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Theoretical Framework for Quasi-One Dimensional Systems

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Contents

1. Introduction

Organic materials have provided theorists with an extraordinary laboratory to study the effects of interactions in solids. Indeed the physics of strongly correlated electrons is one of the most difficult and fascinating subjects of condensed matter. From the theoretical point of view most of our understanding of this problem is based on Landau's Fermi liquid theory.[1–3] However, the great theoretical success of Fermi liquids did not prevent some systems to stubbornly refuse to follow this canonical theory. Such systems, loosely referred to as strongly correlated systems are of course at the heart of today's researches in condensed matter. It was also soon realized that the effects of interactions could be greatly enhanced by reduced dimensionality. Thus, understanding the physics of one and two-dimensional electrons has been the focuss of much theoretical and experimental efforts. In two dimensions, interactions lead to spectacular effects such as the fractional quantum hall effect[4,5] and perhaps to high-temperature superconductivity.[6] We however still mostly lack the theoretical tools to cope with such a two-dimensional situation. One dimension, however, is both more radical and more easy to tackle for the theorist. Here the effects of interactions are at their maximum and the Fermi liquid is destroyed and replaced by a new state of matter, the Luttinger liquid with radically different properties.

Thierry Giamarchi was born in Chamalières, France in 1963. He received his PhD from Paris XI University in 1987. His thesis work, made under the supervision of H. J. Schulz, was on the effects of disorder in Luttinger liquids. He obtained a CNRS researcher position in 1986, at the Laboratoire de Physique des Solides in Orsay, and spent two years at Bell Laboratories in 1990–92. In 2002, he became a professor in the Condensed Matter Department at the University of Geneva. His research work deals with the effects of interactions in low dimensional systems and on the effects of disorder in classical and quantum systems.

Quite naturally, the effort to discover such a state in nature has been intense. Progress in material research and chemistry has provided us with wonderful materials made of chains (see, e.g., ref 7) or ladders (see, e.g., refs 8 and 9), that could be candidates for such a one-dimensional physics. Progress in nanotechnology has given us many realizations of one-dimensional systems such as nanotubes,[10–12] quantum wires (see, e.g., refs 13–15), and edge states in quantum Hall effect.[16–18] Both in the organic materials and in these nanoscopic systems, one could search for one-dimensional effects. However, among all these systems, the organic conductors remain a unique challenge. Indeed, because of their very three-dimensional nature, they provide not a single one-dimensional electron gas but a very large number of such one-dimensional systems coupled together. This allows thus for a unique new physique to emerge where the system is able to crossover from a one-dimensional behavior to a more conventional three-dimensional one.

The organic systems thus offer unique possibilities. Their richness is also a drawback, since too many

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effects are taking place at the same time, making more complex the interpretation of the results. I will thus, in this brief review, make a presentation of the main theoretical concepts for electrons in one dimension and briefly discuss how these concepts are relevant to understanding the quasi-one-dimensional organic conductors. Even with this limitation, the material is enormous: effects of interactions in the normal phase, effects of magnetic fields (field induced spin density waves), ordered states such as the spin-Peierls, charge and spin density waves, and the superconducting phases, the effects of anions and disorder, and it is of course impossible to cover all these aspects in this relatively short review. I have thus chosen to focuss on the properties of the "normal" phase and the effects of interactions in it. This is where the organics, as quasi-one-dimensional systems, are really unique. This is also where the combined action of chemists, experimentalists, and theorists is needed the most given the complexity of the problem. Hopefully, the pursuit of those ideas might stimulate the creation of new materials that would be invaluable in helping to understand this challenging physics. This review is thus written with chemists or experimentalists, rather than theorists, in mind and thus will focuss on concepts and skip all technicalities. I will give references to more technical reports on one-dimensional interacting fermions where the reader can find all of the calculations made in detail.

The plan of this review is as follows. In section 2, I expose the two main concepts for interacting electrons: Fermi liquids in high dimension and Luttinger liquids in one dimension. I discuss in detail the main properties of a Luttinger liquid and the experimental signatures of its existence. In section 3, I discuss the effects of the lattice on the electronic properties, namely the physics of Mott insulators. These three concepts are the essence of what is to be expected in an interacting one-dimensional system such as the organics. In section 4, I consider the physics of many coupled one-dimensional chains and the two main phenomena that it entails: the dimensional crossover and the deconfinement transition. In section 5, I discuss the application of the above concepts to the quasi-one-dimensional organic conductors. Conclusions can be found in section 6 together with a wish list of what new type of compounds could be useful in helping to solve some of the puzzles of these families.

2. Basic Interaction Effects

Let me first expose the main theories describing the effects of interactions. It is of course impossible to cover all of the aspects of the problem. So I will just recall the salient points of the Fermi liquid theory and the Luttinger liquid theory. The experimental consequences specific to the organics will be found in section 5.

2.1 Fermi Liquids

The effects of interactions in "high"-dimensional systems have been masterfully explained by Landau's Fermi liquid theory.[1−3] This theory has been the cornerstone of our understanding of interacting Fermi systems for the last 50 years and has been explained in detail in many textbooks.[19−22] I will thus give here only a sketch of this theory and refer the reader to the aforementioned textbooks for more details.

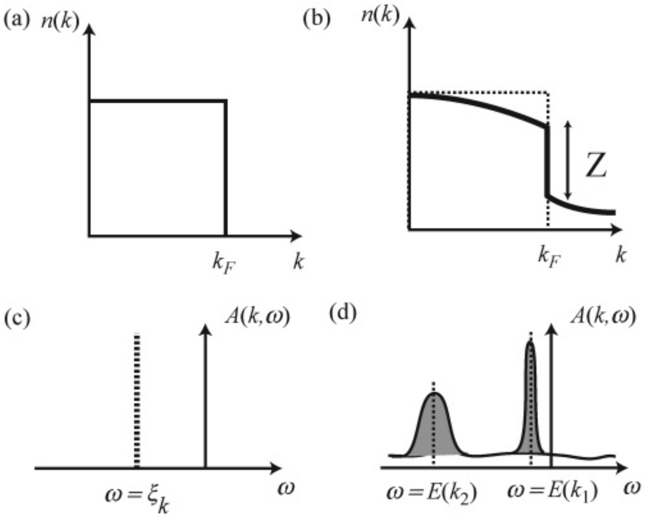

Figure 1. (a) For free electrons the occupation n_k has a discontinuity of amplitude 1 at the Fermi surface. The spectral function $A(k,\omega)$ is a delta function peak (c), showing excitations without damping and a well-defined frequency-momentum relation $\omega = \xi(k)$. The excitations are made of the individual electrons of a given momentum. (b) In a Fermi liquid, the occupation n_k still has a discontinuity at the Fermi wavevector $k = k_F$, but with a reduced amplitude $Z < 1$. The excitations are electrons dressed by density fluctuations. These excitations become sharper (d) when they get closer to the Fermi surface. The total weight in these excitations (quasiparticles) is Z.

The remarkable result of Fermi liquid theory is that not much changes when interactions are present and that the properties of the system remain *essentially* similar to those of *free* fermionic particles. The electrons themselves are strongly interacting; thus, the elementary particles are not the individual electrons anymore but electrons dressed by the density fluctuations around them. Just as in electrodynamics, an electron dresses by surrounding itself by a photon cloud, and here the electrons surround themselves with particle−hole excitations (see Figure 1).

Since these excitations are made of an electron plus density fluctuations, they behave as fermions. These individual objects are called quasiparticles. Only a residual interaction, described by the so-called Landau parameters, exists between the quasiparticles. Thus, the occupation number n_k of a state with momentum k still has a *discontinuity* at the Fermi surface. The amplitude of this discontinuity is not 1 anymore but a number Z that represents the "fraction" of the electron that remains in this quasiparticle state. The more interacting the system is, the more scrambled it is and thus the smaller the discontinuity (see Figure 1).

The quasiparticles have a "well-defined" relation between frequency ω and momentum $\omega = E(k)$, which simply reflects the fact that the wave function of a quasiparticle has a time dependence $e^{-iE(k)t}$. Of course, $E(k)$ is not the bare energy of an electron. The linearization of this new dispersion close to the Fermi level $E(k) \simeq E(k_F) + k_F/m^* (k - k_F)$ defines the effective mass m^* of the quasiparticle. Because they are not completely free the quasiparticle excitations also have a lifetime τ. Thus, the Fourier transform of the time dependence of the wave function of the excitation ($t > 0$)

$$e^{-iE(k)t}e^{-t/\tau} \tag{1}$$

is not just a delta function but a Lorentzian of width $1/\tau$ centered around $\omega = E(k)$ (see Figure 1). Since the lifetime is due to the scattering between quasiparticles close to the Fermi energy, quasiparticles have less and less phase space to scatter and the lifetime *diverges* when one goes closer to the Fermi level. Landau remarkably has shown from simple phase space arguments that the lifetime diverges in $d = 3$ as $1/E(k)^2$. Thus, the lifetime is *always* larger than the period close the Fermi level, and thus, the quasiparticle concept is well defined. In fact, the excitations become better and better defined when one approaches the Fermi level. For most properties, one can simply ignore the lifetime, that is mostly important when dealing with properties such as transport. Because it varies as $1/E(k)^2$, it naturally gives a resistivity proportional to T^2. Note that T^2 holds for dimensions greater or equal to two. In one dimension the same calculation would give $\rho(T) \sim T$ because of phase space restriction. As we will see in section 3, this result is incorrect in one dimension and is strongly modified by the presence of interactions.

The natural separation of energy scales in solids is what makes the concept of quasiparticles so useful. For practical purposes, one is practically always "close" to the Fermi level, since the ratio temperature/Fermi energy is usually about 10^{-2} in a standard metal. At these energies, the quasiparticles are the correct well-defined and "free" excitations, whereas the individual electrons themselves are strongly coupled. The spectral function $A(k,\omega)$, which gives the probability of finding an electron with a momentum k and an energy ω, thus contains (Lorentzian) peaks centered at $\omega = E(k)$ that becomes sharper and sharper when k goes to k_F. The total weight of these peaks is Z and comes from the part of the excitations that is in the quasiparticle state. The rest $1 - Z$ is in a continuous background that has no well-defined structure but can be safely ignored, close to the Fermi surface compared to the well-defined peaks. Since only the sharp excitations have a reason to give a discontinuity in $n(k)$ at k_F (in a similar way than for free electrons), the jump in $n(k)$ at $k = k_F$ has an amplitude Z and not unity any more as for free electrons.

Self-consistent[3] and renormalization[23] proofs of Fermi liquid theory have been given. However, the great strength of Landau's theory resides in the fact that it is *not restricted to weak coupling*. The existence of quasiparticles, which have fermionic nature and the above properties, is extremely robust and relies only on phase space arguments linked to the very existence of the discontinuity at the Fermi surface. It can work even for extremely strong interactions. This is fortunate since for typical systems Coulomb interaction is neither dominant nor negligible compared to the kinetic energy. This is what makes interactions so difficult to treat: strictly speaking one cannot really use any perturbation theory. Fermi liquid theory allows us to circumvent this difficulty. To prevent the theory from applying, one thus needs either exceedingly strong interactions or special conditions such that some other instability occurs (see, e.g., ref 24).

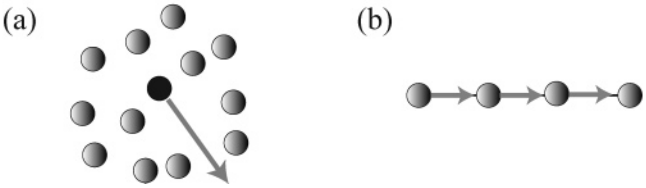

Figure 2. (a) In high dimensions, nearly free quasiparticle excitations, that look nearly as individual fermionic objects exist since a particle can move "between" the others. (b) In a one-dimensional interacting system, an individual electron cannot move without pushing all of the electrons. Thus, only *collective* excitations can exist.

