Sum rule for the optical absorption of an interacting many-polaron gas

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Abstract. A sum rule for the first frequency moment of the optical absorption of a many-polaron system is derived, taking into account many-body effects in the system of constituent charge carriers of the many-polaron system. In our expression for the sum rule, the electron-phonon coupling and the many-body effects in the electron (or hole) system formally decouple, so that the many-body effects can be treated to the desired level of approximation by the choice of the dynamical structure factor of the electron (hole) gas. We calculate correction factors to take into account both low and high experimental cutoff frequencies.

PACS. 74.25.Gz Optical properties – 74.25.Kc Phonons – 74.72.Jt Other cuprates

1 Introduction

Sum rules have proven to be a powerful tool in the analysis of optical spectra [1]. They provide expressions relating a frequency moment of the optical absorption spectrum to characteristic properties of a material such as its plasma frequency. Sum rules provide useful checks (on the optical properties) both for theory and for experiment.

The goal of this paper is to derive a sum rule for the normalized first frequency moment of the optical absorption of a gas of continuum (Fröhlich) polarons, including many-body effects between the charge carriers in this many-polaron gas. The normalized first frequency moment of the optical absorption is obtained from the optical conductivity $\mathrm{Re}[\sigma(\omega)]$ by

$$\langle\omega\rangle = \frac{\displaystyle\int_0^\infty \omega\,\mathrm{Re}[\sigma(\omega)]\mathrm{d}\omega}{\displaystyle\int_0^\infty \mathrm{Re}[\sigma(\omega)]\mathrm{d}\omega}\;. \qquad (1)$$

This quantity is experimentally accessible: for example Calvani and co-workers have determined $\langle\omega\rangle$ as a function of doping (density) for the optical absorption bands attributed to polarons in a family of high-temperature cuprate superconductors [2]. As such, the sum rule (1) for $\langle\omega\rangle$ applied to the polaron gas including many-body effects (such as interaction, screening, occupational effects of Fermi statistics, ...) will prove a useful tool for the analysis of infrared spectra of such high-T_c materials. The

study of infrared spectra in the framework of polaron theory can complement other indications of the presence of Fröhlich polarons and bipolarons [3] in high-T_c materials, thus providing a more solid ground for hypotheses involving polarons and bipolarons in the mechanism of high-temperature superconductivity [4].

In the derivation presented here (in Sect. 2), the many-body effects in the system of constituent electrons (holes) are described through the dynamical structure factor of the charge carrier gas, and they are formally factored out from the charge carrier-phonon interactions, which allows for a great deal of flexibility in treating the many-polaron gas.

We demonstrate how to circumvent two particular difficulties which complicate the practical use of many-polaron sum rules in a comparison with experiment. The first difficulty is related to the presence of a delta function contribution at zero frequency and $T = 0$ in the theoretical optical absorption spectrum of the polaron gas, at temperature zero [5]. Such a sharp zero-frequency feature can be overlooked by experiment, and we derive a correction factor – which turns out to be related to the density dependent effective polaron mass – to compensate for the inability to detect the delta function contribution. This correction factor is the subject of Section 3. A second difficulty is that the theoretical many-polaron optical absorption spectrum contains a slowly decaying high-frequency tail which carries a substantial fraction of the spectral weight of the polaron optical absorption. In Section 4 we discuss the influence of a high-frequency cutoff on the first frequency moment and propose a formula useful to extrapolate experimental data of the polaron optical absorption band at higher frequencies.

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$$\int_0^\infty \mathrm{e}^{\mathrm{i}\omega t}\,\langle[F_x(t),F_x(0)]\rangle\,\mathrm{d}t = \sum_n \int_0^\infty \mathrm{d}t\,\mathrm{e}^{\mathrm{i}\omega t-\delta t}\left(\left\langle\Psi_0\left|\mathrm{e}^{-\mathrm{i}Ht/\hbar}F_x\mathrm{e}^{\mathrm{i}Ht/\hbar}\right|\Psi_n\right\rangle\langle\Psi_n\,|F_x|\,\Psi_0\rangle\right.$$

$$\left.-\langle\Psi_0\,|F_x|\,\Psi_n\rangle\left\langle\Psi_n\left|\mathrm{e}^{-\mathrm{i}Ht/\hbar}F_x\mathrm{e}^{\mathrm{i}Ht/\hbar}\right|\Psi_0\right\rangle\right) \tag{4}$$

$$= \sum_n \mathrm{i}\,|\langle\Psi_0\,|F_x|\,\Psi_n\rangle|^2\left(\frac{1}{\omega+(E_n-E_0)/\hbar+\mathrm{i}\delta}\frac{1}{\omega-(E_n-E_0)/\hbar+\mathrm{i}\delta}\right). \tag{5}$$

