ON THE OPTICAL ABSORPTION SPECTRA OF AN EXTENDED HUBBARD MODEL

By W. Gasser and M. Börner

Sektion Physik der Technischen Universität Dresden*

Starting from the Kubo formula we calculate the optical absorption spectra of an extended Hubbard model including the two-lattice-site Coulomb interaction. The calculations based on the equation of motion formalism of the Green functions are carried out in the limiting cases $U \gg U_1, U_2, t$ and $U, U_1, U_2 \gg t$. It is shown that there are special selection rules for electron transitions which have striking influence on the optical absorption spectra.

1. Introduction

A series of papers devoted to the properties of an extended Hubbard model were recently published Ref. [1]. The Hamiltonian of this extended Hubbard model includes the two-lattice-site Coulomb interaction and is given by

$$H = \sum_{ijs} t_{ij} c_{is}^+ c_{js} + (U/2) \sum_{is} n_{is} n_{i-s} + (\tfrac{1}{2}) \sum_{ijs} U_{1ij} n_{is} n_{js} + (\tfrac{1}{2}) \sum_{ijs} U_{2ij} n_{is} n_{j-s}, \qquad (1)$$

where the sums have to be taken with respect to nearest neighbours only $n_{is} = c_{is}^+ c_{is}$, c_{is}^+ and c_{is} are the creation and annihilation operators respectively for electrons on site i and spin s. t_{ij} is the transfer matrix element, U is the Coulomb interaction potential for electrons on the same lattice-site, and U_1 and U_2 are potentials for electrons at neighbouring lattice-sites.

The main interest of the authors mentioned above was concentrated on the influence of the two-lattice-site Coulomb interaction on the Hubbard splitting of the energy band and on magnetic properties. Here we calculate the optical absorption spectrum which is determined by the real part of the diagonal elements of the conductivity tensor. The starting point of the calculation is the well-known Kubo formula for the conductivity tensor expressed in terms of two-time retarded Green functions. The evaluation of these Green

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* Address: Sektion Physik, Technische Universitat Dresden, Mommsenstrasse 13, 8027 Dresden, DDR.

functions will be performed for both $U, U_1, U_2 \gg t$ and $U \gg U_1, U_2, t$ using the equation of motion method. In the first case a suitable Ansatz for the Green function allows a solution of the equation of motion which is exact in the atomic limit. In the second one, appropriate approximations for the higher-order Green functions arising from the equation of motion method must be introduced. The physical content of the approximations is that of Hubbard's first paper Ref. [2] and corresponds to neglecting the scattering processes of an electron due to temporal and spatial fluctuations in the electron distribution. In both the limits the spectra are quite different from those for the pure Hubbard model. Apart from this we shall see that they are influenced by a selection rule for electron transitions so that they also differ considerably from those which one should expect from the calculations of the one-particle Green function Ref. [1].

In Sec. 2 we give the formulation of the problem and introduce some notations. Strong intraatomic and interatomic interaction ($U, U_1, U_2 \gg t$) is considered in Sec. 3. In Sec. 4 we treat the case $U \gg U_1, U_2, t$ regarding the linear paramagnetic chain and $n = 1$ for the sake of simplicity. Finally, Sec. 5 contains some concluding remarks.

2. Optical absorption

The optical absorption spetrum is given by the real part of the diagonal elements of the conductivity tensor. In the linear response theory the frequency-dependent conductivity tensor $\sigma_{\mu\nu}(\omega)$ is given by the well-known Kubo formula

$$\sigma_{\mu\nu}(\omega) = (1/V) \lim_{\delta \to 0+} \langle\!\langle J_\mu ; P_\nu \rangle\!\rangle_{\omega - i\delta}. \tag{2}$$

P_ν is the ν-component of the polarization operator

$$P_\nu = e \sum_{is} R_{i\nu} n_{is}, \tag{3}$$

where the $R_{i\nu}$ are the components of the lattice-site position vectors. J_μ is the μ-component of the current operator

$$J_\mu = \frac{dP_\mu}{dt} = ie \sum_{ijs} (R_j - R_i)_\mu t_{ij} c_{is}^+ c_{js}. \tag{4}$$

V is the volume of the system and e is the elementary charge. As usual we denote by $\langle\!\langle A; B \rangle\!\rangle_\omega$ the Fourier transform of the retarded Green function

$$\langle\!\langle A; B \rangle\!\rangle_\omega = \int_{-\infty}^{+\infty} e^{-i\omega t} i\Theta(t) \langle [A(t), B] \rangle dt \tag{5}$$

for Im $\omega < 0$. Substituting (4) into (2) we get

$$\sigma_{\mu\nu}(\omega) = ie(1/V) \lim_{\delta \to 0+} \sum_{ijs} (R_j - R_i)_\mu t_{ij} \langle\!\langle c_{is}^+ c_{js} ; P_\nu \rangle\!\rangle_{\omega - i\delta}, \tag{6}$$

so that our task now is to calculate the Green function

$$G_{ijs}(\omega) = \langle\langle c_{is}^+ c_{js}; P_v \rangle\rangle_\omega. \tag{7}$$

This will be done assuming that the Green function $\langle\langle A; B \rangle\rangle_\omega$ satisfies the following equation of motion

$$\langle\langle A; B \rangle\rangle_\omega = \langle [A, B] \rangle - \langle\langle [A, H]; B \rangle\rangle_\omega. \tag{8}$$

In general, Equation (8) contains higher-order Green functions, so that approximations must be performed. These approximations are determined by the physical situation under consideration. Next we study the very large intraatomic and interatomic Coulomb interaction matrix elements U, U_{1ij}, U_{2ij} but small transfer matrix elements t_{ij} and regard the lowest order of σ in t. Under this assumption we shall get an exact expression for the function (7). If $U \gg U_1, U_2, t$ (Sec. 4) we shall derive an approximate equation for the function (7) which allows the determination of σ.

