From Band Insulator to Mott Insulator in One Dimension

Michele Fabrizio,[1,3] Alexander O. Gogolin,[2] and Alexander A. Nersesyan[3,4]
[1]*International School for Advanced Studies and INFM, Via Beirut 4, 34014 Trieste, Italy*
[2]*Department of Mathematics, Imperial College, 180 Queen's Gate, London SW7 2BZ, United Kingdom*
[3]*The Abdus Salam International Centre for Theoretical Physics, P.O. Box 586, 34100, Trieste, Italy*
[4]*Institute of Physics, Tamarashvili 6, 380077, Tbilisi, Georgia*
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We derive the phase diagram for the one-dimensional model of a ferroelectric perovskite recently analyzed by Egami, Ishihara, and Tachiki [Science **261**, 1307 (1993)]. We show that the interplay between covalency, ionicity, and strong correlations results in a spontaneously dimerized phase which separates the weak-coupling band insulator from the strong-coupling Mott insulator. The transition from the band insulator to the dimerized phase is identified as an Ising critical point. The charge gap vanishes at this single point with the optical conductivity diverging as $\sigma(\omega) \sim \omega^{-3/4}$. The spin excitations are gapless above the second transition to the Mott insulator phase.

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Much effort has been devoted to understanding the response of strongly correlated electron systems to lattice distortions. A surprisingly pronounced enhancement of such response has recently been observed in numerical studies of a prototype one-dimensional (1D) model for ferroelectric perovskites [1] (originally this model was introduced in the context of quasi-1D organic materials [2]).

The Hamiltonian of the model is

$$\hat{H} = \sum_{i\sigma}\left[-t\left(c^{\dagger}_{i\sigma}c_{i+1\sigma} + \text{H.c.}\right) + \Delta(-1)^i n_{i\sigma}\right] + U\sum_i n_{i\uparrow}n_{i,\downarrow}, \tag{1}$$

at half filling. The odd and even sites represent oxygen atoms (O) and a generic cation (C), respectively, with the energy difference $E_{\text{C}} - E_{\text{O}} = 2\Delta$. The hopping amplitude t determines the amount of covalency and U stands for the on-site Coulomb repulsion. In spite of its apparent simplicity, model (1) reveals nontrivial physics. At $U = 0$ it describes a band insulator (BI) with a spectral gap for all excitations. At $\Delta = 0$ it is a Mott insulator (MI) with a finite charge gap, m_c, but with gapless spin excitations. In fact, in the strong-coupling limit $U \gg (t, \Delta)$, (1) can be mapped onto a Heisenberg model by projecting out all doubly occupied sites. In the leading order, only a nearest-neighbor exchange $J = 4t^2U/(U^2 - 4\Delta^2)$ is generated [2]. The resulting Heisenberg chain is well known to possess a gapless spin spectrum. The issue of interest is the nature of the crossover from the BI regime to the MI regime which, on general grounds, is expected to occur in the strong-coupling region where the single-particle BI gap Δ becomes comparable with the MI charge gap m_c.

Thus, increasing U at fixed Δ, one expects a transition to a spin-gapless phase. In addition to this *spin transition*, the charge degrees of freedom should be also affected in the course of the crossover. This can easily be understood in the case $t \ll (U, \Delta)$ where, upon increasing U, the system approaches a mixed-valence regime in which the two charge configurations, $\text{O}^{-2}\text{C}^{+2}$ with energy $U - 2\Delta$ and $\text{O}^{-1}\text{C}^{+1}$ with zero energy, become degenerate. Therefore, those excitations responsible for the charge redistribution among these two configurations should soften around some value of U. The finite-size simulations [1,3–6] suggest that the mixed-valence regime is accompanied by a strongly enhanced response to the coupling with zone-center optical phonons ξ,

$$\hat{H}_{e\text{-ph}} = -\lambda\xi\mathcal{D}, \qquad \mathcal{D} = \sum_{i,\sigma}(-1)^i\left[c^{\dagger}_{i\sigma}c_{i+1\sigma} + \text{H.c.}\right], \tag{2}$$