2 First frequency moment of the optical absorption of a many-polaron gas

2.1 Exact expression

The optical absorption coefficient is proportional to the real part of the optical conductivity, $\mathrm{Re}[\sigma(\omega)]$. To evaluate expression (1), both the zeroth and the first frequency moment of the real part of the optical conductivity have to be determined. The zeroth moment is easily related to the plasma frequency through the f-sum rule [6] :

$$\int_0^\infty \mathrm{Re}[\sigma(\omega)]\mathrm{d}\omega = \frac{\pi N e^2}{2m_\mathrm{b}}\,. \tag{2}$$

In this expression, N is the number of polarons per unit volume, m_b is the band mass of the constituent electron (or hole) of the polaron, and e is the electron charge. To obtain the first frequency moment of $\mathrm{Re}[\sigma(\omega)]$, the optical conductivity is expressed as a current-current correlation function according to the Kubo formalism of linear response theory (see *e.g.* [7]). Integrating twice by parts, the current-current correlation function can be transformed into a force-force correlation function [8]. This leads to:

$$\mathrm{Re}[\sigma(\omega)] = \frac{1}{V\hbar\omega^3}\frac{e^2}{m_\mathrm{b}^2}\,\mathrm{Re}\left\{\int_0^\infty \mathrm{e}^{\mathrm{i}\omega t}\,\langle[F_x(t),F_x(0)]\rangle\,\mathrm{d}t\right\}. \tag{3}$$

where V is the volume (chosen equal to unity in what follows) and F_x is the x-component of the force operator $\mathbf{F}=(\mathrm{i}/\hbar)\left[H,\sum_{j=1}^N \mathbf{p}_j\right]$. To formulate the sum rule, the force-force correlation function is rewritten in spectral representation. For this purpose, we introduce a set $\{\Psi_n\}$ of basis states with corresponding energies $\{E_n\}$, of which Ψ_0 is the ground state with ground state energy E_0. The spectral representation then becomes:

see equations (4, 5) above.

Denoting $(E_n - E_0)/\hbar$ as ω_{n0}, one arrives at the following expression for the first moment of the optical conductivity:

$$\int_0^\infty \omega\,\mathrm{Re}[\sigma(\omega)] = \int_0^\infty \mathrm{d}\omega\frac{e^2}{m_\mathrm{b}^2\hbar\omega^2}\,\mathrm{Re}\left[\sum_n \mathrm{i}\,|\langle\Psi_0\,|F_x|\,\Psi_n\rangle|^2\right.$$

$$\times\left(\frac{1}{\omega+(E_n-E_0)/\hbar+\mathrm{i}\delta}\right.$$

$$\left.\left.-\frac{1}{\omega-(E_n-E_0)/\hbar+\mathrm{i}\delta}\right)\right] \tag{6}$$

$$= \frac{\pi e^2}{m_\mathrm{b}^2\hbar}\sum_n\frac{|\langle\Psi_0\,|F_x|\,\Psi_n\rangle|^2}{\omega_{n0}^2}. \tag{7}$$

The result for the normalized first frequency moment of the optical conductivity is found from the expressions (7) and (2):

$$\langle\omega\rangle = \frac{\displaystyle\int_0^\infty \omega\,\mathrm{Re}[\sigma(\omega)]\mathrm{d}\omega}{\displaystyle\int_0^\infty \mathrm{Re}[\sigma(\omega)]\mathrm{d}\omega} = \frac{2}{N\hbar m_\mathrm{b}}\sum_n\left|\frac{\langle\Psi_0\,|F_x|\,\Psi_n\rangle}{\omega_{n0}}\right|^2. \tag{8}$$

To obtain a useful expression, the resummation over the unspecified set of basis functions $\{\Psi_n\}$ should still be performed. This is done by using the integral representation

$$\frac{1}{\omega_{n0}} = \lim_{\delta\to+0}\left(\mathrm{i}\int_0^\infty \mathrm{e}^{\mathrm{i}\omega_{n0}t-\delta t}\mathrm{d}t\right) \tag{9}$$

such that

$$\langle\omega\rangle = \frac{2}{N\hbar m_\mathrm{b}}\lim_{\delta\to+0}\int_0^\infty \mathrm{d}t\int_0^\infty \mathrm{d}s$$

$$\times\sum_n \langle\Psi_0\,|F_x(t)|\,\Psi_n\rangle\langle\Psi_n\,|F_x(s)|\,\Psi_0\rangle\,\mathrm{e}^{-\delta(t+s)}$$

