Optical absorption of strongly correlated half-filled Mott-Hubbard chains

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Abstract

In this last of three articles on the optical absorption of electrons in a half-filled Peierls-distorted chain we address the dimerized extended Hubbard model in the limit of a large on-site interaction $U$. When the Hubbard interaction is large both compared to the band width $W$ and the nearest-neighbor interaction $V$ the charge dynamics is properly described by the Harris-Lange model. This model can be exactly mapped onto a model of free spinless Fermions in parallel (Hubbard-)bands of width $W$ which are eventually Peierls-split.

To determine the coherent absorption features at low temperatures we design and employ the “no-recoil approximation” in which we assume that the momentum transfer to the spin degrees of freedom can only be $\Delta q_S = 0$ or $\Delta q_S = \pi/a$ during an optical excitation. We present explicit analytical results for the optical absorption in the presence of a lattice dimerization $\delta$ and a nearest-neighbor interaction $V$ for the Néel and dimer state. We find
that the coherent part of the optical absorption for $V = 0$ is given by a single peak at $\omega = U$ and broad but weak absorption bands for $W \delta \leq |\omega - U| \leq W$. The central peak at $\omega = U$ only vanishes for $\delta = 0$ in the Néel state. For an appreciable nearest-neighbor interaction $V > W/2$ almost all spectral weight is transferred to the $\Delta q_{C} = 0$-exciton and the $\Delta q_{C} = \pi/a$-exciton whose relative spectral weights very sensitively depend on both the lattice and the spin dimerization of the ground state.

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I. INTRODUCTION

In two previous articles (Gebhard, Bott, Scheidler, Thomas, and Koch I and II 1996) we have studied the optical absorption of non-interacting and strongly interacting tight-binding electrons in Peierls-distorted chains. These two different cases are idealizations of real almost ideal one-dimensional semiconductors (Farges 1994). While polymers like polyacetylen (Heeger, Kivelson, Schrieffer, and Su 1988); (Baeriswyl, Campbell, and Mazumdar 1992); (Schott and Nechtschein 1994) are commonly believed to be Peierls insulators some charge-transfer salts (Alcácer 1994); (Brau and Farges 1994) should be understood as Mott-Hubbard insulators. The extended Hubbard model for interacting electrons on a distorted chain at half-filling is considered appropriate for the latter class of materials (Mazumdar and Dixit 1986); (Fritsch and Ducasse 1991); (Mila 1995). Unfortunately, the study of the optical, i.e., finite frequency properties of correlated electron systems poses a very difficult many-body problem that cannot be solved analytically without further approximations on the Hamiltonian or the calculations (Kohn 1964); (Maldague 1977); (Lyo and Galinar 1977); (Lyo 1978); (Galinar 1979); (Campbell, Gammel, and Loh 1989); (Mahan 1990); (Shastry and Sutherland 1990); (Stafford, Millis, and Shastry 1991); (Fye, Martins, Scalapino, Wagner, and Hanke 1992); (Stafford and Millis 1993).

In our second article (Gebhard et al. II 1996) we focused on the optical absorption in the dimerized extended Harris-Lange model which is equivalent to the Hubbard model to lowest order in the strong-coupling expansion. We could exactly map the charge degrees of freedom onto two parallel (Hubbard-)bands for free spinless Fermions of band-width $W$. The eigenstates of the Harris-Lange model are highly spin-degenerate which allowed us to exactly calculate the optical absorption. The results apply to the “hot-spin” regime of the Hubbard model when the temperature is large compared to the spin exchange energy $J \sim W^2/(U - V)$. Real experiments are carried out at low temperatures for which the system is in an unique ground state with antiferromagnetic correlations. Unfortunately, this problem cannot be solved analytically. In this article we will employ the analogy to an ordinary
semiconductor (electrons and holes in a phonon bath) to design a “no-recoil approximation” for the “chargeons” in a “spinon bath”. It will allow us to determine the coherent absorption features of the Hubbard model at large $U/W$.

The paper is organized as follows. In section II we derive our approximate Hamiltonian from the Hubbard model. The charge dynamics will be governed by the Harris-Lange model while the ground state is determined by the Heisenberg model. Basic results of our articles (Gebhard et al. I and II 1996) for the optical absorption and the current operator are recalled in section III. In section IV we introduce the “no-recoil approximation” which allows us to obtain informations on the coherent part of the absorption spectrum at low temperatures. The corresponding results for the Peierls-distorted extended Harris-Lange model are presented in section V. Summary and conclusions close our presentation.

II. MODEL HAMILTONIANS

A. Charge degrees of freedom at strong coupling: the Harris-Lange model

As shown in (Gebhard et al. II 1996) the Peierls-distorted extended Hubbard model (Hubbard 1963); (Essler and Korepin 1994) can be mapped onto the Harris-Lange model in the limit of strong correlations. In standard notation of second quantization the latter model reads

$$
\hat{H}_{\text{HL},\text{ext}}^{\text{dim}} = \hat{T}_{\text{LHB}}(\delta) + \hat{T}_{\text{UHB}}(\delta) + U \hat{D} + V \hat{V} \quad (1a)
$$

$$
\hat{T}_{\text{LHB}} = (-t) \sum_{l,\sigma} (1 + (-1)^l \delta) \left( \hat{c}_{l,\sigma}^+ \hat{c}_{l+1,\sigma} + \hat{c}_{l+1,\sigma}^+ \hat{c}_{l,\sigma} \right) (1 - \hat{n}_{l+1,-\sigma}) \quad (1b)
$$

$$
\hat{T}_{\text{UHB}} = (-t) \sum_{l,\sigma} (1 + (-1)^l \delta) \hat{n}_{l,-\sigma} \left( \hat{c}_{l,\sigma}^+ \hat{c}_{l+1,\sigma} + \hat{c}_{l+1,\sigma}^+ \hat{c}_{l,\sigma} \right) \hat{n}_{l+1,-\sigma} \quad (1c)
$$

$$
\hat{D} = \sum_{l} \hat{n}_{l,\uparrow} \hat{n}_{l,\downarrow} \quad (1d)
$$

$$
\hat{V} = \sum_{l} (\hat{n}_{l} - 1) (\hat{n}_{l+1} - 1) \quad (1e)
$$
where \( \hat{n}_{t,\sigma} = \hat{c}_{t,\sigma}^{\dagger} \hat{c}_{t,\sigma} \) is the local density of \( \sigma \)-electrons and \( \hat{n}_t = \hat{n}_{t,\uparrow} + \hat{n}_{t,\downarrow} \) is the total local electron density. \( U \hat{D} \) is the Hubbard interaction between electrons on the same site, \( V \hat{V} \) is the nearest-neighbor interaction between charged objects like double occupancies and holes, and \( \hat{T}_{\text{LHB}} \) (\( \hat{T}_{\text{UHB}} \)) describes the motion of holes (double occupancies) in the lower (upper) Hubbard band. The Harris-Lange model is equivalent to the Hubbard model to order \( t(t/U)^{-1}, t(t/U)^0 \), and \( t(V/U)^0 \). Due to the Peierls distortion the electron transfer between two lattice sites is modulated by \( \pm t\delta \). We are only interested in the half-filled case where the number of electrons \( N \) equals the even number of lattice sites \( L \).

**B. Band structure interpretation**

For \( V = 0 \) and in the absence of a lattice distortion the exact eigenenergies of the Harris-Lange model are the same as those of independent spinless Fermions moving in two parallel bands separated by \( U \). For the case of linear optical absorption we may thus work with the effective band structure Hamiltonian

\[
\hat{H}^\text{band}_{\text{HL}} = \sum_{|k| \leq \pi/a} \left[ (U + \epsilon(k))\hat{n}^u_{k} + \epsilon(k)\hat{n}^l_{k} \right]
\]

with \( \epsilon(k) = -2ta \cos(ka) \), and \( \hat{n}^u_{k} = \hat{u}^+_k \hat{u}_k \), \( \hat{n}^l_{k} = \hat{l}^+_k \hat{l}_k \) for our fermionic quasi-particles (chargeons) in the upper and lower Hubbard band. Their band width is \( W = 4t \), the lattice spacing is \( a \), and the \( k \)-values of the first Brillouin zone are spaced by \( \delta k = 2\pi/(La) \).

