Chapter 9
Optical Properties of Correlated Electrons

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Abstract Optical spectra provide a versatile tool for studying the electronic properties of matter. In addition, the absolute spectral weight of an optical spectrum reveals optical sum-rules, which are one of the most powerful tools of experimental and theoretical physics providing access to deeply rooted quantities such as the effective mass of the charge carriers and their kinetic energy. The formalism for the optical conductivity of correlated electrons is presented in this chapter for general values of the inverse wavelength q and general band dispersion ϵ_k of the electrons. The corresponding sumrule is found to have a characteristic q-dependence for the nearest-neighbour tight binding model, causing in this case a vanishing of spectral weight for q at the Brillouin-zone boundary, i.e. for $qa = \pi$. These findings are of possible importance for k-resolved infrared spectroscopy, a technique which is in full development at the moment.

In the treasure trove of correlated matter lurk great opportunities for novel phases of matter, including various different forms of quantum magnetism, unconventional- and high-temperature superconductivity, and many other forms of behavior resulting from correlated motion of electrons. The correlated behavior of electrons in the context of quantum many-body systems constitutes one of the remaining challenges of physics. Characterizing and understanding electronic materials requires sophisticated experimental probes [1]. These include advanced optical techniques, including infrared spectroscopy at low frequency and small wavelength [2–4]. Although seemingly contradictory, near-field techniques with nano-scale resolution are emerging and will open the way toward non-local optical spectroscopy, i.e. optical spectroscopy probing $\sigma(\boldsymbol{q}, \omega)$ with finite $\boldsymbol{q}$.

Excellent texts are on the market treating the experimental optical techniques in the long wavelength limit [5] and the optical conductivity of weakly correlated electrons [6]. Also the various aspects optical conductivity of for $q = 0$ has been described in detail in the literature [7–12]. A step-by-step introduction into the optical conductivity of correlated electrons, for general electron dispersion ϵ_k and wavevector $\boldsymbol{q}$ is to our knowledge not presented in the literature. The purpose of this

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book chapter is to provide such a discussion. Since the purpose is only to provide a fairly complete description of the many-body formalism underlying the optical conductivity at finite wavelength and wave-vectors, no attempt is made here to review the vast literature on optical properties of interacting electrons.

9.1 Reflection of Electromagnetic Waves

In this section we discuss the reflection and transmission of electromagnetic (EM) waves at the interface between vacuum and a substance which could be a solid, a liquid or even a gas. With optical spectroscopy one measures the reflection or transmission as a function of frequency ω. A variety of different experimental geometries can be used, depending on the type of sample under investigation, which can be a reflecting surface of a thick crystal, a free standing thin film, or a thin film supported by a substrate. Two frequently used configurations for measuring the optical constants are shown in Fig. 9.1.

Important factors influencing the type of analysis are also the orientation of the crystal or film surface, the angle of incidence of the ray of photons, and the polarization of the light. In most cases only the amplitude of the reflected or refracted light is measured, but sometimes the phase is measured, or the phase difference between two incident rays with different polarization as in ellipsometry. The task of relating the intensity and/or phase of the reflected or refracted light to the dielectric tensor inside

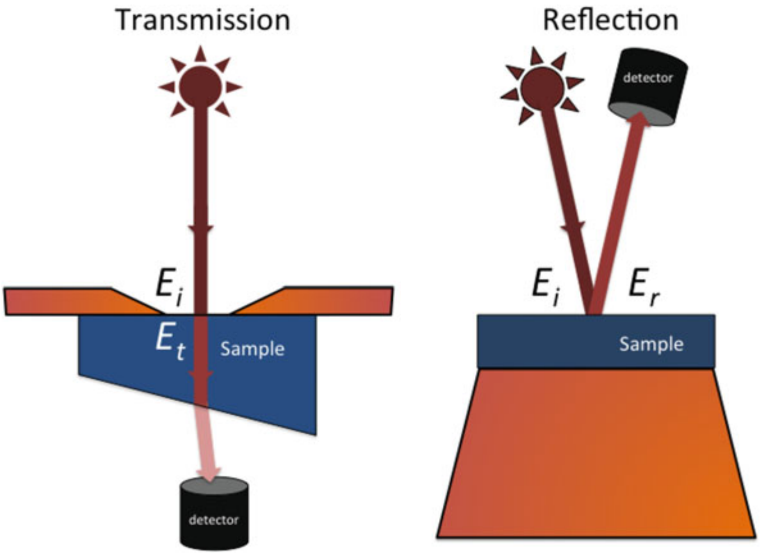

Fig. 9.1 Two commonly used experimental configurations for measuring optical constants

the material boils down to solving the Maxwell equations at the vacuum/sample, sample/substrate interface.

Before attacking the problem of reflection and transmission at an interface, we first give a brief reminder of the macroscopic Maxwell equations inside a medium. The main properties of the medium which controls the propagation of EM-waves are the dielectric constant ϵ, and the magnetic permeability μ

$$\begin{aligned} \boldsymbol{D} &= \epsilon \boldsymbol{E} \\ \boldsymbol{B} &= \mu \boldsymbol{H} \end{aligned} \tag{9.1}$$

We come later in the chapter to the microscopic interpretation of ϵ. We will see among other things, that ϵ depends on $\boldsymbol{q}$ and ω, and that it has a real and imaginary part. The speed of propagation of electromagnetic waves is given by

$$v^2 = \frac{c^2}{\epsilon\mu} \tag{9.2}$$

For $\mathrm{Re}\sqrt{\epsilon\mu} > 1$ the wavelength is compressed compared to what it would be in vacuum for the same frequency, as can be seen from the following expression for the wave-vector inside the medium

$$q^2 = \epsilon\mu\frac{\omega^2}{c^2} \tag{9.3}$$

In vacuum $\mu = \epsilon = 1$. Another consequence of the Maxwell equations is, that the electromagnetic wave has electric and magnetic components, which are given by

$$\begin{aligned} \boldsymbol{E}(z,t) &= \hat{\boldsymbol{x}} E_q e^{i\omega(z\sqrt{\epsilon\mu}/c-t)} \\ \boldsymbol{H}(z,t) &= \hat{\boldsymbol{y}} H_q e^{i\omega(z\sqrt{\epsilon\mu}/c-t)} \\ \mu H_q^2 &= \epsilon E_q^2 \end{aligned} \tag{9.4}$$

We also see now, that the effect of a finite value of $\mathrm{Im}\sqrt{\epsilon\mu}$ is to cause an exponential decay of the wave amplitude from the interface inward to the solid. When one irradiates a perfect interface between the vacuum and a substance with a ray of electromagnetic radiation, part of the light is transmitted to the interior of the substance, and part of the light is reflected. The amplitudes of the incident and reflected rays can measured experimentally. To describe the reflection process one uses the amplitude and phase of the electric field component of the electromagnetic waves just before hitting the sample surface, E_i and just after being reflected E_r, and just after being transmitted inside the solid, E_t. Likewise the corresponding magnetic fields are H_i, H_r and H_t.

In the following table we summarize the consequences of the solution of Maxwell's equations at the interface relevant to this discussion

$$
\begin{aligned}
E_i + E_r &= E_t && \text{(i)} \\
H_i - H_r &= H_t && \text{(ii)} \\
H_i &= E_i && \text{(iii)} \\
H_r &= E_r && \text{(iv)} \\
\mu H_t^2 &= \epsilon E_t^2 && \text{(v)}
\end{aligned}
$$

We divide left and right of (i) and (ii) by E_i and H_i respectively. We then insert (iii), (iv) and (v) in (ii). We furthermore define $r = E_r/E_i$ and $t = E_t/E_i$. The resulting equations are

$$
\begin{aligned}
1 + r &= t && \text{(i)} \\
1 - r &= t\sqrt{\epsilon/\mu} && \text{(ii)}
\end{aligned}
$$

Solution of this system of two equations provides

$$r = \frac{1 - \sqrt{\epsilon/\mu}}{1 + \sqrt{\epsilon/\mu}} \tag{9.5}$$

$$t = \frac{2}{1 + \sqrt{\epsilon/\mu}} \tag{9.6}$$

In the following sections we are going to consider the case of *non-magnetic media* where $\mu(\omega) = 1$. Once $r(\omega)$ has been measured, it then suffices to invert 9.5 to obtain the real and imaginary parts of $\epsilon(\omega)$, which is usually the quantity of interest. An example of this is shown in Fig. 9.2. A similar procedure can be followed when measuring the transmission spectrum through a thin film.

9.2 Optical Conductivity, Current and Electric Field

The optical conductivity expresses the current response to an electric field

$$\boldsymbol{J}(\boldsymbol{r}, t) = \int d^3\boldsymbol{r}' \int_{-\infty}^{t} dt' \boldsymbol{\sigma}(\boldsymbol{r}, \boldsymbol{r}', t - t')\boldsymbol{E}(\boldsymbol{r}', t') \tag{9.7}$$

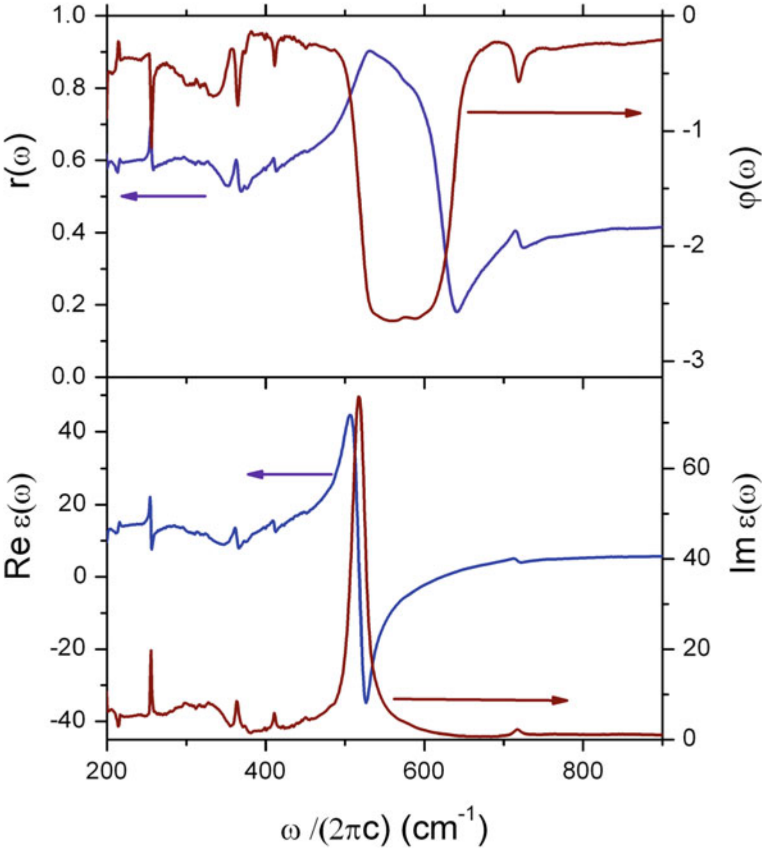

Fig. 9.2 Optical spectrum in the infrared range of the insulating quantum magnet NaV_2O_5. *Top panel* experimental reflectivity and phase. *Lower panel* Real and imaginary parts of the dielectric function $\epsilon(\omega)$. *Source* [13]