where λ is the electron-phonon coupling constant and $\mathcal{D}$ is the dimerization operator. In fact, the so-called average dynamical charge $Z_* = \lim_{\xi\to 0}[P(\xi) - P(0)]/\xi$, where $P(\xi)$ is the polarization, diverges at a particular point, even in finite-size simulations [4,5]. This is due to the *accidental* degeneracy between two singlet ground states with opposite parity which occurs in any finite-size system with the number of sites a multiple of 4 [4,5]. More importantly, Resta and Sorella [6] have recently observed that the localization length of the ground-state wave function diverges at some value of U, which suggests vanishing of the charge gap, hence the *charge transition*.

However, the overall picture which emerges from numerical simulations and the existing mean-field calculations [4], clearly underestimating the crucial role of fluctuations in 1D, is still far from being satisfactory. It remains unclear whether the charge instability occurs at the same point as the spin transition. Even the nature of the transition (transitions?) is not known. Is it a first-order transition or is it a second-order one? In the latter case, what is the universality class it belongs to? To the best of our knowledge, there have been no consistent analytic attempts to resolve these intriguing questions.

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In this Letter we present theoretical arguments showing that there are two continuous transitions: the spin transition of the Kosterlitz-Thouless (KT) type at $U = U_{c2}$, and the charge transition at $U = U_{c1} < U_{c2}$, the latter identified as an Ising critical point where the charge gap vanishes. In the intermediate region, $U_{c1} < U < U_{c2}$, the system occurs in a spontaneously dimerized insulating phase (SDI), with the site parity spontaneously broken, doubly degenerate ground state, and $\langle \mathcal{D} \rangle$ being the order parameter.

We start our discussion by addressing the mechanism of the spin gap generation in the MI phase. For the single-chain Hamiltonian (1), which is SU(2) and site-parity invariant, the only possibility compatible with the symmetries is a spontaneous spin dimerization, like the one occurring at $J_2/J_1 \simeq 0.24$ in the frustrated Heisenberg chain with nearest-neighbor (J_1) and next-nearest-neighbor (J_2) exchanges [7]. The same scenario is realized in our case below a critical value U_{c2}. Moreover, in the J_1-J_2 model, there exists a solvable point [8], $J_2 = 0.5J_1$, where, in any finite chain with an even number of sites, the even and odd parity singlets become exactly degenerate ground states. This is similar to the above mentioned accidental degeneracy which occurs in model (1).

To demonstrate that the same scenario is realized in our case upon decreasing U in the MI phase, we develop the *low-energy* effective field theory for the lattice model (1). Considering the weak-coupling case, $(U, \Delta) \ll t$, we linearize the spectrum and pass to the continuum limit by substituting $a_0^{-1/2} c_{n\sigma} \to i^n \psi_{R\sigma}(x) + (-i)^n \psi_{L\sigma}(x)$, $x = na_0$, where a_0 is the lattice spacing, and $\psi_{R,L}(x)$ are the right and left components of the Fermi field. These fields can be bosonized in a standard way [9]: $\psi_{R,L;\sigma} = (2\pi\alpha)^{-1/2} e^{\pm i\sqrt{4\pi}\phi_{R,L;\sigma}}$, where $\phi_{R(L),\sigma}$ are the right (left)-moving Bose fields. We define $\Phi_\sigma = \phi_{R\sigma} + \phi_{L\sigma}$ and introduce linear combinations, $\Phi_c = (\Phi_\uparrow + \Phi_\downarrow)\sqrt{2}$ and $\Phi_s = (\Phi_\uparrow - \Phi_\downarrow)\sqrt{2}$, to describe the charge and spin degrees of freedom, respectively. Then the Hamiltonian density of the bosonized model is given by $\mathcal{H}_{\text{eff}} = \mathcal{H}_c + \mathcal{H}_s + \mathcal{H}_{cs}$. Here the charge and spin sectors are described by