The lattice distortion results in a Peierls splitting of the upper and lower Hubbard band. In this case the effective Hamiltonian for linear optical absorption becomes (Gebhard et al. II 1996)

\[
\hat{H}^\text{dim,\,band}_{\text{HL}} = \sum_{|k| \leq \pi/(2a), \tau = \pm 1} \left[ (U + \tau E(k))\hat{n}^u_{k,\tau} + \tau E(k)\hat{n}^l_{k,\tau} \right].
\]

with the dispersion relation

\[
E(k) = \sqrt{\epsilon(k)^2 + \Delta(k)^2}
\]

\[
\epsilon(k) = -2t \cos(ka)
\]
where $\hat{n}_{k,\tau}^{u}$ ($\hat{n}_{k,\tau}^{l}$) are the number operators for the quasi-particles for the upper ($\tau = +$) and lower ($\tau = -$) Peierls subband in the upper ($u$) and lower ($l$) Hubbard band.

C. Spin degrees of freedom at strong coupling: the Heisenberg model

At half-filling the ground state of the Harris-Lange model is $2^L$-fold degenerate. We are ultimately interested in the optical properties of the (Peierls-distorted extended) Hubbard model at strong correlations for which the ground state for large $U/W$ is unique. Consequently, we have to go to the next order in the expansion of the Hubbard model in $W/U$ to lift the above degeneracy. The Harris-Lange model is unsatisfactory in yet another aspect: at half filling a finite lattice distortion cannot be sustained within the model because there is no gain in electronic energy. The electronic part of the ground state energy is zero, irrespective of the Peierls parameter $\delta$.

A consistent treatment of the expansion in $W/U$ will also produce corrections to the Harris-Lange model (Harris and Lange 1967); (van Dongen 1994) which would render the problem intractable. Thus we argue that the degeneracy of the ground state will inevitably be lifted by a residual spin-spin interaction which may have its origin in the itinerant exchange in the Hubbard model (next term in the expansion in $W/U$) or in their direct exchange which is not taken into account in the extended Hubbard model where only the direct Coulomb terms were kept ("Zero Differential Overlap Approximation" (Kivelson, Su, Schrieffer, and Heeger 1987); (Wu, Sun, and Nasu 1987); (Baeriswyl, Horsch, and Maki 1988); (Gammel and Campbell 1988); (Kivelson, Su, Schrieffer, and Heeger 1988); (Campbell, Gammel, and Loh 1988); (Kivelson and Girlando 1989); (Campbell, Gammel, and Loh 1990)). The additional “Hubbard-W-terms” might very well be important in strongly-correlated narrow-band materials.

Thus we assume that the charge dynamics is still governed by the Harris-Lange model. In particular, we assume that the chargeons do not scatter from the spinons. For the spin
dynamics we choose the (dimerized, anisotropic) Heisenberg Hamiltonian with an anisotropy into the $z$-direction

$$\hat{H}_{\text{Heis}} = \sum_l \left( 1 + (-1)^l \delta_S \right) \left[ J_\perp (\hat{S}_l^x \hat{S}_{l+1}^x + \hat{S}_l^y \hat{S}_{l+1}^y) + J_z \hat{S}_l^z \hat{S}_{l+1}^z - \frac{1}{4} \right]$$

(5)

where $\hat{S}_l$ is the vector operator for the spin at site $l$, $\hat{S}_l^+ = \hat{S}_l^x + i \hat{S}_l^y = \hat{c}_{l,\uparrow}^\dagger \hat{c}_{l,\downarrow}$, $\hat{S}_l^- = \hat{S}_l^x - i \hat{S}_l^y = \hat{c}_{l,\downarrow}^\dagger \hat{c}_{l,\uparrow}$, and $\hat{S}_l^z = (\hat{n}_{l,\uparrow} - \hat{n}_{l,\downarrow})/2$. $J_\perp > 0$ and $J_z > 0$ are two generally different antiferromagnetic coupling constants, and $0 \leq \delta_S \leq 1$ determines the degree of spin dimerization. It is well-known that a one-dimensional spin-system that is coupled to the lattice degrees of freedom shows the spin-Peierls effect, $\delta_S > 0$.

If the itinerant exchange was responsible for the antiferromagnetic coupling we would have $J \equiv J_\perp = J_z = (1 + \delta^2)2t^2/U$ and $\delta_S = 2\delta/(1 + \delta^2)$. In general, however, we do not assume a simple relation between $J_\perp$, $J_z$ and $t$, or $\delta_S$ and $\delta$. We state, however, that $J_\perp$, $J_z$ are supposed to be small energy scales compared to the Coulomb energies $U$, $V$. Its influence on the exact optical excitation energies will thus be neglected in this work. This implies, for example, that we cannot distinguish between singlet and triplet excitons.

III. OPTICAL ABSORPTION AND EFFECTIVE CURRENT OPERATOR

A. Optical conductivity and optical absorption

The dielectric function $\bar{\epsilon}(\omega)$ and the coefficient for the linear optical absorption $\bar{\alpha}(\omega)$ are given by (Haug and Koch 1990)

$$\bar{\epsilon}(\omega) = 1 + \frac{4\pi i \sigma(\omega)}{\omega}$$

(6a)

$$\bar{\alpha}(\omega) = \frac{4\pi \text{Re}\{\sigma(\omega)\}}{n_b c}$$

(6b)

where $\text{Re}\{\ldots\}$ denotes the real part and $n_b$ is the background refractive index. It is supposed to be frequency independent near a resonance. Hence, the real part of the optical conductivity directly gives the absorption spectrum of the system.
The standard result (Maldague 1977); (Mahan 1990) for the real part of the optical conductivity in terms of the current-current correlation function $\chi(\omega)$ is

$$\text{Re}\{\sigma(\omega)\} = \frac{\text{Im}\{\chi(\omega)\}}{\omega}$$

(7)

$$\chi(\omega) = \frac{N_\perp}{La} i \int_0^\infty dt e^{i\omega t} \langle [\hat{j}(t), \hat{j}]_\perp \rangle$$

(8)

where $N_\perp$ is the number of chains per unit area perpendicular to the chain direction.

The current-current correlation function can be spectrally decomposed in terms of exact eigenstates of the system as

$$\chi(\omega) = \frac{N_\perp}{La} \sum_n |\langle 0 | \hat{j} | n \rangle|^2 \left[ \frac{1}{\omega + (E_n - E_0) + i\gamma} - \frac{1}{\omega - (E_n - E_0) + i\gamma} \right].$$

(9)

Here, $|0\rangle$ is the exact ground state (energy $E_0$), $|n\rangle$ are exact excited states (energy $E_n$), and $|\langle 0 | \hat{j} | n \rangle|^2$ are the oscillator strengths for optical transitions between them. Although $\gamma = 0^+$ is infinitesimal we may introduce $\gamma > 0$ as a phenomenological broadening of the resonances at $\omega = \pm(E_n - E_0)$. The spectral decomposition of the real part of the optical conductivity reads

$$\text{Re}\{\sigma(\omega)\} = \frac{N_\perp \pi}{La \omega} \sum_n |\langle 0 | \hat{j} | n \rangle|^2 \left[ \delta(\omega - (E_n - E_0)) - \delta(\omega + (E_n - E_0)) \right]$$

(10)

which is positive for all $\omega$.

In the following we will always plot the dimensionless reduced optical conductivity

$$\sigma_{\text{red}}(\omega > 0) = \frac{\omega \text{Re}\{\sigma(\omega > 0)\}}{N_\perp ae^2 W}.$$  

(11)

Furthermore we replace the energy conservation $\delta(x)$ by the smeared function

$$\bar{\delta}(x) = \frac{\gamma}{\pi(x^2 + \gamma^2)}$$

(12)

to include effects of phonons and experimental resolution.