$$= \frac{2}{N\hbar m_\mathrm{b}}$$

$$\times\lim_{\delta\to+0}2\,\mathrm{Re}\left[\int_0^\infty \mathrm{d}t\int_0^t \mathrm{d}s\,\langle\Psi_0\,|F_x(t)F_x(s)|\,\Psi_0\rangle\,\mathrm{e}^{-\delta(t+s)}\right], \tag{10}$$

$$\langle\omega\rangle = \frac{2}{N\hbar m_{\mathrm{b}}} \lim_{\delta\to+0} \left\{ \frac{1}{\delta} \int\limits_0^\infty \mathrm{d}t e^{-\delta t} \sum_{\mathbf{k},\mathbf{k}'} k_x k_x' \left\langle \varphi_{\mathrm{el}} \left| \left\langle \phi \left| \begin{array}{c} [V_{\mathbf{k}} b_{\mathbf{k}}(t)\rho_{\mathbf{k}}(t) - V_{\mathbf{k}'}^* b_{\mathbf{k}'}^+(t)\rho_{\mathbf{k}'}^+(t)] \\ \times [V_{\mathbf{k}'} b_{\mathbf{k}'}(0)\rho_{\mathbf{k}'}(0) - V_{\mathbf{k}'}^* b_{\mathbf{k}'}^+(0)\rho_{\mathbf{k}'}^+(0)] \end{array} \right| \phi \right\rangle \right| \varphi_{\mathrm{el}} \right\rangle \right\}. \qquad (14)$$

where the Hermiticity of the force operator was used. If the response is time translational invariant (meaning that the response is only dependent on the time lapse since perturbation), the final result can be written as:

$$\langle\omega\rangle = \frac{2}{N\hbar m_{\mathrm{b}}} \lim_{\delta\to+0} \left[\frac{1}{\delta} \int\limits_0^\infty \langle \Psi_0 | F_x(t) F_x(0) | \Psi_0 \rangle \, e^{-\delta t} \mathrm{d}t \right]. \tag{11}$$

Expression (11) is an exact expression, within the assumptions of linear response theory, for the sum rule for the normalized first frequency moment of the optical absorption of the many-polaron gas.

2.2 The interacting Fröhlich many-polaron gas

Throughout this paper, we consider polarons in the continuum or Fröhlich approximation [9]. The system of N interacting Fröhlich polarons is then described by the Hamiltonian

$$H = \sum_{j=1}^N \frac{\mathbf{p}_j^2}{2m_{\mathrm{b}}} + \sum_{\mathbf{k}} \hbar\omega_{\mathrm{LO}} b_{\mathbf{k}}^+ b_{\mathbf{k}}$$

$$+ \sum_{\mathbf{k}} \left(V_{\mathbf{k}} b_{\mathbf{k}}\rho_{\mathbf{k}} + V_{\mathbf{k}}^* b_{\mathbf{k}}^+ \rho_{\mathbf{k}}^+ \right) + \frac{1}{2}\sum_{j=1}^N \sum_{\substack{l\neq j=1}}^N \frac{e^2/\varepsilon_\infty}{|\mathbf{r}_j - \mathbf{r}_l|} \quad (13)$$

where $\mathbf{r}_j, \mathbf{p}_j$ are the position and momentum operators of charge carrier j with band mass m_{b} and change $\pm e$, and $b_{\mathbf{k}}^+$, $b_{\mathbf{k}}$ are the creation and annihilation operators of a longitudinal optical (LO) phonon with frequency ω_{LO}, $\rho_{\mathbf{k}} = \sum_{j=1}^N \exp\{\mathrm{i}\mathbf{k}\cdot\mathbf{r}_j\}$ is the density operator of the charge carriers, $V_{\mathbf{k}}$ is the interaction amplitude matrix element characterizing the interaction between the charge carriers and the LO phonons, and ε_∞ is the permittivity of the medium. The total force operator for the many-polaron gas, with both charge carrier-phonon interaction and Coulomb interaction between the charge carriers taken into account, takes the form

$$\mathbf{F} = \frac{\mathrm{i}}{\hbar} \left[H, \sum_{j=1}^N \mathbf{p}_j \right] = -\mathrm{i}\sum_{\mathbf{k}} \mathbf{k}(V_{\mathbf{k}} b_{\mathbf{k}}\rho_{\mathbf{k}} - V_{\mathbf{k}}^* b_{\mathbf{k}}^+ \rho_{\mathbf{k}}^+),$$

so that in the force-force correlation $F_x(t)F_x(0)$, a factor $|V_{\mathbf{k}}|^2 \propto \alpha$ is present and formally factors out of the expression for $\langle\omega\rangle$ and $\mathrm{Re}[\sigma(\omega)]$ in equations (11) and (3),

respectively. Herein lies an advantage of working with the force-force correlation as compared to the current-current correlation, especially at weak coupling. Thus, to calculate $\langle\omega\rangle$ (11) to lowest order in the charge carrier-phonon interaction strength α it is sufficient to evaluate the (α-independent) expectation value of the force-force correlation in a product wave function $|\phi\rangle |\varphi_{\mathrm{el}}\rangle$ where $|\varphi_{\mathrm{el}}\rangle$ represents the ground-state wave function for charge carriers and $|\phi\rangle$ is the phonon vacuum:

see equation (14) above.