We consider the situation where the electric field is described by a plane wave with a wavevector $\boldsymbol{q}$ and a frequency ω, hence $\boldsymbol{E}(r, t) = \boldsymbol{E}_q e^{i(\boldsymbol{q}\cdot\boldsymbol{r}-\omega t)}$, with similar definitions, $\boldsymbol{J}_q$, $\boldsymbol{D}_q$, $\boldsymbol{P}_q$, for the current, displacement field and polarization density. We will assume here, that the fields are sufficiently small, so that we may consider only induced electrical currents which are linearly proportional to the electric fields at each coordinate $\boldsymbol{r}$ of the matter. Consequently the currents oscillate at the same frequency and wavelength as the electric field.

Written in frequency and momentum representation the relation between $\boldsymbol{E}_q$ and $\boldsymbol{J}_q$ is

$$\boldsymbol{J}_q = \boldsymbol{\sigma}(\boldsymbol{q}, \omega)\boldsymbol{E}_q \tag{9.8}$$

The electric field of a plane electromagnetic wave is transverse to the photon momentum. The tensor elements of the optical conductivity which can be measured in an optical experiment, are therefore transverse to the direction of propagation of the electromagnetic wave. The component of the dielectric function describing polarization transverse to the propagation of the EM wave can be expressed in terms of the "optical conductivity", which has a real and an imaginary part. The relation linking the optical conductivity to $\boldsymbol{\epsilon}(\boldsymbol{q}, \omega)$ is

$$\boldsymbol{\epsilon}(\boldsymbol{q}, \omega) = \frac{\boldsymbol{D}_q}{\boldsymbol{E}_q} = 1 + \frac{4\pi i}{\omega}\boldsymbol{\sigma}(\boldsymbol{q}, \omega) \tag{9.9}$$

In a typical optical experiment the photon energy is below 6 eV. In vacuum the photon wave number used in optical experiments is therefore 0.0005 Å^{-1}, or smaller, which is at least three orders of magnitude below the reciprocal lattice constant in a solid. Hence with optical spectroscopy one measures the transverse dielectric function -corresponding to the optical conductivity—in the limit of vanishing wavevector.

9.3 Transverse and Longitudinal Dielectric Function

In the previous sections we have seen that optical experiments measure the transverse dielectric function $\epsilon_t(\boldsymbol{q}, \omega)$. This can for example be done by analyzing the reflection coefficient at a sample-vacuum interface. The transverse nature of electromagnetic waves makes that the component of $\epsilon(\boldsymbol{q}, \omega)$ relevant for the optical properties, is polarized transverse to $\boldsymbol{q}$, i.e. to the propagation direction of the EM wave. For the purpose of the discussion in the present section we have written the index t specifically as a reminder of that. However, the general definition of $\boldsymbol{\epsilon}(\boldsymbol{q}, \omega)$ is

$$\boldsymbol{\epsilon}(\boldsymbol{q}, \omega) = \frac{\boldsymbol{D}_q}{\boldsymbol{E}_q} \tag{9.10}$$

where $\boldsymbol{D}(\boldsymbol{r}, t) = \boldsymbol{D}_q \exp i(\boldsymbol{q} \cdot \boldsymbol{r} - \omega t)$ and $\boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{E}_q \exp i(\boldsymbol{q} \cdot \boldsymbol{r} - \omega t)$. For waves traveling in vacuum transverse polarization is the only possibility allowed by Maxwell's equations. Inside a material, on the other hand, longitudinal electromagnetic waves do in fact exist, plasmons for example. Static and dynamic screening of charge inside solids is an important phenomenon which involves the longitudinal component of the dielectric function.

Since we have already seen how to measure $\epsilon_t(\omega)$, we may wonder how one can measure also $\epsilon_l(\boldsymbol{q}, \omega)$. The experimental method allowing to do so is called Electron Energy Loss Spectroscopy (EELS). This technique consists of measuring the inelastic decay of fast electrons passing through a sample. Experimentally one creates a monochromatic beam of high energy electrons, typically with an energy of 170 keV. These electrons are fired through a thin slab (100 nm thick) of the material which one wishes to investigate. Part of the electrons emerge at the other side of the sample with the original energy and momentum, others have lost an amount of their energy, and have transferred momentum to an excitation inside the solid. For each value of transferred momentum, $\boldsymbol{q}$, the number of electrons can be counted for any given value of the energy loss, $\hbar\omega$ by selecting a certain direction in space, and by selecting the energy of the electrons emerging from the sample.

A collision process inside the solid, whereby the momentum changes from $\boldsymbol{p}$ to $\boldsymbol{p}-\boldsymbol{q}$ and the energy from $E_{in} = p^2/(2m)$ to $E_{out} = |\boldsymbol{p}-\boldsymbol{q}|^2/(2m) = p^2/(2m)-\hbar\omega$ generates during the collision process a longitudinal dielectric displacement field, $D(\boldsymbol{r}, t) \propto \exp(i\boldsymbol{q} \cdot \boldsymbol{r} - i\omega t)$. Since inside a material the dielectric displacement is screened by the response of the matter particles, the resulting electric field is

$$\boldsymbol{E}(\boldsymbol{q}, \omega) = \frac{1}{\epsilon(\boldsymbol{q}, \omega)} \boldsymbol{D}(\boldsymbol{q}, \omega) \tag{9.11}$$

The probability per unit time that a fast electron transfers momentum $\boldsymbol{q}$ and energy ω to the electrons was derived by Nozières and Pines for a fluid of interacting electrons

$$P(\boldsymbol{q}, \omega) = \frac{8\pi q_e^2}{|q|^2} \mathrm{Im}\left\{\frac{-1}{\epsilon(\boldsymbol{q}, \omega)}\right\} \tag{9.12}$$

where q_e is the electron charge. Hence this technique provides the longitudinal dielectric function, i.e. the response to a dielectric displacement field which is parallel to the transferred momentum $\boldsymbol{q}$.

Finally we come back to the optical spectroscopy. In the limit $q \to 0$ the distinction between longitudinal and transverse polarization vanishes, and consequently

$$\lim_{q \to 0} \epsilon_t(\boldsymbol{q}, \omega) = \epsilon_l(\boldsymbol{q}, \omega) \tag{9.13}$$

Since optical spectroscopy allows to measure real and imaginary part of $\epsilon_t(\boldsymbol{q}, \omega)$, it is possible to calculate the loss function $\mathrm{Im}(-1/\epsilon(\boldsymbol{q}, \omega))$ for $q \to 0$, and this should correspond exactly to the energy loss spectra measured with EELS. An example where the two techniques are compared for the same material is given in Fig. 9.3. Indeed, we see that the two techniques give the same result for $q \to 0$, as expected.

9.4 Quantum Electrodynamics of Electrons in a Lattice

To keep the notation light, we use Planck units in the remainder of this chapter. In those units $\hbar = 1$, $c = 1$. Since the fine-structure constant is $\alpha = q_e^2/\hbar c$, we automatically have $q_e = -\sqrt{\alpha}$ in these units. We will however continue to use the symbol q_e to indicate the electron charge throughout this chapter. Spin-coordinate plays no role of particular importance in this chapter; we therefore **suppress the spin-labels** in the following subsections in the interest of compactness of notation.

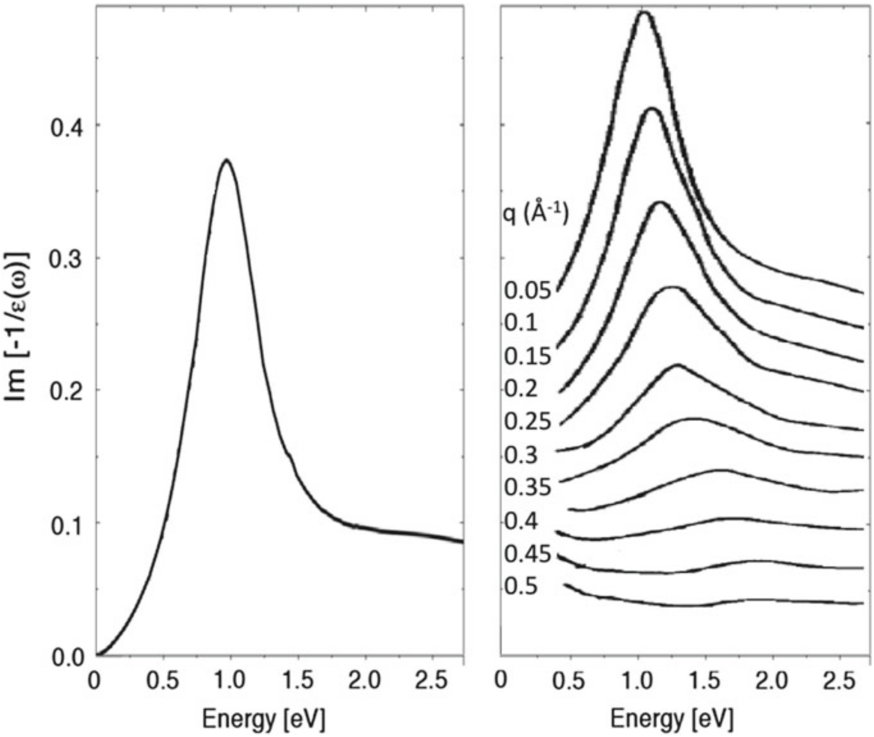

Fig. 9.3 Comparison of the optical data (*left* [14]) and EELS spectra (*right* [15], reproduced from the original figure with permission of the authors.) of $Bi_2Sr_2CaCu_2O_8$, both providing $\mathrm{Im}(-1/\epsilon(\boldsymbol{q}, \omega))$. The peak positions in both spectra correspond to the plasma-resonance frequency

