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Lock-in transitions and charge transfer in one-dimensional fermion systems

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Abstract. The one-dimensional quantum sine–Gordon Hamiltonian with a density ρ_s of solitons is studied. $\rho_s(\mu)$ is the order parameter for the lock-in transition, which happens when the chemical potential μ equals the single soliton energy and temperature is $T = 0$. The soliton density $\rho_s(\mu, T)$ and the critical behaviour at $T = 0$ are studied in the classical limit (quantum coupling $\beta \rightarrow 0$) and in the quantum system with $\beta^2 = 4\pi$; the critical exponent for ρ_s is 0 and $\frac{1}{2}$ respectively. An important application of these results is the temperature dependence of an incommensurate charge transfer ρ_e in one-dimensional conductors due to Umklapp scattering. $\rho_e(T)$ can be determined by the function $\rho_s(\mu, T)$.

1. Introduction

Incommensurate periodicities have recently been observed in many physical systems and have become the subject of considerable theoretical interest. Examples of such systems are: (i) charge density waves (CDW) incommensurate with the underlying lattice (Friend 1978); (ii) exotic crystals with incommensurate sublattices, such as some TTF based salts (Wudl *et al* 1977), some superionic conductors (Beyeler 1976) and $Hg_{2.86}AsF_6$ (Pouget *et al* 1978); (iii) some ferroelectric crystals (Ishibashi 1978); (iv) in the process of some martensitic phase transitions the nucleating clusters are incommensurate with the surrounding matrix; e.g. the ω phase problem (Lin *et al* 1976, Horovitz *et al* 1978); (v) vortex lines in superconducting film pinned by thickness modulation (Daldini *et al* 1974); (vi) thin films which are incommensurate with the substrate (Stephens *et al* 1979 and references therein).

The lock-in transition, also known as the commensurate–incommensurate transition, determines when the interaction between two incommensurate structures can overcome their rigidity and lock them into one common periodicity.

The theory of the lock-in transition has been discussed in the context of dislocation theory (Frank and van der Merwe 1949), theory of CDW (McMillan 1976, Bak 1978, Jackson *et al* 1978, Ohmi and Yamamoto 1977, Horovitz 1980), the incommensurate lattice problem (Ying 1971, Theodorou and Rice 1977), a two-dimensional problem (Pokrovski and Talapov 1979, Bak *et al* 1979), a magnetic system (von Boehm and Bak

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1979) and the general problem of incommensurate periodicities (Bruce and Cowley 1978, Bruce 1980, Aubry 1978, Sokoloff *et al* 1978, Okwamoto *et al* 1979, Batistic and Barisic 1979). A related problem is that of the infinite vortex lattice in a Josephson junction (Kulik 1967, Lebwohl and Stephen 1976, Fetter and Stephen 1968): the phases of the two superconductors are 'incommensurate', i.e. the phase difference is never asymptotically zero (mod 2π).

In most of the physical systems the problem has been reduced to a solution of the one-dimensional (1D) sine–Gordon Hamiltonian. The level of incommensurability is measured by a soliton density $\rho_s(\mu, T)$ where μ is a chemical potential which is proportional to the discrepancy in the periodicities when the interaction is absent and T is the temperature. The lock-in transition is a transition in the ground-state properties ($T = 0$) and happens at $\mu = E_s$, where E_s is the single soliton energy. For $\mu < E_s$ $\rho_s = 0$ and the system is locked, while for $\mu > E_s$ $\rho_s \neq 0$ and the structures are incommensurate. The details of this approach are discussed in § 2.

In the present paper we examine the critical behaviour of the lock-in transition and extend both classical and quantum theories to finite temperatures. In § 3 we evaluate $\rho_s(\mu, T)$ for the classical theory by using results of the transfer matrix theory. In § 4 we consider the quantum theory with a particular coupling which can be solved by a fermion–boson transformation. The critical exponent of the lock-in transition is $\bar{\beta} = 0$ for the classical case and $\bar{\beta} = \frac{1}{2}$ in the quantum case.

In § 5 we show that an incommensurate charge transfer ρ_e in 1D conductors is temperature dependent when Umklapp scattering is present. The method is based on the fermion–boson transformation (Luther and Emery 1974, Emery *et al* 1976, Gutfreund and Klemm 1976) and we show how $\rho_e(T)$ is related to $\rho_s(\mu, T)$. The result is applied to TTF-TCNQ in its 1D regime, i.e. above its CDW instability. In this regime the CDW is a fluctuation effect, and it is not obvious *a priori* that an Umklapp process can couple and shift the electron density ρ_e. Further applications of the temperature-dependent ρ_s are considered in § 6.