The expectation value with respect to the phonon vacuum can be calculated to order α and results in

$$\langle\omega\rangle = -\frac{2}{N\hbar m_{\mathrm{b}}} \sum_{\mathbf{k}} k_x^2 |V_{\mathbf{k}}|^2$$

$$\times \lim_{\delta\to+0} \left\{ \frac{1}{\delta} \int\limits_0^\infty \mathrm{d}t \, e^{\mathrm{i}\omega_{\mathrm{LO}}t} e^{-\delta t} \left\langle \varphi_{\mathrm{el}} \left| \rho_{\mathbf{k}}(t)\rho_{\mathbf{k}}^+(0) \right| \varphi_{\mathrm{el}} \right\rangle \right\}. \tag{15}$$

The remaining expectation value is the density-density Green's function $G(\mathbf{k}, t) = -\mathrm{i}\,\Theta(t) \langle \varphi_{\mathrm{el}} | \rho_{\mathbf{k}}(t) \times \rho_{\mathbf{k}}^+(0) | \varphi_{\mathrm{el}} \rangle / N$ so that

$$\langle\omega\rangle = -\frac{2}{\hbar m_{\mathrm{b}}} \sum_{\mathbf{k}} k_x^2 |V_{\mathbf{k}}|^2 \lim_{\delta\to+0} \left\{ \frac{1}{\delta} G(\mathbf{k}, -\omega_{\mathrm{LO}} + \mathrm{i}\delta) \right\}. \tag{16}$$

Introducing the dynamical structure factor $S(k, \omega)$ as the spectral density function of the retarded density-density Green's function G_{R} of the electron (or hole) gas, we arrive at:

$$\langle\omega\rangle = -\frac{2}{\hbar m_{\mathrm{b}}} \sum_{\mathbf{k}} k_x^2 |V_{\mathbf{k}}|^2 \int\limits_0^\infty \mathrm{d}\omega \, \frac{S(\mathbf{k}, \omega - \omega_{\mathrm{LO}})}{\omega^2}. \tag{17}$$

The modulus square of the Fröhlich interaction amplitude is given by

$$|V_{\mathbf{k}}|^2 = \begin{cases} \dfrac{(\hbar\omega_{\mathrm{LO}})^2}{k^2} \dfrac{4\pi\alpha}{V} \sqrt{\dfrac{\hbar}{2m_b\omega_{\mathrm{LO}}}} & \text{in 3D} \\[3ex] \dfrac{(\hbar\omega_{\mathrm{LO}})^2}{k} \dfrac{2\pi\alpha}{A} \sqrt{\dfrac{\hbar}{2m_b\omega_{\mathrm{LO}}}} & \text{in 2D,} \end{cases} \tag{18}$$

where V (A) is the volume (surface). This leads to

$$\langle\omega\rangle_{\mathrm{3D}} = -\alpha\omega_{\mathrm{LO}} \frac{\hbar\omega_{\mathrm{LO}}}{m_{\mathrm{b}}} \sqrt{\frac{\hbar}{m_{\mathrm{b}}\omega_{\mathrm{LO}}}} \frac{2\sqrt{2}}{3\pi}$$

$$\times \int\limits_0^\infty \mathrm{d}k \int\limits_0^\infty \mathrm{d}\omega \, k^2 \frac{S_{\mathrm{3D}}(\mathbf{k}, \omega - \omega_{\mathrm{LO}})}{\omega^2}, \quad (19\mathrm{a})$$

and

$$\langle\omega\rangle_{2\mathrm{D}} = -\alpha\omega_{\mathrm{LO}}\frac{\hbar\omega_{\mathrm{LO}}}{m_{\mathrm{b}}}\sqrt{\frac{\hbar}{m_{\mathrm{b}}\omega_{\mathrm{LO}}}}\frac{1}{\sqrt{2}}$$
$$\times\int_0^\infty \mathrm{d}k\int_0^\infty \mathrm{d}\omega\; k^2\frac{S_{2\mathrm{D}}(\mathbf{k},\omega-\omega_{\mathrm{LO}})}{\omega^2}. \quad (19\mathrm{b})$$