9.4.1 Coupling of Interacting Electrons in Solids to an Electromagnetic Field

We begin by defining the system in the absence of an external electromagnetic field. We consider a tight-binding model for the conduction band, where the tunneling of an electron on a given site $\boldsymbol{r}_m$ to surrounding sites $\boldsymbol{r}_{m+j}$ is described by the hopping matrix element t_j. In addition we have to take into account the interaction between the electrons, which in general is a function of their mutual distance. Although the interaction may in principle also depend on the spin, this aspect plays no role in the present discussion. The full matter Hamiltonian including interactions is then

$$\begin{aligned}\hat{H}^0 &= \hat{H}^{kin} + \hat{H}^{int}\\ \hat{H}^{kin} &= -\sum_{m,j} t_j (c^\dagger_{m+j} c_m + c^\dagger_m c_{m+j})\\ \hat{H}^{int} &= \frac{1}{2}\sum_{m,n} V_{m-n}\hat{\rho}(\boldsymbol{r}_m)\hat{\rho}(\boldsymbol{r}_n)\end{aligned} \tag{9.14}$$

where $\hat{\rho}(\boldsymbol{r_j}) = c^\dagger_j c_j$ is the density operator at lattice site j. For later use we point out, that the first term is diagonal on the basis of Bloch-states represented by the operators

$$c^\dagger_k = \frac{1}{\sqrt{N_s}}\sum_m e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_m} c^\dagger_m$$

In terms of which

$$\begin{aligned}\hat{H}^{kin} &= \sum_k \epsilon_k c^\dagger_k c_k\\ \epsilon_k &= -2\sum_j t_j e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j}\\ \boldsymbol{v}_k &= \frac{\partial \epsilon_k}{\partial \boldsymbol{k}} = -2i\sum_j t_j \boldsymbol{r}_j e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j}\end{aligned} \tag{9.15}$$

Let us now turn to the microscopic quantum mechanical expression for the optical conductivity. We will follow closely the treatment of Mahan [9], and by Nozières and Pines [7]. We consider the effect of a time-varying electric field, which is the time derivative of the vector potential, i.e. the relation between $\boldsymbol{A} = \boldsymbol{A}_q e^{-i\omega t}$ and $\boldsymbol{E} = \boldsymbol{E}_q e^{-i\omega t}$ is

$$\boldsymbol{E}(t) = -\frac{\partial \boldsymbol{A}(t)}{\partial t} = i\omega \boldsymbol{A}(t). \tag{9.16}$$

In quantum electrodynamics the coupling between the electromagnetic field and the electrons is introduced by making the "minimal substitution" $\boldsymbol{p} \to \boldsymbol{p} - q_e\boldsymbol{A}$ in the kinetic energy term of the Hamiltonian. Since here we are concerned with the tight-binding model on a lattice, we need some form of "course graining" of the vector potential. The relation between direct- and reciprocal space representations is then

$$\boldsymbol{A}_q = \frac{1}{\sqrt{N_s}}\sum_m e^{-i\boldsymbol{q}\cdot\boldsymbol{r}_m}\boldsymbol{A}_m \tag{9.17}$$

If an electron is transferred from lattice coordinate $\boldsymbol{r}_m$ to $\boldsymbol{r}_n$, the wavefunction picks up a phase due to the vector potential, described by the integral $(q_e/\hbar)\int_m^{m+j}\boldsymbol{A}(\boldsymbol{s})\cdot d\boldsymbol{s}$, which depends on the path if $\nabla\times\boldsymbol{A}\neq 0$. A difficulty is, that with the course-graining procedure we have introduced some ambiguity regarding the path followed by the electron. Since the path of a hopping term t_j in the tight-binding Hamiltonian is of the order of a lattice constant, the variation of $\boldsymbol{A}(\boldsymbol{s})$ along the path is very small. We follow the Peierls coupling scheme, and substitute a constant value in the integral, $\boldsymbol{A}_{m,j}=(\boldsymbol{A}_m+\boldsymbol{A}_{m+j})/2$, corresponding to the average over the tunneling path. The integral than becomes $\int_m^{m+j}\boldsymbol{A}(\boldsymbol{s})\cdot d\boldsymbol{s}=\boldsymbol{A}_{m,j}\cdot\boldsymbol{r}_j$. The corresponding Peierls substitution

$$t_j \rightarrow t_j e^{-iq_e\boldsymbol{A}_{m,j}\cdot\boldsymbol{r}} \tag{9.18}$$

does not affect the interaction part of the Hamiltonian (9.14), but the kinetic energy term picks up the extra phase. The Hamiltonian in the presence of $\boldsymbol{A}_{m,j}$ is, according to the "Peierls coupling" scheme

$$\hat{H}=-\sum_{m,j}t_j\left(e^{-iq_e\boldsymbol{A}_{m,j}\cdot\boldsymbol{r}}c_{m+j}^{\dagger}c_m+e^{iq_e\boldsymbol{A}_{m,j}\cdot\boldsymbol{r}}c_m^{\dagger}c_{m+j}\right)+\hat{H}^{int} \tag{9.19}$$

9.4.2 General Consideration About the Calculation of the Linear Response

We are interested to calculate the linear response of the current density $\boldsymbol{J}_q$ to the electric field $\boldsymbol{E}_q$, which is related to the vector potential $\boldsymbol{A}_q$ through $\boldsymbol{E}_q=i\omega\boldsymbol{A}_q$. The current density is related to the local velocities of all electrons, described by the expectation value of the current operator $\hat{\boldsymbol{j}}(\boldsymbol{r})$ through $\boldsymbol{J}(\boldsymbol{r})=\langle\hat{\boldsymbol{j}}(\boldsymbol{r})\rangle$. Note, that $\boldsymbol{J}(\boldsymbol{r})$ refers to the amount charge passing per unit of time per unit area perpendicular to $\boldsymbol{J}(\boldsymbol{r})$. The optical conductivity is then

$$\boldsymbol{\sigma}(\boldsymbol{q},\omega)=\frac{\boldsymbol{J}_q}{\boldsymbol{E}_q}=\frac{\langle\hat{\boldsymbol{j}}_q\rangle}{i\omega\boldsymbol{A}_q}\qquad(\lim\boldsymbol{A}_q\rightarrow 0) \tag{9.20}$$

In general terms our task is to compute the time-dependent expectation value of the current operator in the presence of the vector potential. The vector potential enters the problem on two levels: First, the time-independent current operator in the presence of the vector potential is already different from the current operator without vector potential. Secondly the time evolution of the current operator is described by the Hamiltonian (9.19), which also contains the vector potential. Taken together

$$\boldsymbol{\sigma}(\boldsymbol{q},\omega)=\frac{1}{i\omega\boldsymbol{A}_q}\left\langle e^{i\hat{H}(A_q)t}\hat{\boldsymbol{j}}_q(A_q)e^{-i\hat{H}(A_q)t}\right\rangle\qquad(\lim\boldsymbol{A}_q\rightarrow 0) \tag{9.21}$$

We see, that the vector potential enters in various different positions in the expression. One can proceed by making Taylor series expansions of all terms, and collect at the end all terms linear in $\boldsymbol{A}_q$. Each step takes care and precision, since the operators in this expression don't commute. These steps are treated in the following three subsections.

9.4.3 Expansion of the Current Operator in Powers of the Vector Potential

In quantum electrodynamics the current density is obtained from the relation $\boldsymbol{J} = -\Omega^{-1}\partial\hat{H}/\partial\boldsymbol{A}$. For our tight-binding Hamiltonian (9.14) this implies

$$\begin{aligned}\hat{\boldsymbol{j}}(\boldsymbol{r}_m) &= -\frac{1}{\Omega}\left[\frac{\partial\hat{H}}{\partial\boldsymbol{A}_m}\right] \\ &= \frac{1}{\Omega}\sum_j t_j\boldsymbol{r}_j(-iq_e e^{-iq_e\boldsymbol{A}_{m,j}\cdot\boldsymbol{r}_j}c^{\dagger}_{m+j}c_m + iq_e e^{iq_e\boldsymbol{A}_{m,j}\cdot\boldsymbol{r}_j}c^{\dagger}_m c_{m+j})\end{aligned} \tag{9.22}$$

which we expand in powers of $\boldsymbol{A}_{m,j}$, retaining only terms up to first order

$$\begin{aligned}\hat{\boldsymbol{j}}(\boldsymbol{r}_m) &= \hat{\boldsymbol{j}}^r(\boldsymbol{r}_m) + \hat{\boldsymbol{j}}^d(\boldsymbol{r}_m) \\ \hat{\boldsymbol{j}}^r(\boldsymbol{r}_m) &= -i\frac{q_e}{\Omega}\sum_j t_j\boldsymbol{r}_j(c^{\dagger}_{m+j}c_m - c^{\dagger}_m c_{m+j}) \\ \hat{\boldsymbol{j}}^d(\boldsymbol{r}_m) &= -\frac{q_e^2}{\Omega}\sum_j t_j\boldsymbol{r}_j(\boldsymbol{r}_j\cdot\boldsymbol{A}_{m,j})(c^{\dagger}_{m+j}c_m + c^{\dagger}_m c_{m+j})\end{aligned} \tag{9.23}$$

The first term in (9.23) is the so-called "regular" term of the current operator, since it is independent of the vector potential. We draw attention to the physical interpretation of the velocity operator. The matrix element $\left\langle -iq_e t_j\boldsymbol{r}_j c^{\dagger}_{m+j}c_m\right\rangle$ describes a process whereby an electron tunnels from position $\boldsymbol{r}_m$ to $\boldsymbol{r}_{m+j}$. The tunneling time of this process is $1/t_j$, and the displacement is $\boldsymbol{r}_j$. The effective velocity of this process is therefore $\boldsymbol{r}_j t_j$. The second term proportional to $< c^{\dagger}_m c_{m+j} >$ describes the same event, except that it occurs in the opposite direction. The operator $\hat{\boldsymbol{j}}^r(\boldsymbol{r}_m)$ therefore describes exactly the current passing through lattice position $\boldsymbol{r}_m$. While the Peierls substitution is burdened by ambiguities (except if $\boldsymbol{A}(r)$ is uniform), the regular part of the current operator in (9.23) is a robust result, which satisfies our intuitive understanding of a local current. Its momentum-space representation is

$$\hat{\boldsymbol{j}}^r_q = \frac{1}{\sqrt{N_s}} \sum_m e^{-i\boldsymbol{q}\cdot\boldsymbol{r}_m} \hat{\boldsymbol{j}}^r(\boldsymbol{r}_m) = \frac{q_e}{\Omega} \sum_k \frac{\boldsymbol{v}_k + \boldsymbol{v}_{k-q}}{2} c^\dagger_k c_{k-q} \tag{9.24}$$

where $\boldsymbol{v}_k = \partial\epsilon_k/\partial\boldsymbol{k}$ is the group velocity.

