mathbf{r}+\boldsymbol{\delta}_\alpha,\sigma} c_{\mathbf{r},\sigma} + h.c.)$.

To make contact with the experimental situation, let us now specialize to the case where $\mathbf{q}$ is parallel to a lattice vector, say $\boldsymbol{\delta}_a$. In the limit $q_a \to 0, 4\sin^2(q_a/2) \cong q_a^2$. Besides, regardless of the nature of local interactions, the Fourier transform of the two-body interaction is dominated by the long-range Coulomb interaction in this limit, and $V(\mathbf{q}) \cong 4\pi e/q^2 = 4\pi e/q_a^2$.

So in this small q_a limit we get:

$$2\lim_{q_a\to 0}\int_0^\infty \omega Im\left(\frac{-1}{\varepsilon^{\parallel}(\mathbf{q},\omega)}\right) d\omega = -\frac{4\pi^2 e^2}{\Omega}\langle\Psi_0|H_{kin,a}|\Psi_0\rangle. \tag{7}$$

Finally, we note that the longitudinal and transverse dielectric functions are equal in the $\mathbf{q} \to \mathbf{0}$ limit. Therefore, the sum rule of equation (2) is valid for a single band. In other words, the same *partial* sum rule is valid for the energy loss function and for the real part of the optical conductivity.

1.2 On the experimental verification of the sum rules

The consistency of these *partial* sum rules has been checked experimentally for a number of metals quite some time ago [4,5]. In principle, the optical sum rules provide a useful guide to interpreting the experimental results and could be used in order to check the consistency of optical data. Indeed, the optical functions can be experimentally determined over a limited frequency range and the redistribution of spectral weight in dispersive (*i.e.*, the real part of the dielectric function, ε_1) and absorptive (*i.e.*, imaginary part of the dielectric function, ε_2) processes might be affected by the finite cutoff frequency used as integration limit in the Kramers-Kronig relation between ε_1 and ε_2.

However, there is one subtlety in this analysis. One is never in the situation where a band is alone, and the sum rule can only be valid if one includes high energy bands in the integration. Now if the conduction band is well separated from the empty bands, the real part of the dielectric function $\varepsilon_1(\omega)$ will have a plateau above the conduction band at a value usually called ε_c, and the following sum rule can be proven:

$$\varepsilon_c^2\int_0^{\omega^+} \omega Im\left(\frac{-1}{\varepsilon^{\perp}(\omega)}\right) d\omega = 4\pi\int_0^{\omega^+} \sigma_1(\omega) d\omega \tag{8}$$

where the frequency ω^+ should be above the screened plasma frequency Ω_p, at which the main contribution to the loss function is located, but below the beginning of the next contribution coming from high energy bands. From now on, I_σ will refer to the quantity defined in equation (1) but with a cut-off ω^+, and I_p will refer to the quantity of equation (2) multiplied by ε_c^2 and with a cut-off ω^+. Note that the screened plasma frequency is related to the unscreened plasma frequency ω_p by $\omega_p = \sqrt{\varepsilon_c}\Omega_p$.

This is the ideal situation where the optical conductivity of the conduction electrons is given by a simple Drude peak. Now, if the electron-electron interactions are strong enough, some spectral weight can be transferred to frequencies of the order of the band width and/or of the local correlation parameters. Suppose that this weight extends up to a frequency ω_c. As long as these contributions are still well separated from the high energy bands, one can still hope to verify the consistency of the *partial* sum rules by including ε_c, but one should now integrate up to an energy larger than ω_c. Let us now assume that ω_c is larger than the screened plasma frequency Ω_p. The sum rule on the loss function will still have a plateau above Ω_p, but some weight will still be missing both in the conductivity and in the loss function. Now, we have verified on a number of test cases that the following inequality holds:

$$\varepsilon_c^2\int_0^{\omega_0} \omega Im\left(\frac{-1}{\varepsilon^{\perp}(\omega)}\right) d\omega \le 4\pi\int_0^{\omega_0} \sigma_1(\omega) d\omega. \tag{9}$$

The equality is satisfied when $\omega_0 \ge \omega_c$. So, if we compare the sum rules for the conductivity and the loss function, we expect them to saturate at the same value just above Ω_p if all the weight of the conduction band is in the Drude peak, and we expect the plateau in the sum rule of the loss function just above Ω_p to be below the sum rule on the conductivity if some spectral weight coming from the conduction band is distributed over a frequency range extending beyond Ω_p. Interestingly enough, this result seems to be in clear contradiction with the results reported by Jacobsen [2].