$$\mathcal{H}_c = \frac{v_c}{2} \left[\Pi_c^2 + (\partial_x \Phi_c)^2\right] - \frac{g_3}{2(\pi\alpha)^2} \cos\sqrt{8\pi}\, \Phi_c + \frac{2g_2 - g_1}{\pi}\, \partial_x \phi_{cR} \partial_x \phi_{cL}\,, \tag{3}$$

$$\mathcal{H}_s = \frac{v_s}{2} \left[\Pi_s^2 + (\partial_x \Phi_s)^2\right] + \frac{g_1}{2(\pi\alpha)^2} \cos\sqrt{8\pi}\, \Phi_s - \frac{g_1}{\pi}\, \partial_x \phi_{sR} \partial_x \phi_{sL}\,, \tag{4}$$

where $\Pi_{c,s}$ are the momenta conjugate to $\Phi_{c,s}$, and $v_{c,s}$ are the velocities of the charge and spin excitations. Actually, $\mathcal{H}_s$ in (4) is an Abelian bosonized version of the $SU(2)_1$-symmetric Wess-Zumino-Novikov-Witten model with a marginally irrelevant ($g_1 > 0$) current-current perturbation, $-2g_1 \mathbf{J}_R \cdot \mathbf{J}_L$, given by the last two terms in (4). The two sectors of the theory are coupled by the Δ term,

$$\mathcal{H}_{cs} = -(2\Delta/\pi\alpha) \sin\sqrt{2\pi}\, \Phi_c \cos\sqrt{2\pi}\, \Phi_s. \tag{5}$$

Assuming that the spin sector is gapless while the charge one is gapped, one can integrate out the charge degrees of freedom to obtain in the second order in Δ a nontrivial renormalization of g_1: $\tilde{g}_1 = g_1 - C(\Delta/m_c)^2 v_c$, where $C \sim 1$ is a nonuniversal numerical constant. Until $\tilde{g}_1 > 0$, the spin spectrum remains gapless. However, when U is decreased, $\tilde{g}_1$ eventually becomes negative, and the system undergoes a continuous (KT) transition to the SDI phase with a nonzero spin gap (notice that the charge gap stays finite across the transition). Using the exact result for the small-U Hubbard model, $m_c \sim \sqrt{Ut}\, e^{-2\pi t/U}$, one concludes that

$$U_{c2} = \frac{2\pi t}{\ln(t/\Delta)} \left[1 + O\left(\frac{\ln\ln(t/\Delta)}{\ln(t/\Delta)}\right)\right]. \tag{6}$$

The appearance of the SDI phase can be also inferred starting from the BI phase. In the absence of interaction, the electrons are decribed by the Hamiltonian

$$H_0 = \sum_{k,\sigma} \Psi_{k\sigma}^\dagger [\varepsilon_k \tau_3 + \Delta \tau_1] \Psi_{k\sigma}\,, \tag{7}$$

where $|k| < \pi/2$, $\varepsilon_k = -2t \cos k$, $\Psi_{k\sigma}^\dagger = (c_{k\sigma}^\dagger, c_{k+\pi,\sigma}^\dagger)$ is a two-component Fermi field, and $\tau_{1,2,3}$ are the Pauli matrices. We also introduce particle-hole operators $\hat{\tau}_{a\sigma}(q) = \sum_k \Psi_{k\sigma}^\dagger \tau_a \Psi_{k+q\sigma}$, $(a = 0, 1, 2, 3)$, where $\tau_0 = I$. The staggered charge/spin density wave (CDW)/(SDW) order parameters are given by $\hat{\tau}_{1\uparrow}(0) \pm \hat{\tau}_{1\downarrow}(0)$, while the dimerization $\mathcal{D} = \hat{\tau}_{2\uparrow}(0) + \hat{\tau}_{2\downarrow}(0)$. A generic interaction compatible with the symmetry of (1) can be written as

$$\hat{H}_{\text{int}} = (1/4L) \sum_{q,\sigma,\sigma'} \sum_{a=0}^{3} \lambda_a \hat{\tau}_{a\sigma}(q) \hat{\tau}_{q\sigma'}(-q)\,. \tag{8}$$