To complete our brief tour of Fermi liquid, let us note two additional facts. First, in addition to these individual quasiparticle excitations, other types of excitations exist in an interacting system. One can define collective excitations that describe the response of the system to a disturbance of the density or the spin density. For example, for the charge, if only short-range interactions are present (as is the case, e.g., in helium) this collective excitation is the zero sound and represents the way a density wave can propagate (with or without damping) in the electron gas. When long-range (Coulomb) interactions are present, this is the plasmon excitation.[20,21] Thus, three main classes of excitations exist: the charge and spin collective modes, bosonic in nature since they are simple fluctuations of density or spin density *and* the individual collective excitations, fermionic in nature.

2.2. Luttinger Liquids

Fermi liquids are thus the paradigm to describe interacting electrons in three-dimensional systems. If the motion of the electrons is one-dimensional, drastically new effects occur. In one dimension, as shown in Figure 2, an electron that tries to propagate has to push its neighbors because of electron–electron interactions. So no individual motion is possible. Any individual excitation has to become a collective one. Thus, no single particle excitations can exist. This "collectivization" of excitations is obviously a major difference between the one-dimensional world and higher dimensions and invalidates any possibility to have a Fermi liquid theory work. For fermions with spin, this is even worse. Because only collective excitations exist, it implies that a single fermionic excitation has to split into a collective excitation carrying charge (like a sound wave) and a collective excitation carrying spin (like a spin wave). These excitations have in general different velocities,

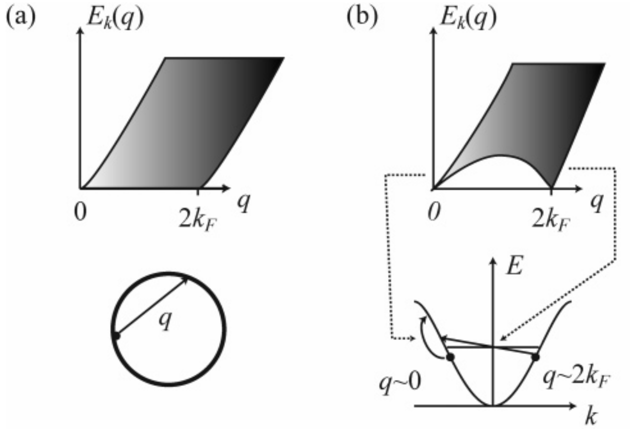

Figure 3. Particle−hole spectrum for two- or three-dimensional systems (a) and for one-dimensional ones (b). In one dimension, contrary to higher dimensions, particle−hole excitations have asymptotically for small q both a well-defined momentum and energy. Low energy modes exists around $q \sim 0$ and $q \sim 2k_{\mathrm{F}}$.

so the electron has to "break" into two elementary excitations. These properties, quite different from the ones of a Fermi liquid, are the essence of the Luttinger liquid, which I now discuss.

I will not give here a full derivation of the Luttinger liquid properties since many good reviews[25−31] and textbooks[32,33] exist in the literature but will recall only the salient points.

2.2.1. Basic Concepts

The collective excitations forming the basis of the Luttinger liquid are the particle−hole excitations where an electron is taken from below the Fermi level and promoted above. Since one destroys a particle with momentum k and creates some particle with momentum $k + q$, the momentum of the excitation is well fixed and equal to q. The energy of such an excitation, on the contrary, depends in general on *both* k and q. If we look at excitations at small q, the energy of a particle−hole excitation is

$$E_k(q) = \xi(k + q) - \xi(k) \tag{2}$$

where $\xi(k)$ should be occupied and $\xi(k + q)$ empty. For a standard quadratic dispersion

$$\xi(k) = \frac{k^2 - k_{\mathrm{F}}^2}{2m} \tag{3}$$

It is easy to check that for $k \in [k_{\mathrm{F}} - q, k_{\mathrm{F}}]$ the average value $E(q)$ of $E_k(q)$ and the dispersion $\delta E(q) = \max(E_k(q)) - \min(E_k(q))$ are

$$\begin{aligned} E(q) &= \frac{k_{\mathrm{F}} q}{m} = v_{\mathrm{F}} q \\ \delta E(q) &= \frac{q^2}{m} = \frac{E(q)^2}{m v_{\mathrm{F}}^2} \end{aligned} \tag{4}$$

The behavior of the particle−hole spectrum in one dimension is shown in Figure 3. Note the difference with the one in $d \geq 2$. Regardless of the dispersion relation $\xi(k)$ provided it has a finite slope at the Fermi level: (i) the average energy of a particle−hole excitation is *only* dependent on its momentum q thus the particle−hole excitations are excitations with well-defined momentum q and energy $E(q)$; (ii) the dispersion in energy $\delta E(q)$ goes to zero much faster than the average energy. This is the same situation as the one we discussed for the Fermi liquid quasiparticles. It means that in one dimension the particle−hole excitations are well-defined "particles" (that is, objects with well-defined momentum and energy), which become longer and longer lived when the energy tends to zero. Because these excitations are made of the destruction and creation of a fermion, they are bosonic in nature. These bosonic "quasiparticles" that represent density (or spin density) fluctuations are thus the good excitations in one dimension. These collective excitations are at the heart of the properties of the Luttinger liquid.

All excitations of the system are thus sound waves of density and spin density. Their energy is given by a standard elastic-like Hamiltonian. The Hamiltonian of the system is the sum of a part containing only charge excitations and one containing only spin excitations

$$H = H_\rho + H_\sigma \tag{5}$$

where H_ν ($\nu = \rho,\sigma$) is of the form

$$H = \frac{1}{2\pi} \int \mathrm{d}x \left[u_\nu K_\nu (\pi \Pi_\nu(x))^2 + \frac{u_\nu}{K_\nu} (\nabla \phi_\nu(x))^2 \right] \tag{6}$$

where ϕ_ν and Π_ν are conjugate variables $[\Pi_\nu(x), \phi_\nu(x')] = i\hbar\delta(x - x')$. The form (5) immediately shows that an excitation that is looking like a free electron (i.e., that carries both charge and spin) cannot exist. This charge−spin separation is one important property of the Luttinger liquids. $u_{\rho,\sigma}$ are the velocities of these collective excitations. In the absence of interactions $u_\rho = u_\sigma = v_{\mathrm{F}}$. Interactions of course renormalize the velocities of charge and spin excitations, as in higher dimensions. $K_{\rho,\sigma}$ are dimensionless parameters depending on the interactions. For systems with spin rotation symmetry, $K_\sigma = 1$ (for repulsive interactions), whereas the spin excitations are gapped for attractive interactions. $K_\rho = 1$ in the absence of interactions and quite generally $K_\rho < 1$ for repulsive ones.

The three parameters u_ρ, u_σ, and K_ρ completely characterize the low energy properties of a one-dimensional system. They can of course be computed for a given microscopic model as a function of the interactions,[34−39] but as was shown by Haldane,[40−42] the form (5) is the generic low energy form. This means that (5) and the parameters u_ρ, u_σ, and K_ρ play a role similar to the one of the Landau Fermi liquid Hamiltonian (and Landau parameters) in higher dimensions. The fact that one has again in one dimension a concept equivalent to the Fermi liquid, i.e., a generic description of the low energy physics of the interacting problem, is of course an extremely important point. This removes part of the caricatural aspects of any modelization of a true experimental system. This use of the Luttinger liquid is reminiscent of the one made of Fermi liquid theory. Very

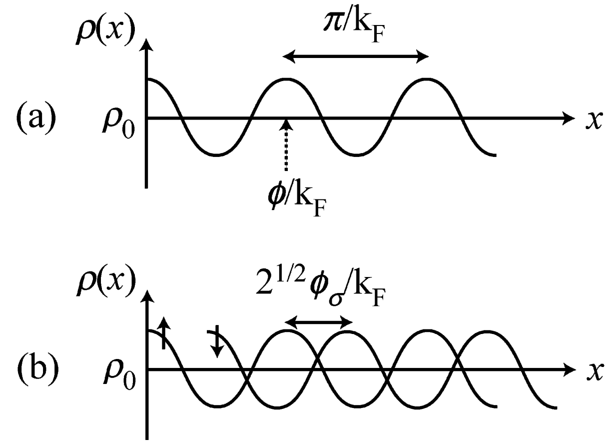

Figure 4. Semiclassical representation of the density. (a) For a spinless system, the field ϕ gives the position of the maxima of a wave of density. (b) For systems with spins, ϕ_ρ gives the maxima of charge, whereas ϕ_σ is the offset between the spin ↑ and ↓ density waves. Of course, the fields ϕ usually fluctuate in space and time so the perfect order in the density waves is destroyed (usually algebraically).

often calculations are performed in solids starting from "free" electrons and adding important perturbations (such as the BCS attractive interaction to obtain superconductivity). The justification of such a procedure is rooted in the Fermi liquid theory, where one does not deal with "real" electrons but with the quasiparticles, which are intrinsically fermionic in nature. The mass m^* and the Fermi velocity v_F are then some parameters. The calculations in $d = 1$ proceed in the same spirit with the Luttinger liquid replacing the Fermi liquid. The Luttinger liquid theory is thus an invaluable tool to tackle the effect of perturbations on an interacting one-dimensional electron gas (such as the effect of lattice, impurities, coupling between chains, etc.) as we will see in the following sections.

Let me now turn to a summary of the properties of the excitations in the Luttinger liquid. The field ϕ_ρ (respectively ϕ_σ) is related to the density (respectively spin density) of particles. Since the low energy modes are for $q \sim 0$ and $q \sim 2k_F$ (see Figure 3), the important density modes will be around these two wavevectors. The long wavelength part ($q \sim 0$) of the density is simply

$$\begin{aligned}\rho_{q\sim 0}(x) &= -\sqrt{2}\nabla\phi_\rho(x)/\pi \\ \sigma_{q\sim 0}(x) &= -\nabla\phi_\sigma(x)/(\pi\sqrt{2})\end{aligned} \tag{7}$$

whereas the $2k_F$ part is given by

$$\begin{aligned}\rho_\uparrow(x) &\propto \cos\,(2k_F x - \sqrt{2}\phi_\rho - \sqrt{2}\phi_\sigma) \\ \rho_\downarrow(x) &\propto \cos\,(2k_F x - \sqrt{2}\phi_\rho + \sqrt{2}\phi_\sigma)\end{aligned} \tag{8}$$

Thus, if ϕ was a constant, one would have density waves of wavevector $2k_F$ for the up and down spins. ϕ_ρ gives the phase of the charge density whereas ϕ_σ is the dephasing between the ↑ and ↓ density waves. As shown in Figure 4, if $\phi_\sigma = 0$, the two waves are in phase. The spin density then is zero and the charge is modulated. On the contrary, if $\phi_\sigma = \pi/\sqrt{8}$, the two densities are in opposite phase, and thus, the total charge is constant whereas the spin is modulated.

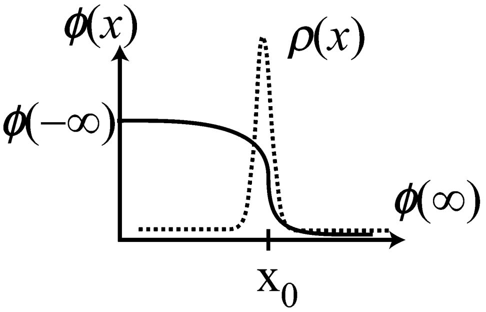

Figure 5. A kink in ϕ at point x_0 corresponds to the creation of a particle at this point. The amplitude of the kink gives the charge (or the spin for ϕ_σ) of the particle.

Of course, this perfect wave exists only if the field ϕ orders. For the Hamiltonian (6), the field ϕ fluctuates, which means that the precise phase of the wave changes with space and time and thus the correlation between different points of the wave are lost. As we will see in the next section, this gives rise to a power law decay of the various density correlations.