3. Optical absorption of a system with strong intraatomic and two-lattice-site Coulomb interaction

In the following we seek an expression for $\sigma_{\mu v}(\omega)$ which is of first order in t. Since (6) is proportional to t we shall omit the influence of the kinetic energy of the Hamiltonian on the equation of motion for the Green function $\langle\langle c_{is}^+ c_{js}; P_v \rangle\rangle_\omega$. However, the parameters U_1 and U_2 will be taken into account exactly. We shall get a nonsymmetric dependence of the absorption spectrum on these parameters. To see this we resolve the Green function
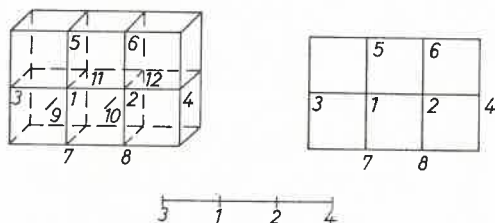

Fig. 1

(6) into components using the projection operators $n_{ls}^+ = n_{ls}$ and $n_{ls}^- = 1 - n_{ls}$. Of course, the resolution depends on the lattice type. Here we consider the simple cubic lattice, the square lattice and the linear chain. Now we denote the atoms i and j by 1 and 2. For the resolution of the Green functions we need all the atoms being nearest neighbours of 1 and 2 as indicated in Fig. 1. This enables us to write in the case of the simple cubic lattice

$$\langle\langle c_{1s}^+ c_{2s}; P_v \rangle\rangle_\omega = \sum_{a_1} \cdots \sum_{a_{12}} \sum_{b_3} \cdots \sum_{b_{12}} \langle\langle c_{1s}^+ c_{2s} \prod_{n=1}^{12} \prod_{m=3}^{12} n_{n-s}^{a_n} n_{ms}^{b_m}; P_v \rangle\rangle_\omega \tag{9}$$

with $a, b = +$ or $-$.

The equation of motion gives for the functions on the right-hand side of (9)

$$\{\omega-(U-U_2)(\delta_{a_1+}-\delta_{a_2+})-\sum_{n=2}^{6}[U_1(\delta_{b_{2n-1}+}-\delta_{b_{2n}+})-U_2(\delta_{a_{2n-1}+}-\delta_{a_{2n}+})]\}$$

$$\times\langle\!\langle c_{1s}^+c_{2s}\prod_{n=1}^{12}\prod_{m=3}^{12}n_{n-s}^{a_n}n_{ms}^{b_m};P_v\rangle\!\rangle_\omega = e(R_2-R_1)_v\langle c_{1s}^+c_{2s}\prod_{n=1}^{12}\prod_{m=3}^{12}n_{n-s}^{a_n}n_{ms}^{b_m}\rangle. \qquad (10)$$
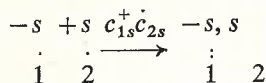
If we consider the square lattice or the linear chain the summation and product indices in (9) are limited to 8 or 4 resp. Correspondingly, the summation indices in (10) are limited to 4 or 2 resp. and the product indices in (10) are limited to 8 or 4 resp.

The set of all the poles of function (7) is represented in Table I. Typical transitions are shown in Fig. 3.

From Equation (10) one sees that there is a nonsymmetric dependence of the function (7) on the parameters U_1 and U_2. This nonsymmetry caused by the Pauli principle is expressed by the term

$$(U-U_2)(\delta_{a_1+}-\delta_{a_2+}), \qquad (11)$$

in (10) whereas the sum of all the remaining terms in (10) exhibits a complete symmetry due to these parameters. To understand this we neglect the surroundings of atoms 1 and 2 and regard both the atoms as occupied by two electrons of opposite spins.

$$\begin{array}{ccc} -s\ \ +s & c_{1s}^+c_{2s} & -s,\ s \\ \overset{\cdot}{1}\ \ \overset{\cdot}{2} & \longrightarrow & \overset{\vdots}{1}\ \ 2 \end{array}$$

The energy of the configuration on the left is $E^{(i)}=U_2$. The operator $c_{1s}^+c_{2s}$ entering in the expression for the current operator causes a jump of the s-spin electron from site 2 to site 1 so that we get a new configuration (on the right) with the energy $E^{(f)}=U$. If both the atoms were occupied by electrons with parallel spins the energy of that configuration would be $E^{(i)}=U_1$. But here an electron jump cannot take place because the operator $c_{1s}^+c_{2s}$ does not involve spin-flip. Consequently it results in the non-symmetric behaviour. In other words, a selection rule for the optical transition produced by the Pauli principle is obtained. To see the forbidden transitions we calculate the positions of the energy levels of our system. These positions of the energy levels come from the Green one-particle function

$$\langle\!\langle c_{is};c_{is}^+\rangle\!\rangle_\omega. \qquad (12)$$

We denote the ith atom by 1 and regard its nearest neighbours sphere as indicated in Fig. 2.

