Peierls transition in the strong-coupling Hubbard chain

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It is demonstrated that in the limit of infinitely strong intra-atomic Coulomb repulsion ($U \to \infty$) the Hubbard chain can distort to become a Peierls band insulator with a low-temperature Curie law. The wavelength of this Peierls distortion is different from that commonly obtained for the case $U = 0$.

In a recent publication in this journal, Beni, Holstein, and Pincus[1] have considered the thermodynamic properties of the Hubbard chain in the limit of infinitely strong intra-atomic Coulomb repulsion U. We point out here that their study may be extended to obtain some interesting rigorous results for the effects of a strong intra-atomic Coulomb repulsion on the *Peierls transition*[2-5] in a one-dimensional band conductor. Since our treatment will not include fluctuation effects,[6,7] the present discussion of the distorted state will be restricted to low temperatures ($T \to 0$).

We consider a linear chain of N atoms, not necessarily regularly spaced, described by the Hamiltonian

$$H = -\sum_{\substack{i,j=1 \\ |i-j|=1}}^{N} \sum_{\sigma} t_{ij} c_{i\sigma}^{*} c_{j\sigma} + U \sum_{i=1}^{N} n_{i\uparrow} n_{i\downarrow} - h \sum_{i=1}^{N} (n_{i\uparrow} - n_{i\downarrow}), \tag{1}$$

where $c_{i\sigma}^{*}$, $c_{i\sigma}$, and $n_{i\sigma} = c_{i\sigma}^{*} c_{i\sigma}$ are, respectively, the creation, annihilation, and number operators for electrons with spin σ at site i. The first two terms of H represent a one-dimensional Hubbard Hamiltonian,[8] in which the matrix element t_{ij} (hopping integral) for the transfer of an electron from the ith to the jth atomic site is nonvanishing only for pairs of sites that are nearest neighbors. U denotes the intra-atomic Coulomb repulsion that acts whenever two electrons occupy the same site. The last term of H describes the interaction with a constant external magnetic field of magnitude h/μ_B, where μ_B is the Bohr magneton. Finally, the number of electrons per atom will be denoted by ν ($0 < \nu < 2$).

For the case of a constant nearest-neighbor hopping integral (regularly-spaced static linear chain), the thermodynamic properties of the chain in the limit $U \to \infty$ have been investigated, with different techniques, by Sokoloff,[9] Beni *et al.*,[1] and Klein.[10] They noted that in the limit $U \to \infty$ there is a complete decoupling of the translational and spin degrees of freedom. Specifically, the thermodynamic properties of the chain in the limit $U \to \infty$ are equivalent to those of a composite system consisting of $|1-\nu|N$ noninteracting spinless tight-binding fermions and N (for $\nu < 1$) or $(2-\nu)N$ (for $\nu > 1$) independent spins. Their argument depended on the fact that, in the $U \to \infty$ limit, no reordering of the electronic spins can occur by nearest-neighbor hopping in one dimension. We stress here that it is actually the *restriction to nearest-neighbor hopping* that is crucial, and not the specific form of the nearest-neighbor hopping integral. Consequently, the arguments leading to the decoupling of the translational and spin degrees of freedom are valid for an arbitrary form of hopping integral (linear chain of arbitrarily spaced atoms).

For the partition function corresponding to our Hamiltonian H of Eq. (1), the limit $U \to \infty$ can be treated rigorously.[11] (A counterterm has to be added to H which shifts the lowest eigenvalue of the Coulomb repulsion term in each k-particle subspace F_k to zero.) Let Z_k denote the canonical partition function for $k = \nu N$ electrons. Then

$$Z_k(\beta, h, U) = \mathrm{Tr}_k e^{-\beta P_k H P_k}, \tag{2}$$

where Tr_k denotes the trace in the k-electron subspace F_k, P_k the projector onto F_k, and $\beta = 1/k_B T$. The first exact result[11] can now be expressed as follows:

$$Z_k(\beta, h, \infty) = \mathrm{Tr}_k P_k^{(\min)} e^{-\beta P_k^{(\min)} H_0 P_k^{(\min)}}, \tag{3}$$

where

$$H_0 = -\sum_{\substack{i,j=1 \\ |i-j|=1}}^{N} \sum_{\sigma} t_{ij} c_{i\sigma}^{*} c_{i\sigma} - h \sum_{i=1}^{N} (n_{i\uparrow} - n_{i\downarrow}), \tag{4}$$

and $P_k^{(\min)}$ is the projector onto the subspace of states in F_k with a minimum number of doubly occupied sites. For the calculation of thermodynamic quantities in the limit $U \to \infty$, H may thus be replaced by the projected Hamiltonian $P_k^{(\min)} H_0 P_k^{(\min)}$. Furthermore, it can be shown[11] that this projected Hamiltonian is a unitary equivalent to the Hamiltonian

$$\tilde{H} = P_m \tilde{H}_0 P_m + \tilde{H}_s, \tag{5}$$

with

$$\tilde{H}_0 = -\sum_{\substack{i,j=1 \\ |i-j|=1}}^{N} t_{ij} d_i^* d_j , \tag{6}$$