The 2D and 3D results are related by the scaling relation

$$\langle\omega\rangle_{2\mathrm{D}}\,(\alpha, S_{2\mathrm{D}}) = \langle\omega\rangle_{3\mathrm{D}}\,(3\pi\alpha/4, S_{3\mathrm{D}}) \quad (19)$$

provided the corresponding 2D or 3D dynamical structure factor of the electron (hole) system is used. Taking the low-density limit of (19), the correct one-polaron scaling relation [10] is retrieved. The present analysis shows that the generalization (19) of the one-polaron scaling relation holds in the many-polaron case, provided that the corresponding 2D or 3D dynamical structure factor is used. Both expressions (19a) and (19b) contain the Fröhlich electron-phonon (or hole-phonon) coupling constant α only as a prefactor: the remaining integrations of (19a) and (19b) depend only on the details of the electron-electron (or hole-hole) many-body effects.

2.3 Results and discussion

For one polaron, the normalized first frequency moments are $\langle\omega\rangle_{3\mathrm{D}} = (2/3)\,\langle\omega\rangle_{2\mathrm{D}}$ and $\langle\omega\rangle_{2\mathrm{D}} = (\pi/2)\alpha\omega_{\mathrm{LO}}$ [11]. These limits correspond to the low density limit of the expressions (19a) and (19b), which is taken by letting $k_{\mathrm{F}} \to 0$. In this limit the dynamical structure factor $S(\mathbf{k},w)$ becomes strongly peaked along $w = \hbar k^2/(2m_{\mathrm{b}})$. In the case of many polarons, many-body effects such as the electron-electron interaction, screening effects, and the occupational effect due to Fermi statistics will play a role.

Figure 1 shows the result obtained from (19a, 19b) for the first frequency moment of the optical absorption in 3D and 2D as a function of density, expressed through the Fermi wave vector k_{F}. For the 3D case, the material parameters of ZnO were used ($\omega_{\mathrm{LO}} = 73.27$ meV, $\varepsilon_\infty = 4.0$, $m_{\mathrm{b}} = 0.24\,m_{\mathrm{e}}$) and for the 2D case, the material parameters for GaAs were taken ($\omega_{\mathrm{LO}} = 36.77$ meV, $\varepsilon_\infty = 10.9$, $m_{\mathrm{b}} = 0.0657\,m_{\mathrm{e}}$) [12]. The limit of low density corresponds to the one polaron-results indicated by arrows starting on the y-axis.

The dashed curve (for 2D) and the dotted curve (for 3D) were calculated with the Hartree-Fock structure factor of the electron (hole) system. The increase of $\langle\omega\rangle$ in the region $k_{\mathrm{F}} \lesssim 1\,\sqrt{m_{\mathrm{b}}\omega_{\mathrm{LO}}/\hbar}$ shows an initial shift in spectral density towards higher frequencies. The full curve (for 2D) and the dash-dotted curve (for 3D) are the result obtained by substituting the RPA structure factor of the electron (hole) system in expressions (19a, 19b). When the many-body effects are taken into account on the level of the RPA, they result in a monotonously decreasing value
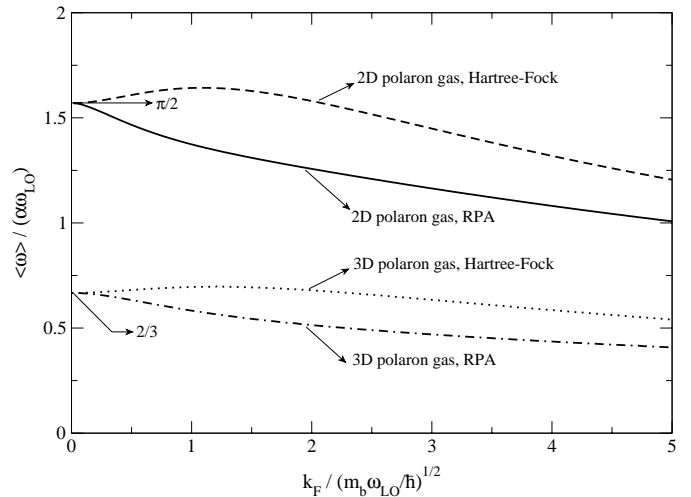

Fig. 1. The normalized first frequency moment of the optical absorption of a gas of polarons including many-body effects in the system of constituent charge carriers, as obtained with the present treatment, is shown as a function of density, expressed through the Fermi wave vector k_{F} in polaron units. The top two curves are for the 2D polaron gas (with parameters corresponding to GaAs: $\omega_{\mathrm{LO}} = 36.77$ meV, $\varepsilon_\infty = 10.9$, $m_{\mathrm{b}} = 0.0657\,m_{\mathrm{e}}$), and the bottom two curves are the results for the 3D case (with parameters representative of ZnO: $\omega_{\mathrm{LO}} = 73.27$ meV, $\varepsilon_\infty = 4.0$, $m_{\mathrm{b}} = 0.24\,m_{\mathrm{e}}$).

of the normalized first frequency moment as a function of density.