Fig. 2

TABLE I

$$
\begin{array}{c}
+6U_1+6U_2 \;\; +5U_1+6U_2 \;\; +4U_1+6U_2 \;\; +3U_1+6U_2 \;\; +2U_1+6U_2 \;\; +U_1+6U_2 \;\; +6U_2 \;\; -U_1+6U_2 \;\; -2U_1+6U_2 \;\; -3U_1+6U_2 \;\; -4U_1+6U_2 \;\; -5U_1+6U_2 \\
+6U_1+5U_2 \;\; +5U_1+5U_2 \;\; +4U_1+5U_2 \;\; +3U_1+5U_2 \;\; +2U_1+5U_2 \;\; +U_1+5U_2 \;\; +5U_2 \;\; -U_1+5U_2 \;\; -2U_1+5U_2 \;\; -3U_1+5U_2 \;\; -4U_1+5U_2 \;\; -5U_1+5U_2 \\
+6U_1+4U_2 \;\; +5U_1+4U_2 \;\; +4U_1+4U_2 \;\; +3U_1+4U_2 \;\; +2U_1+4U_2 \;\; +U_1+4U_2 \;\; +4U_2 \;\; -U_1+4U_2 \;\; -2U_1+4U_2 \;\; -3U_1+4U_2 \;\; -4U_1+4U_2 \;\; -5U_1+4U_2 \\
+6U_1+3U_2 \;\; +5U_1+3U_2 \;\; +4U_1+3U_2 \;\; +3U_1+3U_2 \;\; +2U_1+3U_2 \;\; +U_1+3U_2 \;\; +3U_2 \;\; -U_1+3U_2 \;\; -2U_1+3U_2 \;\; -3U_1+3U_2 \;\; -4U_1+3U_2 \;\; -5U_1+3U_2 \\
+6U_1+2U_2 \;\; +5U_1+2U_2 \;\; +4U_1+2U_2 \;\; +3U_1+2U_2 \;\; +2U_1+2U_2 \;\; +U_1+2U_2 \;\; +2U_2 \;\; -U_1+2U_2 \;\; -2U_1+2U_2 \;\; -3U_1+2U_2 \;\; -4U_1+2U_2 \;\; -5U_1+2U_2 \\
+6U_1+U_2 \;\; +5U_1+U_2 \;\; +4U_1+U_2 \;\; +3U_1+U_2 \;\; +2U_1+U_2 \;\; +U_1+U_2 \;\; +U_2 \;\; -U_1+U_2 \;\; -2U_1+U_2 \;\; -3U_1+U_2 \;\; -4U_1+U_2 \;\; -5U_1+U_2 \\
+6U_1 \;\; +5U_1 \;\; +4U_1 \;\; +3U_1 \;\; +2U_1 \;\; +U_1 \;\; 0 \;\; -U_1 \;\; -2U_1 \;\; -3U_1 \;\; -4U_1 \;\; -5U_1 \\
+5U_1-U_2 \;\; +5U_1-U_2 \;\; +4U_1-U_2 \;\; +3U_1-U_2 \;\; +2U_1-U_2 \;\; +U_1-U_2 \;\; -U_2 \;\; -U_1-U_2 \;\; -2U_1-U_2 \;\; -3U_1-U_2 \;\; -4U_1-U_2 \;\; -5U_1-U_2 \\
+5U_1-2U_2 \;\; +5U_1-2U_2 \;\; +4U_1-2U_2 \;\; +3U_1-2U_2 \;\; +2U_1-2U_2 \;\; +U_1-2U_2 \;\; -2U_2 \;\; -U_1-2U_2 \;\; -2U_1-2U_2 \;\; -3U_1-2U_2 \;\; -4U_1-2U_2 \;\; -5U_1-2U_2 \\
+5U_1-3U_2 \;\; +5U_1-3U_2 \;\; +4U_1-3U_2 \;\; +3U_1-3U_2 \;\; +2U_1-3U_2 \;\; +U_1-3U_2 \;\; -3U_2 \;\; -U_1-3U_2 \;\; -2U_1-3U_2 \;\; -3U_1-3U_2 \;\; -4U_1-3U_2 \;\; -5U_1-3U_2 \\
+5U_1-4U_2 \;\; +5U_1-4U_2 \;\; +4U_1-4U_2 \;\; +3U_1-4U_2 \;\; +2U_1-4U_2 \;\; +U_1-4U_2 \;\; -4U_2 \;\; -U_1-4U_2 \;\; -2U_1-4U_2 \;\; -3U_1-4U_2 \;\; -4U_1-4U_2 \;\; -5U_1-4U_2 \\
+5U_1-5U_2 \;\; +5U_1-5U_2 \;\; +4U_1-5U_2 \;\; +3U_1-5U_2 \;\; +2U_1-5U_2 \;\; +U_1-5U_2 \;\; -5U_2 \;\; -U_1-5U_2 \;\; -2U_1-5U_2 \;\; -3U_1-5U_2 \;\; -4U_1-5U_2 \;\; -5U_1-5U_2
\end{array}
$$

Regions with poles of the Green two–particle function of the linear chain, the square lattice and the s.c. lattice. These regions represent a part of the poles. The whole set of all the poles is got adding to every element of the matrix $\pm(U-U_2)$