The purpose of this paper is to revisit the sum rules' concept, particularly for the quasi-one-dimensional (1D) systems. In the present analysis of this problem, we will focus our attention on the quasi-one-dimensional organic (TMTSF)$_2$PF$_6$ compound on which a complete set of thorough optical data has been recently collected [6–8]. The main goal of this paper is to use the electrodynamic response of (TMTSF)$_2$PF$_6$ with the scope of checking the validity of the *partial* sum rules for a one-dimensional system, where the presence of an upper Hubbard band due to correlations has to be taken into account. The effect of such correlations in the frequency dependence of the electrodynamic response should lead to some incoherent spectral weight above but not too far from the plasma frequency, which should be considered in the analysis of the *partial* sum rules. First of all, we will briefly present the investigated system together with some details about the optical measurements and the corresponding evaluation of the optical functions. Then, we will address the problem concerning the estimation of the sum rules from

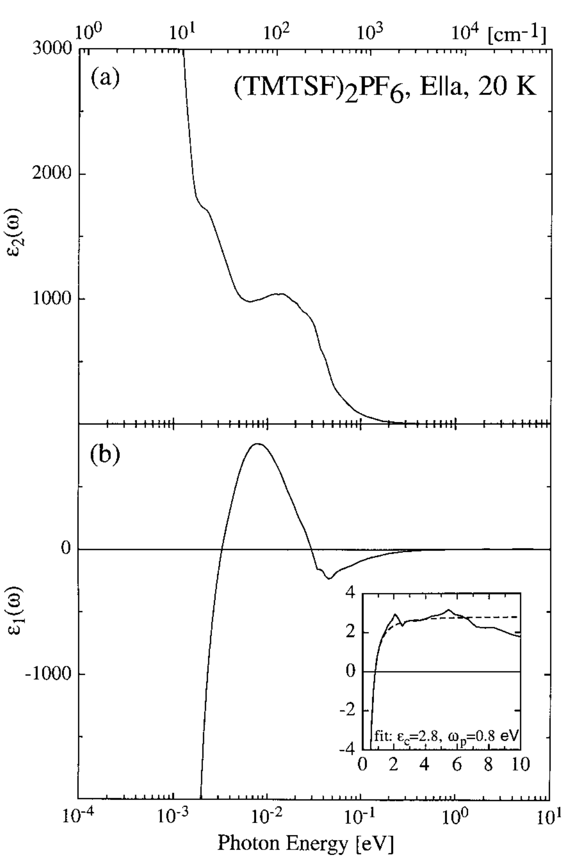

Fig. 1. Components (a) $\varepsilon_2(\omega)$ and (b) $\varepsilon_1(\omega)$ on a logarithmic energy scale of the complex dielectric function obtained from Kramers-Kronig analysis of the reflectivity measurements of $(TMTSF)_2PF_6$ along the highly conducting axis at $T = 20$ K [6]. The inset shows $\varepsilon_1(\omega)$ on a linear energy scale and the result of the fit after equation (11).

the experimental data. Finally, a discussion of the relevant results will be performed in the perspective of a microscopic model, based on a 1D extended Hubbard model.

2 Experiment and results

2.1 The data

The Bechgaard salts $(TMTSF)_2X$, where X is an inorganic monovalent anion PF_6, ClO_4, SbF_6, ReO_4, BF_4 or NO_3, have been found to have extremely high conductivity along at least one crystallographic axis and highly anisotropic band structures, leading to Fermi surface instabilities and therefore to phase transitions [9]. The organic conductor $(TMTSF)_2PF_6$, on which we will focus our attention, undergoes a transition to a SDW state at $T_{SDW} = 12$ K, and it was found to be superconducting under pressure [10].