B. Effective current operator

In (Gebhard et al. II 1996) we showed that the current operator for an excitation from the filled lower Hubbard band to the empty upper Hubbard band can be written as
\[ \hat{j}_{\text{inter},+}^H = -(\text{itea}) \sum_{l,\sigma} \left( 1 + (-1)^l \delta \right) \left( 1 + (-1)^l \eta \right) \left[ \hat{n}_{l+1,-\sigma} \hat{c}_{l+1,\sigma}^+ (1 - \hat{n}_{l,-\sigma}) - \hat{n}_{l,-\sigma} \hat{c}_{l,\sigma}^+ \hat{c}_{l+1,\sigma} (1 - \hat{n}_{l+1,-\sigma}) \right] \]  

where \( \eta = -|R_{l+1} - R_l|/a < 0 \) is the relative change of lattice distances due to the Peierls distortion (Gebhard et al. I 1996). \( \hat{j}_{\text{inter},+}^H \) creates a neighboring pair of a double occupancy and a hole.

As a central result of (Gebhard et al. II 1996) we found the band structure representation of the current operator as

\[ \hat{j}_{\text{inter},+}^{\text{band}} = \sum_{|k|,|q| \leq \pi/a} i\text{eae}(k) \hat{n}_{k+q/2}^+ \hat{\hat{I}}_{k-q/2} \hat{x}_q. \]  

for the translational invariant case, and

\[ \hat{j}_{\text{inter},+}^{\text{band, dim}} = \sum_{|q|,|k| \leq \pi/(2a)} \left\{ i\text{eae}(k) \left[ \hat{n}_{k+q/2}^+ \hat{\hat{I}}_{k-q/2} - \hat{n}_{k+q/2}^+ \hat{\hat{I}}_{k-q/2+\pi/a} + \hat{n}_{k+q/2}^+ \hat{\hat{I}}_{k-q/2+\pi/a} - \hat{n}_{k+q/2}^+ \hat{\hat{I}}_{k-q/2} \right] \hat{x}_q + ea \Delta(k) \left[ \hat{n}_{k+q/2}^+ \hat{\hat{I}}_{k-q/2} - \hat{n}_{k+q/2}^+ \hat{\hat{I}}_{k-q/2+\pi/a} \right] \hat{x}_{q+\pi/a} \right\} \]  

for a Peierls-distorted lattice. The diagonalized form of the interband current operator is given in (Gebhard et al. II 1996).

Since the excitation energy only depends on the charge configuration the complicated spin problem could be hidden in the operators \( \hat{x}_q \) which are defined only in terms of their product,

\[ \hat{x}_q^+ (\delta, \eta) \hat{x}_q^-(\delta, \eta) = \sum_{S_1' \cdots S_{L-2}'} \frac{1}{L^2} \sum_{l,r} \exp(iq_l q_r) (1 + \eta \delta + (-1)^l (\delta + \eta))(1 + \eta \delta + (-1)^r (\delta + \eta)) \langle 0| S_1' \cdots S_{L-2}', (\uparrow_{l+1}, \downarrow_{l+1} - \uparrow_l \downarrow_{l+1}) , S_l' , \ldots S_{L-2} ' \rangle \]  

\[ \langle S_{L-2}' , \ldots S_r' , (\downarrow_{r+1} \uparrow_r - \uparrow_{r+1} \downarrow_r) , S_{r-1}' , \ldots S_1' | 0 \rangle. \]  

In practice, \( q' = q \) or \( q' = q + \pi/a \). In (Gebhard et al. II 1996) we studied the “hot-spin” case where all \( 2^L \) spin configurations were equally possible ground states. Now we will set up the “no-recoil” approximation for the case of the (unique) ground state of the Heisenberg model, equation (15).
IV. NO-RECOIL APPROXIMATION

A. Basic ideas

Although the eigenenergies and even the eigenstates of the Hubbard model at strong coupling nicely display charge-spin separation (Ogata and Shiba 1990); (Parola and Sorella 1990) the two dynamical degrees of freedom are coupled again by the current operator that creates an optical excitation. The oscillator strength for such an excitation, however, factorizes into a charge and a spin part. For the charge part our band structure picture applies. The spin part is determined by a ground state correlation function for nearest-neighbor singlets at all lattice distances, see below. Hence, the calculation of the optical absorption spectrum remains a difficult many-particle problem even for a formally charge-spin separated system.

If we want to make further progress we have to make assumptions on the behavior of the spin system. In (Gebhard et al. II 1996) we treated the case of a degenerate spin background (“hot-spin case”) where all spin configurations were equivalent. Now we are interested in the more realistic case where the optical excitation starts from the unique ground state of the Heisenberg model, equation (5).

Our band structure interpretation suggests an analogy between our strongly correlated Mott-Hubbard system and the situation in an intrinsic semiconductor with a direct gap. The Hubbard bands for the charges correspond to conduction and valence bands, while the spin degrees of freedom play the role of phonons in a semiconductor or metal. In both cases the energy scales are well separated, the spinon (phonon) energy being much smaller than the chargeon (electron) energy. The spin spectrum is gaped in the presence of dimerization ($\delta S \neq 0$) such that the spin excitations correspond to optical rather than acoustical phonons in that case. This appealing analogy is also the basic idea behind the tJ-model approach to high-Tc superconductivity (Zhang and Rice 1988); (Dagotto 1994).

In electron-phonon systems one may often ignore phonon emission or absorption during
an excitation at low temperatures. This is an approximation even at zero temperatures since phonons can always be emitted even if phonon absorption is impossible. Effectively there is a finite probability (“Debye-Waller factor”) that there is no momentum transferred to the phonons during an excitation by photons. This constitutes the “no-recoil approximation”. It has many applications in solid state science (e.g., X-ray scattering, Mößbauer-effect), and it is also very successfully applied for the explanation of optical spectra of semiconductors (Haug and Koch 1990). Thus we are confident that a (modified) “no-recoil approximation” will also be valid for the strongly correlated Hubbard model where chargeons and spinons are energetically well separated.

**B. Formulation of the approximation**

To formulate the approximation for our case we will scrutinize equation (15) for the spin sector. Recall that the charge sector has already been mapped onto a standard band structure picture. We apply

\[ |S_1', \ldots S_L', (\uparrow_1, \downarrow_{l+1} - \downarrow_l, \uparrow_{l+1}), S_1', \ldots S_L' - 1 \rangle = \hat{T}^{(l-2)}_S \hat{T}^{-(l-2)}_{S'} |S_1'(\uparrow_2, \downarrow_3 - \downarrow_2, \uparrow_3), S_2', \ldots S_L' - 2 \rangle \]

for both expectation values in equation (15) where \( \hat{T}_S \) shifts all spins by one site and \( \hat{T}_{S'} \) shifts all spins but those at sites 2 and 3.

Furthermore, we use the identity

\[ 2 \sum \left( \frac{1}{4} - S_2 \hat{S}_3 \right) |S_2, S_3 \rangle \langle S_3, S_2 | \left( \frac{1}{4} - \hat{S}_2 \hat{S}_3 \right) = |(\uparrow_2, \downarrow_3 - \uparrow_3, \downarrow_2) \rangle \langle (\downarrow_3, \uparrow_2 - \downarrow_2, \uparrow_3) |. \]  

(17)

Now that we have eliminated the restrictions on the intermediate spin sum we can carry it out and are left with a complicated ground state expectation value,

\[ \hat{x}_q^+ \hat{x}_{q'} = 2 \langle 0 | \hat{Z}_{2,3}^+(q) \left( \frac{1}{4} - \hat{S}_2 \hat{S}_3 \right) \hat{Z}_{2,3}^-(q') |0 \rangle \]

(18a)

\[ \hat{Z}_{2,3}(q) = \frac{1}{L} \sum_i e^{-iqla} \hat{T}^{(l-2)}_S \hat{T}^{-(l-2)}_{S'} (1 + (-1)^i \delta) (1 + (-1)^i \eta) . \]

(18b)
Thus far the expressions are exact. Note that \( \hat{x}_q^+ \hat{x}_{q'} \) is almost—but not quite—the double Fourier transform of the correlation function of nearest-neighbor singlets at site \( l \) and \( r \).