Regions with poles of the Green one–particle function of the linear chain, the square lattice and the s.c. lattice. These regions represent one part of the set of the poles. The whole set is got adding to every element of the matrices $+U$
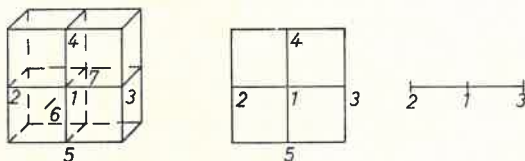
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For the simple cubic lattice we can write

$$\langle\langle c_{1s}; c_{1s}^+ \rangle\rangle_\omega = \sum_{a_1} \cdots \sum_{a_7} \sum_{b_2} \cdots \sum_{b_7} \langle\langle c_{1s} \prod_{n=1}^{7} \prod_{m=2}^{7} n_{n-s}^{a_n} n_{ms}^{b_m}; c_{1s}^+ \rangle\rangle_\omega. \tag{13}$$

The Green functions on the right hand side of (13) satisfy the equation of motion

$$\left[\omega - U\delta_{a_1+} - \sum_{n=2}^{7} (U_1\delta_{a_n+} - U_2\delta_{b_n+})\right] \langle\langle c_{1s} \prod_{n=1}^{7} \prod_{m=2}^{7} n_{n-s}^{a_n} n_{ms}^{b_m}; c_{1s}^+ \rangle\rangle_\omega$$

$$= \langle \prod_{n=1}^{7} \prod_{m=2}^{7} n_{n-s}^{a_n} n_{ms}^{b_m} \rangle. \tag{14}$$

For the square lattice or the linear chain, the summation and product indices in (13) and (14) are limited to 5 or 3 respectively.
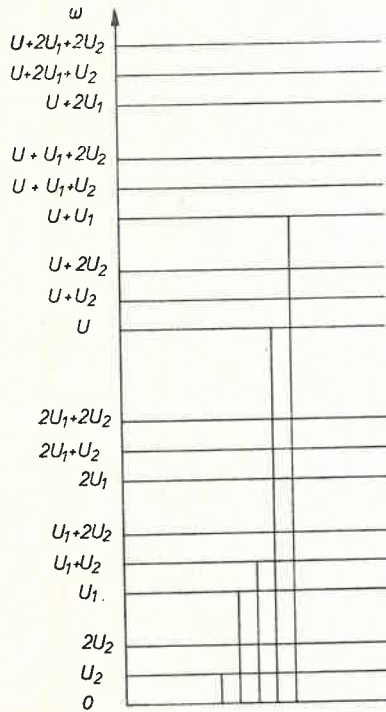
Fig. 3. Energy levels of the one-dimensional chain and the possible transitions between the zero level and the higher ones. Other transitions from higher levels to the zero level are forbidden due to the selection rule

We see from (15) that the set of energy levels is symmetric with respect to the parameters U_1 and U_2. The set of all the poles of function (13) is represented in Table I.

4. The absorption spectrum of the linear paramagnetic chain in the case of a weak interatomic Coulomb interaction

In this Section we regard the limit of strong intra-site Coulomb interaction U and small but finite matrix elements U_1, U_2 and t. In order to treat the influence of the largest parameter exactly we use a decomposition of the function (7) into components following Hubbard III Ref. [3]:

$$G_{ijs}(\omega) = \sum_{ab} G_{ijs}^{ab}(\omega) \quad \text{with} \quad G_{ijs}^{ab}(\omega) = \langle\!\langle c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b; P_v \rangle\!\rangle_\omega, \tag{15}$$

where a, b are equal to $+$ or $-$.

These components satisfy the equation of motion

$$[\omega - U(\delta_{a+} - \delta_{b+})] G_{ijs}^{ab}(\omega) = e(R_j - R_i)_v \Phi_{ijs}^{ab}$$

$$+ \sum_{cl} t_{il} \langle\!\langle c_{is}^+ n_{l-s}^c n_{i-s}^a c_{js} n_{j-s}^b; P_v \rangle\!\rangle_\omega$$

$$- \sum_{cl} t_{jl} \langle\!\langle c_{is}^+ n_{i-s}^a c_{ls} n_{l-s}^c n_{j-s}^b; P_v \rangle\!\rangle_\omega$$

$$- q_a \sum_l t_{il} \langle\!\langle (c_{i-s}^+ c_{l-s} - c_{l-s}^+ c_{i-s}) c_{is}^+ n_{j-s}^b c_{js}; P_v \rangle\!\rangle_\omega$$

$$+ q_b \sum_l t_{jl} \langle\!\langle c_{is}^+ n_{i-s}^a (c_{j-s}^+ c_{l-s} - c_{l-s}^+ c_{j-s}) c_{js}; P_v \rangle\!\rangle_\omega$$

$$+ \sum_l \langle\!\langle (U_{1il} n_{ls} + U_{2il} n_{l-s}) c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b; P_v \rangle\!\rangle_\omega$$

$$- \sum_l \langle\!\langle c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b (U_{1jl} n_{ls} + U_{2jl} n_{l-s}); P_v \rangle\!\rangle_\omega, \tag{16}$$

with $q_\pm = \pm 1$ and $\Phi_{ijs}^{ab} = \langle c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b \rangle$.