Figure 1 shows $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ on a logarithmic energy scale obtained by Kramers-Kronig transformation of the measured optical reflectivity $R(\omega)$ with the polarization direction parallel to the highly conducting axis ($E||a$) in the normal metallic state at 20 K (*i.e.*, $T > T_{SDW}$) [6,8]. In this respect, it is worth noting that the Kramers-Kronig transformation, which yields the real and imaginary part of the complex dielectric function $\varepsilon(\omega)$ necessary to evaluate I_σ and I_p, requires the knowledge of one of the optical function for all frequencies from zero to infinity. In order to address this issue the reflectivity $R(\omega)$ of a large single crystal of $(TMTSF)_2PF_6$ was measured over an extremely broad spectral range combining the results from different spectrometers in the submillimeter, infrared, optical and ultraviolet frequency ranges [6]. The Hagen-Rubens extrapolation for the low frequency limit and the power law $R(\omega) \propto \omega^{-4}$, representing the free-electron behaviour for the high energy region above 12 eV were assumed. A detailed report on the optical investigation of the system in its normal ($T > T_{SDW}$) and SDW-ground state ($T < T_{SDW}$) can be found elsewhere [6–8].

In this paper we do not want to give a physical interpretation of the optical response in the normal state, but refer to reference [6,8] for a comprehensive discussion. Nevertheless, it is worthwhile to remind a few general features. The spectra are characterized by a very narrow Drude-like term ascribed to free carriers at zero energy (ZE) and a rather broad finite energy (FE) excitation around 3×10^{-2} eV (see $\varepsilon_2(\omega)$). Moreover, the sum of the spectral weight of the two components

$$\int_0^{\omega_i} \sigma_1^{ZE}(\omega)d\omega + \int_{\omega_i}^{\infty} \sigma_1^{FE}(\omega)d\omega = \frac{\omega_p^2}{8}, \qquad (10)$$

where $\omega_i \sim 20$ cm^{-1} is a cut-off frequency between the ZE and FE modes, leads to a total plasma frequency of $\omega_p = 13.6$ eV. The ZE mode contains only 1% of the total spectral weight, the remaining part being encountered by the FE mode [6].

Finally, Figure 2a displays the real part $\sigma_1(\omega)$ of the complex optical conductivity $\sigma(\omega) = \dfrac{\omega}{4\pi i}(\varepsilon(\omega) - 1)$ and the loss function $Im\left(\dfrac{-1}{\varepsilon^{\perp}(\omega)}\right)$ on a linear energy scale. In view of the coming discussion we note the peak at about 0.9 eV of the loss function, which will be ascribed to the screened plasmon.

2.2 The optical sum rules

The sum rules I_σ and I_p differ significantly as a function of energy, because they describe somewhat different processes. The right hand side of equation (8), the sum rule on the optical conductivity I_σ, is related to the rate of energy absorption by the system from transverse fields (photons) and the left hand side of equation (8), the sum rule on the electron loss function I_p, to the rate of energy absorption by the system from longitudinal fields (electrons) [11]. $I_\sigma(\omega)$ thus measures the intraband oscillator strength, while $I_p(\omega)$ is related to plasma excitations. Note that the saturation value of both $I_\sigma(\omega)$ and $I_p(\omega)$ should be the plasma frequency ω_p^2 (Eq. (10)).