The "diamagnetic part" of the current, $\hat{\boldsymbol{j}}^d$, is the term which is proportional to A_m. Its Fourier transform is

$$\hat{\boldsymbol{j}}^d_q = -q_e^2 \sum_{q'} \boldsymbol{A}_{q'} \cdot \hat{\boldsymbol{K}}_{q,q'} \tag{9.25}$$

$$\hat{\boldsymbol{K}}_{q,q'} \equiv \frac{1}{2\Omega} \sum_{j,k} t_j \boldsymbol{r}_j \boldsymbol{r}_j (2e^{i\boldsymbol{q}'\cdot\boldsymbol{r}_j} \cos\boldsymbol{k}\cdot\boldsymbol{r}_j + e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j} + e^{i(\boldsymbol{k}+\boldsymbol{q}-\boldsymbol{q}')\cdot\boldsymbol{r}_j}) c^\dagger_k c_{k+q-q'}$$

The expectation value has finite contributions only from $q = q'$ in the above expression, hence

$$\boldsymbol{J}^d_q = \left\langle \hat{\boldsymbol{j}}^d_q \right\rangle = -q_e^2 \boldsymbol{A}_q \cdot \boldsymbol{K}(\boldsymbol{q}) \tag{9.26}$$

where the tensor $\boldsymbol{K}(\boldsymbol{q})$ is proportional to the spectral weight in the optical conductivity function

$$\boldsymbol{K}(\boldsymbol{q}) = \frac{2}{\Omega} \sum_{k,j} t_j \boldsymbol{r}_j \boldsymbol{r}_j e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j} \cos^2\left(\frac{\boldsymbol{q}\cdot\boldsymbol{r}_j}{2}\right) \left\langle c^\dagger_k c_k \right\rangle \tag{9.27}$$

We see, that (9.27) indicates that $\boldsymbol{K}(\boldsymbol{q})$ depends on the wavevector $\boldsymbol{q}$ of the perturbing field. For the special case of the nearest-neighbour tight-binding model we will find confirmation of exactly this $\boldsymbol{q}$-dependence from a different approach in subsection 9.5.4. Nevertheless, we have to keep in mind that the course-graining procedure used in the Peierls substitution is only fully accurate for a uniform vector potential, i.e. $q = 0$. For a general type of dispersion $\epsilon(k)$ non-negligible corrections of order q^2 to (9.27) are to be expected. The limit for $q \to 0$ of (9.27) gives

$$\boldsymbol{K} = \frac{1}{\Omega} \sum_k \frac{\partial^2 \epsilon_k}{\partial \boldsymbol{k}^2} \left\langle c^\dagger_k c_k \right\rangle \tag{9.28}$$

The only exception where $\boldsymbol{K}(\boldsymbol{q})$ is $\boldsymbol{q}$-independent occurs for a parabolic energy-momentum dispersion relation of the electrons, $\epsilon_k = k^2/2m$. We then obtain the result

$$K(q) = K = \frac{n}{m} \tag{9.29}$$

For an entirely filled band at zero temperature

$$\frac{(2\pi)^3}{\Omega}\sum_k \frac{\partial^2 \epsilon_k}{\partial \boldsymbol{k}^2} n_k = \int_{1^t BZ} \frac{\partial \epsilon_k}{\partial \boldsymbol{k}} d\boldsymbol{s}_k = 0 \tag{9.30}$$

where the integral is over the surface of the first Brillouin zone. Since $\partial\epsilon_k/\partial\boldsymbol{k} = 0$ on the BZ surface, this integral is zero, and consequently $K = 0$ in this case.

Combining (9.24) and (9.26) we obtain the full expression for the current operator

$$\hat{\boldsymbol{j}}_q = \hat{\boldsymbol{j}}_q^d + \hat{\boldsymbol{j}}_q^r = -q_e^2 \sum_{q'} \boldsymbol{A}_{q'} \cdot \hat{\boldsymbol{K}}_{q,q'} + \frac{q_e}{\Omega}\sum_k \frac{\boldsymbol{v}_k + \boldsymbol{v}_{k-q}}{2} c_k^\dagger c_{k-q} \tag{9.31}$$

9.4.4 Expansion of the Hamiltonian in Leading Order of the Vector Potential

We now turn to the Hamiltonian in (9.19). Since we are interested in the linear response to the vector potential, we use a Taylor expansion in powers of A_m

$$\hat{H}^{kin}(\boldsymbol{A}_q) = \hat{H}^{kin} + iq_e \sum_{m,j} t_j \boldsymbol{A}_{m,j} \cdot \boldsymbol{r}_j (c_{m+j}^\dagger c_m - c_m^\dagger c_{m+j} + \cdots) \tag{9.32}$$

The terms proportional to $\boldsymbol{A}_{m,j}$ can be worked out as follows:

$$\begin{aligned} & iq_e \sum_{m,j} t_j \boldsymbol{A}_{m,j} \cdot \boldsymbol{r}_j (c_{m+j}^\dagger c_m - c_m^\dagger c_{m+j}) \\ &= iq_e \sum_{k,q,j} t_j \boldsymbol{A}_q \cdot \boldsymbol{r}_j (e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j} - e^{-i(\boldsymbol{k}+\boldsymbol{q})\cdot\boldsymbol{r}_j}) c_k^\dagger c_{k+q} \\ &= -q_e \sum_{k,q,j} \boldsymbol{A}_q \cdot \frac{\boldsymbol{v}_k + \boldsymbol{v}_{k+q}}{2} c_k^\dagger c_{k+q} = -\Omega \sum_q \boldsymbol{A}_q \cdot \hat{\boldsymbol{j}}_{-q}^r \end{aligned}$$

where, in order to write the last equation, we use the result for the regular term of the current operator, (9.24). In leading order of $\boldsymbol{A}_q$ the Hamiltonian can then be written as the sum of two terms, $\hat{H}^0$ representing the full matter Hamiltonian including interactions (9.14), and $\hat{H}^p$ representing the perturbation due to a time-varying vector potential

$$\begin{aligned} \hat{H} &= \hat{H}^0 + \hat{H}^p \\ \hat{H}^p &= -\Omega \sum_q \boldsymbol{A}_q \cdot \hat{\boldsymbol{j}}_{-q}^r \end{aligned} \tag{9.33}$$

9.4.5 Current Response to an Applied Field

The response to the vector potential is described by expectation value of the Heisenberg representation of the current operator, taking the full Hamiltonion including the vector potential

$$\boldsymbol{J}_q(t) = \left\langle e^{i\hat{H}t}\hat{\boldsymbol{j}}_q e^{-i\hat{H}t}\right\rangle = \left\langle \hat{\boldsymbol{j}}_q^d(t)\right\rangle + \left\langle \hat{\boldsymbol{j}}_q^r(t)\right\rangle = -q_e^2\boldsymbol{A}_q\cdot\boldsymbol{K}(\boldsymbol{q}) + \boldsymbol{J}_q^r(t) \quad (9.34)$$

The diamagnetic contribution to the conductivity is now easily obtained using the definition (9.20)

$$\boldsymbol{\sigma}^d(\omega, q) = \frac{\boldsymbol{J}_q^d}{i\omega\boldsymbol{A}_q} = \frac{iq_e^2\boldsymbol{K}(\boldsymbol{q})}{\omega} \quad (9.35)$$

This term expresses that in the absence of a potential binding the electrons to the lattice, the response will be that of a plasma of freely moving charged particles with mass m, charge q_e, and density $n = N/\Omega$.

The second contribution to the current in (9.34) represents the combined effect of the crystal potential in which the electrons move, and their mutual interactions. The corresponding "regular" contribution to the optical conductivity is

$$\boldsymbol{\sigma}^r(\omega, q) = \frac{\boldsymbol{J}_q^r}{i\omega\boldsymbol{A}_q} \quad (9.36)$$

At this point it will be useful to introduce the Heisenberg representation of the current operator *in the absence* of the external field described by the term $\hat{H}^p$ in the hamiltonian.

$$\hat{\boldsymbol{j}}_q^0(t) = e^{i\hat{H}^0t}\hat{\boldsymbol{j}}_q e^{-i\hat{H}^0t} \quad (9.37)$$

The vector potential $\boldsymbol{A}(t)$ has been switched on at a certain time t_0. Without loss of generality we can choose this time to be $t_0 = 0$. To describe the time-evolution of the current operator caused by $\boldsymbol{A}(t)$, we introduce the time-evolution operator

$$\begin{aligned} \hat{U}(t) &= e^{i\hat{H}^0t}e^{-i\hat{H}t} \quad && t \geq 0 \\ \hat{U}(t) &= 1 && t < 0 \end{aligned} \quad (9.38)$$

With the help of (9.37), (9.38) we can rewrite $\hat{\boldsymbol{j}}_q^r(t)$ in the expression of $\boldsymbol{J}_q^r(t)$

$$\hat{\boldsymbol{j}}_q^r(t) = e^{i\hat{H}t}\hat{\boldsymbol{j}}_q e^{-i\hat{H}t} = \hat{U}^\dagger(t)\hat{\boldsymbol{j}}_q^0(t)\hat{U}(t) \quad (9.39)$$