Excitations corresponding to the addition or extraction of a single fermion (single particle excitations) have quite special properties in a Luttinger liquid since they cannot exist as nearly free excitations. Because of (7), adding a charge (or a spin) at point x_0 (i.e., $\rho(x) = \delta(x - x_0)$) corresponds to creating a kink in ϕ_ρ (ϕ_σ) as shown in Figure 5.

Indeed, since

$$\int_{-\infty}^{x>x_0} \mathrm{d}x'\,\rho(x') = 1 = \frac{-1}{\pi}[\phi(x) - \phi(-\infty)] \tag{9}$$

one sees that the step in ϕ is quantized and is a measure of the total charge added in the system. The converse is of course true: kink-like excitations for ϕ can be interpreted as some charge carrying excitations whose charge (not necessarily integer) is given by (9). This will be specially important to describe Mott insulators in section 3.

As is physically obvious, for repulsive interactions, the system tends to a state with antiferromagnetic (also called spin density wave SDW) correlations. Let me again caricature such a state by considering that the ϕ are constants. If now one removes a fermion, it means that at a single site one creates a kink *both* in ϕ_ρ and ϕ_σ. These two excitations are free to propagate completely independently. In the fermion language, let us see what happens if we let the hole propagate as shown in Figure 6a.

In that case, one reaches the state of Figure 6b. At one point, there is a site where a charge is lacking, but the spin environment is purely antiferromagnetic. This corresponds to an excitation that is a kink in ϕ_ρ but no disturbance in ϕ_σ. This excitation is known as a holon. In another part of the system, there is a place where no charge is missing but we have two neighboring spin up. This is a spin excitation with a spin 1/2 compared to the ground state. This excitation is known as a spinon, and corresponds to a kink in ϕ_σ. In one dimension, we see that the spinon and holon are free to separate, and thus, any

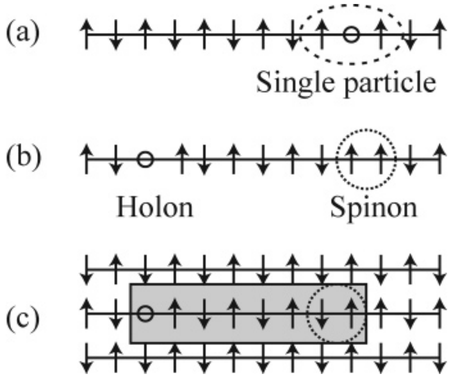

Figure 6. In a one-dimensional system, a single-particle excitation (a) is converted into an excitation that contains only charge degrees of freedom (holon) and spin degrees of freedom (spinon). This is represented schematically in (b). In (c) one sees that in a higher-dimensional system the spinon and holon would be held together by a string whose energy cost grows with distance, because of the strings of ferromagnetic links. These two excitations thus form a bound that is the single-particle fermionic excitation.

single-particle excitation dissociates into these elementary excitations. In higher dimension, our cartoon shows that because of the presence of other chains the spinon and the holon would be separated by a string of frustrated bonds. Indeed, all of the spins between the spinon and the holon are parallel to the ones of the neighboring chains. This costs the antiferromagnetic exchange, and thus, the spinon and holon are held together by a string whose energy grows linearly with the distance. They are thus confined and their bound state is the single-particle fermionic excitation, showing that in dimension greater than one single particle like excitations are good excitations.

2.2.2. Physical Properties

Let us now examine the physical properties of a Luttinger liquid. I refer the reader to the above-mentioned reviews for derivations of the results and a complete description. The compressibility of the system is constant and given by

$$\kappa_\rho/\kappa_\rho^0 = \frac{K_\rho v_{\mathrm{F}}}{u_\rho} \tag{10}$$

where κ_ρ^0 is the compressibility of the free electron gas. As can be expected it depends only on the charge sector. The uniform magnetic susceptibility is also constant and given by

$$\kappa_\sigma/\kappa_\sigma^0 = \frac{K_\sigma v_{\mathrm{F}}}{u_\sigma} \tag{11}$$

For spin isotropic systems, the Luttinger parameter K_σ is equal to one. Non analytic corrections to the magnetic susceptibility are present at finite magnetic field.[43−45] The results of the thermodynamics quantities are very similar to the Fermi liquid ones, where the constant compressibility and susceptibility are simply renormalized independently by the interactions.

The difference between Fermi liquids and Luttinger liquids shows in its full glory when one looks at correlation functions. Indeed in that case, the fact that the free excitations of the system are the collective charge and spin bosonic modes and not the individual fermion-like excitations shows clearly. First, as we already discussed, these excitations have in general two different velocities so one can expect singularities in the correlation functions both for $\omega = v_\rho k$ and $\omega = v_\sigma k$. Second, because the system is fighting between particle−particle correlations and particle−holes ones, it is on the brink of an instability without really being able to order and thus exhibits power-law correlations. Different regimes exist depending on the interactions. From now on, I focuss on repulsive interactions that are invariant by spin rotation (for the effects of spin anisotropy in the interactions see, e.g., ref 46). In that case, the spin sector is not gapped and $K_\sigma = 1$. For example, the total density ($\delta\rho = \rho - \rho_0$) or spin correlation functions in a Luttinger liquid with spins are given by (for simplicity I take here $u_\rho = u_\sigma = u$)

$$\begin{aligned}\langle \delta\rho(x,\tau)\,\delta\rho(0)\rangle &= \frac{K_\rho}{\pi^2}\frac{y_\alpha^2 - x^2}{(x^2+y_\alpha^2)^2} + \\ &\rho_0^2 A_2 \cos(2k_{\mathrm{F}}x)\left(\frac{\alpha}{\mathscr{R}}\right)^{K_\rho+1} \log^{-3/2}(\alpha/\mathscr{R}) + \\ &\rho_0^2 A_4 \cos(4k_{\mathrm{F}}x)\left(\frac{\alpha}{\mathscr{R}}\right)^{4K_\rho} + \cdots \\ \langle S_\mu(x,\tau) S_\mu(0)\rangle &= \frac{K_\sigma}{4\pi^2}\frac{y_\alpha^2 - x^2}{(x^2+y_\alpha^2)^2} + \\ &A_2' \cos(2k_{\mathrm{F}}x)\left(\frac{\alpha}{\mathscr{R}}\right)^{K_\rho+1} \log^{1/2}(\alpha/\mathscr{R}) + \cdots\end{aligned} \tag{12}$$

where x and τ are the space and (imaginary) time coordinates. α is a short distance cutoff of the order of the lattice spacing, $y_\alpha = u|\tau| + \alpha$ and $\mathscr{R} = [x^2 + y_\alpha{}^2]^{1/2}$. ρ_0 is the average density of electrons, and the A are non universal amplitudes. The logarithmic corrections come from the presence of a marginal operator at the spin isotropic point.[45−50] Since $K_\rho < 1$, we see from eq 12 that charge/spin fluctuations lead to divergent susceptibilities at $2k_{\mathrm{F}}$. The dominant correlation is thus a $2k_{\mathrm{F}}$ spin density wave. This is physically very satisfying since antiferromagnetism is the physics that one would expect for electrons with repulsion. Other phases are of course possible for more complex interactions. Note that the modulation of the spin density wave is $2k_{\mathrm{F}}$, which is an incommensurate modulation. This is the wavevector that one would get by simply assuming an antiferromagnetic order between the holes.[51]

Each harmonic ($q \sim 2k_{\mathrm{F}}$, $q \sim 4k_{\mathrm{F}}$) decays with a different power-law. The amplitudes A_i are nonuniversal objects.[52] They depend on the precise microscopic model and even on the parameters of the model. For example, for the so-called Tomonaga-Luttinger model with a strictly linear dispersion relation, all amplitudes except A_2 are strictly zero, since only $q = 0$ and $q = 2k_{\mathrm{F}}$ terms exist. Contrary to the amplitudes A_n, which depend on the precise microscopic model, the power-law decay of the various terms are *universal*. They *all* depend on the unique Luttinger coefficient K_ρ.

These powerlaw singularities, that are characteristic of the Luttinger liquid behavior, can be directly probed in experiments. The divergence of the spin correlations at $Q = 2k_F$ is given by

$$\chi(Q = 2k_F, \omega = 0, T) \sim T^{K_\rho - 1} \log^{1/2}(W/T) \quad (13)$$

where W is the bandwidth. Note that the charge susceptibility at $2k_F$ has in fact the same power law divergence and is only suppressed by logarithmic factors compared to the spin susceptibility. This divergence in the spin correlations can be probed for example in an NMR experiment. Indeed, the relaxation time T_1 in an NMR experiment is sensitive to the local spin correlation function

$$1/T_1 = A_h^2 T \sum_q \frac{\mathscr{I}\chi(q,\omega)}{\omega} \quad (14)$$

where $\mathscr{I}\chi$ is the imaginary part of the spin susceptibility and A_h is the hyperfine coupling constant. The local spin−spin correlation function has two contributions (one from the $q \sim 0$ modes the other from the $q \sim 2k_F$ ones) that give[53]

$$1/T_1 \propto T + T^{K_\rho} \quad (15)$$

The first term is nothing but the Korringa law that holds in Fermi liquids, for a susceptibility that becomes a constant at low temperatures. This is indeed the case even in $d = 1$ for the $q \sim 0$ modes. In a Fermi liquid, this is all there is. In a Luttinger liquid, the $2k_F$ contribution dominates ($K_\rho < 1$) at low temperatures and one expects strong deviations compared to the Korringa law. Neutrons experiments can also be used to probe the powerlaw divergence, but they are not yet a useful probe for organic compounds. Such neutron scattering experiments have probed with success the power-law correlations in spin chains.[54]

The retarded single-particle Green's function, whose imaginary part gives the spectral function, is a direct measure of single particle excitations. In a Luttinger liquid, it shows also a powerlaw decay at Fourier components around $\pm k_F$, $\pm 3k_F$, $\pm 5k_F$.[51,55−58] Its expression is (only the dominant $\pm k_F$ singularity is shown here)

$$\begin{aligned} G^{\text{ret}}_{r,s}(x, t) &= -iY(t)\langle[\psi_{rs}(x, t), \psi^\dagger_{rs}(0, 0)]_+\rangle \\ &= -i\frac{Y(t)}{2\pi} e^{irk_F x} \lim_{\epsilon \to 0} \left\{ \frac{\alpha + i(v_F t - rx)}{\epsilon + i(v_F t - rx)} \times \right. \\ &\prod_{\nu=\rho,\sigma} \frac{1}{\sqrt{\alpha + i(u_\nu t - rx)}} \left(\frac{\alpha^2}{(\alpha + iu_\nu t)^2 + x^2} \right)^{\gamma_\nu} + \\ &\left. \begin{pmatrix} x & \to & -x \\ t & \to & -t \end{pmatrix} \right\} \end{aligned} \quad (16)$$

where $[\,]_+$ is the anticommutator, $Y(t)$ is the Heaviside-

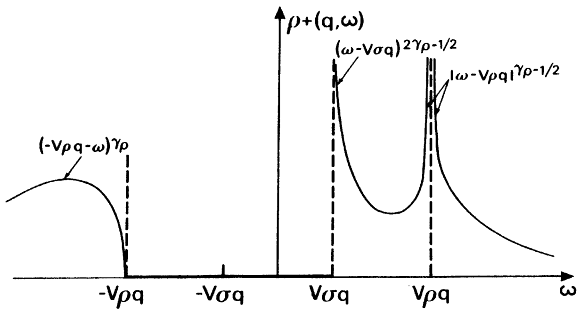

Figure 7. Spectral function (denoted $\rho_+(q,\omega)$ in this figure) of right movers in a LL. In this figure, V_ρ (respectively V_σ) denotes the charge (respectively spin) velocity (denoted u_ρ and u_σ in this review). Instead of a Lorentzian peak, one has power law divergences with an exponent that depends on the Luttinger liquid parameter. There are two peaks: one corresponding to the holons ($\omega \sim u_\rho q$) and one to the spinons ($\omega \sim u_\sigma q$). Each peak has a power law structure. This is markedly different from the spectral function in a Fermi liquid. (From ref 56; Copyright 1993 by the American Physical Society.)

function, $r = \pm 1$ denotes particles with momentum close to $\pm k_F$, and s is the spin. $\psi^\dagger_{rs}(x, t)$ is the creation operator of a particle at point x and time t. The exponent is

$$\gamma_\nu = (K_\nu + K_\nu^{-1} - 2)/8 > 0 \quad (17)$$

For a spin rotation invariant system, $K^*_\sigma = 1$ and $\gamma_\sigma = 0$. The imaginary part of the retarded correlation function can be directly measured in a photoemission experiment. I discussed the spectral function of a Fermi liquid in section 2.1, which is essentially a Lorentzian. The spectral function of a Luttinger liquid (e.g., for right movers $r = 1$) is quite different and given by

$$A_{R,s}(q,\omega) = -\frac{1}{\pi}\operatorname{Im} G^{\text{ret}}_{R,s}(k_F + q,\omega) \quad (18)$$

The Fourier transform gives the structure

$$A_{R,s}(q,\omega) \sim (\omega - u_\sigma q)^{\zeta - 1/2}|\omega - u_\rho q|^{(\zeta - 1)/2}(\omega + u_\rho q)^{\zeta/2} \quad (19)$$

where $\zeta = \sum_\nu \gamma_\nu$. Complete expressions can be found in refs 55 and 56. The spectral function is shown in Figure 7.