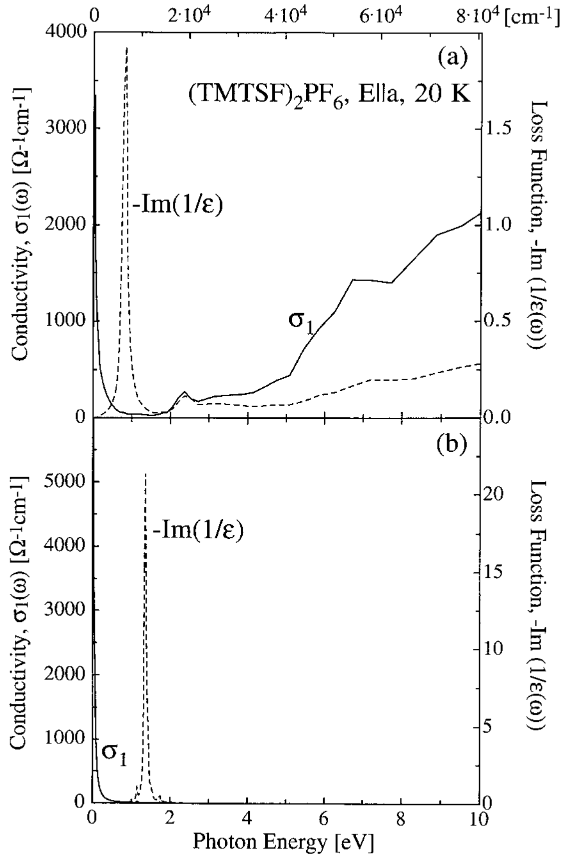

Fig. 2. (a) Real part of the optical conductivity $\sigma_1(\omega)$ and energy loss function $-Im(1/\varepsilon^{\perp})$ given on a linear energy scale [6]. (b) Optical conductivity and energy loss function of the one-dimensional extended Hubbard model at quarter-filling with $U/t = 5$ and $V/t = 2$ for a 16-site cluster. The peaks have been given an arbitrary width of $0.1t$.

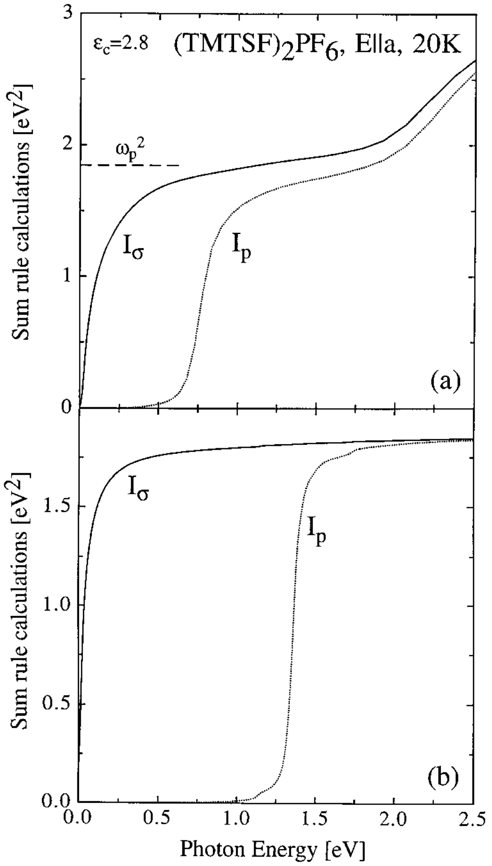

Fig. 3. (a) Sum rule calculations based on the data of Figure 2 for (TMTSF)$_2$PF$_6$ at 20 K. (b) Optical sum rules of the optical conductivity (I_σ) and of the energy loss function (I_p) for the model of Figure 2b.

In view of the experimental verification of the *partial* sum rules (Eq. (8)), the first step in our analysis is to find the most precise way of determining ε_c. This is essential since different determination of the optical dielectric constant may lead to very strong deviations of the saturation values of I_σ and I_p. The different ways employed in determining ε_c could probably explain the difference in the sum-rules analysis between our results and those of Jacobsen [2]. The best way to determine ε_c is to fit the real part of the dielectric constant $\varepsilon_1(\omega)$ around Ω_p, in other words at the zero crossing of $\varepsilon_1(\omega)$ (*i.e.*, $\varepsilon_1(\Omega_p) = 0$) by the expression:

$$\varepsilon_1(\omega) = \varepsilon_c \left[1 - \left(\frac{\Omega_p}{\omega}\right)^2\right] \tag{11}$$

where Ω_p and ε_c are free parameters. The result of such a fit is given in the inset of Figure 1a. The value found for ε_c is 2.8.

It is worth noting that the maximum of the loss function is found at 0.86 eV, in agreement with $\Omega_p = 0.8$ eV obtained from our fit (Eq. (11)). Apart from the free carrier contribution and the contribution coming from high energy bands, there is clear evidence for some residual spectral weight above Ω_p, which can have important consequences on the *partial* sum rules.

The result of the analysis for (TMTSF)$_2$PF$_6$ at 20 K is shown in Figure 3a. We integrated both functions in equation (8) up to $\omega^+ = 10$ eV. For energies higher than this cut-off frequency absorptions no longer take place and $\varepsilon_2(\omega)$ is zero. Both I_σ and I_p saturate near ω_p^2, even though the saturation value of the integral of the loss function remains below the integral of the optical conductivity over the whole investigated spectral range.