To obtain further insight into the problem we first analyze two special cases which can exactly be solved before we formulate the general approximation.

1. Néel state

The Néel state is the ground state of the anisotropic Heisenberg model, equation (5), for \( J_\perp = 0 \) (Ising model),

\[
|\text{AF}\rangle = \prod_{l=1}^{L/2} \hat{c}_{2l,1}^+ \hat{c}_{2l+1,1}^+ |\text{vacuum}\rangle.
\]  

(19)

It displays a perfect double-periodic structure, i. e., if we shift all spins by two lattice sites we recover \( |\text{AF}\rangle \). Hence,

\[
\hat{T}_\perp^{(l-2)} \hat{T}_\perp^{(l-2)} |\text{AF}\rangle = \frac{1}{2} \left( (1 + (1-1)^l) + (1 - (1-1)^l) \hat{T}_\perp \hat{T}_\perp^{-1} \right) |\text{AF}\rangle.
\]  

(20)

The wave function renormalization factors become

\[
\hat{Z}_{2,3}(q)|\text{AF}\rangle = \frac{\delta_{q,0} + \delta_{q,\pi/a}}{2} |\text{AF}(q)\rangle
\]  

(21a)

\[
|\text{AF}(q)\rangle = \left( (1 + \delta)(1 + \eta) + e^{iqa}(1 - \delta)(1 - \eta) \hat{T}_\perp \hat{T}_\perp^{-1} \right) |\text{AF}\rangle.
\]  

(21b)

Thus we may write

\[
\hat{x}_q^+ \hat{x}_{q'} |\text{AF}\rangle = \frac{1}{2} \left( \delta_{q,0} + \delta_{q,\pi/a} \right) \left( \delta_{q',0} + \delta_{q',\pi/a} \right) \langle \text{AF}(q)| \hat{S}_2 \hat{S}_3 |\text{AF}(q')\rangle.
\]  

(22)

For later use we define

\[
Z_0^{\text{AF}} = \langle \text{AF}(0)| \hat{S}_2 \hat{S}_3 |\text{AF}(0)\rangle
\]  

(23a)

\[
Z_\pi^{\text{AF}} = \langle \text{AF}(\pi/a)| \hat{S}_2 \hat{S}_3 |\text{AF}(\pi/a)\rangle
\]  

(23b)

\[
Z_M^{\text{AF}} = \left[ \langle \text{AF}(0)| \hat{S}_2 \hat{S}_3 |\text{AF}(\pi/a)\rangle + \text{h.c.} \right]
\]  

(23c)
such that $\hat{x}_0^+\hat{x}_0 \rvert_{AF} = Z_0^\text{AF}/2$, $\hat{x}_\pi^+/\hat{x}_\pi/\alpha \rvert_{AF} = Z_\pi^\text{AF}/2$, and $(\hat{x}_\pi^+/\hat{x}_\pi/\alpha \hat{x}_0^+\hat{x}_\pi/\alpha) \rvert_{AF} = Z_\pi^\text{AF}/2$. Explicitly,

$$Z_0^\text{AF} = 2(\delta + \eta)^2$$
$$Z_\pi^\text{AF} = 2(1 + \delta \eta)^2$$
$$Z_M^\text{AF} = 4(\delta + \eta)(1 + \delta \eta)$$

The Néel state dominantly provides the momentum $\Delta q_S = \pi/\alpha$ to the charge system during an optical excitation since $Z_\pi^\text{AF} > Z_M^\text{AF} > Z_0^\text{AF}$ for generic values for $\delta, \eta$.

We note that for $\delta = \eta = 0$ only $Z_\pi^\text{AF}$ contributes. This implies that for a Néel state one of the two Hubbard bands can be thought as being shifted by $\pi/\alpha$ which results in vertical transitions between antiparallel bands as for non-interacting electrons in a Peierls-distorted lattice (Gebhard et al. 1996).

2. Dimer state

The ground state of the fully dimerized Heisenberg model, equation (5) for $J_\perp = J_z \equiv J$ and $\delta_S = 1$, is the dimer state

$$|\text{DIM}⟩ = \prod_{l=1}^{\lfloor L/2 \rfloor} \sqrt{\frac{1}{2}} \left( \hat{c}_{2l,\uparrow}^+ \hat{c}_{2l+1,\downarrow} - \hat{c}_{2l,\downarrow}^+ \hat{c}_{2l+1,\uparrow} \right) |\text{vacuum}⟩.$$  

(25)

Again, the dimer state is invariant under a shift of all spins by two lattice sites. Hence, equations (20), (21), (22), and (23) analogously hold for the dimer state. In particular,

$$\left( \frac{1}{4} - \hat{S}_2 \hat{S}_3 \right) |\text{DIM}⟩ = |\text{DIM}⟩$$

(26a)

$$\left( \frac{1}{4} - \hat{S}_2 \hat{S}_3 \right) \hat{T}_S \hat{T}_S^{-1} |\text{DIM}⟩ = -\frac{1}{2} |\text{DIM}⟩$$

(26b)

holds such that the $Z$-factors for the dimer state become

$$Z_0^\text{DIM} = [1 + \delta \eta + 3(\delta + \eta)]^2 / 4$$
$$Z_\pi^\text{DIM} = [3(1 + \delta \eta) + \delta + \eta]^2 / 4$$
$$Z_M^\text{DIM} = [1 + \delta \eta + 3(\delta + \eta)] [3(1 + \delta \eta) + \delta + \eta] / 2$$

(27)

We see again that momentum transfer by $\Delta q_S = \pi/\alpha$ dominates in the dimer state but that there is zero momentum transfer even for $\delta = \eta = 0$, in contrast to the Néel state.
3. General case

The above examples are limiting cases for the ground state of the general Heisenberg model in equation (3). The correlation function for nearest-neighbor spin singlets is long-ranged ordered such that the spin system only allows for momentum transfers $\Delta q_S = 0, \pi/a$. It has recently been shown by (Talstra, Strong, and Anderson 1995) that this order persists in the standard Heisenberg model ($J_\perp = J_z = J, \delta_S = 0$) as “hidden” long-range order although there is neither dimer nor antiferromagnetic order in the ground state.

These observations make us confident that the following “no-recoil” approximation will give the coherent features of the optical absorption spectrum for all values of $J_\perp/J_z > 0$ and $\delta_S$ in the dimerized Heisenberg model.

$$\hat{T}_S\hat{T}_{S'}^{\dagger}|0\rangle = \left[w_{DW} \left(1 + \frac{(-1)^l}{2}\right) + \overline{w}_{DW} \left(1 - \frac{(-1)^l}{2}\right)\right] |0\rangle + \text{rest}. \quad (28)$$

Due to the “hidden” long-range order of spin singlets in the ground state we expect that we need two different “Debye-Waller factors” $w_{DW}, \overline{w}_{DW}$ as in the cases of the Néel and dimer state where they both were unity. In general, the square of their absolute value is smaller than one as can be checked from sum rules. It is clear that more elegant approximations than this can be designed, e.g., an $l$-dependent factor $w_{DW}(l)$ could be introduced to mimic a finite correlation length for finite temperatures as in (Gebhard et al. II 1996). We will not follow the latter ideas here.