The results obtained from the sum rule expressions (19a, 19b) are in agreement with the result [13] obtained by direct integration of the many-polaron optical absorption spectrum calculated with the variational ground state wave function of Lemmens, Brosens and Devreese [14]. Note that the derivation presented in the present paper does not rely on the variational wave function used by LDB. The use of the force-force correlation function introduces already a factor α in expression (14), so that it is sufficient to calculate the expectation value in (14) with respect to the phonon vacuum in order to obtain the first frequency moment up to order α in the charge carrier-phonon coupling strength.

Concerning the electron-phonon coupling strength dependence, it is clear that our results are valid at low coupling (i.e. to order α). Yet this does not mean that the present theory would also be restricted to low charge carrier density. The density dependent effects are described through the dynamical structure factor of the electron (hole) gas. In as far as the *density dependence* of the results are concerned, the validity of our treatment is determined by the validity of the dynamical structure factor used in the calculation. The dynamical structure factors used here are valid for electron gases and do not describe electron solids – thus polaronic effects in such electron solids (the polaronic Wigner crystal [15]) are not described here.

3 Zeroth frequency moment and many-body effects in the polaron mass

In the polaron optical absorption spectra at zero temperature, a delta-function contribution is present at $\omega = 0$ [5]. This feature of the polaron spectrum will, at low temperatures, elude experimental detection since all infrared absorption experiments have naturally a lowest measurable frequency. Nevertheless, this delta-function contribution will play a role in the zeroth frequency moment of the optical absorption and hence in all normalized frequency moments. Note that as the temperature is raised, this DC delta-function contribution will substantially widen and become detectable [16]. At temperature zero, we must rely on an adaptation of the f-sum rule for one polaron, derived in [5]:

$$\frac{N\pi e^2}{2m^*} + \int_{\omega_{LO}}^{\infty} \text{Re}[\sigma(\omega)]d\omega = \frac{N\pi e^2}{2m_b}. \tag{20}$$

In this expression, m^* is the polaron effective mass. At very low temperature, the integral in the left-hand-side of (20) can be determined experimentally or used to derive the effective polaron mass from many-polaron optical absorption theories such as [13,17]. The LO phonon frequency ω_{LO} is typically a few hundred cm^{-1}, obviously within the range of far infrared spectroscopy. Expression (20) allows then to determine the polaron mass from the optical conductivity:

$$1 - \frac{m_b}{m^*} = \frac{2m_b}{N\pi e^2} \int_{\omega_{LO}}^{\infty} \text{Re}[\sigma(\omega)]d\omega. \tag{21}$$

The evolution of the polaron effective mass as a function of the density of the interacting many-polaron gas is shown in Figure 2. To obtain the polaron effective mass, the sum rule (21) was applied to the many-polaron optical absorption spectrum calculated in [13]:

$$\text{Re}[\sigma(\omega)] = \frac{n}{\hbar\omega^3} \frac{e^2}{m_b^2} \sum_{\mathbf{k}} k_x^2 |V_{\mathbf{k}}|^2 S(\mathbf{k}, \omega - \omega_{LO}). \tag{22}$$

The result for $(m^*/m_b - 1)/\alpha$ is shown in Figure 2 as a function of density expressed *via* k_F. For low density the effective mass naturally converges to its one-polaron value in the weak and intermediate electron-phonon coupling limit, given by $m^* = m_b(1 + \alpha/6)$ in 3D and by $m^* = m_b(1 + \pi\alpha/8)$ in 2D [11]. Both dashed curves were calculated using the Hartree-Fock structure factor. The shift of spectral weight towards higher frequencies, due to occupation effects, is reflected here as an initial increase in the effective polaron mass. The full and dash-dotted curves show the result using the RPA structure factor. The resulting effective polaron mass is a monotonously decreasing function of density.