There are two important things to note in this figure. First, there is not a single peak but two, at frequencies $\omega = u_\rho k$ and $\omega = u_\sigma k$. There are only peaks for the holon and the spinon. This clearly shows that no individual excitation exists in the LL but that the individual fermion decomposes in these elementary objects as discussed in section 2.2.1. More importantly, since by accident the two velocities u_ρ and u_σ could be equal, the shape of the peak is *not* Lorentzian. It corresponds to the usual power law divergence with an exponent controlled by the LL parameter K_ρ.

The integrated (over k) intensity can also be computed and measured. For a Fermi liquid, it coincides with the density of states of single particle excitations at the Fermi level and has a discontinuity

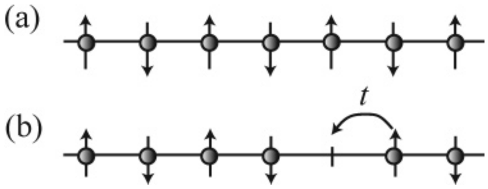

Figure 8. (a) If there is one particle per lattice site and the repulsion among particles is strong, a plane wave state for the particles is energetically unfavorable since the density is uniform. It is better to localize the particles on each site. Such a state is a Mott insulator since hopping would cost an energy of the order of the interaction U among particles. All of these effects concern the charge. For the spin degrees of freedom, virtual hopping leads to superexchange and favors antiferromagnetic order. (b) If the system is doped the extra electrons or holes can propagate without any energy cost from the interactions and gain some kinetic energy $\sim 2t$. The system is then metallic.

of amplitude Z at the Fermi level. For a Luttinger liquid, one finds

$$n(\epsilon) \propto |\epsilon|^{1/4[K_\rho + K_\rho^{-1}] - 1/2} \tag{20}$$

The integrated intensity goes to zero at the Fermi level, which signals again that no single-particle excitations do survive in the LL. Note that this does *not* mean that the system is an insulator since charge carrying excitations do exist as we will see in the next section. Besides photoemission experiments, this quantity can be probed by scanning tunneling microscope. Transport through highly resistive contacts is also a direct measure of the single particle density of states. Such probes have been used for nanotubes[11,12] and quantum wires.[14,15] For the organics, a way to probe this quantity is to look at the transport along the least conducting axis (see, e.g., ref 7). I will come back to the experimental consequences for the organic compounds in section 5.

3. Mott Insulators

So far I have dealt with Luttinger liquids. However, in the case when the density of carriers is commensurate with the lattice, another interaction-induced phenomenon occurs. Indeed, in that case, the system can become an insulator. This is the mechanism known as Mott transition[59,60] and is a metal−insulator transition induced by the interactions since a free electron gas would remain metallic with a partly filled band. The physics of a Mott insulator is well-known and illustrated in Figure 8.

If the repulsion U among the particles is much larger than the kinetic energy t, then the plane wave state is not very favorable since it leads to a uniform density where particles experience the maximum repulsion. It is more favorable to localize the particles on the lattice sites to minimize the repulsion and the system is an insulator for one particle per site. If the system is weakly doped compared to a state with one particle per site the holes can propagate without experiencing repulsion, the system is thus a metal again but with a number of carriers proportional to the doping. The above argument shows that, in high dimensions, one usually needs a finite (and in general of the order of the bandwidth) repulsion to reach that state. For further details on the Mott transition in higher dimension see refs 61 and 62.

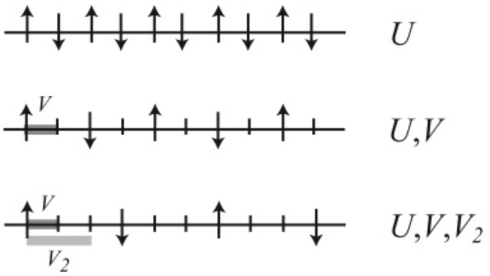

Figure 9. In a model with onsite interactions (U), a Mott insulator only exists for one particle per site (half filling). Nearest neighbor interactions V can stabilize the insulating state for up to one particle every two sites (quarter filling). And so on with longer ranger interactions (e.g., next nearest neighbor interactions V_2). The longer the range of the interaction, the higher the commensurability for which one can have a Mott insulator.

It is important to note that one particle per site is not the only commensurate filling where one can in principle get a Mott insulator but that every commensurate filling can do in principle depending on the interactions. This is illustrated in Figure 9.

It is indeed easy to see that for large enough onsite (U) and nearest neighbor (V) repulsion a quarter-filled system is an ordered Mott insulator.

3.1. Basic Ideas

Although one can of course work out the Mott transition from microscopic models such as the Hubbard model,[63] the Luttinger liquid theory provides a remarkable framework to take into account the effects of a lattice and describe the Mott transition. To incorporate the Mott transition in Luttinger liquid description, we just need to remember that in the presence of a lattice the wavevector is in fact defined modulo a vector of the reciprocal lattice (that is, in one dimension a multiple of $2\pi/a$ with a the lattice spacing). It means that in an interaction process the momentum should now only be conserved modulo a vector of the reciprocal lattice. Thus, in addition to the interaction processes that truly conserve momentum $k_1 + k_2 = k_3 + k_4$, one can now have processes such that $k_1 + k_2 - k_3 - k_4 = Q$ where Q is a vector of the reciprocal lattice. The electronic system can transfer momentum to the lattice and get it back. These processes are well-known and called umklapp processes.[43] Since umklapps do not conserve momentum, they are the only ones that can lead to a finite resistivity and are responsible for the T^2 law in a Fermi liquid[64] (or T in one dimension in a simple (and incorrect) Boltzmann approximation). For these processes to contribute at low-energy, one should of course look whether they occur for electrons on the Fermi surface. As shown in Figure 10, in high dimension, it is relatively easy to get such processes.

The situation is quite different in $d = 1$. Since the Fermi surface is reduced to two points, conserving both momentum and interactions is not easy. If one wants the particles at the Fermi surface, imposing $k_1 + k_2 - k_3 - k_4 = 2\pi/a$ can only be realized for $4k_F$

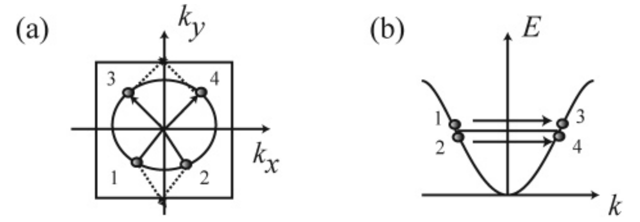

Figure 10. (a) In high dimensions, one can adjust angles and easily get umklapp processes, practically regardless of the filling (provided $|k_F|$ is large enough). (b) In one dimension, since there is no angle to play with, one gets an umklapp process where two particles are scattered from one side of the Fermi surface to the other only if $4k_F = 2\pi/a$, that is, for half-filling. Higher order umklapps for other commensurate fillings are also possible.

$= 2\pi/a$, that is, at half-filling. Physically these terms do correspond to the scattering of two fermions that have a momentum $-k_F$ on the other side of the Fermi surface with a momentum transfer of $4k_F$ from the lattice. This process is shown in Figure 10b. For one fermion per site the charge part of the Hamiltonian becomes[65,66] because of the umklapp process, instead of eq 6

$$H = H_0 + \frac{2g_3}{(2\pi\alpha)^2}\int \mathrm{d}x \cos(\sqrt{8}\phi_\rho(x)) \tag{21}$$

whereas the spin part is unchanged. The coefficient g_3 is the strength of the umklapp process and of course depends on the microscopic model used. Although the coefficient g_3 depends on the precise microscopic model, the form of the Hamiltonian (sine-Gordon) is universal.

In fact, umklapps are not restricted to one particle per site[67,68] but occur for any commensurate fillings. The umklapp terms can be viewed as the action of the periodic potential due to the Lattice on the density of electrons. For one particle per site the lattice has a periodicity of $4k_F$

$$H = \int \mathrm{d}x\, V(x)\rho(x) = V_0\int \cos(4k_F x)\rho(x) \tag{22}$$

which, keeping only the nonoscillating terms, gives back the umklapp term (21). In addition to recovering the umklapp for one particle per site, the full formula for the density[52,67] shows us that we can get an umklapp term for any *commensurate* density. Indeed, if $2pk_F = 2\pi q/a$ (where p and q are integers) then the corresponding term stays in the Hamiltonian. For even commensurabilities ($p = 2n$) this gives a term

$$H_{1/2n} = g_{1/2n}\int \mathrm{d}x \cos(n\sqrt{8}\phi_\rho(x)) \tag{23}$$

Doping causes a slight deviation from this commensurate value of k_F so in the presence of doping one can thus write quite generally the umklapp as

$$H_{1/2n} = g_{1/2n}\int \mathrm{d}x \cos(n\sqrt{8}\phi_\rho(x) - \delta x) \tag{24}$$

where n is the order of the commensurability ($n = 1$ for half-filling, one particle per site; $n = 2$ for quarter-filling, one particle every two sites and so on). The coupling constant $g_{1/2n}$ is the umklapp process corresponding to the commensurability n and δ the deviation (doping) from the commensurate filling. Odd commensurabilities can be treated in the same way. In that case, the spin part cannot be eliminated.[68] I concentrate here on even commensurabilities for simplicity and since it is the case corresponding to the Bechgaard salts.

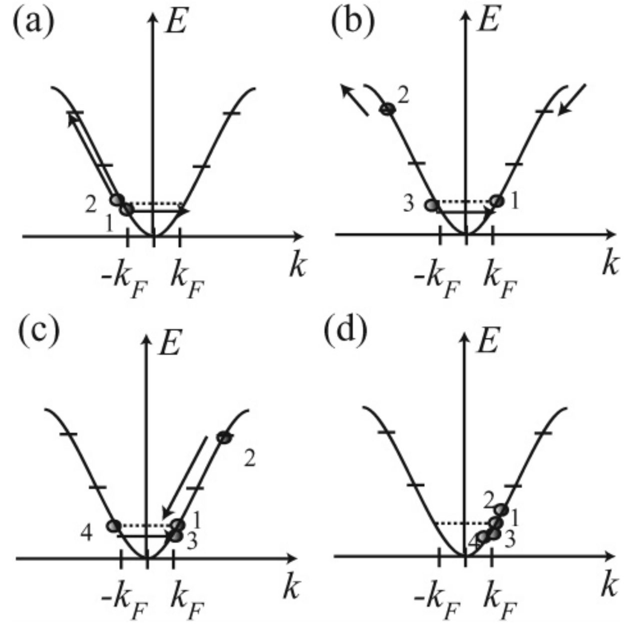

Figure 11. A quarter-filled umklapp can be constructed from a third-order perturbation theory in the interaction U. The processus needs two intermediate states of high-energy W of the order of the bandwidth. Thus, the amplitude for such a process is of order $U(U/W)^2$.