The coupling constants λ_a are related to the backward (g_1), forward (g_2), and umklapp (g_3) scattering amplitudes as follows [10]: $\lambda_0 = -\lambda_3 = g_2$, $\lambda_1 = g_1 + g_3$, and $\lambda_2 = g_1 - g_3$. In the special case of (1), $g_1 = g_2 = g_3 = Ua_0 \equiv g_0$, and the $\hat{\tau}_2$-$\hat{\tau}_2$ term vanishes. Therefore, in any mean-field treatment of the original Hamiltonian, only variational states with finite average values of $\hat{\tau}_{1\sigma}(0)$, i.e., the CDW and SDW densities, can be explored. However, renormalization of the coupling constants taking place in the energy range $\max(\Delta, m_c) < |\varepsilon| \ll t$ enhances g_3 and g_2 but suppresses g_1 [10]. This implies the generation of an effective coupling λ_2 which may lead to the appearance of a finite average value of $\hat{\tau}_2(0)$.

To understand which of the two competing instabilities, SDW vs $\mathcal{D}$, develops first upon increasing U in the BI phase, we consider singularities of the dynamical

susceptibilities $\chi_a(\omega,0)$ ($a = \mathrm{SDW}, \mathcal{D}$), associated with triplet and singlet excitonic modes in the $\hat{\tau}_1$ and $\hat{\tau}_2$ channels, respectively. In the noninteracting case, the susceptibilities are

$$\chi_a^0(\omega,0) = (1/2\pi v_F)[\ln(2t/\Delta) + Q_a(\omega)].$$

The functions $Q_a(\omega)$ are given by $Q_{\mathcal{D}} = \theta\tan\theta$, $Q_{\mathrm{SDW}} = -(\theta/2)\sin 2\theta$, where $\sin\theta = \omega/2\Delta$. Vertex corrections to $\chi_a^0(\omega,0)$ can be included by separating in all diagrams the low-energy parts of the particle-hole loops, $Q_a(\omega)$, which are connected by the leading logarithmic parquet vertices $\Gamma_a(\xi)$ emerging upon integrating out the higher-energy region $\Delta < |\varepsilon| \ll t$ and depending on the logarithmic parameter $\xi = \ln(t/\Delta)$. The excitonic poles are then determined by the equations

$$1 - (1/2\pi v_F)\Gamma_a(\xi)Q_a(\omega) = 0,$$

where $\Gamma_{\mathcal{D}}(\xi) = g_2(\xi) - 2g_1(\xi) + g_3(\xi)$, $\Gamma_{\mathrm{SDW}}(\xi) = g_2(\xi) + g_3(\xi)$ [explicit expressions for renormalized couplings $g_i(\xi)$ can be found in Ref. [10]]. The above equation has no solution in the SDW case, implying that, as long as the effective interaction is weak enough ($\xi < \xi_0 = \pi v_F/2g$), the triplet exciton is not formed at all. On the other hand, we find that the singlet exciton appears at an infinitesimal interaction strength; moreover, it becomes soft on approaching the strong-coupling region $\xi \sim \xi_0$, where the excitonic gap is very small: $\omega \sim \Delta(\xi_0 - \xi)^{1/2}$. Such softening of the singlet excitonic mode unambiguously indicates that interaction drives the BI phase to the SDI phase via a charge transition at which the spin degrees of freedom do not undergo qualitative changes.

With the mechanism of the spin transition already discussed, it becomes clear that the two transitions are of an entirely different physical nature. We can safely state that $U_{c1} < U_{c2}$ implying that the SDI phase occupies a finite range of U. Namely, we find that $U_{c2}/U_{c1} - 1 = \mathrm{const}/\ln(t/\Delta)$, where the positive constant in the right-hand side is of the order of 1, once the leading logarithmic accuracy is adopted. Next-to-leading corrections may change the value of this constant but not its sign.