3 Discussion

From equation (8) it clearly appears, that the effect of ε_c on the electron energy loss function sum rule is very strong. Thus, one of the crucial issues for the consistency of the sum rules seems to be the value of ε_c. With an accurate estimate for the dielectric constant, the two sum rules should agree, if there is no spectral weight at higher energy due to correlations. This is not the case in (TMTSF)$_2$PF$_6$, as can be seen in Figure 2, where, due to electron-electron interactions, some incoherent weight shows up in the conductivity between the screened plasma frequency Ω_p and higher frequency contributions. It is clear from Figure 2a, that I_σ will saturate for $\omega < \omega_p$, while the saturation value for I_p will be reached only at higher frequency, since the contribution to I_p comes from a narrow range around Ω_p at which the main contribution of the loss function is located. A poor estimation of ε_c, or, in other words an estimation taken at too high energies with respect to Ω_p, can thus lead to a situation where the saturation value of I_p is too high or too low in comparison with I_σ. It is therefore important to stick to an estimate of ε_c obtained close to the value where $\varepsilon_1(\omega)$ vanishes, as we did in our fit (Eq. (11)).

If now the conductivity has some weight above Ω_p, our deduction of ε_c close to Ω_p should lead to a saturation plateau of the sum of the energy loss function I_p at a value smaller than the saturation value of I_σ. As we can see in Figure 3a this corresponds to the encountered situation in (TMTSF)$_2$PF$_6$, where I_p saturates at about 1.7 eV^2 and I_σ at around 1.85 eV^2 just above Ω_p. This result is in clear contradiction to the trend found by Jacobsen [2], where: $I_\sigma^{sat} < I_p^{sat} \cong \omega_p^2$.

Our value of ω_p is significantly larger than the value reported by previous authors. Given the overall consistency of our results, we believe that it is more accurate. This implies that the actual hopping integral along the stacks is larger than that deduced from previous studies (250 meV). For instance, in the context of a simple tight binding Hamiltonian, this would lead to a hopping integral of 310 meV. However, this description is too simple, and we now turn to a theoretical description based on a more realistic one-dimensional microscopic model. The relevance of such a model, as well as a discussion of reasonable values of the parameters, can be found in [12]. The basic Hamiltonian can be written

$$H = -t_1 \sum_{i_{even},\sigma} (c^\dagger_{i,\sigma} c_{i+1,\sigma} + h.c.) - t_2 \sum_{i_{odd},\sigma} (c^\dagger_{i,\sigma} c_{i+1,\sigma} + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow} + \sum_i \sum_{l\geq 1} V_l n_i n_{i+l}. \tag{12}$$

$c^\dagger_{i,\sigma}, c_{i,\sigma}$ are fermionic operators creating and annihilating holes in the HOMO of the TMTSF molecules, $n_{i,\sigma} = c^\dagger_{i,\sigma} c_{i,\sigma}$ and $n_i = n_{i\uparrow} + n_{i\downarrow}$. The parameters of this model are: i) a hopping integral t_1 for the short bonds; ii) a hopping integral $t_2 \leq t_1$ for the long bonds; iii) an on-site repulsion U; iv) inter-site repulsions V_l. Besides, there is one electron missing for each pair of TMTSF molecules. So the model is quarter-filled in terms of holes.

For the present purpose, the dimerization can be neglected because its effect is just to shift the spectral weight of the Drude peak at finite but small frequencies, so that this has no detectable effect on the sum rule at energy scales of the order of the bandwidth. Besides, these sum rules are mostly dependent on parameters that influence the kinetic energy, and for a quarter-filled system, the main effect comes from the on-site repulsion U and the repulsion between first neighbors $V \equiv V_1$. So in this section we will work with the extended Hubbard model defined by:

$$H = -t \sum_{i,\sigma} (c^\dagger_{i,\sigma} c_{i+1,\sigma} + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow} + V \sum_i n_i n_{i+1}. \tag{13}$$

Following the previous remark concerning the value of the hopping integral, t should be considered as an adjustable parameter. The other parameters are not known accurately. In particular, the determination based on Jacobsen's analysis of the sum rules [13] must be abandoned on the basis of the new data and interpretation presented in this paper. However, a number of other experiments point toward the following estimates: $U/t \cong 5$ and $V/t \cong 2$. These parameters put the non-dimerized system close to the metal-insulator transition [14] and are thus consistent with a Luttinger liquid exponent K_ρ close to 0.25, in agreement with the interpretation of NMR and transport data [15].