The derivation using the LL expression for the density cannot give access to the amplitudes and thus to the coefficients $g_{1/2n}$. The amplitude of the umklapp depends on the precise microscopic interaction. For the half-filled case, $g_{1/2}$ is of the order of the interaction U. The higher commensurabilities can also be understood physically. Let us consider a quarter-filled band such that $8k_F = 2\pi/a$ (this corresponds to $n = 2$ in the above notations). To produce an umklapp, one needs to transfer *four* particles from one side of the Fermi surface to the other to get the proper $8k_F$ momentum transfer. This can be done in higher-order perturbation terms by doing three scatterings as shown in Figure 11.

For weak interactions the amplitude of such a process would thus be of order $U(U/W)^2$, where W is the bandwidth. In addition to the above two simple processes, there is a additional one that is quite important for the Bechgaard salt family. Indeed, in these systems, the stack is slightly dimerized.[7] This dimerization opens a gap in the middle of the band as indicated in Figure 12.

Thus, although the system is originally quarter filled, the dimerization turns the system into a half-filled band. This means that, even if the system is quarter filled, a nonzero $g_{1/2}$ exists in addition to $g_{1/4}$. If Δ_d is the dimerization gap, the strength of such umklapp is $g^{\mathrm{d}}_{1/2} = U(\Delta_d/W)$. Note that contrary to what happens in a true half filled system, the umklapp coefficient is now much smaller than the typical interaction U. This allows us to get a small

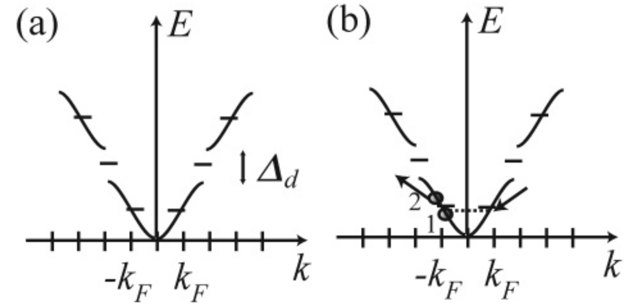

Figure 12. Dimerization opens a gap in the band (a). Because of this gap, the quarter-filled band becomes effectively half filled. The dimerization gap Δ_d thus creates even for a quarter-filled system an half filling umklapp (b).

Mott gap even if the interactions are large. In the presence of dimerization, a quarter filled system can thus be a Mott insulator either because of the half-filling umklapp (that exists now because of the dimerization) or because of the quarter-filled one. Which process is dominant depends of course of the strength of the dimerization and of the interactions and has important consequences on the physics of the system.[69]

The Hamiltonian (24) thus provides a complete description of the Mott transition and the Mott insulating state in one dimension. To change the physical properties of a commensurate system, one has thus two control parameters. One can vary the strength of the interactions while staying at commensurate filling or vary the chemical potential (or filling) while keeping the interactions constant. One can thus expect two different classes of transition to occur.

3.2. Physical Properties

Let us first deal with the transition where the filling is kept commensurate and interaction strength is varied (Mott-U transition). In that case, $\delta = 0$ and eq 24 is just a sine-Gordon Hamiltonian. As is well-known, this Hamiltonian has a quantum phase transition at $T = 0$ as a function of the Luttinger parameter K_ρ and thus as a function of the strength (and range) of the interactions. This transition is a Berezinskii-Kosterlitz-Thouless (BKT) transition.[47] The critical value is $K_\rho^* = 1/n^2$ where n is the order of the commensurability. For larger values of K_ρ, the cosine is irrelevant and the system is massless. For $K_\rho < K_\rho^*$, the cosine is relevant and the system is massive. This opening of a gap corresponds to the Mott transition and the system becomes an insulator. The larger the commensurability the smaller K_ρ needs to be for the system to become insulating. For a commensurability $n = 1$, that is, half-filling, the critical value is $K_\rho = 1$. This means that, contrary to the higher dimensional case, *any* repulsive interactions turn the system into an insulator. For a quarter-filled band ($n = 2$), the critical value is $K_\rho = 1/4$. To get the insulator, one needs both pretty strong interactions *and* interactions of a finite range, since the minimum value of K_ρ for a local interaction is $K_\rho = 1/2$.[70] This is physically obvious: in order to stabilize a structure in which there is a particle every two sites, one cannot do it with purely local interactions (see Figure 9). The range of the interactions in addition of their strength and thus the precise chemistry of the compound controls the range of values of K_ρ that one is able to explore.

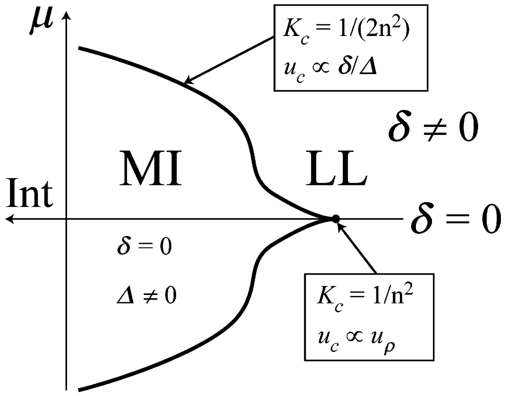

Figure 13. Phase diagram close to a commensurability of order n ($n = 1$ for half-filling and $n = 2$ for quarter-filling). Int denotes a general (that is, not necessarily local) repulsive interaction. μ is the chemical potential, δ is the doping, and Δ is the Mott gap. MI and LL are respectively the Mott insulator the Luttinger liquid (metallic) phases. The critical exponent K_c and velocity v_c at the transition depend on whether it is a Mott-U or Mott-δ transition. (After ref 69.)

The critical properties of the transitions are the ones of the BKT transition: K jumps discontinuously from the universal value K_c^* at the transition in the metallic (nongapped) regime to zero in the Mott phase (since there is a gap). Since the velocity is not renormalized, it means, using eq 10, that the charge compressibility goes to a constant at the transition and then drops discontinuously to zero inside the Mott phase. Note that the Mott transition involves *only* the charge degrees of freedom in one dimension, and thus, *all* spin properties (susceptibility, etc.) are totally unaffected by the transition. This means that the transport properties can vary wildly while at the same time the spin susceptibility is practically unaffected, a behavior indeed observed.[7] Spectral functions in the Mott insulator can be worked out.[71,72] A summary of the critical properties of the Mott transition is given in Figure 13.

Since the Bechgaard salts are commensurate systems I will not comment in details on the physics of the doped system. A possible way to tackle this problem is provided by the Luther-Emery solution.[66,73−76] Other methods can also be used with success.[77,78] To study the transition as a function of doping (Mott-δ transition),[79] it is useful to map the sine-Gordon Hamiltonian $H_0 + H_{1/2n}$ to a spinless fermion model (known as massive Thiring model[25,26]), describing the charge excitations (solitons) of the sine-gordon model. The remarkable fact is that close to the Mott-δ transition the solitons become noninteracting, and one is simply led to a simple semiconductor picture of two bands separated by a gap (see Figure 14). The Mott-δ transition is of the commensurate−incommensurate type.[75,76,80,81]

This image has to be used with caution since the solitons are only noninteracting for infinitesimal doping (or for a very special value of the initial

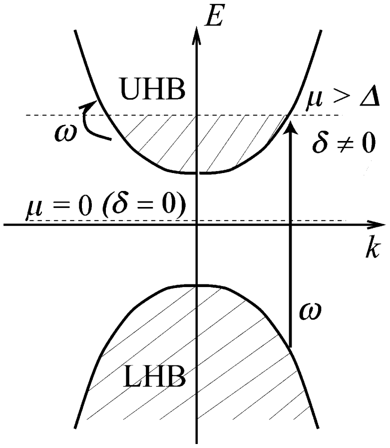

Figure 14. The repulsion splits the band into two "bands" known as lower Hubbard band (LHB) and upper Hubbard band (UHB). This concept can be made rigorous in one dimension by mapping the full interacting system to a massive Thiring model. The excitations in these bands are solitons (kinks) of charge along the chains. For a commensurate system, the chemical potential is between the two bands $\mu = 0$ and the system is a Mott insulator. A finite doping, the chemical potential is on one of the bands are charge carrying excitations exist. Optical transitions can be made either within or between the two "bands".

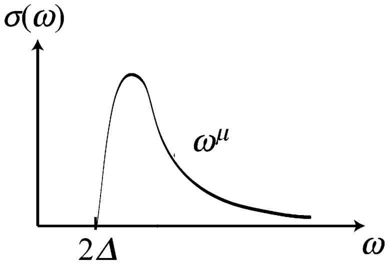

Figure 15. ac conductivity for $\delta = 0$ for a commensurability of order n. Δ is the Mott gap. The full line is the conductivity in the Mott insulator. Above the optical gap (twice the thermodynamic one Δ), the conductivity decays as a power law with an exponent characteristic of the Luttinger liquid behavior. A simple band insulator would give ω^{-3}. For a Luttinger liquid $\mu = 4n^2K_\rho - 5$ where n is the order of commensurability.

interaction) and has to be supplemented by other techniques.[79] Nevertheless, it provides a very appealing description of the excitations and a good guide to understand the phase diagram and transport properties. The universal (independent of the interactions) value of the exponents $K_c^\delta = 1/(2n^2)$ is half of the one of the Mott-U transition. Since at the Mott-δ transition the chemical potential is at the bottom of a band, the velocity goes to zero with doping. This leads to a continuous vanishing of the Drude weight and a divergent compressibility. The dynamical exponent is now $z = 2$. For more details see refs 67, 68, 79, and 82.

Of course, the Mott transition has drastic consequences on the transport properties. In a Luttinger liquid, one can thus expect quite drastically different properties than in Fermi liquid and thus to use the transport as a probe of the Luttinger properties.[67,79,83-86] The ac conductivity (at $T = 0$) for $\delta = 0$ is shown in Figure 15. In the Mott insulator, σ is zero until ω can make transitions between the lower Hubbard band (LHB) and the upper Hubbard band (UHB). For frequencies larger than the Mott gap, interactions dress the umklapps and give a nonuniversal (i.e., interaction-dependent) power-law-like decay. Such a power law can be described by renormalization group calculations[79] or by form factors calculations.[84] If one ignores the renormalization of K_ρ by the umklapp (for the effect of the renormalization of K_ρ see ref 79) one gets for the ac conductivity, for frequencies larger than the Mott gap

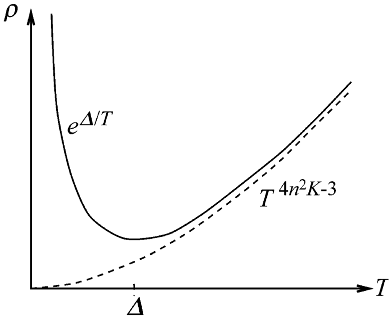

Figure 16. dc conductivity as a function of T. Δ is the Mott gap. Above the Mott gap, the dc transport shows an exponent characteristic of the Luttinger liquid and simply relate to the one in the optical conductivity. Below the Mott gap, the number of carriers is exponentially small, and any scattering will give an exponentially small conductivity (see text). This is the Mott insulating regime.

$$\sigma(\omega) \sim \omega^{4n^2K_\rho - 5} \tag{25}$$

where n is the order of commensurability.