However, separation between U_{c1} and U_{c2} becomes significantly stronger when the electron-phonon interaction, always present in realistic situations, is taken into account. Assume for simplicity that the frequency of the zone-center optical phonons, ω_0, is the largest energy scale in the problem. Such quantum phonons will mediate an instantaneous interaction between the electrons which adds to the Hubbard repulsion and leads to a renormalization of the "bare" coupling constants: $g_3 \to g_3 + \zeta$, $g_1 \to g_1 - \zeta$, where $\zeta \sim \lambda^2/\omega_0$. On increasing ζ, the transition points U_{c1} and U_{c2} will start moving apart, and if the electron-phonon coupling is large enough ($\zeta > g_1$), a finite spin gap will be always present, and the model will display only BI and SDI phases. This situation is realized in mixed-stack organic compounds [11].

In what follows, we shall focus on the nature of excitations in the SDI phase and a nonperturbative description of the charge transition to the BI phase. The Hamiltonian $\mathcal{H}_{\mathrm{eff}}$ given by (3)–(5) will be regarded as a *phenomenological* Landau-Ginzburg energy functional, in the sense that all the couplings are effective ones obtained by integrating out high-energy degrees of freedom. The saddle points of the effective potential in $\mathcal{H}_{\mathrm{eff}}$ are determined by the equations (here $\varphi_{c,s} = \sqrt{2\pi}\,\Phi_{c,s}$)

$$\cos\varphi_c(4\tilde{g}_3\sin\varphi_c - 2\pi\tilde{\Delta}\cos\varphi_s) = 0, \qquad (9)$$

$$\sin\varphi_s[2\pi\tilde{\Delta}\sin\varphi_c - 4\tilde{g}_1\cos\varphi_s] = 0, \qquad (10)$$

supplemented by stability conditions. At $\tilde{g}_3 < \pi\tilde{\Delta}/2 < \sqrt{\tilde{g}_3\tilde{g}_1}$ there are two sets of minima (defined modulo 2π) located at $\varphi_s = 0$, $\varphi_c = \pi/2$, and $\varphi_s = \pi$, $\varphi_c = -\pi/2$. These sets characterize the BI phase. Indeed, the vacuum-vacuum transitions, $\Delta\varphi_{s(c)} = \pm\pi$, describe stable topological excitations carrying the charge $Q = \Delta\varphi_c/\pi = \pm 1$ and spin $S^z = \Delta\varphi_s/2\pi = \pm 1/2$ and therefore coinciding with "massive" single-fermion excitations of the BI. The situation changes at $\tilde{g}_3 > \pi\tilde{\Delta}/2$ where each minimum in the charge sector splits into two degenerate minima and thus transforms to a local *double-well* potential. The new minima are given by $\varphi_s = 0$, $\varphi_c = \phi_0$, $\pi - \phi_0$, and $\varphi_s = \pi$, $\varphi_c = -\phi_0$, $-\pi + \phi_0$, where $\phi_0 = \arcsin(\pi\tilde{\Delta}/2\tilde{g}_3)$. They describe the SDI phase where the dimerization operator $\mathcal{D}(x) \sim \cos\varphi_c(x)\cos\varphi_s(x)$ acquires a finite expectation value. Notice that the location of the minima in the spin sector, and hence the spin quantum numbers of the topological excitations, are the same as in the BI phase ($\tilde{g}_3 < \pi\tilde{\Delta}/2$). Therefore the spin part of the spectrum formed in the SDI phase smoothly transforms to that of the BI phase. The transition at $\tilde{g}_3 = \pi\tilde{\Delta}/2$ mainly involves the charge degrees of freedom which undergo dramatic changes. In particular, the charge quantum numbers become *fractional*, depending on ϕ_0 [12]. The Z_2 degeneracy of the SDI state implies the existence of topological kinks carrying the spin $S = 1/2$ and charge $Q = \pm 2\varphi_0/2$. These massive excitations, described by interset vacuum-vacuum transition, interpolate between the neutral spinons of the MI phase and single-fermion excitations of the BI phase. However, the double-well local structure of the effective potential in the charge sector gives rise to *singlet* kink excitations as well, which lose their mass and charge $Q = 1 - 2\phi_0/\pi$ at $\tilde{g}_3 = \pi\tilde{\Delta}/2$ and transform to the neutral excitons of the BI phase. Precisely at $\tilde{g}_3 = \pi\tilde{\Delta}/2$, the effective potential becomes $\sim\varphi_c^4$, which is well known for describing the Ising universality class.