With the help of this we obtain

$$\boldsymbol{J}_q^r(t) = \left\langle \hat{U}^\dagger(t)\hat{\boldsymbol{j}}_q^0(t)\hat{U}(t) \right\rangle \tag{9.40}$$

We like to expand the expression for the current as a function of $\hat{H}^p$ and look for the leading terms for the linear response. Since the definition of $\hat{\boldsymbol{j}}^0(\boldsymbol{r}, t)$ contains $\hat{H}^0$ instead of $\hat{H}$, it does not depend on the vector potential $\hat{H}^p$. It is therefor only necessary to expand $U(t)$ as a function of $\hat{H}^p$. To do so we need to know the properties of $\hat{U}(t)$. First of all, according to (9.38) we have $\hat{U}(-\infty) = 1$. In addition, $\hat{U}(t)$ satisfies the following expression for the derivative with respect to time

$$\begin{aligned}\frac{\partial \hat{U}}{\partial t} &= i\hat{H}^0 e^{i\hat{H}^0 t} e^{-i\hat{H}t} - ie^{i\hat{H}^0 t}\hat{H}e^{-i\hat{H}t} = e^{i\hat{H}^0 t}\hat{H}^p e^{-i\hat{H}t} \\ &= ie^{i\hat{H}^0 t}\hat{H}^p e^{i\hat{H}^0 t} e^{-i\hat{H}^0 t} e^{-i\hat{H}t} = -i\hat{H}^p(t)\hat{U}(t)\end{aligned}$$

To solve this equation, we integrate from the lower limit $\hat{U}(-\infty) = 1$ to finite time t:

$$\hat{U}(t) = 1 - i\int_{-\infty}^{t} dt'\hat{H}^p(t')\hat{U}(t')$$

Iterative solution yields

$$\hat{U}(t) = 1 - i\int_{-\infty}^{t} dt'\hat{H}^p(t') + \frac{1}{2}\int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt''\hat{H}^p(t')\hat{H}^p(t'') + \cdots$$

Since we restrict the discussion here to linear response, we need only the first two terms. Substituting those in (9.40) yields

$$\boldsymbol{J}_q^r(t) = \left\langle \left(1 + i\int_{-\infty}^{t} dt'\hat{H}^p(t')\right)\hat{\boldsymbol{j}}_q^0(t)\left(1 - i\int_{-\infty}^{t} dt'\hat{H}^p(t')\right)\right\rangle \tag{9.41}$$

The matrix element $\left\langle \hat{\boldsymbol{j}}^0(\boldsymbol{r}, t)\right\rangle = 0$ for a system in equilibrium. Furthermore terms proportional to $(\hat{H}^p)^2$ contribute to the quadratic response. We now substitute $\hat{H}^p = -\Omega\sum_q \boldsymbol{A}_q \cdot \hat{\boldsymbol{j}}_{-q}^r$ (9.33). Retaining only the linear terms gives

$$\boldsymbol{J}_q^r(t) = i\Omega \int\limits_{-\infty}^{t} \left\langle \left[\hat{\boldsymbol{j}}_q^0(t), \hat{\boldsymbol{j}}_{-q}^0(t') \cdot \boldsymbol{A}_q(t') \right] \right\rangle dt'$$

We substitute $\boldsymbol{J}_q^r(t) = \boldsymbol{J}_q^r e^{-i\omega t}$ and $\boldsymbol{A}_q(t) = \boldsymbol{A}_q e^{-i\omega t}$ and multiply both sides with $e^{i\omega t}$

$$\boldsymbol{J}_q^r = i\Omega \int\limits_{-\infty}^{t} \left\langle \left[\hat{\boldsymbol{j}}_q^0(t), \hat{\boldsymbol{j}}_{-q}^0(t') \cdot \boldsymbol{A}_q \right] \right\rangle e^{i\omega(t-t')} dt'$$

Due to time-invariance, we have

$$\left[\hat{\boldsymbol{j}}_q^0(t), \hat{\boldsymbol{j}}_{-q}^0(t') \right] = \left[\hat{\boldsymbol{j}}_q^0(t-t'), \hat{\boldsymbol{j}}_{-q}^0(0) \right] \tag{9.42}$$

We substitute $t - t' \to t''$, so that

$$\boldsymbol{J}_q^r = i\Omega \int\limits_{0}^{\infty} \left\langle \left[\hat{\boldsymbol{j}}_q^0(t''), \hat{\boldsymbol{j}}_{-q}^0(0) \cdot \boldsymbol{A}_q \right] \right\rangle e^{i\omega t''} dt''$$

We now divide both sides by $\boldsymbol{A}_q$

$$\frac{\boldsymbol{J}_q^r}{\boldsymbol{A}_q} = -\chi_{jj}(\boldsymbol{q}, \omega) \tag{9.43}$$

where $\chi_{jj}(\boldsymbol{q}, t)$ is the current-current correlation function defined as

$$\chi_{jj}(\boldsymbol{q}, t) = -i\theta(t)\Omega \left\langle \left[\hat{\boldsymbol{j}}_q(t), \hat{\boldsymbol{j}}_{-q}(0) \right] \right\rangle \tag{9.44}$$

and

$$\chi_{jj}(\boldsymbol{q}, \omega) = \int\limits_{-\infty}^{\infty} \chi_{jj}(\boldsymbol{q}, t) e^{i\omega t} dt \tag{9.45}$$

Comparing this to (9.36) we see, that the regular part of the optical conductivity is

$$\boldsymbol{\sigma}^r(\omega, q) = \frac{i}{\omega + i\eta} \chi_{jj}(\boldsymbol{q}, \omega) \tag{9.46}$$

where $i\eta = i0^+$ moves the pole infinitesimally away from the real axis, such as to assure causality in the time-dependence of the relation between current and electric field.

9.4.6 Frequency and Temperature Dependent Optical Conductivity

We see, that the regular par of the optical conductivity is proportional to the current-current correlation function, divided by the frequency. We continue the discussion by considering a well-defined many-particle state $|n\rangle$, for which we calculate the optical conductivity

$$\boldsymbol{\sigma}^r(\boldsymbol{q},\omega) = \frac{\Omega}{\omega+i\eta}\int_0^\infty \left\langle n \left| \left\{ e^{i\hat{H}^0 t}\hat{\boldsymbol{j}}_q e^{-i\hat{H}^0 t}\hat{\boldsymbol{j}}_{-q} - \hat{\boldsymbol{j}}_{-q} e^{i\hat{H}^0 t}\hat{\boldsymbol{j}}_q e^{-i\hat{H}^0 t} \right\} \right| n \right\rangle e^{i\omega t} dt$$

After some manipulation with the operators $e^{i\hat{H}^0 t}$ and $e^{-i\hat{H}^0 t}$ we obtain

$$\boldsymbol{\sigma}^r(\omega,q) = \frac{\Omega}{\omega+i\eta}\int_0^\infty \sum_{m\neq n}\left\{ \boldsymbol{j}_q^{nm}\boldsymbol{j}_{-q}^{mn} e^{i(\omega+E_n-E_m)t} - \boldsymbol{j}_{-q}^{nm}\boldsymbol{j}_q^{mn} e^{i(\omega+E_m-E_n)t} \right\} dt$$

where for compactness of notation we represent the matrix elements of the current operators as

$$\boldsymbol{j}_q^{nm} \equiv \langle n|\hat{\boldsymbol{j}}_q^r|m\rangle \tag{9.47}$$

In the remainder we will assume a basis on which the conductivity tensor is diagonal, so that $\boldsymbol{j}_q^{nm} // \boldsymbol{j}_{-q}^{mn}$. We can than drop the tensor notation and replace $\boldsymbol{j}_q^{nm}\boldsymbol{j}_{-q}^{mn}$ with $|\boldsymbol{j}_q^{nm}|^2$. We also introduce the short-hand notation for the energy differences $E_m - E_n = \omega_{mn}$. Carrying out the integrations over time we obtain

$$\sigma^r(\omega,q) = i\sum_{m\neq n}\frac{\Omega|\boldsymbol{j}_q^{nm}|^2}{\omega+i\eta}\left\{\frac{1}{\omega-\omega_{mn}+i\eta_1} - \frac{1}{\omega+\omega_{mn}+i\eta_1}\right\} \tag{9.48}$$

Note, that both η and η_1 are to be taken in the limit $\eta \to 0$, but that these limits are independent from one another, i.e. $\eta_1 \neq \eta$.

The expression for the total conductivity (diamagnetic + regular) is obtained by combining this with (9.35).

$$\begin{aligned}\sigma(\boldsymbol{q},\omega) &= \sigma^d(\boldsymbol{q},\omega) + \sigma^r(\boldsymbol{q},\omega) \qquad (9.49)\\ &= \frac{i}{\omega+i\eta}\left\{ q_e^2 K + \sum_{m\neq n}\left[\frac{\Omega|\boldsymbol{j}_q^{nm}|^2}{\omega-\omega_{mn}+i\eta_1} - \frac{\Omega|\boldsymbol{j}_q^{nm}|^2}{\omega+\omega_{mn}+i\eta_1}\right]\right\}\end{aligned}$$

In (9.49) $\sigma(\omega)$ is represented by two separate terms, a δ-function for $\omega = 0$ and a summation over excited many-body eigen-states. The δ-function is a diamagnetic contribution of *all* electrons in the system, the presence of which is a consequence of the gauge invariant treatment of the optical conductivity. Note however, that according to (9.30) for an insulator (the bands of which are either fully occupied or entirely empty) we have $K = 0$. The presence of this term is at first glance rather confusing, since left by itself this δ-function would imply that all metals are ideal conductors! However, the second term has, besides a series of poles corresponding to the optical transitions, also a pole for $\omega = 0$, corresponding to a *negative* δ-function of $\mathrm{Re}\sigma(\omega)$. It turns out, that for all materials except ideal conductors this δ-function compensates exactly the first (diamagnetic) term of (9.49). This exact compensation is a consequence of the relation.[1]

$$\text{For every } n\text{: } q_e^2 K = 2 \sum_{m \neq n} \frac{\Omega |\boldsymbol{j}_q^{nm}|^2}{\omega_{mn}} \tag{9.50}$$

Experimentally truly 'ideal' conductivity is only seen in superconductors. In ordinary conducting materials the diamagnetic term broadens to a Lorentzian peak due to elastic and/or inelastic scattering. The width of this peak is the inverse life-time of the charge carriers. Often in the theoretical literature the broadening is not important, and the Drude peak is counted to the Dirac-function in the origin. The infrared properties of superconductors are characterized by the presence of both a purely reactive diamagnetic response, and a regular dissipative conductivity. The sum of these contributions counts the partial intra-band spectral weight which we discussed previously in relation to the "kinetic energy sum rule". With the help of (9.50), the diamagnetic term of (9.49) can now be absorbed in the summation on the right-hand side, so that after combining all terms

$$\sigma(\boldsymbol{q}, \omega) = 2i\omega \sum_{m \neq n} \frac{\Omega |\boldsymbol{j}_q^{nm}|^2}{\omega_{mn}} \frac{1}{\omega(\omega + i\eta) - \omega_{mn}^2} \tag{9.51}$$

where we have to keep in mind, that the equivalence with (9.49) holds under condition that η $(= 2\eta_1)$ is infinitesimally small. Note, that in deriving (9.51) from (9.49), an $\omega + i\eta$ term in the numerator has been replaced by ω since, unlike for the denominators containing $i\eta$, the effect of $i\eta$ vanishes if we consider the limit $\eta \to 0$.