Away from commensurate filling ($\delta \neq 0$), features above the Mott gap are unchanged (the system has no way to know it is or not at half filling at high frequencies). The two new features are a Drude peak with a weight proportional to δ/Δ, and an ω^3 absorption[67] at small frequency (see, e.g., Figure 35 in ref 7). Features above the Mott gap come from inter (hubbard)-band transitions, whereas they come from intra-band processes below the Mott gap (see Figure 14). The dc conductivity can be computed by the same methods[69,86] and is shown in Figure 16.

Here again the dressing of umklapps by the other interactions results in a nonuniversal power law dependence. For temperatures larger than the Mott gap and for the same approximations than for eq 25, one obtains

$$\rho(T) \sim T^{4n^2K_\rho - 3} \tag{26}$$

Note that close to the transition itself one recovers universal exponents: at the Mott-U transition, one has $\rho(T) \sim T/\ln(1/T)^2$ and $\sigma(\omega) \sim 1/(\omega \ln(\omega)^2)$, whereas at the Mott-$\delta$ transition, due to the different K_c, one expects $\rho(T) \sim 1/T$.

All of this results are completely general and apply to any one-dimensional systems for which Δ is smaller than the scale above which all interactions can be treated perturbatively (typically U), a situation that covers most of the experimentally relevant cases for the organics (see Section 5). These nonuniversal power law temperature and frequency dependence are quite different from what happens in a Fermi liquid and can thus be used to directly show the existence of the Luttinger liquid/one-dimensional

Mott insulator nature of the system. The transport also allows a direct access to the Luttinger liquid parameter K_ρ.

4. Interchain Coupling

Now that we have a good understanding of the properties of a single electronic chain, we can investigate how the one-dimensional physics is changed when one goes from a purely one-dimensional system to a two- or three-dimensional situation. This is of course very relevant to the physics of quasi-one-dimensional systems such as the organics, which contrary to the nanoscopic realizations of one-dimensional systems are made of a large number of such coupled chains.

The most important term describing the coupling between the chains is the interchain tunneling traducing, the fact that single electrons are able to hop from one chain to the next

$$H_\perp = -\int \mathrm{d}x \sum_{\langle\mu,\nu\rangle} t_{\perp,\mu,\nu}[\psi^\dagger_\mu(x)\psi_\nu(x) + \mathrm{hc}] \qquad (27)$$

where $\langle\mu,\nu\rangle$ denotes a pair of chains, and $t_{\perp,\mu,\nu}$ is the hopping integral between these two chains. These hopping integrals are of course directly determined by the overlap of the orbitals of the various chains. In addition to the single particle hopping, there are of course also direct interaction terms between the chains. Such terms can be density−density or spin−spin exchange. However, they are easy to treat using mean field approximation. For example a spin−spin term $S_\mu S_\nu$ can be viewed, in a mean field approximation, as an effective "classical" field acting on chain ν: $S_\mu S_\nu \rightarrow \langle S_\mu\rangle S_\nu$. Thus, at least for an infinite number of chains for which one could expect a mean field approach to be qualitatively correct, the physics of such a term is transparent: it pushes the system to an ordered state. For the fermionic single-particle hopping (27), no such mean field description is possible since a single fermion operator has no classical limit. It is thus impossible to approximate $\psi^\dagger_\mu(x)\psi_\nu(x)$ as $\langle\psi^\dagger_\mu(x)\rangle\psi_\nu(x)$, which makes the solution of the problem of coupled chains quite complicated.

This extremely difficult problem is relevant to a large variety of experimental systems, for which there is a strong anisotropy in the hopping integrals. This includes for example high-Tc superconductors (see, e.g., ref 87). Of course, for more isotropic systems such that $t_\perp \sim t_\|$ (where $t_\|$ is the intrachain hopping), one is in a high-dimensional situation to start with, and one should consider the effect of the interactions in this high dimensional situation (weak interactions will most likely lead back to a Fermi liquid state). I consider here the other limit where $t_\perp$ is much smaller than the intrachain characteristic energy scales (e.g., the kinetic energy or the interactions). In that case, the chains have a well-defined Luttinger liquid regime before the processes due to interchain hopping can spoil the pure one-dimensional physics. This is summarized in Figure 17.

4.1. Dimensional Crossover

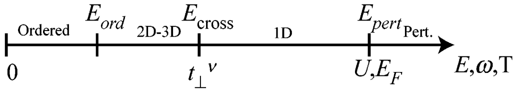

Figure 17. Separation of energy scales if the interchain hopping $t_\perp$ is much smaller than the intrachain one $t_\|$. At energies larger than the intrachain hopping (or equivalently the Fermi energy E_F) and interactions (denoted generically U), simple perturbation theory is valid. Below this scale, the system is in a one-dimensional regime. The interchain hopping couples the chains at an energy E_cr and destroys the one-dimensional physics. For noninteracting particles $E_\mathrm{cr} \sim t_\perp$ but this scale is renormalized by interactions into $t_\perp^\nu$ in a LL. In the coupled chains (two- or three-dimensional) regime, the system can have a transition to an ordered state at an energy E_0. If the one-dimensional crossover takes place before this transition should be described from the two- or three- dimensional interacting theory.

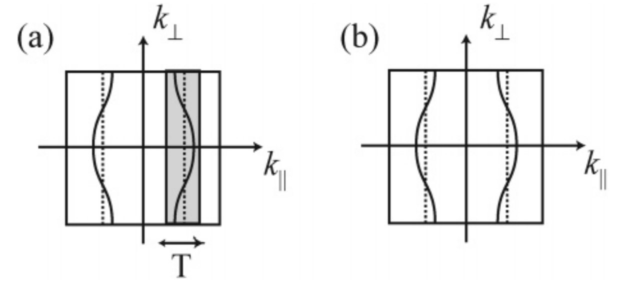

Figure 18. (a) If the temperature (or any other external energy scale) is larger than the warping of the Fermi surface due to interchain hopping, the system cannot feel the warping. It is thus behaving as a one-dimensional system. (b) At a lower temperature/energy, the system feels the two (or three) dimensional nature of the dispersion and thus behaves as a full two (or three) dimensional system. There is thus a dimensional crossover as the temperature/energy is lowered.

The first effect of the single particle hopping is thus to induce a dimensional crossover between a one-dimensional situation and a higher dimensional one. Understanding this dimensional crossover is of course one of the key questions of the physics of quasi-one-dimensional systems. In the absence of interactions, such dimensional crossover is easy to understand. In Fourier space, the kinetic energy becomes

$$\epsilon(k_\|, k_\perp) = -t_\| \cos(k_\| a) - t_\perp \cos(k_\perp b) \qquad (28)$$

where b denotes a perpendicular direction. If the perpendicular hopping $t_\perp$ is much smaller than the parallel one $t_\|$, then eq 28 leads to the open Fermi surface of Figure 18.

We can see already on this simple example the effect of the interchain hopping. If one is at an energy scale (let us say, e.g., at a given temperature T) larger than the warping of the Fermi surface, then the warping is washed out. In that case the system is indistinguishable from one with a flat Fermi surface. It can thus be considered as a one-dimensional system. There is no coherent hopping between the chains. On the other hand, if the temperature is much smaller than the warping of the Fermi surface, all correlation functions are sensitive to the presence of the warping, and the system is two- or three-dimensional. Since we considered free electrons in the above example, this crossover occurs at an energy

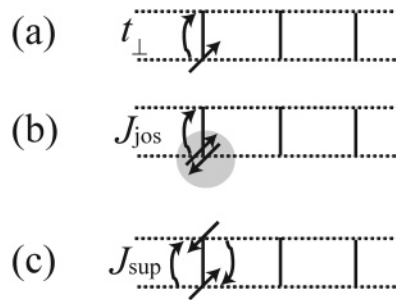

Figure 19. (a) The single-particle hopping is renormalized by interactions since a particle that hops leaves behind particles (or holes) with which it interacts. The single-particle hopping generates at second-order particle−particle (pair) hopping (Josephson coupling) shown in (b) and particle−hole (density−density interaction and superexchange) shown in (c). These couplings can become relevant even if the single-particle hopping itself is irrelevant.

scale of the order of the interchain hopping. This is summarized in Figure 18.

An important question is how the interactions change this dimensional crossover. This can be determined by looking at the renormalization of the interchain hopping.[88−94] If one neglects the renormalization of ζ, the single particle exponent (19), by the interchain hopping, then one has[89]

$$E^* \sim W\left(\frac{t_\perp}{W}\right)^{1/1-\zeta} \tag{29}$$

For the noninteracting case $\zeta = 0$ and one recovers $E^* \sim t_\perp$. Since ζ for an interacting system is always positive (see section 2.2.1), we see that the scale at which the dimensional crossover takes place is always *smaller* than for free fermions. Interactions thus tend to make the system more one-dimensional. This reduction of the crossover scale comes of course from the fact that in a Luttinger liquid *single particle excitations* are strongly suppressed. Since the objects hopping are fermions and *not* the excitations leaving in the one-dimensional chains (spinons and holons, see section 2.2) one has to "recombine" the elementary excitations to create a particle able to hop. Such processes can easily be visualized if the interactions are attractive, in that case, an electron that tries to hop leaves behind a particle to which it was attracted. This makes the hopping more difficult. With repulsive interactions, it is the same mechanism, the attraction being between an electron and a hole rather than between two electrons. This is shown in Figure 19.

One could naively conclude from the previous analysis that if the LL parameter K_ρ is such that one can reach $\zeta > 1$ then eq 29 shows that $t_\perp$ is an irrelevant perturbation and the system would remain one-dimensional as the temperature is lowered. This is perfectly true as far as *single-particle* hopping is concerned, but it is not the end of the story. Indeed among the terms that are generated at second order in the hopping between chains, one find pair hopping (Josephson coupling) and spin−spin and density−density couplings (particle−hole hopping). These terms are shown in Figure 19. Since they scale differently than single particle hopping, they can be relevant even if the single particle hopping itself is not. Let me illustrate it with the case of attractive interactions. In that case, it is perfectly possible and now much better for the particles to jump in pairs. Although this process is much smaller to start with since it is second order in $t_\perp$, it is relevant. This would remain perfectly true even if there is a gap in the proper sector, for example, a spin gap caused by attractive interactions since it will not prevent (in fact it helps) the Josephson term from becoming relevant. For repulsive interactions, one has an identical mechanism. It corresponds to particle−hole hopping. It can easily be recognized as either a density−density coupling or a spin−spin exchange (superexchange) term of the form

$$\sum_{\langle\mu,\nu\rangle}\int \mathrm{d}x\ [\vec{S}_\mu(x)\cdot\vec{S}_\nu(x) + \rho_\mu(x)\rho_\nu(x)] \tag{30}$$

These processes would lead to an ordered state. For example for a Josephson coupling J, the transition temperature to the ordered state (here a superconducting phase) is[95−97]

$$T_{\mathrm{x}2} \simeq W\left(\frac{J}{W}\right)^{1/(2-1/K_\rho)} \tag{31}$$

We thus have two different phenomena taking place.[88−94] On one hand, the single-particle hopping induces a dimensional crossover at an energy scale determined by eq 29, which I call from now on T_{x1}. On the other hand, the interchain hopping generates couplings that want to induce an order state at a temperature T_{x2}. We only know how to compute T_{x2} by the above method if the chains stay in the one-dimensional limit, that is, if $T_{x2} > T_{x1}$. Otherwise, if the dimensional crossover happens first, the transition to an ordered state should be computed from the proper two- or three-dimensional limit and the above result for T_{x2} has no meaning any more. Usually, these two-particle processes being of order $t_\perp{}^2$ are much smaller than the single-particle hopping and thus one is dominated by the single-particle hopping. The system has a dimensional crossover way before these processes can play a role ($T_{x1} > T_{x2}$). However, this depends on the strength of interactions as shown in Figure 20.