The normalized first frequency moment of the optical absorption, *excluding* the delta-function contribution,
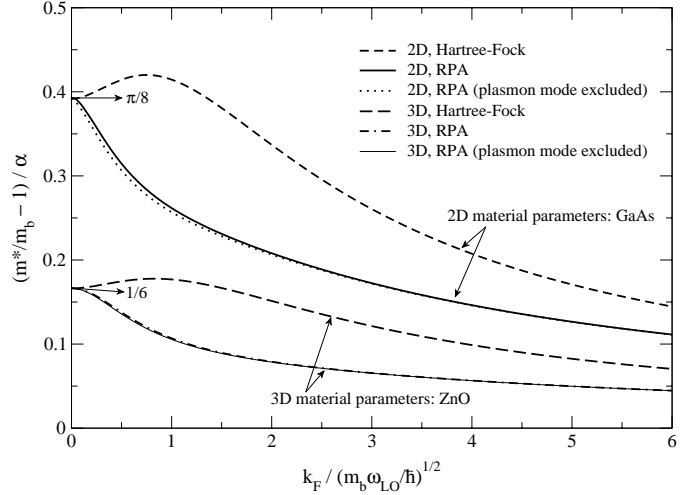

Fig. 2. The effective polaron mass in the many-polaron system is shown as a function of density, for the 2D case (GaAs parameters: $\omega_{LO} = 36.77$ meV, $\varepsilon_\infty = 10.9$, $m_b = 0.0657\,m_e$) and the 3D case (ZnO parameters: $\omega_{LO} = 73.27$ meV, $\varepsilon_\infty = 4.0$, $m_b = 0.24\,m_e$), and for different approximations (Hartree-Fock and RPA) to take the many-body effects into account.

$\langle\omega\rangle_0$, is accessible to experiment, using:

$$\langle\omega\rangle_0 = \frac{\int_{\omega_{LO}}^{\infty} \omega\, \text{Re}[\sigma(\omega)]d\omega}{\int_{\omega_{LO}}^{\infty} \text{Re}[\sigma(\omega)]d\omega}. \tag{23}$$

The question of the high-frequency cutoff in the experiment will be discussed as a further refinement in the next section. The numerator in expression (23) is still coincident with the numerator in (1) for many-polaron optical conductivity. Based on (21) for the denominator, we obtain

$$\langle\omega\rangle = \left(1 - \frac{m_b}{m^*}\right)\langle\omega\rangle_0. \tag{24}$$

As a consequence, to obtain the normalized first frequency moment $\langle\omega\rangle$ of the optical absorption including the delta function contribution, one has to correct the result $\langle\omega\rangle_0$ obtained without the delta function, by multiplying it with a correction factor $\left(1 - \frac{m_b}{m^*}\right)$.

4 High frequency tail of the many-polaron optical absorption spectrum

An upper bound ω_{max} of the integration domain (cutoff frequency) is necessarily present in the experimental determination of the frequency integrals in (23). Thus the
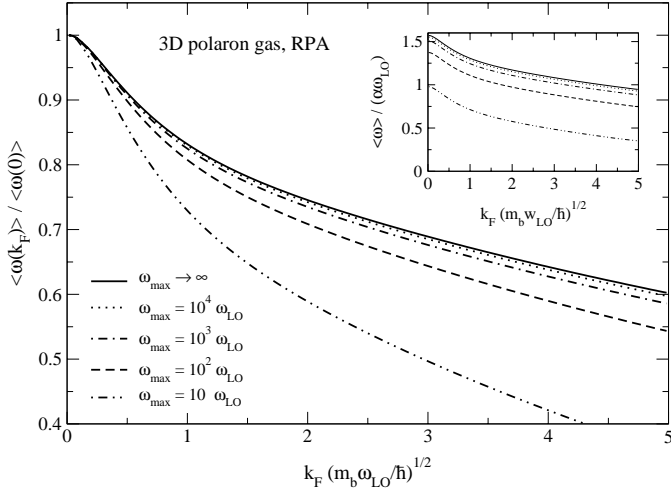
Fig. 3. The effect of introducing an upper limit (a cutoff, ω_{max}) to the integration domain in the expression for the first frequency moment $\langle\omega\rangle$ of the optical absorption of the polaron gas is shown in this figure. The different curves show $\langle\omega\rangle / \langle\omega\rangle_{k_F=0}$ as a function of the Fermi wave vector for the set of cutoff values listed in the legend, obtained from direct integration of the many-polaron spectrum as calculated in [13]. The full curve shows the sum rule result (without cutoff) in the present treatment. In the inset, the absolute value of $\langle\omega\rangle$ is shown as a function of the Fermi wave vector, for the same set of cutoff values.

experimentally accessible quantity for the experiment is

$$\langle\omega\rangle_{\exp} = \frac{\displaystyle\int_{\omega_{LO}}^{\omega_{max}} \omega \operatorname{Re}[\sigma(\omega)]\mathrm{d}\omega}{\displaystyle\int_{\omega_{LO}}^{\omega_{max}} \operatorname{Re}[\sigma(\omega)]\mathrm{d}\omega}, \tag{25}$$

where ω_{max} is typically of the order of $10\,000$ cm^{-1} [2]. This upper bound will influence the experimental value of the first moment $\langle\omega\rangle$ since a high-frequency tail ($\omega > \omega_{max}$) is present in the optical absorption spectrum of polarons.