In (9.51) we have calculated the optical conductivity assuming that, under the influence of the external potential, the system evolves as a function of time from an eigenstate $|n\rangle$. In the most common experimental situation the sample is in thermal equilibrium with heat bath with temperature T. Following the approach of Kubo we calculate the conductivity using thermodynamical weight factors $e^{-\beta E_n}/Z$ for each

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[1] Equation (9.50) is obtained if one represents the current operators as commutators of the hamiltonian with the dipole operator defined in (9.68). The expectation value of the hamiltonian is used to cancel out the factor ω_{mn} in the denominator of the expression. In the final step the commutator of the dipole operator and the current operator is calculated, which completes the derivation of (9.50).

many-body eigenstate (see [9] for a discussion on the justification of this approach). Here Z is the partition function, and $\beta = 1/k_B T$. The expression for the optical conductivity at finite temperature becomes then

$$\sigma(\boldsymbol{q}, \omega) = 2i\omega \sum_{n,m\neq n} \frac{\Omega|\boldsymbol{j}_q^{nm}|^2}{\omega_{mn}} \frac{e^{-\beta E_n}}{Z} \frac{1}{\omega(\omega + i\eta) - \omega_{mn}^2} \tag{9.52}$$

The imaginary parts of the terms are Dirac δ-functions, so that

$$\mathrm{Re}\sigma(\boldsymbol{q}, \omega) = \pi \sum_{n,m\neq n} \frac{\Omega|\boldsymbol{j}_q^{nm}|^2}{\omega_{mn}} \frac{e^{-\beta E_n}}{Z} \{\delta(\omega - \omega_{mn}) + \delta(\omega + \omega_{mn})\} \tag{9.53}$$

The spectrum thus consists of a series of peaks, each representing an excitation from the ground state $|n\rangle$ to an excited many-body state $|m\rangle$ at an energy cost ω_{mn}. In atoms and molecules one observes indeed a discrete set of lines. In solids the excitations broaden into bands due to the fact that the excitations in different parts of the lattice are coupled, resulting in bands of excited states. The most commonly observed excited states are the creation of one hole in the occupied band and one electron in the states above E_F. The resulting optical spectrum is than something like a joint density of states of the bands below and above the Fermi energy. Examples of this are shown for a metal (MnSi), a semi-comductor (FeSi), a semimetal (CoSi) and a

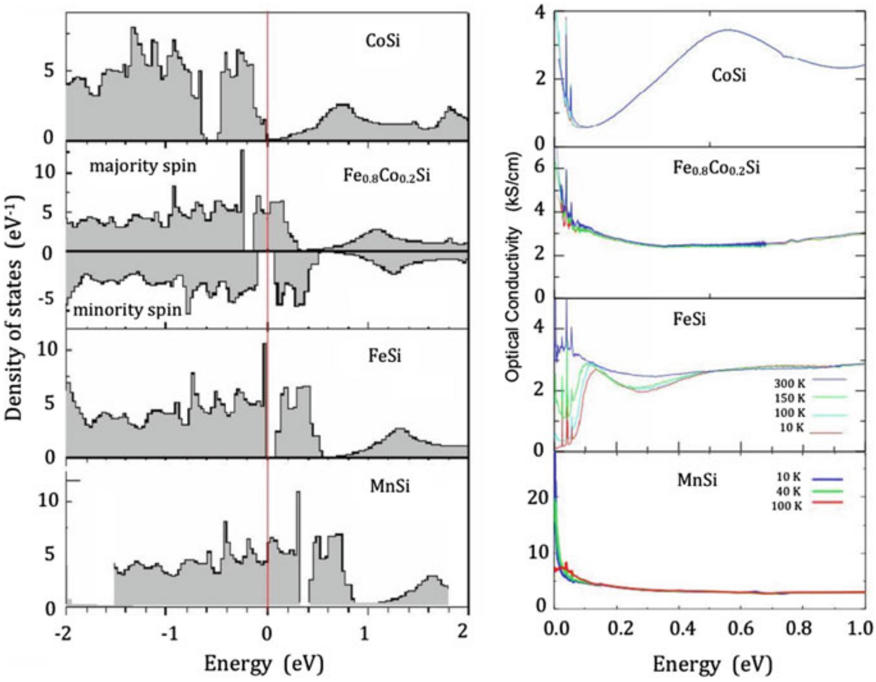

Fig. 9.4 Density of states (*left*) and real part of the optical conductivity (*right*) of a number of transition-metal silicides. *Source* [16]

doped semiconductor (Co-doped FeSi) are shown in Fig. 9.4. The optical conductivity of FeSi reveals the gap around the Fermi energy visible in the density of states of the left panel.

9.4.7 *The Drude-Lorentz Expression*

It is useful at this stage to relabel the transitions $m, n \rightarrow j$, where j is a generalized index, and to introduce a plasma frequencies for each transition with energy $\omega_{mn} = \omega_j$

$$\Omega_j(\boldsymbol{q})^2 = 8\pi \frac{\Omega |j_q^{nm}|^2}{\omega_{mn}} \frac{e^{-\beta E_n}}{Z} \tag{9.54}$$

with the help of which we obtain the following compact expression for the optical conductivity tensor

$$\sigma(\boldsymbol{q}, \omega) = \frac{i\omega}{4\pi} \sum_j \frac{\Omega_j(\boldsymbol{q})^2}{\omega(\omega + i\gamma_j) - \omega_j^2} \tag{9.55}$$

Although formally $\gamma_j = \eta = 0^+$, a natural modification of (9.55) consists of limiting the summation to a set of oscillators representing the main optical transitions and inserting a finite value for γ_j, which in zero'th approximation represents the inverse lifetime of the corresponding excited state (*e.g.* calculated using Fermi's Golden Rule). With this modification (9.55) is one of the most commonly used phenomenological representations of the optical conductivity, generally known as the Drude-Lorentz expression. An example is shown in Fig. 9.5. In its original incarnation the Drude-Lorentz expression is obtained from a model of classical damped oscillators.

The simple recipe of broadening the δ-functions by a life-time broadening is prone to pitfalls: For example, different line shapes of the oscillators are obtained if we make the substitution $\eta_1 = \eta = \gamma$ in (9.49). Apparently it is important to combine all (positive and negative) δ-functions coming from diamagnetic (σ^d) and regular (σ^r) terms in (9.49), not only in order to cancel out the negative δ-function in the origin, but also to make a connection to the Drude-Lorentz expression.

9.5 Spectral Weight Sum Rules

9.5.1 *K-Sum Rule*

We define the total spectral weight of the optical conductivity as follows

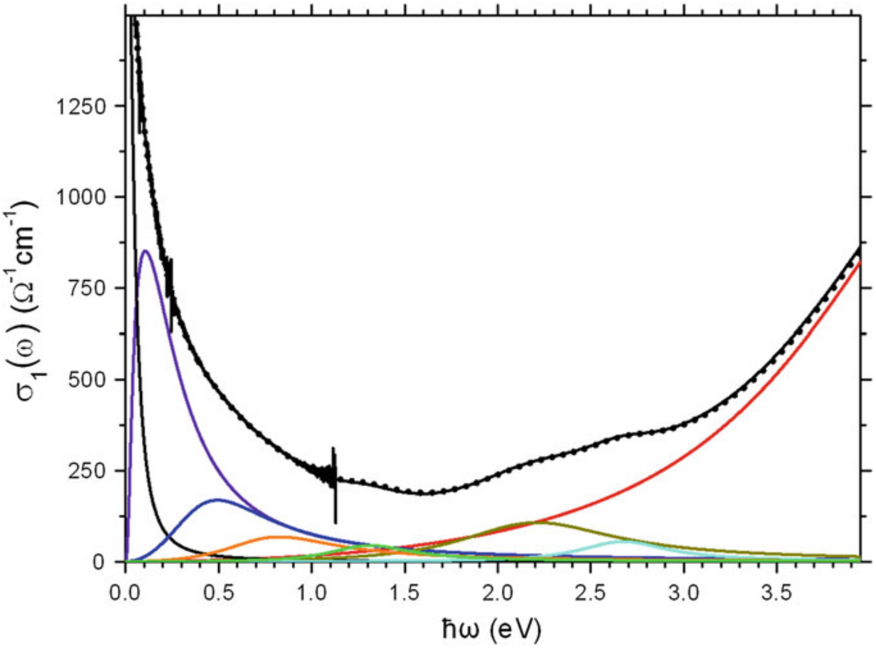

Fig. 9.5 Optical conductivity of $HgBa_2CuO_4$, shown together with a fit to (9.55). Each of the colored curves represents a separate term defined by its position ω_{mn}, strength Ω^2_{mn} and width γ_{mn}. *Source* [17]

$$W(\boldsymbol{q}) \equiv \mathrm{Re} \int_{-\infty}^{\infty} \sigma(\boldsymbol{q}, \omega) d\omega \tag{9.56}$$