This is the difficult case since we now flow to a two- or three-dimensional system that is still in the "normal state". Since in such a system the interactions can still be strong, the resulting low-dimensional phase is a complicated problem. Even if it is a Fermi liquid, since this Fermi liquid stems from the high temperature non-Fermi liquid phase, its features are certainly quite special. In particular, the quasiparticle residue Z and lifetime of the quasiparticles could in principle retain the memory of the strong correlations that existed in the one-dimensional phase. More importantly, since the strength of the hopping depends on the transverse momentum $k_\perp$, these quantities could be varying on the Fermi surface and lead to the presence of hot spots.[98] Determining the characteristics of the resulting low energy phase is thus a major challenge. Note that

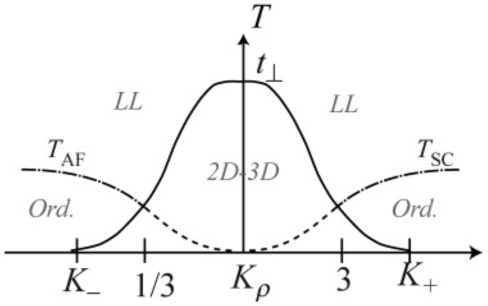

Figure 20. Crossover temperatures. For small $t_\perp$ and not too strong interactions the crossover due to single-particle hopping (full line) occurs first. The system thus stops to be one-dimensional before being able to order. If the interactions are strong enough, the single particle hopping is weakened and can even become irrelevant both for attractive ($K > K_+$) or repulsive enough ($K < K_-$) interactions. However, single particle hopping always generates a two particle (or particle−hole) hopping that drives the system to an ordered state. For repulsive interactions, the system becomes antiferromagnetic at the temperature T_{af} and for attractive ones, superconducting at the temperature T_{sc}. (After ref 94.)

the RG analysis is quite useless in this respect since the flow goes to strong coupling so one needs other techniques below T_{x1}. Various methods have been used such as RPA,[99] and more recently a mean field approach (ch-DMFT) treating the chains as an effective bath.[100−104] Note that I mostly talked here about temperature, but any energy scale will do. In particular, if one studies optical conductivity, the frequency also allows to scan through the dimensional crossover.

4.2. Deconfinement Transition

The effects of interactions that lead to the reduction of the dimensional crossover scale in a Luttinger liquid are in fact much more drastic in the case where the isolated chains are commensurate and in a Mott insulating state. Indeed, if each chain develops a gap, either, for example, by having a spin gap (e.g., for attractive interactions) or in the charge part (e.g., for commensurate filling by being a Mott insulator), it means that the single-particle Green's function decays *exponentially*. The single-particle hopping is now an irrelevant variable. Physically this means that a single particle cannot hop from one chain to the other without breaking a pair. The formation of a gap is thus in direct competition with the interchain hopping. Of course, by increasing the interchain hopping to a critical value, one can break the one-dimensional Mott gap and drive the system to a gapless higher dimensional state (in general a metallic one). Thus, the simple dimensional crossover is now replaced by a quantum phase transition for a critical value of the interchain hopping, where the system goes from a one-dimensional Mott insulator to a (in general) three-dimensional metal. Such a transition, known as a deconfinement transition since the electrons are able at the same time to leave the chains and to conduct, is of course more complex to study. Quite interestingly such a situation is the generic situation for the organic conductors such as the Bechgaard salts due to their commensurate nature.

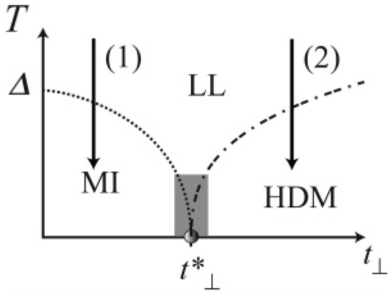

Figure 21. Schematic representation of the deconfinement. This is a quantum phase transition that takes place at $T = 0$ as a function of the interchain hopping. Note that, although the gap Δ corresponding to activation in the dc conductivity decreases, the gap observed in the ac conductivity remains finite even for $t_\perp > t_\perp^*$, so the precise critical behavior is still to be understood. At finite temperature, the presence of this quantum critical point leads to various crossovers between a Mott insulator (MI), Luttinger liquid (LL), and high dimensional metallic (HDM) phase. These two crossovers can be observed in different materials by changing the temperature as indicated by the two arrows (1) and (2) (see text). Whether the HDM phase is a Fermi liquid and what are its properties is one of the main questions.

A schematic phase diagram of this transition is shown in Figure 21. Compared to the case of the dimensional crossover, much less is known about such a deconfinement transition. It has been studied by simple scaling arguments,[69] study of two chain systems[105,106,107,108] or using approximations such as RPA[99] or ch-DMFT methods.[103] A rule of thumb to get the position of this deconfinement transition is to compare the two scales T_{x1} and Δ. Thus, roughly if $T_{x1} > \Delta$, one is deconfined, whereas for $T_{x1} < \Delta$, the gap wins and the chains are confined, only allowing for two particles hopping. Of course, this is only a rule of thumb and one should, in principle, solve the full coupled problem to obtain the critical value $t_\perp^*$ at which deconfinement occurs. For systems with $t_\perp < t_\perp^*$, the single particle hopping is irrelevant and as a function of temperature one observes a crossover between a LL at high temperature and a Mott insulator at low temperatures (trajectory (1) in Figure 21). For systems with $t_\perp > t_\perp^*$, the single particle hopping is relevant and the system is deconfined. As a function of the temperature (trajectory (2) in Figure 21), one now observes a dimensional crossover between a one-dimensional LL and a high dimensional metallic phase.

Besides the phase diagram, it is difficult to extract the physical properties in the deconfined phase. The transverse conductivity can be computed in the high-temperature high-frequency regime by an expansion in the perpendicular hopping.[101] One finds a power-law either in frequency of temperature, controlled by the single particle Green's function exponent. At finite temperatures $\mathcal{R}\sigma_\perp(T \gg \omega) \propto T^{2\zeta-1}$ for $kT \gg E^*$, whereas at high-frequency ($\hbar\omega \gg E^*$), one gets

$$\sigma_\perp(\omega \gg T) \propto \omega^{2\zeta-1} \tag{32}$$

E^* is the scale at which this expansion breaks down (roughly the dotted and dashed lines in Figure 21). The Hall effect is much more difficult to obtain and so far could only be obtained either at very high fields or in the absence of scattering along the chains[109,110] or for phenomenological models.[111,112]

The shape of the Fermi surface and quasiparticle properties in the deconfined phase still cannot be determined unambiguously. The RPA approximation[99] shows pockets close to the deconfinement transition, whereas the more sophisticated ch-DMFT approach[103] gives an open Fermi surface quite similar to the naive tight binding one, although the presence of hot spots at the transition is possible.[113] This important question is thus still largely left open.

5. Application to Bechgaard and Fabre Salts

Let us now look how these concepts can be applied and help to understand the physics of real quasi-one-dimensional compounds. I will focuss here on the physics of the Bechgaard (TMTSF) and Fabre (TMTTF) salts. It is of course not my goal to make in this chapter an experimental review of this family nor to touch the whole cornucopia of physics that they brought. I refer the reader to refs 114−116 and the other papers of this volume, in particular ref 7 for a complete description of these systems and in particular for their structure and basic properties. I will mostly concentrate here on the properties of the "normal" phase, without touching the various ordered phase that occur in these systems.

These one-dimensional systems share many common features. In particular, they are very good realization of quasi-one-dimensional systems with hopping integrals of the order of $t_a = 3000K$, $t_b = 300K$, $t_c = 20K$, leading to relatively well separated energy scales in which one is indeed dominated by the intrachain hopping. Experimentally, at ambient pressure, the $(TMTTF)_2PF_6$ compound displays an insulating behavior (MI). A transition to a metallic phase is found, with increasing pressure and the properties of the TMTTF compounds evolve toward those of the compounds of the TMTSF family, which are good conductors. This evolution is clear[117] from the a-axis resistivity measurements (see e.g., Figure 23 of ref 7). Such an insulating behavior is well consistent with what one would expect for a one-dimensional Mott insulator (see section 3). The minimum of the resistivity (followed by an activated law as temperature is lowered) defines the onset of the MI regime on Figure 21. Since the chains are dimerized, with a small dimerization of the order of $\Delta_d \sim 100K$, such a Mott insulating behavior could come either from the half-filled nature of the band (because of the dimerization) or if the interactions are large enough from the quarter-filled nature of the band (see section 3).

It is thus clear that the interactions play a crucial role in the TMTTF family even at relatively high energies. For the TMTSF, the question is more subtle in view of the metallic behavior at ambient pressure and it was even suggested that such compounds could be described by a FL behavior with weak interactions.[118] The TMTTF and TMTSF family thus prompts for very fundamental questions in connection with one-dimensional physics:

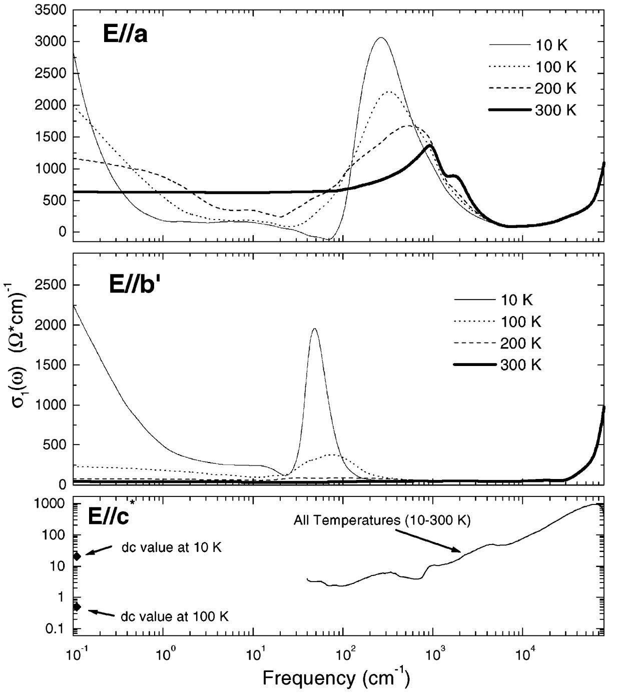

Figure 22. Optical conductivity along the three axis for $TMTSF_2(ClO_4)$, at various temperatures. Although such compound seems to have a rather "standard" dc conductivity, all of the dc transport is in fact due to a very narrow Drude peak containing only 1% of the spectral weight, whereas 99% of the spectral weight is above an energy gap (of the order of 200 cm^{-1}). This is reminiscent of a Mott insulating structure. (From ref 121; Copyright 1999 by Springer.)

1. Are interactions also important in the TMTSF family or can the compounds of this family simply be regarded as a Fermi liquid with an anisotropic Fermi surface?

2. If indeed interactions are important, what is their strength and can one identify Luttinger liquid or Mott insulating behavior? What are the Luttinger parameters?

3. If the system is a Mott insulator, is this mostly due to the dimerization of the band or is the quarter filling commensurability sufficient?

4. What are the reason for such a difference between the very close families TMTTF and TMTSF, for which the various characteristics (bandwidth, dimerization, and interactions) vary relatively little?

5. What is the dimensional crossover scale at which the system stops to be essentially a one-dimensional system? What are the properties of the phase below this crossover scale? Of course many other questions are open, such as the mechanism and nature of superconductivity in these materials, on which I will not touch here.