The wave function renormalization factors become

$$\hat{Z}_{2,3}(q)|0\rangle = \frac{\delta_{q,0} + \delta_{q,\pi/a}}{2} |\Psi(q)\rangle + \text{rest} \quad (29a)$$

$$|\Psi(q)\rangle = \left[w_{DW}(1 + \delta)(1 + \eta) + e^{iqa}\overline{w}_{DW}(1 - \delta)(1 - \eta)\hat{T}_S\hat{T}_{S'}^{\dagger}\right]|0\rangle. \quad (29b)$$

Thus we may write

$$\hat{x}_q^{\dagger}\hat{x}_{q'}|_{\text{coh}} = \frac{1}{2} \left(\delta_{q,0} + \delta_{q,\pi/a}\right) \left(\delta_{q',0} + \delta_{q',\pi/a}\right) \langle\Psi(q)|\hat{1}/4 - \hat{S}_2\hat{S}_3|\Psi(q')\rangle. \quad (30)$$

In the presence of a Peierls distortion there is no recoil from the spin system since the reciprocal lattice vector is given by $Q = \pi/a$. Even if the lattice distortion was absent, however, the
translational symmetry is broken by the “hidden” long-range order in the nearest-neighbor singlet correlation function. The coherent part of the spin contribution can then be expressed with the help of \( \hat{x}_0^+ \hat{x}_0 \) \( |_{\text{coh}} = Z_0/2 \), \( \hat{x}_{\pi/a}^+ \hat{x}_0^+ \) \( |_{\text{coh}} = Z_{\pi}/2 \), and \( \left( \hat{x}_{\pi/a}^+ \hat{x}_0 + \hat{x}_0^+ \hat{x}_{\pi/a} \right) \) \( |_{\text{coh}} = Z_{M}/2 \).

V. OPTICAL CONDUCTIVITY IN THE HUBBARD MODEL AT STRONG COUPLING

In (Gebhard et al. II 1996) we calculated the optical absorption for the case of an incoherent spin background. In the present case we only have to replace the expressions for the operator products \( \hat{x}_0^+ \hat{x}_0^+ \) by those for the “no-recoil approximation”, equations (28) and (30).

A. Band case: \( V = 0 \)

Since the ground state implicitly breaks the translational symmetry of our system even for \( \delta = \eta = 0 \) we directly approach the case of non-vanishing \( \delta, \eta \).

For the dimerized case we have to diagonalize the current operator, equation (14b), in terms of the quasi-particle operators for the respective Peierls bands. The result for the optical conductivity can be written as (Gebhard et al. II 1996)

\[
\text{Re}\{\sigma(\omega > 0, \delta, \eta)\} = \frac{\pi N_\perp}{La \omega} \sum_{\tau,\tau'=\pm 1} \sum_{|q|,|k| \leq \pi/(2a)} |\lambda_{\tau,\tau'}(k, q)|^2 \delta(\omega - E_{\tau,\tau'}(k, q))
\]

with the absorption energies between the respective Peierls subbands

\[
E_{\tau,\tau'}(k, q) = U + \tau' \delta E(k + q/2) - \tau \delta E(k - q/2)
\]

The transition matrix elements are given by \( \lambda_{+,+}(k, q) = -\lambda_{-,-}(k, q) \), \( \lambda_{-,+}(k, q) = \lambda_{+,+}(k, q) \), and

\[
\lambda_{+,+}(k, q) = i e a \left[ \epsilon(k)(\alpha_+ \alpha_-^* - \beta_+ \beta_-^*) \hat{x}_q^+ + \frac{\Delta(k)}{\delta}(\alpha_+ \beta_-^* + \beta_+ \alpha_-^*) \hat{x}_{q+\pi/a}^+ \right]
\]

\[
\lambda_{+,+}(k, q) = i e a \left[ -\epsilon(k)(\alpha_+ \beta_-^* + \beta_+ \alpha_-^*) \hat{x}_q^+ + \frac{\Delta(k)}{\delta}(\alpha_+ \alpha_-^* - \beta_+ \beta_-^*) \hat{x}_{q+\pi/a}^+ \right].
\]
Here we used the short-hand notation $\alpha_{\pm} = \alpha_{k\pm q/2}$ etc. for the mixing amplitudes in the standard Bogoliubov transformation (Gebhard et al. I and II 1996).

For the coherent part of the optical absorption we only need their values at $q = 0$. In this case, $\alpha_{k}^{2} - \beta_{k}^{2} = -\epsilon(k)/E(k)$ and $2\alpha_{k}\beta_{k} = -\Delta(k)/E(k)$. We thus find $|\lambda_{\pm}(k, q)|^{2}|_{\text{coh}} = \delta_{q, 0}|\lambda_{\pm}(k)|^{2}$ with

$$\begin{align*}
|\lambda_{+,+}(k)|^{2} &= \frac{(ea)^{2}}{2} \left[ Z_{0} \frac{\epsilon(k)^{4}}{E(k)^{2}} + \delta^{2} Z_{\pi} \frac{\Delta(k)^{4}}{\delta^{4} E(k)^{2}} + \delta Z_{M} \left( \frac{\epsilon(k)\Delta(k)}{\delta E(k)} \right)^{2} \right], \\
|\lambda_{+,-}(k)|^{2} &= \frac{(ea)^{2}}{2} \left( \frac{\epsilon(k)\Delta(k)}{\delta E(k)} \right)^{2} \left[ \delta^{2} Z_{0} + Z_{\pi} - \delta Z_{M} \right].
\end{align*}$$

Each of the two quantities above can be expressed in terms of an expectation value of the projection operator $(1/4 - \hat{S}_{2}\hat{S}_{3})$. Hence we can be sure that the result is positive or zero.

For vanishing nearest-neighbor interaction we may directly use equation (31) to arrive at

$$\text{Re}\{\sigma_{\text{coh}}(\omega > 0, \delta, \eta)\} = \frac{\pi N_{1}}{a\omega} \left[ \frac{2}{L} \sum_{|k| \leq \pi/(2a)} \delta(\omega - U) |\lambda_{+,+}(k)|^{2} \right. \left. + \frac{1}{L} \sum_{|k| \leq \pi/(2a)} |\lambda_{+,-}(k)|^{2} \left( \delta(\omega - U - 2E(k)) + \delta(\omega - U + 2E(k)) \right) \right].$$

For later use we define the abbreviations

$$\begin{align*}
H_{1} &= \frac{16}{W^{2}L} \sum_{|k| \leq \pi/(2a)} \frac{\epsilon(k)^{4}}{E(k)^{2}} = 1 - \delta^{2} H_{3}, \\
H_{2} &= \frac{16}{W^{2}L} \sum_{|k| \leq \pi/(2a)} \frac{\Delta(k)^{4}}{\delta^{4} E(k)^{2}} = \frac{1}{\delta^{2}} (1 - H_{3}), \\
H_{3} &= \frac{16}{W^{2}L} \sum_{|k| \leq \pi/(2a)} \left( \frac{\epsilon(k)\Delta(k)}{\delta E(k)} \right)^{2} = \left( \frac{1}{1 + |\delta|} \right)^{2}, \\
X(\omega) &= -\frac{1}{\pi} \frac{16}{W^{2}L} \sum_{|k| \leq \pi/(2a)} \left( \frac{\epsilon(k)\Delta(k)}{\delta E(k)} \right)^{2} \frac{\omega - U}{(\omega - U)^{2} - 4E(k)^{2}}.
\end{align*}$$

For vanishing nearest-neighbor interaction we only need the imaginary part of the function $X(\omega)$ which can easily be calculated. The real part of the coherent optical conductivity finally becomes
\[
\text{Re}\{\sigma_{\text{coh}}(\omega > 0, \delta, \eta)\} = \frac{\pi N \pi}{10848 \omega} \left[ (Z_0 H_1 + \delta^2 Z_\pi H_2 + \delta Z_M H_3) \delta(\omega - U) \right] \\
+ \frac{2(\delta^2 Z_0 + Z_\pi - \delta Z_M) \sqrt{[(\omega - U)^2 - (W \delta)^2] [W^2 - (\omega - U)^2]}}{\pi W^2 (1 - \delta^2)^2 |\omega - U|} 
\]

in the regions \(\omega = U\) and \(W \delta \leq |\omega - U| \leq W\). Thus the generic coherent features are (i) a \(\delta\)-peak at \(\omega = U\) which results from vertical transitions between the parallel Hubbard bands \((\Delta q_C = \Delta q_S = 0)\), and (ii) a band-to-band transition feature for \(W \delta \leq |\omega - U| \leq W\) which results from vertical transitions between two antiparallel bands \((\Delta q_C = \Delta q_S = \pi/a)\).