Obviously, the last two terms on the right hand side of (16) are caused by the terms added to the Hubbard model. To solve this equation we make use of the decoupling procedure of Hubbard's first paper (Ref. [2]). These approximations lead in all terms arising from the pure Hubbard model to the result of K. Kubo (Ref. [4], Eq. (3.8)). The remaining terms are decoupled in the same manner:

$$\sum_l \langle\!\langle (U_{1li} n_{ls} + U_{2li} n_{l-s}) c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b; P_v \rangle\!\rangle_\omega$$

$$= \langle\!\langle \Big[\sum_{l \neq j} (U_{1li} n_{ls} + U_{2li} n_{l-s}) + U_{2ij} \delta_{b+} \Big] c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b; P_v \rangle\!\rangle_\omega$$

$$\simeq \Big[\sum_l (U_{1li} n_s + U_{2li} n_{-s}) - U_{1ij} n_s - U_{2ij} n_{-s} + U_{2ij} \delta_{b+} \Big] \langle\!\langle c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b; P_v \rangle\!\rangle_\omega. \tag{17}$$

Correspondingly we approximate

$$\sum_l \langle\!\langle c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b (U_{1lj} n_{ls} + U_{2lj} n_{l-s}); P_v \rangle\!\rangle_\omega$$

$$= \langle\!\langle c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b \Big[\sum_{l \neq i} (U_{1lj} n_{ls} + U_{2lj} n_{l-s}) + U_{2ij} \delta_{a+} \Big]; P_v \rangle\!\rangle_\omega$$

$$\simeq \Big[\sum_l (U_{1lj} n_s + U_{2lj} n_{-s}) - U_{1ij} n_s - U_{2ij} n_{-s} + U_{2ij} \delta_{a+} \Big] \langle\!\langle c_{is}^+ n_{i-s}^a c_{js} n_{j-s}^b; P_v \rangle\!\rangle_\omega. \tag{18}$$

Substituting the approximations (17) and (18) into (16) we see that the terms proportional to U_1 cancel each other. Therefore we obtain:

$$[\omega - (U - U_{2ij})(\delta_{a+} - \delta_{b+})]G_{ijs}^{ab}(\omega) = e(R_j - R_i)_v \Phi_{ijs}^{ab}$$

$$+ n_{-s}^a \sum_{lc} t_{il} G_{ljs}^{cb}(\omega) - n_{-s}^b \sum_{lc} t_{jl} G_{ils}^{ac}(\omega), \tag{19}$$

with $n_{-s}^a = \langle n_{i-s}^a \rangle$ in virtue of translational invariance. Comparing this equation with Equation (10) of Sec. 3 one sees that (19) can be interpreted as a two-atomic cluster approximation with respect to the two-lattice-site Coulomb interaction. Moreover, U_1 does not occur explicitly in (19). This is caused by a general selection rule for the electronic transitions which we have discussed in detail in Sec. 3. Neglecting in U_{2ij} the dependence on lattice-sites i and j, Equation (19) can be solved easily by means of Fourier transformation. In this case both the solution and the discussion can be got from that of K. Kubo, Ref. [5], if we replace there the parameter U by $U - U_2$. But this corresponds to a nonrealistic situation since it implies that the Coulomb interaction potential U_2 has the same value for all distances between lattice-sites i and j. This error will be avoided in the next Section where Equation (19) will be solved taking into consideration the dependence of U_{2ij} on i and j.

From the term proportional to U_{2ij} considerable difficulties arise for the solution of Equation (19): it prevents a simple solution of (19) by means of a Fourier transform since it leads to a convolution integral in the momentum space. However, we shall make use of the fact that we only need the integral (6) rather than the full information contained in $G_{ijs}(\omega)$. For the sake of simplicity we restrict ourselves to the one-dimensional case and show in the Appendix how the following procedure can be generalized to cubic lattices. Next we rewrite (6) in the form

$$\sigma_{\mu\nu}(\omega) = -e(1/V) \lim_{\delta \to 0+} \sum_{ab} \sum_{ks} (\nabla_\mu t_k) G_{ks}^{ab}(\omega - i\delta), \tag{20}$$

where we have introduced the Fourier transforms

$$t_{ij} = (1/N) \sum_k t_k e^{ik(R_i - R_j)} \tag{21}$$

and

$$G_{ijs}^{ab} = (1/N) \sum_k G_{ks}^{ab} e^{ik(R_i - R_j)}. \tag{22}$$

For the one-dimensional chain we have $t_k = 2t \cos(ka)$ and $V = Na$ and therefore

$$\sigma(\omega) = \frac{2et}{N} \lim_{\delta \to 0+} \sum_{ab} \sum_{ks} \sin(ka) G_{ks}^{ab}(\omega - i\delta). \tag{23}$$

Obviously, to know $\sigma(\omega)$ it is sufficient to calculate $\sum_k \sin(ka) G_{ks}^{ab}$ instead of G_{ks}^{ab}

Now we return to Equation (19). The Fourier transform gives

$$(\omega - U(\delta_{a+} - \delta_{b+}))G_{ks}^{ab} + t_k \sum_c (n_{-s}^b G_{ks}^{ac} - n_{-s}^a G_{ks}^{cb})$$

$$= -ie(d/dk_v)\Phi_{ks}^{ab} + (\delta_{b+} - \delta_{a+})(1/N)\sum_p U_{2,p-k}G_{ps}^{ab}, \tag{24}$$

where we have introduced

$$\Phi_{ijs}^{ab} = (1/N)\sum_k \Phi_{ks}^{ab} e^{ik(Ri-Ri)} \tag{25}$$

and

$$U_{2ij} = (1/N)\sum_k U_{2,k} e^{ik(Ri-Ri)}. \tag{26}$$

For the one-dimensional chain we have $U_{2,k} = 2U_2 \cos(ka)$. It is convenient to define still the matrices

$$G_{ks} = \begin{bmatrix} G_{ks}^{++} \\ G_{ks}^{-+} \\ G_{ks}^{+-} \\ G_{ks}^{--} \end{bmatrix}; \quad B = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}; \quad \Phi_{ks} = \begin{bmatrix} \Phi_{ks}^{++} \\ \Phi_{ks}^{-+} \\ \Phi_{ks}^{+-} \\ \Phi_{ks}^{--} \end{bmatrix} \tag{27}$$

and

$$A_{ks} = \begin{bmatrix} \omega & -n_{-s}t_k & +n_{-s}t_k & 0 \\ -n_{-s}^- t_k & \omega + U + (2n_{-s}-1)t_k & 0 & +n_{-s}t_k \\ +n_{-s}^- t_k & 0 & \omega - U - (2n_{-s}-1)t_k & -n_{-s}t_k \\ 0 & +n_{-s}^- t_k & -n_{-s}^- t_k & \omega \end{bmatrix}. \tag{28}$$