Figure 3 illustrates this cutoff problem. The full curve in Figure 3 shows the sum rule result for the normalized first frequency moment of the optical absorption of the many-polaron system as a function of the Fermi wave vector. The other curves show the results, obtained by direct integration of the spectrum calculated with the variational ground state wave function of reference [13], using different values for the cutoff parameter ω_{max}. Already at $\omega_{max} = 10\omega_{LO}$ a considerable difference is found between the result with cutoff and the result without cutoff.

For energy transfers $\hbar\omega$ much larger than the Hartree-Fock exchange energy of the many-polaron gas, the optical absorption will not be strongly modified by many-body effects. This property is used in the optical absorption measurements to extrapolate the high-frequency measurements of the reflectance by using a free-electron response

(as described *e.g.* in Ref. [18]). For the many-polaron optical absorption in the high frequency limit (for example in 3D) we find [13]:

$$\lim_{\omega\to\infty} \operatorname{Re}[\sigma(\omega)] = \frac{Ne^2}{m_b}\frac{2}{3}\alpha\frac{\sqrt{\omega-1}}{\omega^3}, \tag{26}$$

so that

$$\lim_{\omega_{max}\to\infty} \int_{\omega_{max}}^{\infty} \omega \operatorname{Re}[\sigma(\omega)] =$$
$$\frac{Ne^2}{m_b}\frac{2}{3}\alpha\left[\frac{\sqrt{\omega_{max}-1}}{\omega_{max}} + \arcsin\left(\frac{1}{\sqrt{\omega_{max}}}\right)\right]. \tag{27}$$

For the first frequency moment this leads to

$$\langle\omega\rangle = \lim_{\omega_{max}\to\infty}\left\{ \langle\omega\rangle_{\exp}(\omega_{max})\right.$$
$$\left. + \frac{4}{3\pi}\alpha\left[\frac{\sqrt{\omega_{max}-1}}{\omega_{max}} + \arcsin\left(\frac{1}{\sqrt{\omega_{max}}}\right)\right]\right\}. \tag{28}$$

Expression (28) allows one to correct the error made by using a cutoff frequency in determining the experimental value for the first frequency moment of the polaron optical absorption. How large does ω_{max} have to be for the limit (28) to be accurate? Naturally, this depends on the density. The dynamical structure factor of the electron (hole) system reduces to the dynamical structure factor of a one-particle system for $k \gg 2k_F$, as does the static structure factor [19]. At these values of the wave vector, the only frequency for which the dynamical structure factor $S(k, \omega - \omega_{LO})$ differs substantially from zero is for $\omega - \omega_{LO} = k^2/2$. Hence, we can estimate that the many-polaron optical absorption will be well approximated by the one-polaron optical absorption only for frequencies $\omega \gg 2k_F^2 + \omega_{LO}$. This is also the value of ω_{max} which should be used as high-frequency cutoff in the experiments determining $\langle\omega\rangle_{\exp}$. For example, in ZnO ($\omega_{LO} = 73.27$ meV, $\varepsilon_\infty = 4.0$, $m_b = 0.24\,m_e$) with a charge carrier density of $n = 10^{20}$ cm^{-3}, this would correspond to $\omega_{max} \approx 18.9\,\omega_{LO}$ or $\omega_{max} \approx 11\,000$ cm^{-1}.

5 Conclusion

Expressing the optical conductivity as a force-force correlation function, a closed expression is obtained for the sum rule of the normalized first moment of a many-polaron system, in such a way that the electron-phonon coupling and the many-body effects of the electron (hole) system formally decouple. This procedure allows one to treat the many-body effects in the system of charge carriers using any desired approximation (Hartree-Fock, RPA, ...). The results obtained by the sum rule derived here are valid for a many-polaron system with weak electron-phonon coupling and at temperature zero.

Experimental data are characterized by a cutoff frequency both for low and high frequencies This in general complicates the use of any sum rule, since its evaluation in principle requires a summation over all frequencies, $\omega = 0...\infty$. To overcome this problem we introduced formula (28) for many-polaron optical absorption spectra at high frequencies, and discussed up to which frequency experimental data have to be available so that this extrapolation formula is useful. At zero temperature, a delta function contribution is present at $\omega = 0$ in the many-polaron optical absorption spectrum [5]. The spectral weight of this delta function contribution is related to the polaron mass [5], which in its turn can be derived from a measurement of the zeroth frequency moment of the many-polaron optical absorption.

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