To check the sum rule we integrate over the reduced coherent optical conductivity and obtain

\[
\int_0^\infty \frac{d\omega}{W} \sigma_{\text{red,coh}}(\omega, \delta, \eta) = \frac{\pi}{16} (Z_0 + Z_\pi) \\
= \frac{\pi}{4} \left[ |w_{\text{DW}}|^2 (1 + \delta)^2 (1 + \eta)^2 C_{S}^{\text{even}} + |w_{\text{DW}}|^2 (1 - \delta)^2 (1 - \eta)^2 C_{S}^{\text{odd}} \right] 
\]

where \(C_{S}^{\text{even,odd}}\) is nearest-neighbor spin-correlation function on even/odd sites, see equation (A3) of the appendix. The comparison with the sum rule, equation (A5), shows that \(|w_{\text{DW}}|^2 \leq 1\) and \(|w_{\text{DW}}|^2 \leq 1\) have to hold which indeed allows to interpret them as Debye-Waller factors.

For \(\delta = 1, \eta = 0\) (\(L/2\) independent two-site systems) we have \(Z_0 = Z_\pi = Z_M/2 = 8|w_{\text{DW}}|^2 C_{S}^{\text{even}}\). The contribution of the band vanishes again, and we obtain a single peak at \(\omega = U\) with the coherent fraction \(|w_{\text{DW}}|^2 = 1\) of the total oscillator strength.

The case of a vanishing Peierls distortion \((\delta = \eta = 0)\) is particularly interesting. Recall that in the case of an incoherent spin background ("hot-spin" case) (Gebhard et al. II) we observed a logarithmic divergence at \(\omega = U\). Now we find from equation (36) that

\[
\text{Re}\{\sigma_{\text{red,coh}}(\omega > 0)\} = \frac{1}{16} \left[ W \pi Z_0 \delta(\omega - U) + 2Z_\pi \sqrt{1 - \left(\frac{\omega - U}{W}\right)^2} \right] 
\]

in which the proper \(Z\)-values for \(\delta = \eta = 0\) have to be inserted. In general, the absorption consists of a \(\delta\)-peak at \(\omega = U\) and a semielliptic contribution which come from vertical transitions between the parallel \((\Delta q_C = \Delta q_S = 0)\) and antiparallel \((\Delta q_S = \Delta q_C = \pi/a)\) bands.
For the Néel state $Z^\text{AF}_0(\delta = \eta = 0) = 0$, and $Z^\text{AF}_\pi(\delta = \eta = 0) = 2$, and the result of (Lyo and Galinar 1977); (Lyo 1978) is recovered. It is only the Néel state that suppresses the physics of the parallel Hubbard bands for $\delta = \eta = 0$. If we had only considered this state as reference state, the physics of the parallel Hubbard bands would have been missed. The dimer state with $Z^\text{DIM}_0(\delta = \eta = 0) = 1/4$, and $Z^\text{DIM}_\pi(\delta = \eta = 0) = 9/4$ does show the generic features. The exact results for both states are depicted in figure 1.

For nonzero lattice dimerization ($\delta \neq 0, \eta \neq 0$) even the Néel state reproduces the generic situation. The coherent peak at $\omega = U$ is not at all a consequence of the lattice distortion but a consequence of the parallel Hubbard bands. This has been overlooked in previous analytical investigations (Lyo and Galinar 1977); (Lyo 1978); (Galinar 1979). Numerical calculations (Campbell, Gammel, and Loh 1989) for $\delta = \eta = 0$ essentially give the results from the Néel ground state since this state dominates for small system sizes. Small traces of a peak at $\omega = U$ may have been washed out by the adopted smoothing procedure. For non-zero lattice distortion, however, the $\delta$-resonance at $\omega = U$ becomes clearly visible even in the numerical simulations.

The exact results for the optical absorption of the Néel and dimer state in the presence of a lattice distortion are shown in figure 2. For realistic states the $Z$-factors will not be too different such that the figure should reproduce the generic situation for the Peierls-distorted Hubbard model at strong correlations and low temperatures. This observation further supports our “no-recoil approximation”. Figure 2 has to be compared to figure 4 of (Gebhard et al. II 1996). It is seen that the linear absorption at the threshold $\omega = U - W$ now shows a square-root behavior. This is also the case in the “hot-spin” case for large enough $\delta$ when the Peierls gap has not been smeared out. It is again seen that the significant features of the absorption spectrum have not changed much when we go from the Harris-Lange model to the strongly correlated Hubbard model.
B. Exciton case: $V \neq 0$

In the presence of a nearest-neighbor interaction we have to solve an integral equation when the lattice is distorted. Note that only the absolute position of the resonances will be determined by the Coulomb parameter $U/t$ while their relative position depends on $V/t$. For illustrative purposes we tune $V/t$ independently of $U/t$ and put aside the question of the stability of the ground state against the formation of a charge density wave.

The calculations are lengthy and will not be redone here, see appendix B of (Gebhard et al. II 1996) for details. The result can be expressed in terms of two operator-valued functions $G_{1,2}(q),$

$$G_1(q) = itea \left[ \hat{x}^+_q F_1(q) - \hat{x}^+_{q+\pi/a} F_3(q) \right]$$

$$G_2(q) = itea \left[ -\hat{x}^+_q F_3(q) + \hat{x}^+_{q+\pi/a} F_2(q) \right],$$

and three $q$-dependent functions $F_{1,2,3}(q)$ as

$$\text{Re}\{\sigma(\omega > 0, V, \delta, \eta)\} = \text{Re}\{\sigma(\omega > 0, \delta, \eta)\} + \frac{2VN_{\perp}}{a\omega}$$

$$\text{Im}\left\{ \sum_{|q| \leq \pi/(2a)} \frac{1}{(1 + VF_1)(1 + VF_2) - (VF_3)^2} \left[ G_1 G_1^+ + G_2 G_2^+ + V \left( G_1 G_1^+ F_2 + G_2 G_2^+ F_1 + (G_1 G_2^+ + G_2 G_1^+) F_3 \right) \right] \right\}.$$ 

In the “no-recoil approximation” we only need the $q = 0$ value of the functions $F_{1,2,3}$. We have

$$F_1(q = 0) = \frac{H_1}{\omega - U} - \pi \delta^2 X(\omega)$$

$$F_2(q = 0) = \frac{\delta^2 H_2}{\omega - U} - \pi X(\omega)$$

$$F_3(q = 0) = -\delta \left[ \frac{H_3}{\omega - U} + \pi X(\omega) \right],$$

see eqs. (35). The definitions (23) allow us to reduce equation (40) to
\[
\text{Re}\{\sigma_{\text{coh}}(\omega > 0, V, \delta, \eta)\} = \text{Re}\{\sigma_{\text{coh}}(\omega > 0, \delta, \eta)\}
\]

\[
+ \frac{VN_{\perp}(W_{ea})^2}{16a\omega} \text{Im} \left\{ \frac{1}{(1 + VF_1)(1 + VF_2) - (VF_3)^2} \right\}
\]

\[
\left[ Z_0(F_1^2 + F_3^2) + Z_\pi(F_2^2 + F_3^2) - Z_M(F_1 + F_2)F_3
\]

\[
+ V (F_1F_2 - F_3^2)(Z_0F_1 + Z_\piF_2 - Z_MF_3) \right\}.
\]

Note that the \textit{positions} of the excitons are determined by the zeros of the denominator which is a function of \(\omega, V,\) and \(\delta\) but is independent of the \(Z\)-factors and \(\eta\). The oscillator strength of an exciton, however, strongly depends on the \(Z\)-factors.

For the translational invariant case, \(\delta = \eta = 0\), equation (42) can be simplified to \((F_3 = 0, F_1 = 1/(\omega - U), F_2 = -\pi X(\omega))\)

\[
\text{Re}\{\sigma(\omega > 0, V)\} = -\frac{(W_{ea})^2N_{\perp}}{16a\omega} \text{Im} \left\{ \frac{Z_0F_1}{1 + VF_1} + \frac{Z_\piF_2}{1 + VF_2} \right\}.
\]

From the first part we see that the peak at \(\omega = U\) for \(V = 0\) becomes a \(q = 0\)-exciton at frequency \(\omega_0 = U - V\). For the Néel state this exciton is absent while it is present in all generic cases, e. g., in the dimer state. The second part describes the formation of the \(q = \pi/a\)-exciton from the semielliptic “band absorption” part for \(V = 0\). The formation of this exciton leads to a redshift in the “band absorption” part as already noticed by (Galinar 1979).