Using (27) and (28) Equation (24) can be written in the following matrix form

$$A_{ks}G_{ks} = -ie(\partial/\partial k_v)\Phi_{ks} + B(1/N)\sum_p U_{2,p-k}G_{ps} \tag{29}$$

or

$$G_{ks} = -ieA_{ks}^{-1}(\partial/\partial k_v)\Phi_{ks} + A_{ks}^{-1}B(1/N)\sum_p U_{2,p-k}G_{ps}, \tag{30}$$

and finally for the linear chain

$$\sum_k \sin(ka)G_{ks} = ie\left[(1/N)\sum_q A_{qs}^{-1}B2U_2 \sin^2(qa) - 1\right]^{-1} \sum_k \sin(ka)A_{ks}^{-1}\frac{d}{dk}\Phi_{ks}. \tag{31}$$

After straightforward calculation we get in the case of the neutral half-filled band model the following solution

$$\sigma(\omega) = \lim_{\delta \to 0+} \frac{ie^2}{\omega - i\delta} \left[\frac{a}{N} \sum_k t_k \left(\Phi_{ks}^{++} + \Phi_{ks}^{--} + \frac{2UJ\Phi_{ks}^{+-}}{2 + J(2U - U_2)} \right) \right.$$

$$\left. + \frac{4t}{N} \sum_k \frac{\sin(ka) \left\{ 2(U - U_2)t_k \dfrac{d}{dk}(\Phi_{ks}^{++} - \Phi_{ks}^{--}) + (UU_2 - t_k^2 - (\omega - i\delta)^2) \dfrac{d}{dk} \Phi_{ks}^{+-} \right\}}{(2 + J(2U - U_2))((\omega - i\delta)^2 - U^2 - t_k^2)} \right] \quad (32)$$

with

$$J = \frac{U_2}{t^2} \left[1 - \sqrt{\frac{(\omega - i\delta)^2 - U^2 - 4t^2}{(\omega - i\delta)^2 - U^2}} \right].$$

The correlation functions Φ_{ks}^{ab} are given in the Appendix and it will be seen there that they are independent of U_1 and U_2 in the case $n = 1$ considered here.

The optical absorption spectrum is given by the real part of the conductivity. Here we do not give a detailed resolution of σ into real and imaginary part since it would lead to voluminous expressions. However, it can be easily seen that there occur two fundamental types of electron transitions. After a simple manipulation the energy-denominator of the second term in (32) takes the form:

$$\frac{1}{\omega_s^2(\omega - \omega_s)} + \frac{1}{\omega_s^2(\omega + \omega_s)} - \frac{1}{\omega_k^2(\omega - \omega_k)} - \frac{1}{\omega_k^2(\omega + \omega_k)} + \frac{2}{\omega_s^2 \omega_k^2 \omega} \quad (33)$$

with

$$\omega_k = \sqrt{U^2 - t^2} \quad \text{and} \quad \omega_s = \sqrt{U^2 - \frac{16[U_2(2U - U_2)]^2}{4t^2 + 16U_2(2U - U_2)}}.$$

The first and the second term of (33) represent electron transitions of the transfer energy ω_s. A further contribution to this type comes from the energy-denominator of the first term
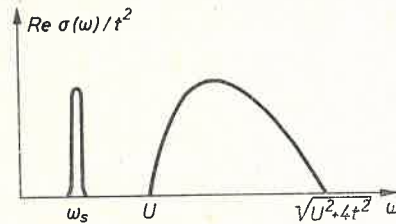

Fig. 4

of (32). The third and the forth term of (33) describe transitions of transfer energy ω_k and correspond to electron transitions between the split-band of the Hubbard model. Since k runs over the first Brillouin zone, the sharp lines $\delta(\omega - \omega_k)$ form an absorption

band of band width $\sqrt{U^2+4t^2}-U$. The absorption line $\delta(\omega-\omega_s)$ is well-seperated from this band. The separation distance is about U_2. Hence the reduced absorption spectrum has the shape qualitatively represented in Fig. 4. The reduced absorption spectrum is defined by σ/t^2. This quantity is of more interest than σ as we shall see in the following discussion of the limiting behaviour. If we keep fixed U_2 and regard the limit $t \to 0$ of σ/t^2, the absorption line $\delta(\omega-\omega_s)$ will be shifted only toward the position $(U-U_2)$ whereas
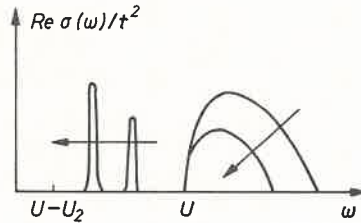
Fig. 5

the band will be both narrowed and lowered because the oscillator strength for the transitions $\delta(\omega-\omega_k)$ is proportional to t^2 for fixed U_2. Moreover, the lower edge of the band is unchanged if t goes to zero. In Fig. 5 the behaviour of the absorption spectrum for decreasing band width is represented. The arrow indicates the direction in which t decreases.
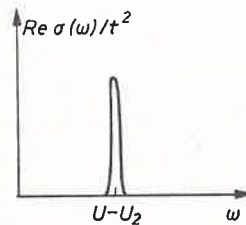

Fig. 6

In the limit $t = 0$ of σ/t^2 the band disappears and the sharp line $\delta(\omega-\omega_s)$ is now in the position $U-U_2$ (Fig. 6).