The Ising scenario at $U = U_{c1}$ can be more rigorously proven by studying the charge degrees of freedom with the spin bosonic field locked at $\Phi_s = 0$ or $\sqrt{\pi/2}$. Such an approach will certainly be valid in a small vicinity of U_{c1} where the charge sector is described by a *double*

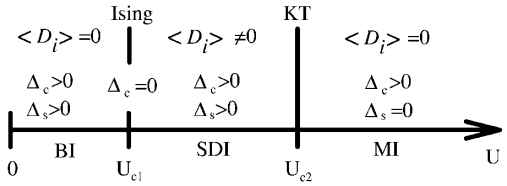

FIG. 1. Phase diagram of (1) as a function of U at fixed Δ and t. $\Delta_{c(s)}$ stands for the charge (spin) gap.

sine-Gordon (DSG) Hamiltonian

$$\hat{H}_c(x) = \frac{v_c}{2}\{\Pi_c(x)^2 + [\partial\Phi_c(x)]^2\} - \frac{g}{\pi^2 a_0}\cos\sqrt{8\pi K_c}\,\Phi_c(x) - \frac{2\Delta}{\pi a_0}\sin\sqrt{2\pi K_c}\,\Phi_c(x)\,. \quad (11)$$

We stress once more that all the parameters are purely phenomenological, and part of the interaction has been absorbed in the rescaling parameter $K_c < 1$. A quantum phase transition of an Ising type in the DSG model was recently discovered by Delfino and Mussardo [13]. Hamiltonian (11) can be mapped onto two coupled 2D Ising models, or equivalently, two coupled quantum Ising chains in a transverse magnetic field. One of the two chains always remains off critical, while the other one can pass through a critical point by fine-tuning the parameters of the model. The details will be presented elsewhere [14]. Here we just quote our main findings, which can be easily understood once the Ising scenario is assumed. The most relevant perturbation at an Ising transition is the magnetic field h, which, in the ordered phase, selects one of the two equivalent vacua. By analogy, the role of h is played in our case by the explicit dimerization ξ, Eq. (2). The dimerization operator $\mathcal{D}$, which also plays the role of the charge polarization operator, is therefore proportional to the order parameter σ of the quantum critical Ising model. The Ising mapping allows us to identify the electron current operator, $j \sim \partial_t\sigma$, and the average electron charge operator $\rho \sim -\partial_x\sigma$. This is consistent with the general relations $\rho \sim -\partial_x\mathcal{D}$ and $\partial_t\rho + \partial_x j = 0$ (continuity) valid for insulators. The real part of ac conductivity is related to the imaginary part of polarizability $\chi(\omega)$, the latter being proportional to the dynamical spin susceptibility of the Ising model: $\sigma(\omega) \sim -\omega\Im m\chi(\omega)$. From known results on the correlation functions at the Ising transition we conclude that $\Im m\chi(\omega) \sim \omega^{-7/4}$, implying that at zero temperature the optical conductivity, $\sigma(\omega) \sim \omega^{-3/4}$ (no Drude peak), is "semimetallic." For $T \neq 0$, this behavior changes to $\sigma(\omega) \sim \omega^2/T^{11/4}$ at $\omega \ll T$, and crosses over onto the quantum-critical asymptotics ($\omega^{-3/4}$) at $\omega \gg T$.

Our findings are summarized in Fig. 1, where the phase diagram is shown at fixed Δ and t as a function of U. Between the BI ($U < U_{c1}$) and MI ($U > U_{c2}$) phases there exists a spontaneously dimerized phase even in the absence of an explicit electron-phonon interaction [15]. The transition from the MI to the spontaneously dimerized insulator is of the KT type, accompanied by the opening of the spin gap. The transition from the SDI to the BI has been shown to be of the Ising type, with the singlet exciton binding energy vanishing at this single point [15].

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