The exact result for \(\delta = \eta = 0\) for the dimer state is displayed in figure 3 for various values of \(V/t\). For \(V > W/2\) the \(q = \pi/a\)-exciton becomes a true bound state at \(\omega_{\pi/a} = U - V - W^2/(4V)\) below the band absorption edge at \(\omega_{\text{edge}} = U - W\). For large enough \(V/t\) the fact that \(Z_\pi > Z_0\) implies that the \(q = \pi/a\)-exciton resonance is stronger than the one from the \(q = 0\)-exciton. Since they are only separated by an energy difference \(\delta\omega = W^2/(4V)\) thermal and disorder broadening could merge the two exciton lines into a single asymmetric absorption line.

The general case of the optical absorption of the dimer state in the presence of a lattice distortion is shown in figure 4. The Néel state results in a very similar curve such that
we are confident that it reproduces the generic features for the coherent absorption in the Hubbard model at strong correlations. At $V = 0$ we had the resonance at $\omega = U$ and a Peierls-split “band absorption” in the range $W\delta \leq |\omega - U| \leq W$. For moderate $V/t$ the two bands evolve into two peaks such that three lines are visible. The peak at energy $\omega_{\pi/a} = U - V - W^2/(4V)$ corresponds to the $q = \pi/a$-exciton while the dominant peak is the $q = 0$-exciton at $\omega_0 = U - V$. The peak near $\omega = U$ corresponds to the anti-bound exciton which quickly looses oscillator strength as $V/t$ increases. For large $V/t$ the $q = 0$- and $q = \pi/a$-excitons dominate the absorption spectrum with a preference of the $q = \pi/a$-exciton since $Z_{\pi} > Z_0$. In addition, a weak structure remains near $\omega = U$ which does not play a role for the optical absorption but may become visible in electroabsorption, i.e., optical absorption in the presence of a static electrical field.

The dimer state underestimates the factors $Z_{\pi}, Z_M$ as compared to $Z_0$. For the Néel state the strength of the $q = 0$-exciton is much weaker for realistic values for the lattice distortion. The result for the Néel state is shown in figure 5. It is now seen that the $q = \pi/a$-exciton strongly dominates the $q = 0$-exciton. This is to be expected since the components which imply a momentum transfer $q = 0$ to the spin system $(Z_0, Z_M)$ are much weaker now. Even a large value of $\delta$ does not drastically change this situation. It should be clear that the strengths of all exciton peaks above the exciton at $\omega_{\pi/a} = U - V - W^2/(4V)$ are a direct measure for the dimerization degree both of the charge ($\delta, \eta \neq 0$) and the spin ($\delta_S \neq 0$) system.

VI. SUMMARY AND CONCLUSIONS

In this paper we have further studied the optical absorption for strongly-correlated electrons in half-filled Peierls-distorted chains. In (Gebhard et al. II 1996) we exactly solved the case of the Harris-Lange model with its incoherent spin background ("hot-spin case"). For the more realistic situation of low temperatures, i.e., a unique ground state, exact solutions are impossible for the generic cases. To make further progress we assumed that the spin
dynamics is irrelevant: (i) we neglected corrections of the order $J$ to the optical excitation energies and thus a singlet/triplet-splitting of the excitations, and (ii) we assumed that no spin excitations are created during an optical excitation (“no-recoil approximation”). Then we could analytically investigate the problem even in the presence of a lattice dimerization $\delta$ and a nearest-neighbor interaction $V$ between the electrons.

In the “no-recoil approximation” for the strongly correlated Hubbard model all the features already seen in the “hot-spin case” become more prominent since now only transitions with $\Delta q_S = 0$ and $\Delta q_S = \pi/a$ are allowed. This corresponds to vertical transitions between parallel Hubbard bands ($\Delta q_S = \Delta q_C = 0$) and antiparallel bands ($\Delta q_S = \Delta q_C = \pi/a$). The relative strength of the two processes can be measured by three parameters ($Z_0$, $Z_\pi$, $Z_M$) which depend on both the spin structure of the ground state and the lattice distortion. In the generic case the parallel Hubbard bands result in a single peak at $\omega = U$. The $\Delta q_S = \Delta q_C = \pi/a$ part supplements the optical absorption spectrum by two absorption bands for $W \delta \leq |\omega - U| \leq W$. An additional nearest-neighbor interaction results in a tightly bound exciton at $\omega_0 = U - V$ and another exciton at $\omega_{\pi/a} = U - V - W^2/(4V)$. Simpson’s exciton band has seized to exist since the exciton’s center of mass momentum has to be $\Delta q_C = 0$ or $\Delta q_C = \pi/a$. Again, the excitons draw the oscillator strength from the “band absorption” part. Although the location of the exciton resonances is independent of the spin configuration of the ground state their relative amplitude is a very sensitive measure of the lattice and spin dimerization. We thus come to the important conclusion that the linear optical absorption in strongly correlated electron systems might serve as a subtle probe for the magnetic structure of the ground state. In particular, optical absorption allows to measure the “hidden” long-range order of nearest-neighbor spin singlets (Talstra, Strong, and Anderson 1995).

One might think that it is fairly simple to experimentally distinguish between a Peierls and a Mott-Hubbard insulator on the basis of their optical absorption. At low temperatures we expect a Peierls insulator to show a broad band-to-band transition while a Mott-Hubbard insulator with appreciable values for the Coulomb interaction ($U \gg W$, $V > W/2$) should
display sharp exciton lines in the optical absorption spectrum. Unfortunately, a direct comparison to experiment is difficult for two reasons. First, disorder effects can inhomogeneously broaden single lines. Hence, an experimentally observed band can very well be a sign of disorder rather than an argument against a Mott-Hubbard insulator. Secondly, when a residual electron-electron interaction is included in a Peierls insulator (Abe, Yu, and Su 1992); (Abe, Schreiber, Su, and Yu 1992) one can equally well obtain excitons which draw the oscillator strength form the band transitions. Hence, single exciton lines are not a clear-cut indication against the presence of a Peierls insulator either.

It is clear that one needs to compare model calculations and experimental observations on many more physical quantities than just the linear optical absorption before definite conclusions can be reached.

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APPENDIX: SUM RULE

We briefly account for the sum rule. We have

$$\int_0^\infty d\omega \text{Im} \{\chi(\omega)\} = \pi \frac{N_\bot}{La} \sum_n \left| \langle 0 | j^2 | n \rangle \right|^2 = \pi \frac{N_\bot}{La} \langle 0 | j^2 | 0 \rangle . \quad (A1)$$

It is a standard exercise to show that

$$\langle 0 | j^2 | 0 \rangle = (2\text{tea})^2 \sum_l \left( 1 + (-1)^l \delta \right)^2 \left( 1 + (-1)^l \eta \right)^2 \langle 0 | \left( \frac{1}{4} - \hat{S}_l \hat{S}_{l+1} \right) | 0 \rangle . \quad (A2)$$

We define the positive quantities

$$C_{S\text{even,odd}} = \frac{1}{L} \sum_l \frac{1 \pm (-1)^l}{2} \langle 0 | \left( \frac{1}{4} - \hat{S}_l \hat{S}_{l+1} \right) | 0 \rangle \quad (A3)$$

such that

$$C_S = C_{S\text{odd}} + C_{S\text{odd}}$$

is the value of the nearest-neighbor spin-spin correlation function. Then we may write

$$\int_0^\infty d\omega \text{Im} \{\chi(\omega)\} = \pi N_\bot a (2\text{tea})^2 \left[ (1 + \delta)^2 (1 + \eta)^2 C_{S\text{even}} + (1 - \delta)^2 (1 - \eta)^2 C_{S\text{odd}} \right] . \quad (A4)$$

The area under the curves for $\sigma_{\text{red}}(\omega)$, equation (11), is thus given by

$$\int_0^\infty \frac{d\omega}{W} \sigma_{\text{red}}(\omega) = \frac{\pi}{4} \left[ (1 + \delta)^2 (1 + \eta)^2 C_{S\text{even}} + (1 - \delta)^2 (1 - \eta)^2 C_{S\text{odd}} \right] . \quad (A5)$$
REFERENCES

• S. Abe, M. Schreiber, W. P. Su, and J. Yu, Phys. Rev. B 45, 9432 (1992).