and

$$\tilde{H}_s = -h \sum_{l=1}^{N_s} \sigma_l^z . \tag{7}$$

$\tilde{H}$ describes the composition of two very simple mutually independent systems. $\tilde{H}_0$ is a tight-binding Hamiltonian for a linear chain of *spinless fermions*, and $\tilde{H}_s$ is the Hamiltonian for a system of $N_s = \nu N$ (for $\nu < 1$) or $N_s = (2-\nu)N$ (for $\nu > 1$) noninteracting spins in a constant magnetic field. d_i^* and d_i denote creation and annihilation operators for a spinless fermion at the site i, and P_m is the projector on the space of states with $m = N\tilde{\nu}$ ($\tilde{\nu} = |1-\nu|$) spinless particles. $\sigma_l^{(z)}$ denotes the usual Pauli matrix.

These rigorous results tell us that in the $U \to \infty$ limit, our Hubbard system may be described (in each k-particle subspace F_k separately) as a composition of a system of spinless fermions with a system of independent spins. The Hamiltonian $\tilde{H}$ of the composite system contains no interaction between the two subsystems, i.e., translational and spin degrees of freedom are completely decoupled.

We note that the independent spins imply a Curie law for the susceptibility and that with spinless fermions a BCS ground state with singlet pairing will, of course, never be possible.

In general, the spinless fermions will be coupled to the linear lattice through the dependence of t_{ij} on the distance between sites i and j. In the limit $N \to \infty$ and at low temperatures, we may then rigorously apply the usual Peierls argument[2,3] to this system of spinless fermions. This tells us that a regularly-spaced chain structure of lattice constant a will become unstable with respect to a periodic distortion of period

$$k_P = \begin{cases} 2\tilde{k}_F & \text{if } \tilde{k}_F < \pi/2a \\ 2(\pi/a - \tilde{k}_F) & \text{if } \tilde{k}_F > \pi/2a , \end{cases} \tag{8}$$

where $\tilde{k}_F = (\pi/a)\tilde{\nu}$ is the Fermi wave vector of the spinless fermions. We thus obtain for the wavelength $\lambda_\infty = 2\pi/k_P$ of the Peierls distortion in the limit of strong intra-atomic Coulomb repulsion ($U \to \infty$)

$$\lambda_\infty = \begin{cases} a/|1-\nu| & \text{if } |1-\nu| \le \frac{1}{2} \\ a/(1-|1-\nu|) & \text{if } |1-\nu| \ge \frac{1}{2} . \end{cases} \tag{9}$$

These results are significantly different from those obtained in the usual $U = 0$ case,[2]

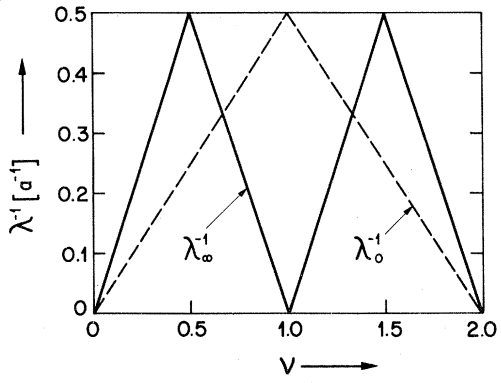

FIG. 1. Inverse wavelengths λ_∞^{-1} and λ_0^{-1} of the Peierls distortion of a Hubbard chain with $U \to \infty$ and $U = 0$, respectively, as functions of the electron density ν [Eqs. (9) and (10)].

$$\lambda_0 = \begin{cases} 2a/\nu & \text{if } 0 < \nu < 1 \\ 2a/(2-\nu) & \text{if } 1 < \nu < 2. \end{cases} \tag{10}$$

A plot of λ_0^{-1} and λ_∞^{-1} as a function of the electron density ν is shown in Fig. 1 and gives a clear illustration of the essential difference between the two limiting cases. For $U \to \infty$, we have to deal with $|1-\nu|$ *spinless* fermions, whereas for $U = 0$ we consider ν fermions with spin.

The results for λ_∞ may be of relevance to actual systems of experimental interest. It is known[12–16] that some quasi-one-dimensional tetracyanoquinodimethane (TCNQ) crystals, e.g., triethylamine $(TCNQ)_2$ and diethyltiacarbocyanine $(TCNQ)_2$, with a quarter-filled band ($\nu = \frac{1}{2}$) consist of distorted chains with wavelength $\lambda = 2a$. If this distortion is interpreted as a Peierls instability, the usual arguments ($U = 0$) would lead to a wrong wavelength of $\lambda_0 = 4a$, whereas our results for strong intra-atomic Coulomb repulsion ($U \to \infty$) give the correct (observed) wavelength of $\lambda_\infty = 2a$. These experimental observations could therefore be regarded as evidence for the existence of strong Coulomb repulsion. We note that this interpretation of the distortion (Peierls instability) would be entirely different from the usual one,[13,16] where an *a priori* dimerized Hubbard chain (alternating values of the hopping integrals) is assumed.

We finally remark that from the present exact analysis of the limit $U \to \infty$ nothing can be concluded concerning the value of the Peierls wavelength for large but finite U.

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