If we keep fixed t and consider the limit $U_2 \to 0$ of σ/t^2, the band grows in height while the band edges are preserved. The line $\delta(\omega-\omega_s)$ will be lowered and shifted from
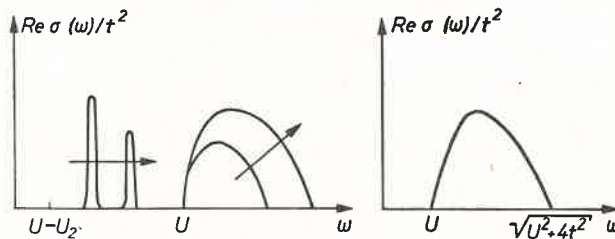

Fig. 7

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the left to the right and vanishes at U since the oscillator strength for the transitions of the frequency ω_s is proportional to U_2^2 for fixed t. This behaviour is plotted qualitatively in Fig. 7. The arrows there indicate the direction of decreasing U_2. Finally at $U_2 = 0 = t$ the reduced absorption spectrum consists in only one sharp line at the position U.

5. Concluding remarks

In the present paper the optical absorption spectrum of an extended Hubbard model was discussed. It was shown that the inter-atomic Coulomb interaction is of striking influence on the spectra at least with regard to the cases considered here: $U, U_1, U_2 \gg t$ and $U \gg U_1, U_2\, t$. In the first one, studied in Sec. 3, the resulting spectra consist of a number of dens-lying sharp lines between the edges U_2 and $U+U_1$ (for the one-dimensional chain — for the s. c. lattice the edges are U_2 and $U+5U_1+4U_2$). The distribution of these lines depends in a nonsymmetric way on the parameters U_1 and U_2 whereas one could expect a symmetric dependence since the poles of the one-particle Green function depend symmetrically on these parameters. The nonsymmetry of the two-particle Green function concerning U_1 and U_2 is caused by a selection rule for electron transitions in external electrical fields. This is illustrated in Fig. 3 for the one-dimensional chain. There we have plotted all the energy levels and the set of the possible transitions between the zero level and the others, and it can be seen that this set is smaller than that of all transitions which could have been imagined from the calculation of the one-particle function.

The selection rule is of importance in the second case studied in Sec. 4 too: it operates in such a way that for $n = 1$ the spectra will not depend on the parameter U_1. Moreover, the spectrum differs from that of the pure Hubbard model not only by a change in the shape of the Hubbard absorption band but also in the occurence of a new sharp line at a frequency well-separated from the band. This situation should be realized in materials like NiO, CoO or MnO and one should observe in experiments spectra similar to that qualitatively represented in Fig. 4. Optical absorption experiments in these materials should at least enable us to estimate the order of magnitude of the two-lattice-site Coulomb interaction matrix element U_2 and therefore allow us to answer the question whether this model or the pure Hubbard model is more suitable for the description of these materials.

The authors thank Professor Dr Haubenreisser, Professor Dr Ziesche and Dr Schneider for useful remarks.

APPENDIX

1. Equations for the conductivity in the case of cubic lattices

In the case of cubic lattices Equation (20) can be written in the form s. c.

$$\sigma_{xx} = -\frac{2tea}{V} \lim_{\delta \to 0+} \sum_{ks} \sin(k_x a) \sum_{ab} G_{ks}^{ab}(\omega - i\delta),$$

f. c. c.:

$$\sigma_{xx} = -\frac{4tea}{V}\lim_{\delta\to 0+}\sum_{ks}\sin\frac{k_x a}{2}\left[\cos\frac{k_y a}{2}+\cos\frac{k_z a}{2}\right]\sum_{ab}G_{ks}^{ab}(\omega - i\delta),$$

b. c. c.:

$$\sigma_{xx} = -\frac{4tea}{V}\lim_{\delta\to 0+}\sum_{ks}\sin\frac{k_x a}{2}\cos\frac{k_y a}{2}\cos\frac{k_z a}{2}\sum_{ab}G_{ks}^{ab}(\omega - i\delta).$$

Correspondingly we get instead of (31) the equations,

s. c.:

$$\sum_{k}\sin(k_x a)G_{ks} = -ie\sum_{k}\sin(k_x a)A_{ks}^{-1}\frac{\partial}{\partial k_x}\Phi_{ks}$$

$$+2U_2\sum_{k}\sin^2 k_x a\, A_{ks}^{-1}B\sum_{p}\sin(p_x a)G_{ps},$$

f. c. c.:

$$\sum_{k}\sin\frac{k_x a}{2}\cos\frac{k_y a}{2}G_{ks} = -ie\sum_{k}\sin\frac{k_x a}{2}\cos\frac{k_y a}{2}A_{ks}^{-1}\frac{\partial}{\partial k_x}\Phi_{ks}$$

$$+4U_2(1/N)\sum_{kp}\sin^2\frac{k_x a}{2}\cos\frac{k_z a}{2}\sin\frac{p_x a}{2}A_{ks}^{-1}BG_{ps}$$