The optical conductivity has, as we have seen in the previous subsection, a diamagnetic part and a regular part. We split up the corresponding contributions to the spectral weight accordingly, i.e.

$$\begin{aligned} W(\boldsymbol{q}) &= W^d(\boldsymbol{q}) + W^r(\boldsymbol{q}) \\ W^d(\boldsymbol{q}) &= \mathrm{Re} \int_{-\infty}^{\infty} \sigma^d(\boldsymbol{q}, \omega) d\omega \\ W^r(\boldsymbol{q}) &= \mathrm{Re} \int_{-\infty}^{\infty} \sigma^r(\boldsymbol{q}, \omega) d\omega \end{aligned} \tag{9.57}$$

To calculate $W^d(\boldsymbol{q})$ we use (9.35)

$$W^d(\boldsymbol{q}) = \mathrm{Re} \int_{-\infty}^{\infty} \frac{iK(\boldsymbol{q})}{\omega + i\eta} d\omega = \pi q_e^2 K(\boldsymbol{q}) \tag{9.58}$$

where $\boldsymbol{K}(\boldsymbol{q})$ is defined in (9.27). The substitution $\omega \rightarrow \omega + i\eta$ in the denominator is needed to ensure causality and convergence of the integrals. The same substitution is used in (9.46) from which we calculate $W^r(\boldsymbol{q})$.

$$W^r(\boldsymbol{q}) = \mathrm{Re} \int_{-\infty}^{\infty} d\omega \frac{i}{\omega + i\eta} \int_{0}^{\infty} \chi_{jj}(\boldsymbol{q}, t) e^{i\omega t} dt \tag{9.59}$$

It is easy to see that the total regular spectral weight is zero. We first interchange the order of integration over frequency and time:

$$W^r(\boldsymbol{q}) = \mathrm{Re} \int_{0}^{\infty} dt \chi_{jj}(\boldsymbol{q}, t) \int_{-\infty}^{\infty} \frac{i}{\omega + i\eta} e^{i\omega t} d\omega \tag{9.60}$$

We can calculate this by a contour integral. Since $t > 0$ the term $e^{i\omega t}$ converges exponentially to zero for $|\omega| \rightarrow \infty$ provided that ω is in the upper half of the complex plane. The contour integral along the half-circle in the upper half of the complex plane than also converges to zero for $|\omega| \rightarrow \infty$. The integral along the real axis has the same value as the contour consisting of the integral along the real axis and the half-circle in the upper half plane. To calculate the latter contour, we can apply Cauchy's residue theorem. However, the only pole in the integrand occurs for for $\omega = -i\eta$, and this is in the *lower half* of the complex plane. Since the contour-integral encloses no poles, the integral over ω has a vanishing result. Consequently

$$W^r(\boldsymbol{q}) = 0 \tag{9.61}$$

Apparantly there is an exact compensation going on between positive and negative contributions to the "regular" optical conductivity. In the previous subsection we discussed this in relation to (9.49): The regular optical conductivity has a negative δ-function at zero frequency, which has exactly the same spectral weight as the conductivity integrated over all finite frequencies. Moreover, for all metals except superconductors there is an exact cancellation between the positive δ-function coming from the diamagnetic response, and the negative one from the regular part of the conductivity.

This completes the general discussion of the spectral weight sum rule. Since this is a central theorem of many-body physics, we write here in full glory the limit for $q \rightarrow 0$

$$\mathrm{Re}\int_{-\infty}^{\infty}\boldsymbol{\sigma}(0,\omega)d\omega=\frac{\pi q_e^2}{\Omega}\sum_k\frac{\partial^2\epsilon_k}{\partial\boldsymbol{k}^2}\left\langle c_k^\dagger c_k\right\rangle \tag{9.62}$$

Two cases of the spectral weight sumrule, (9.62) are of particular importance, which we detail in the following two subsections.

9.5.2 F-Sum Rule

The free electron dispersion $\epsilon_k = k^2/(2m)$ gives

$$\mathrm{Re}\int_{-\infty}^{\infty}\sigma(\boldsymbol{q},\omega)d\omega=\frac{\pi n q_e^2}{m} \tag{9.63}$$

This is the f-sum rule, or Thomas-Reich-Kuhn rule. It is a cornerstone for optical studies of materials, since it relates the integrated optical conductivity directly to the density of charged objects, and the absolute value of their charge and mass. It reflects the fundamental property that also in strongly correlated matter the number of electrons is conserved.

9.5.3 Kinetic Energy Sum Rule

The nearest-neighbour tight-binding model has a dispersion relation $\epsilon_k = -2t \cos(ka)$, where a is the lattice constant. The implication for the K-sum rule is

$$\mathrm{Re}\int_{-\infty}^{\infty}\sigma(q,\omega)d\omega=\pi q_e^2 K(q) \tag{9.64}$$

$$K(q)=\frac{1}{\Omega}\sum_k 2ta^2\cos(ka)\cos^2\left(\frac{qa}{2}\right)\langle c_k^\dagger c_k\rangle=-\cos^2\left(\frac{qa}{2}\right)\frac{a^2}{\Omega}\langle\hat{H}^{kin}\rangle$$

This is also known as the “kinetic energy sum rule”, since the spectral weight is proportional to the minus the average kinetic energy of the electrons. An example of such a measurement is shown in Fig. 9.6 for a high temperature superconductor. Besides a gradual change of the kinetic energy (spectral weight) as a function of temperature, one observes a sudden change at the superconducting phase transition at 110 K.

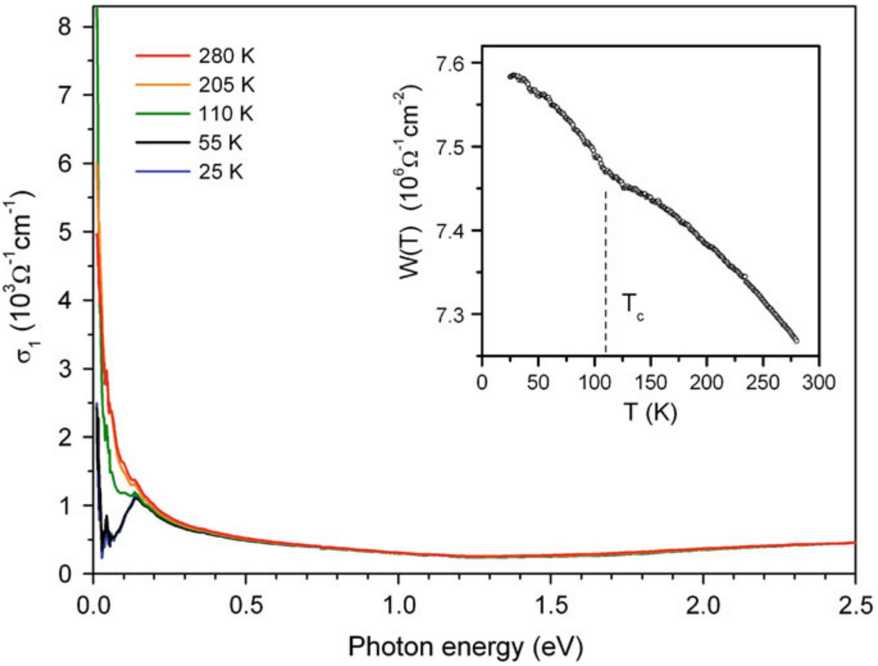

Fig. 9.6 Re$\sigma(\omega)$ of the high T_c superconductor $Bi_2Sr_2Ca_2Cu_3O_{10}$ for some selected temperatures. Inset: Free carrier spectral weight as a function of T. The *dotted line* signals the critical temperature of the sample ($T_c = 110$ K). Note that in the superconducting state the optical conductivity has a δ-function at $\omega = 0$. The spectral weight of this δ-function has been experimentally determined from the imaginary part of the optical conductivity (not shown), and it's contribution is taken into account in $W(T)$. *Source* [18]

9.5.4 Regular Part of the Spectral Weight

The exact compensation between the negative *zero*-frequency mode and the finite frequency spectral weight of the regular spectral weight (9.61) deserves some extra attention. It is still interesting to verify the amount present at zero frequency. This is easily obtained. From inspection of (9.59), we notice that the real part of $\frac{i}{\omega+i\eta}$ represents a δ-function $\pi\delta(\omega)$. The spectral weight of the zero-frequency mode is then

$$\boldsymbol{W}^{r,0}(\boldsymbol{q}) = \pi\mathrm{Re}\int_{-\infty}^{\infty} \chi_{jj}(\boldsymbol{q},t)dt = \pi\Omega\mathrm{Im}\int_{0}^{\infty} \left\langle\left[\hat{\boldsymbol{j}}^{r}_{q}(t), \hat{\boldsymbol{j}}^{r}_{-q}(0)\right]\right\rangle dt \tag{9.65}$$

where (9.44) was substituted to obtain the righthand part of the equation. We first define the "dipole" field operator with the property

$$\frac{\partial \hat{\boldsymbol{p}}_q(t)}{\partial t} = \hat{\boldsymbol{j}}^r_q(t) \tag{9.66}$$

We can now integrate (9.65) and obtain

$$\boldsymbol{W}^{r,0}(\boldsymbol{q}) = \pi\Omega\text{Im}\left(\left\langle\left[\hat{\boldsymbol{p}}_q(\infty), \hat{\boldsymbol{j}}^r_{-q}(0)\right]\right\rangle - \left\langle\left[\hat{\boldsymbol{p}}_q(0), \hat{\boldsymbol{j}}^r_{-q}(0)\right]\right\rangle\right) \tag{9.67}$$