The optical conductivity can be calculated using standard numerical techniques based on the Lanczos algorithm that allows to diagonalize the Hamiltonian on small clusters [16] and to calculate the current-current correlation function. As we use a Hamiltonian without the long-range part of the Coulomb repulsion, *i.e.* with a screened interaction, this current-current correlation function gives a good estimate of the response to the total field, hence of the real part of the optical conductivity. The results for a 16-site cluster are presented in Figure 2b, together with the loss function deduced from a Kramers-Kronig analysis of the real part of the conductivity. The hopping integral was adjusted to reproduce the correct value of ω_p, leading to the estimate $t = 355$ meV. Note that this value is larger than the value one would get for a non-interacting system (310 meV) because the correlations reduce the kinetic energy, hence the plasma frequency. The similarity with the main features of the experimental results of Figure 2a is rather striking. Some incoherent weight shows up however in the conductivity around the plasma edge frequency, and according to the discussion of the previous section, this can have important consequences for the comparison of the *partial* sum rules.

To know whether the 16-site cluster is representative of the thermodynamic limit, we have made a finite-size scaling analysis of the relative weight of the Drude peak in the optical conductivity. The results are shown in Figure 4. As can be seen, the scaling is reasonably accurate, and the Drude peak should represent 96% of the spectral weight in the thermodynamic limit, to be compared to 96.4% for the 16-site cluster. So the 16-site cluster should provide a reasonable description of the thermodynamic limit. Note that the inclusion of the repulsion between neighbours is quite important to get a non-negligible incoherent weight.

The integral of the conductivity I_σ and of the energy loss function I_p are depicted in Figure 3b. There is of course no need to include an ε_c in this analysis because we are considering a single band. These results can be

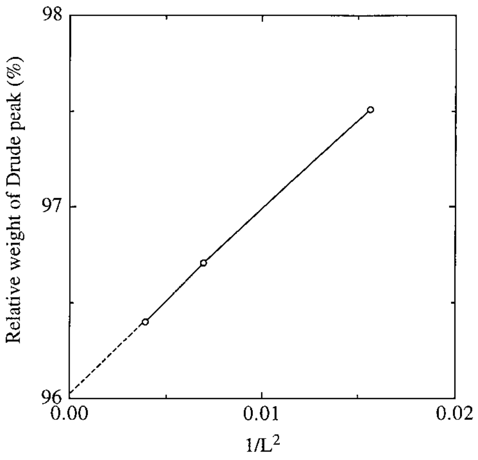

Fig. 4. Finite-size analysis of the relative weight of the Drude peak for the model of Figure 2b.

compared to the experimental data (Fig. 3a) if the experimental energy loss function is multiplied by the appropriate ε_c^2 to account for the high energy contributions to the optical conductivity. As we can see, the sum rules are only consistent at a frequency about twice as large as Ω_p. Just above Ω_p, the integral of the energy loss function is below the integral of the conductivity, in qualitative agreement with the experimental results. This gives further support to the model of equation (13) with values of the ratios $U/t = 5$ and $V/t = 2$ since this progressive saturation between ω_p and $2\omega_p$ of the *partial* sum rule of the loss function is a direct consequence of the presence of weight around ω_p due to these correlation terms.

We conclude that both longitudinal and transverse optical sum rules can be satisfied. Crucial seems to be the correct and precise determination of the contribution to the optical dielectric function due to the high frequency residual spectral weight, which is incorporated by the parameter ε_c. The meaning of the suggested theoretical comparison could be further enhanced by reducing the uncertainty in the determination of ε_c. We have shown with the example on $(TMTSF)_2PF_6$ that this can be in principle achieved by the investigation of the optical excitation spectrum on a frequency spectral range extending over several decades.

The authors wish to thank T. Giamarchi, G. Grüner, K. Penc and P. Wachter for helpful discussions. L.D. and V.V. acknowledge the important financial support by the Swiss National Foundation for the Scientific Research. The numerical calculations have been performed on the C-94 and C-98 of IDRIS (Orsay).

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