$$\times\left[\cos\frac{k_y a}{2}\cos\frac{p_y a}{2}+\cos\frac{k_z a}{2}\cos\frac{p_z a}{2}\right]$$

and

$$\sum_{k}\sin\frac{k_x a}{2}\cos\frac{k_z a}{2}G_{ks} = -ie\sum_{k}\sin\frac{k_x a}{2}\cos\frac{k_z a}{2}A_{ks}^{-1}\frac{\partial}{\partial k_x}\Phi_{ks}$$

$$+4U_2(1/N)\sum_{kp}\sin^2\frac{k_x a}{2}\cos\frac{k_z a}{2}\sin\frac{p_x a}{2}A_{ks}^{-1}BG_{ps}$$

$$\times\left[\cos\frac{k_y a}{2}\cos\frac{p_y a}{2}+\cos\frac{k_z a}{2}\cos\frac{p_z a}{2}\right],$$

b. c. c.:

$$\sum_k \sin \frac{k_x a}{2} \cos \frac{k_y a}{2} \cos \frac{k_z a}{2} \, G_{ks}$$

$$= -ie \sum_k \sin \frac{k_x a}{2} \cos \frac{k_y a}{2} \cos \frac{k_z a}{2} \, A_{ks}^{-1} \frac{\partial}{\partial_{kx}} \, \Phi_{ks}$$

$$+ \frac{8U_2}{N} \sum_{kp} \sin^2 \frac{k_x a}{2} \cos^2 \frac{k_y a}{2} \cos^2 \frac{k_z a}{2} \sin \frac{p_x a}{2} \cos \frac{p_y a}{2} \cos \frac{p_z a}{2} \, A_{ks}^{-1} G_{ps}.$$

These equations can be solved directly with respect to the xx-components of the conductivity tensor.

2. Correlation functions

The correlation functions Φ_{ijs}^{ab} are calculated by Sakurai (Ref. [6]) in the case of the pure Hubbard model. For this purpose he has introduced the Green functions $\langle\!\langle c_{js} n_{j-s}^b; c_{is}^+ n_{i-s}^a \rangle\!\rangle$ which allow the determination of the functions Φ_{ijs}^{ab} by means of the spectrum theorem. In our case now we can write the following system of equations

$$(\omega - U\delta_{a+} + Z(U_1 n_s + U_2 n_{-s})) \langle\!\langle c_{js} n_{j-s}^b; c_{is}^+ n_{i-s}^a \rangle\!\rangle$$

$$= \delta_{ij}\delta_{ab} n_{-s}^b/2 + n_{-s}^b \sum_{lc} \langle\!\langle c_{ls} n_{i-s}^c; c_{is}^+ n_{i-s}^a \rangle\!\rangle t_{lj},$$

where we have made use of a similar decoupling procedure as in Sec. 4. The solution of this system comes from Ref. [6] substituting $\omega + U'$ for E in Ref. [6] with $U' = Z(U_1 n_s + U_2 n_{-s})$:

$$G_{ks}^{++} = \frac{n_{-s}}{2\pi} \frac{\omega + U' - n_{-s} t_k}{(\omega + U' - t_k)(\omega + U' - U) - n_{-s} U t_k},$$

$$G_{ks}^{+-} = G_{ks}^{-+} = \frac{n_{-s} n_{-s}}{2\pi} \frac{t_k}{(\omega + U' - t_k)(\omega + U' - U) - n_{-s} t_k U},$$

$$G_{ks}^{--} = \frac{n_{-s}}{2\pi} \frac{\omega + U' - U - n_{-s} t_k}{(\omega + U' - t_k)(\omega + U' - U) - n_{-s} t_k U}.$$

Of course, the sum $\sum_{ab} \langle\!\langle c_{js} n_{j-s}^b; c_{is}^+ n_{i-s}^a \rangle\!\rangle$ gives the function $\langle\!\langle c_{js}; c_{is}^+ \rangle\!\rangle$. For the correlation functions we get

$$\Phi_{ks}^{++} = \frac{n_{-s}}{E_{ks}^U - E_{ks}^L} [(E_{ks}^U - n_{-s} t_k) f(E_{ks}^U - U') - (E_{ks}^L - n_{-s} t_k) f(E_{ks}^L - U')],$$

$$\Phi_{ks}^{--} = \frac{n_{-s}^-}{E_{ks}^U - E_{ks}^L}\left[(U + n_{-s}t_k - E_{ks}^L)f(E_{ks}^L - U') - (U + n_{-s}t_k - E_{ks}^U)f(E_{ks}^U = U')\right],$$

$$\Phi_{ks}^{+-} = \Phi_{ks}^{-+} = \frac{n_{-s}^- n_{-s}t_k}{E_{ks}^U - E_{ks}^L}\left[f(E_{ks}^U - U') - f(E_{ks}^L - U')\right],$$

with

$$f(E_{ks}^{U,L} - U') = 1/(e^{\beta(E_{ks}^{U,L} - U' - \mu)} + 1)$$

and

$$E_{ks}^{U,L} = \tfrac{1}{2}\left(t_k + U \pm \sqrt{t_k^2 + 2(2n_{-s} - 1)t_k U + U^2}\right).$$

In the case solved here ($n_\uparrow = n_\downarrow = 1/2$) we have $\mu = U/2 - U'$ so that the functions Φ_{ks}^{ab} are independently on U_1 and U_2 respectively.

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