The dipole (current) operator is the position (velocity) operator times the electron charge. Maldague [8] used the position operator for $q = 0$ defined as $\hat{\boldsymbol{x}} = \sum_m \boldsymbol{r}_m c^\dagger_m c_m$, and calculated the commutator with the velocity operator, resulting in the K-sum rule for $q = 0$. Here we generalize this result to $q \neq 0$. It is tempting to use $\hat{\boldsymbol{x}}_q = \sum_m e^{-i\boldsymbol{q}\cdot\boldsymbol{r}_m}\boldsymbol{r}_m c^\dagger_m c_m$. While the time derivative has the required property, that $\partial\hat{\boldsymbol{x}}_q/\partial t = i[\hat{H}^{kin}, \hat{\boldsymbol{x}}_q] = \hat{\boldsymbol{v}}_q$, there are obvious difficulties with this definition due to the divergence of $\boldsymbol{r}_m$ in the thermodynamic limit. However, those difficulties can be avoided. If we consider the following operator

$$\hat{\boldsymbol{p}}_q \equiv -\frac{iq_e}{2\Omega}\sum_k \frac{\boldsymbol{v}_k + \boldsymbol{v}_{k-q}}{\epsilon_k - \epsilon_{k-q}} c^\dagger_k c_{k-q}, \tag{9.68}$$

then we notice that there are no divergencies of the expression in the thermodynamic limit for any $q \neq 0$. Moreover, it is easy to verify, that

$$\partial\hat{\boldsymbol{p}}_q/\partial t = i\left[\hat{H}^0, \hat{\boldsymbol{p}}_q\right] = \hat{\boldsymbol{j}}^r_q \tag{9.69}$$

which implies that $\hat{\boldsymbol{p}}_q$ corresponds to the momentum-space Fourier transform of the dipole operator. The first term in (9.67) represents the response in the $\boldsymbol{p}_q$-channel after an infinite amount of time, which is certainly zero. The second term can be calculated directly from the commutation relations

$$\Omega\left\langle\left[\hat{\boldsymbol{p}}_q, \hat{\boldsymbol{j}}^r_{-q}\right]\right\rangle = iq_e^2\boldsymbol{K}^r(\boldsymbol{q})$$

where

$$\boldsymbol{K}^r(\boldsymbol{q}) = \frac{1}{4\Omega}\sum_k\left\{\frac{(\boldsymbol{v}_{k+q} + \boldsymbol{v}_k)^2}{\epsilon_{k+q} - \epsilon_k} + \frac{(\boldsymbol{v}_{k-q} + \boldsymbol{v}_k)^2}{\epsilon_{k-q} - \epsilon_k}\right\}\left\langle c^\dagger_k c_k\right\rangle \tag{9.70}$$

Consequently

$$\boldsymbol{W}^{r,0}(\boldsymbol{q}) = -\pi q_e^2\boldsymbol{K}^r(\boldsymbol{q}) \tag{9.71}$$

The amount of spectral weight at finite frequencies exactly balances this amount, hence

$$\boldsymbol{W}^{r,+}(\boldsymbol{q}) = \int_{0^+}^{\infty} \mathrm{Re}\boldsymbol{\sigma}(\boldsymbol{q},\omega)d\omega = \frac{\pi q_e^2}{2}\boldsymbol{K}^r(\boldsymbol{q}) \tag{9.72}$$

Let us now consider the properties of $K^r(\boldsymbol{q})$. In the first place it is interesting to look at the limit $q \to 0$. We can use that $\boldsymbol{v}_{k+q} - \boldsymbol{v}_k = \boldsymbol{q} \cdot \partial^2\epsilon_k/\partial\boldsymbol{k}^2$ for the terms in the numerators, and $\epsilon_{k+q} - \epsilon_k = \boldsymbol{q} \cdot \partial\epsilon_k/\partial\boldsymbol{k} + (q^2/2)\partial^2\epsilon_k/\partial k^2$ in the denominators to show that

$$\lim_{q\to 0}\left\{\frac{(\boldsymbol{v}_{k+q}+\boldsymbol{v}_k)^2}{\epsilon_{k+q}-\epsilon_k} + \frac{(\boldsymbol{v}_{k-q}+\boldsymbol{v}_k)^2}{\epsilon_{k-q}-\epsilon_k}\right\} = 4\frac{\partial^2\epsilon_k}{\partial\boldsymbol{k}^2}. \tag{9.73}$$

Consequently by comparing to (9.28) we see, that for $q = 0$ there is a perfect compensation of the zero-frequency spectral weights of the diamagnetic and the regular parts of the conductivity:

$$\boldsymbol{K}^r(0) = \frac{1}{\Omega}\sum_k \frac{\partial^2\epsilon_k}{\partial\boldsymbol{k}^2}\langle c_k^\dagger c_k\rangle = \boldsymbol{K} \tag{9.74}$$

Again we consider the most commonly encountered case: The free electron dispersion $\epsilon_k = k^2/(2m)$ gives

$$\int_{0^+}^{\infty} \mathrm{Re}\sigma(q,\omega)d\omega = \frac{\pi q_e^2}{2m}\sum_k \langle c_k^\dagger c_k\rangle \tag{9.75}$$

which is the same expression as the familiar f-sum rule, (9.63), and we see that in this case $K^r(q) = K$ is independent of q.

With the tightbinding formula $\epsilon_k = -2t\cos(ka)$ one obtains after some goniometric manipulations

$$K^r(q) = \cos^2\left(\frac{qa}{2}\right)\frac{1}{\Omega}\sum_k 2\,Ta^2\cos(ka)\left\langle c_k^\dagger c_k\right\rangle = K\cos^2\left(\frac{qa}{2}\right) \tag{9.76}$$

which corresponds exactly to the result in (9.64).

The fact that the same expression for $K(q)$ is found in (9.76) and (9.64) indicates the perfect compensation of the positive (diamagnetic) and negative (regular) zero-frequency delta-functions. This result makes perfect sense physically; the implication is that no dissipation-less DC currents can flow for any wave-vector. Presumably the implementation of the Peierls substitution used here is quite accurate, despite some ambiguity for any $q \neq 0$ due to the course-graining procedure of the tight-binding form.[2]

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[2] One can pose the question whether the corresponding expression for the current density satisfies the continuity equation. On a fundamental level this relation expresses the conservation of the number

Following (9.72) the spectral weight of the optical conductivity **decreases as a function of increasing q and vanishes at the Brillouin-zone boundary**. From inspection of (9.68) it is clear why this is the case: for $q = \pi$ there is an exact cancellation of terms in the numerator, i.e. $v_k + v_{k+\pi} = 0$. In other words, all optical matrix elements are zero for $q = \pi$ and consequently the intensity of the optical spectrum vanishes in this limit. While for general $\epsilon(k)$ dispersion such an exact cancellation is not expected, yet this indicates that the trend that $W(q)$ diminishes for increasing q is the rule rather than the exception. With the advent of new experimental techniques which allow the exploration of the optical conductivity at finite $\boldsymbol{q}$ [2–4] the $\boldsymbol{q}$-dependent optical conductivity sumrule in (9.72) provides a lower bound on the intensity of the free carrier optical response of correlated electrons.

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References

1. D.N. Basov, R.D. Averitt, D. van der Marel, M. Dressel, K. Haule, Rev. Mod. Phys. **83**, 471 (2011)
2. F. Keilmann, R. Hillenbrand, Phil. Trans. A **362**, 787 (2004)
3. F. Huth, A. Govyadinov, S. Amarie, W. Nuansing, F. Keilmann, R. Hilenbrand, Nano Lett. **12**, 3973 (2012)
4. J. Chen, M. Badioli, p Alonso-Gonzlez, S. Thongrattanasiri, F. Huth, J. Osmond, M. Spasenović, A. Centeno, A. Pesquera, P. Godignon, A.Z. Elorza, N. Camara, F. Javier Garca de Abajo, R. Hillenbrand, F.H.L. Koppens, Nature **487**, 77 (2012)
5. M. Dressel, G.G. Grüner, *Electrodynamics of Solids: Optical Properties of Electrons in Matter* (Cambridge University Press, Cambridge, 2002)
6. C.F. Klingshirn, *Semiconductor Optics* (Springer, Berlin, 1995)
7. D. Pines, P. Nozières, *The Theory of Quantum Liquids, Part I* (Benjamin, New York, 1966)
8. F.P. Maldague, Phys. Rev. B **16**, 2437 (1977)
9. G.D. Mahan, *Many-Particle Physics*, 2nd edn. (Plenum, New York, 1990), pp. 207–223
10. A. Millis, in *Strong Interactions in Low Dimensions*, Physics and Chemistry of Materials with Low-Dimensions, Vol. 25, (Springer, Dordrecht, 2004)
11. D. van der Marel, in *Strong Interactions in Low Dimensions*, Physics and Chemistry of Materials with Low-Dimensions, Vol. 25, (Springer, Dordrecht, 2004)
12. E. van Heumen, D. van der Marel, in *Lectures on the Physics of Strongly Correlated Systems XI*, ed. by A. Avella, F. Manzini, (American Institute of Physics, New York, 2007)
13. A. Damascelli, D. van der Marel, M. Grueninger, C. Presura, T.T.M. Palstra, J. Jegoudez, A. Revcolevschi, Phys. Rev. Lett. **81**, 918 (1998)

(Footnote 2 continued)
of particles. This condition corresponds to the local constraint $\nabla \cdot \boldsymbol{J} + \partial\rho/\partial t = 0$ in continuous space. Although there is no obvious way to define a quantity equivalent to $\nabla \cdot \boldsymbol{J}$ for a lattice, the situation is in fact somewhat simpler. It is sufficient to verify that removal of an electron from a given site $\boldsymbol{r}_m$ is always compensated by the creation of an electron elsewhere in the lattice. Since the current operator in (9.23) swaps electrons between different sites, the conservation of particle number is therefor built in the definition of the current operator

14. C.N. Presura, Ph.D. thesis, Rijksuniversiteit Groningen, 2003
15. S. Nakai, N. Nuecker, H. Romberg, M. Alexander, J. Fink, Phys. Scripta **41**, 596 (1990)
16. F.P. Mena, J.F. DiTusa, D. van der Marel, G. Aeppli, D.P. Young, A. Damascelli, J.A. Mydosh, Phys. Rev. B **73**, 085205 (2006)
17. S. I. Mirzaei, D. Stricker, J. N. Hancock, C. Berthod, A. Georges, E. van Heumen, M. K. Chan, X. Zhao, Y. Li, M. Greven, N. Barisic, D. van der Marel, Proc. Natl. Acad. Sci. USA **110**, 5774 (2013)
18. F. Carbone, A.B. Kuzmenko, H.J.A. Molegraaf, E. van Heumen, E. Giannini, D. van der Marel, Phys. Rev. B **74**, 024502 (2006)