• S. Abe, J. Yu, and W. P. Su, Phys. Rev. B 45, 8264 (1992).

• L. Alcácer, in: Organic Conductors, ed. by J.-P. Farges, (Marcel Dekker, New York (1994)).

• D. Baeriswyl, P. Horsch, and K. Maki, Phys. Rev. Lett. 60 (C), 70 (1988).

• D. Baeriswyl, D. K. Campbell, and S. Mazumdar, in: Conjugated Conducting Polymers, ed. by H. Kiess, (Springer Series in Solid State Sciences 102, Springer, Berlin (1992)).

• A. Brau and J.-P. Farges, in: Organic Conductors, ed. by J.-P. Farges, (Marcel Dekker, New York (1994)).

• D. K. Campbell, J. T. Gammel, and E. Y. Loh, Phys. Rev. B 38, 12043 (1988).

• D. K. Campbell, J. T. Gammel, and E. Y. Loh, Int. J. Mod. Phys. B 3, 2131 (1989).

• D. K. Campbell, J. T. Gammel, and E. Y. Loh, in: Interacting Electrons in Reduced Dimensions, ed. by D. Baeriswyl and D. K. Campbell, (NATO ASI Series B 213, Plenum Press, New York (1989)), p. 171.

• D. K. Campbell, J. T. Gammel, and E. Y. Loh, Phys. Rev. B 42, 475 (1990).

• E. Dagotto, Rev. Mod. Phys. 66, 763 (1994).

• P. G. J. van Dongen, Phys. Rev. B 49, 7904 (1994).

• P. G. J. van Dongen, Phys. Rev. B 50, 14016 (1994).

• F. H. L. Eßler and V. E. Korepin (ed.), Exactly Solvable Models of Strongly Correlated Electrons, (World Scientific, Singapore (1994)).
• J.-P. Farges (ed.), *Organic Conductors*, (Marcel Dekker, New York (1994)).

• J. L. Fave, in: *Electronic Properties of Polymers*, ed. by H. Kuzmany, M. Mehring, and S. Roth, (Springer Series in Solid State Sciences 107, Springer, Berlin (1992)).

• A. Fritsch and L. Ducasse, J. Physique I 1, 855 (1991).

• R. M. Fye, M. J. Martins, D. J. Scalapino, J. Wagner, and W. Hanke, Phys. Rev. B 45, 7311 (1992).

• J.-P. Galinar, J. Phys. C 12, L335 (1979).

• J. T. Gammel and D. K. Campbell, Phys. Rev. Lett. 60 (C), 71 (1988).

• F. Gebhard, K. Bott, M. Scheidler, P. Thomas, and S. W. Koch, next to last article, referred to as I.

• F. Gebhard, K. Bott, M. Scheidler, P. Thomas, and S. W. Koch, last article, referred to as II.

• A. B. Harris and R. V. Lange, Phys. Rev. 157, 295 (1967).

• H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, (World Scientific, Singapore (1990)).

• A. J. Heeger, S. Kivelson, J. R. Schrieffer, and W.-P. Su, Rev. Mod. Phys. 60, 781 (1988).

• J. Hubbard, Proc. R. Soc. London, Ser. A 276, 238 (1963).

• S. Kivelson, W.-P. Su, J. R. Schrieffer, and A. J. Heeger, Phys. Rev. Lett. 58, 1899 (1987).

• S. Kivelson, W.-P. Su, J. R. Schrieffer, and A. J. Heeger, Phys. Rev. Lett. 60 (C), 72 (1988).
• W. Kohn, Phys. Rev. 133, A171 (1964).

• S. K. Lyo and J.-P. Galinar, J. Phys. C 10, 1693 (1977).

• S. K. Lyo, Phys. Rev. B 18, 1854 (1978).

• G. D. Mahan, Many-Particle Physics, (2nd edition, Plenum Press, New York (1990)).

• P. F. Maldague, Phys. Rev. B 16, 2437 (1977).

• S. Mazumdar and S. N. Dixit, Phys. Rev. B 34, 3683 (1986).

• F. Mila, Phys. Rev. B 52, 4788 (1995).

• M. Ogata and H. Shiba, Phys. Rev. B 41, 2326 (1990).

• A. Painelli and A. Girlando, Synth. Met. 27, A15 (1988).

• A. Painelli and A. Girlando, in: Interacting Electrons in Reduced Dimensions, ed. by D. Baeriswyl and D. K. Campbell, (NATO ASI Series B 213, Plenum Press, New York (1989)), p. 165.

• A. Painelli and A. Girlando, Phys. Rev. B 39, 2830 (1989).

• A. Parola and S. Sorella, Phys. Rev. Lett. 60, 1831 (1990).

• L. Salem, Molecular Orbital Theory of Conjugated Systems, (Benjamin, London (1966)).

• M. Schott and M. Nechtschein, in: Organic Conductors, ed. by J.-P. Farges, (Marcel Dekker, New York (1994)).

• B. S. Shastry and B. Sutherland, Phys. Rev. Lett. 65, 243 (1990).

• W. T. Simpson, J. Am. Chem. Soc. 73, 5363 (1951).

• W. T. Simpson, J. Am. Chem. Soc. 77, 6164 (1955).
• C. A. Stafford and A. J. Millis, Phys. Rev. B 48, 1409 (1993).

• C. A. Stafford, A. J. Millis, and B. S. Shastry, Phys. Rev. B 43, 13660 (1991).

• J. C. Talstra, S. P. Strong, and P. W. Anderson Phys. Rev. Lett. 74, 5256 (1995).

• C.-Q. Wu, X. Sun, and K. Nasu, Phys. Rev. Lett. 59, 831 (1987).

• F. C. Zhang and T. M. Rice, Phys. Rev. B 37, 3759 (1988).
FIGURES

FIG. 1. Reduced optical conductivity, $\sigma_{\text{red}}(\omega > 0)$, for the dimer and the Néel state for $U = 2W$. A broadening of $\gamma = 0.01W$ has been included.

FIG. 2. Reduced optical conductivity, $\sigma_{\text{red}}(\omega > 0, \delta, \eta)$, for the dimer and the Néel state for $U = 2W$ in the presence of a lattice distortion, $\delta = 0.2$, $\eta = -0.06$. A broadening of $\gamma = 0.01W$ has been included.

FIG. 3. Reduced optical conductivity, $\sigma_{\text{red}}(\omega > 0, V)$, for the dimer state for $U = 2W$ in the presence of a nearest-neighbor interaction, $V = 0, W/2, W$. A broadening of $\gamma = 0.01W$ has been included.

FIG. 4. Reduced optical conductivity, $\sigma_{\text{red}}(\omega > 0, V, \delta, \eta)$, for the dimer state for $U = 2W$ in the presence of a lattice distortion, $\delta = 0.2$, $\eta = -0.06$, and a nearest-neighbor interaction, $V = 0, W/2, W$. A broadening of $\gamma = 0.01W$ has been included.

FIG. 5. Reduced optical conductivity, $\sigma_{\text{red}}(\omega > 0, V, \delta, \eta)$, for the Néel state for $U = 2W$ in the presence of a lattice distortion, $\delta = 0.2$, $\eta = -0.06$, and a nearest-neighbor interaction, $V = 0, W/2, W$. A broadening of $\gamma = 0.01W$ has been included.
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Optical absorption of strongly correlated ...
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