5.1. Interactions and LL/MI Physics

A clear proof of the importance of interactions for *both* the TF and SF compounds is provided by the optical conductivity,[119,120] as shown in Figure 22. The optical conductivity clearly shows a decreasing gap (of the order of 2000 cm^{-1} for the $TMTTF_2(PF_6)$ to 200 cm^{-1} for $TMTSF_2(PF_6)$. Nearly (99%) of the spectral weight is in this high-energy structure. In

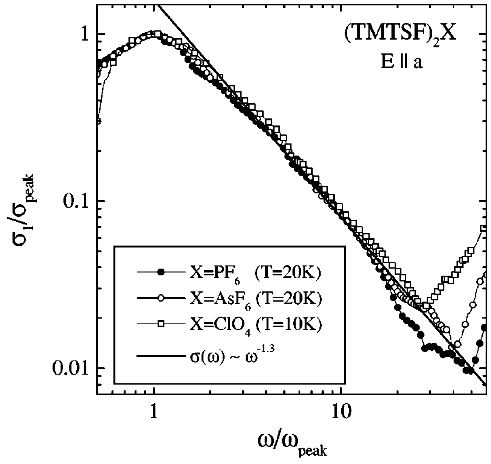

Figure 23. Optical conductivity along the chain axis in the TMTSF family. The conductivity is rescaled by the gap in various samples. A fit of the ω dependence of the conductivity above the gap is well consistent with Luttinger liquid behavior. (From ref 120; Copyright 1998 by the American Physical Society.)

the metallic compounds, there is in addition a very narrow Drude peak. This clearly indicates that these compounds are very far from simple Fermi liquids. The data of optical conductivity can be compared with the expectations for a one-dimensional Mott insulator (see Figure 15) as shown in Figure 23.

The data above the gap fits the power law LL behavior very well and thus shows quite convincingly that these compounds are indeed well described by a LL theory down to a scale of a few hundred Kelvin (temperature or frequency). This was the first direct proof of a Luttinger liquid behavior in an electronic system. The a-axis optical measurements described above even allow for a quantitative determination[120] of the LL parameter K_ρ. A fit of the frequency dependence of the longitudinal conductivity (see Figure 23) can be performed using eq 25. A commensurability of order one ($n = 1$) does not allow for a consistent fit of both the exponent and the gap.[120] This indicates that the dominant umklapp comes from the quarter filled nature of the band. Formula 25 with $n = 2$ thus yields $K_\rho \simeq 0.23$, indicating quite strong electron−electron interactions. Moreover, this indicates that the finite range nature of the interactions should be taken into account, with interactions extending at least to nearest neighbors. A modelization of the organics thus should not be done with a purely local Hubbard model, with an interaction U, but also take into account at least the nearest neighbor interaction V. This estimate of the LL parameter agrees reasonably well with measurements of the longitudinal resistivity in the range 100−300 K.[115] The optical data is thus consistent with an interpretation of the insulating state as a quarter-filled Mott insulator, suggesting that the dimerization plays a little role at least in the TMTSF family. In the TMTTF family, dimerization is larger, and it is unclear there which process is dominant. Note that because of the anions other transitions can exist such as a ferroelectric transition.[122,123]

These results are in good agreement with other measurements. The measured values of K_ρ are consistent with the photoemission data.[117,124,125] However, photoemission in these system should be looked at with a grain of salt because of possible surface problems due to the ionic nature of the system (for other systems see, e.g., refs 126 and 127). The optical data along the c axis[121] is consistent with the predictions of LL. These measurements directly probe the density of excited states in the $a-b$ plane. However, clearly the data along the c axis is of poor quality compared to the two other axes, so new measurements along this axis would clearly be needed. Note also that along the b axis the big bump in mid-spectra is yet unexplained.

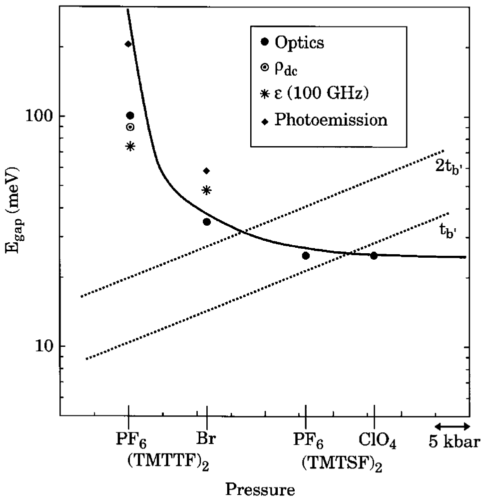

Figure 24. Comparison of the measured gap in the optical conductivity with the interchain hopping. The change of behavior from insulator to metallic occurs when the two quantities are of the same order of magnitude showing that the difference between the various members of the TM families is indeed linked to a deconfinement transition. (From ref 117; Copyright 2000 by Springer.)

5.2. Deconfinement

These compounds are of course excellent systems to study the deconfinement transition, as well as the associated dimensional crossovers discussed in the previous section (see Figure 21). Since the isolated chains would be insulators, the system will thus crossover from a regime where one has essentially uncoupled (insulating) chains to one of metallic planes. A direct evidence that the change of behavior between the insulating and metallic regimes is due to such deconfinement transition[69] is provided by the optical data (see Figure 24). A measure of the gap extracted from the optical conductivity shows that the change of nature occurs when the observed gap is roughly of the order of the magnitude of the interchain hopping.[128]

For systems that are deconfined, a change in temperature leads to a dimensional crossover between a high-temperature one-dimensional phase, in which hopping between the chains is incoherent, to a low-temperature two-dimensional metallic phase, where the chains are coupled along the b axis (intermediate axis perpendicular to the chains; see Figure 21). This dimensional crossover can be seen by the appearance of a Drude peak in the b axis conductivity, when temperature is lowered, signaling coherent transport along the *b* axis. Such a behavior would be consistent with the observed optical conductivity along the *b* direction as shown in Figure 22, but obviously, more data and data on other materials would be needed to clarify this complex phenomenon. Transport along the *c*-axis would be also be consistent with eq 32. Note however that, although clearly revealing that electrons are confined in the chains above ∼100 K, the measurements of dc transport along the *c* axis[129] are not yet fully understood theoretically from a LL picture and most likely need to take into account the Mott nature of the system (see ref 101 for a discussion). From these experiments, we see that the crossover takes place around 100 K in $(TMTSF)_2PF_6$. This also is in agreement with the change of behavior from T (LL behavior) to T^2 observed in dc transport along the *a* axis.[115,7]

The physics below the dimensional crossover scale is still a mystery. Indeed, although the system is not one-dimensional any more, the NMR is quite anomalous.[53,130] Whether this is simply due to antiferromagnetic fluctuations or whether this is a more profound sign of a nonfermi liquid behavior is still an open issue.

6. Conclusions, Theoretical Dreams, and Chemical Realities

I have presented in this review the main concepts and questions relevant to tackle the normal phase physics of quasi-one-dimensional systems. The most important ones for isolated chains are the Luttinger liquid theory and the Mott insulating physics which is quite special in one dimension. For quasi-one-dimensional systems, an extremely rich physics stems from the coupling between the chains. Its most spectacular expression is the presence of a deconfinement transition between a one-dimensional insulator and a high dimensional metal.

The organic conductors, such as the Bechgaard and Fabre salts, provide wonderful systems to investigate these phenomena. Some agreement between the theoretical studies and the experimental observations can be reached even if many open questions remain. In particular the role of the quarter filling of the band seems crucial in these systems, and determines the physics of the whole metallic phase. However, the Bechgaard salts are in some sense too interesting materials, with too many different phenomena occurring at once. To better understand these phenomena, it would be good to have other compounds in which some degree of simplification or new physics occurs. This is where the interplay between chemistry and the theoretical concepts explained here can play a major role. Now that we understand better the main points and questions in the physics of the Bechgaard salts, we can examine three main directions in which having new compounds would drastically help the theoretical understanding of these materials. So I would like to conclude this review with a plea for new materials.

6.1. Non-Dimerized Systems

One complication of the Bechgaard salts is the presence of the dimerization. As we saw in section 3, this introduces two umklapps corresponding to the half- and quarter-filling of the band and makes it hard to separate the two effects. Having a compound without dimerization is thus of the utmost importance. In such a compound the band is purely quarter filled. It is thus clear in that case that if the compound becomes a Mott insulator the underlying mechanism can only come from the quarter-filling umklapp and thus be caused by taking into account at least nearest neighbor interactions as explained in section 3.

In fact some of such compounds have been synthesized[131−134] and turn out to be (Mott) insulators. The fact that these compounds are indeed insulators and with a structure similar to the Bechgaard salts strongly confirms the interpretation that the dominant mechanism is also in these systems the quarter filling of the band. It would be of course very interesting to investigate the phase diagram and the physical properties under pressure of these compounds. Since they share the same basis microscopic features, it is crucial to assert whether these quarter filled systems also exhibit superconductivity under pressure as in the Bechgaard salts.

6.2. Interchain Coupling

As we saw in section 4, the interchain coupling plays a crucial role in regard to the deconfinement transition. In the Bechgaard salts, given the hierarchy of the interchain couplings 3000K, 300K, 30K, a complication occurs. When the system stops to be one-dimensional at about 100 K it enters a two-dimensional regime. As we discussed in section 5, it is unclear whether such a state is a canonical Fermi liquid or not. On the other hand, at temperatures of about 20 K, a more conventional three-dimensional ordering is observed. To disentangle these effects, it would be very interesting to have a family of compounds with one or both of the following properties:

1. Smaller interchain hopping: This would allow us to have a broader range where a one-dimensional behavior could be observed. Some compounds have been synthesized with a very small interchain hopping, but because of the commensurate filling, they are just one-dimensional Mott insulators. There should thus be the proper balance between the interchain hopping and smaller intramolecular interactions, to weaken also the Mott mechanism. This most certainly means using other building blocks than the TMTTF or TMTSF molecules.

2. More isotropic interchain hopping (even more interesting): Having such a compound would make a direct transition between a one-dimensional system and a three-dimensional one. This would allow us to decide whether some of the low temperature properties such as the anomalous NMR and even the superconductivity are related to the existence of the two-dimensional structure or not. With the Bechgaard salts, it is of course difficult because of the presence of the anions, so a different and more isotropic construction of the crystal would probably be needed.

6.3. Mott Gap and Doping

Of course it would be interesting to play with the Mott gap. As we saw in section 3, the Mott gap is mostly controlled by the on-site and nearest neighbor interactions. Thus, changing the local repulsion probably means changing the building block of the one-dimensional salts, a difficult feat. If this could be achieved, this would be extremely interesting since it would allow us to observe the Luttinger liquid behavior on a broader energy scale. One could thus probe the dimensional crossover rather than the deconfinement transition.

Even more interesting would, of course, be the ability to *dope* the compound. This is a difficult feat chemically and has been attempted several times, with no success so far. This is a Holy Grail. Note that, because the charge transfer between the cations an the chains is total, the full phase diagram so far corresponds only to the commensurate filling. Since the advent of high Tc superconductors, that are doped Mott insulators, there has been a considerable interest in understanding such problems. Indeed one of the important question is whether the doping of a Mott insulator could be good for superconductivity, as some theoretical studies indicate. Being able to reach a similar situation in a one-dimensional geometry (rather than a two-dimensional one) would of course be of considerable importance. This would allow us to better understand the similarities and the differences of the two systems. Would the superconductivity that appears under pressure be also present under doping?

Of course many other directions are possible. The ball in this field is clearly now in the chemist's camp. Physical measurements and theories have made enormous progress. Most of the simple questions have been explored, and the remaining ones are extremely difficult and central to many domains of strongly correlated systems. Only new materials can help unlocking further the richness of these quasi-one-dimensional materials. Of course many of the proposals above are most likely theoretical dreams. Some of them have already been partly explored, some will probably be quite tough to do, but all of them are definitely worthwhile trying. Who knows, in this quest for new materials, another unexpected piece of physics might emerge.

7. Acknowledgments

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