2. The model

In this work we consider the sine–Gordon Hamiltonian, which is written in terms of a boson field $\varphi(x, t)$ with bare mass m^* and coupling constant β (in the classical problem β can be scaled out and is not relevant):

$$H_{SG} = \int dx[\tfrac{1}{2}\pi^2(x, t) + \tfrac{1}{2}\varphi'^2(x, t) + (m^{*2}/\beta^2)(1 - \cos\beta\varphi)] \qquad (1)$$

where $\pi = \dot{\varphi}$, and $\dot{\varphi}$, φ' are time and space derivatives respectively.

The derivation of this Hamiltonian for the physical systems mentioned in the Introduction has been done by various methods: phenomenological arguments (Frank and van der Merwe 1949), Landau type expansions (McMillan 1976), perturbation theory on a microscopic Hamiltonian (Ohmi and Yamamoto 1977, Horovitz 1980) or by the fermion–boson transformation for the 1D fermion problem (Emery *et al* 1976) (see also § 6). In order to discuss the basic ingredients of the theory, we present here the simplest 'classical' derivation.

Assume that the interaction between two incommensurate structures is dominated by a single pair of reciprocal wavevectors G_i ($i = 1, 2$) such that $G_1 - G_2 = \bar{q}$ is small, $|\bar{q}| \ll G_i$. The relevant Fourier component of each structure is proportional to $\exp(G_i x + \varphi_i)$ and a space dependence of the phase φ_i represents deviation from exact periodicity. The interaction between these structures is $\sim \cos(\varphi_1 - \varphi_2 + \bar{q}x)$ while the elastic and kinetic energies are $\varphi_i'^2$ and $\dot{\varphi}_i^2$. Energy is gained from this interaction if the phases are space dependent, and this compensates the loss from the elastic energies. Usually one of the structures is much more rigid than the other and then the problem involves a single field. In addition the coefficient of the cosine interaction may also depend on space (the coupled phase–amplitude problem (Jackson *et al* 1978, Horovitz 1980)) which is important in some cases. Here we consider just the basic problem of a single scalar field φ_1 and $\varphi_2 = 0$. Define now

$$\beta\varphi = \varphi_1 + \bar{q}x \tag{2}$$

so that the total Hamiltonian has the form ($\mu \sim \bar{q}$)

$$H = H_{\text{SG}} - \frac{\mu}{2\pi}\int \mathrm{d}x\beta\varphi'(x,t). \tag{3}$$

Thus, except for the boundary term, the problem is essentially that of solving equation (1).

The sine–Gordon Hamiltonian has been extensively studied in recent years including classical solutions (Scott *et al* 1973), classical statistical mechanics (Gupta and Sutherland 1976, Currie *et al* 1980) and the corresponding quantum theory (Dashen *et al* 1975a, b, Coleman 1977, Jackiw 1977, Maki and Takayama 1979). The basic nonlinear solution is the soliton whose boundary conditions require a change of 2π in the field $\beta\varphi(x,t)$. The soliton is a localised, stable and finite-energy object. The classical solution for two solitons (Scott *et al* 1973) shows that they repel each other and the solution must be time dependent; the same is true for any finite number of solitons. However, an infinite number of solitons form a periodic static solution which may be called the 'soliton lattice' solution (Frank and van der Merwe 1949, Gupta and Sutherland 1976). The soliton density ρ_s is determined by the boundary condition, i.e.

$$\rho_s = \frac{1}{2\pi}\int \frac{\mathrm{d}x}{L}\beta\varphi' \tag{4}$$

where L is the length of the system.

Since ρ_s is a thermodynamic variable, the parameter μ in equation (3) is the corresponding chemical potential and equation (3) is the grand canonical sine–Gordon Hamiltonian. In our problem of the incommensurate structures μ is a constant which measures the discrepancy $\bar{q}$ of the two periodicities. If E_s is the energy of a single soliton in equation (1) (in either a classical or quantum theory) then its energy in equation (3) is $E_s - \mu$. Thus for $\mu < E_s$ the ground state has $\rho_s = 0$, while for $\mu > E_s$ $\rho_s \neq 0$. This is the basic feature of the lock-in transition: if the discrepancy in periodicities is large enough to overcome the soliton energy, the ground state is a soliton lattice and the structures are incommensurate, otherwise the ground state has no solitons and the structures are locked. The soliton density ρ_s is the order parameter of the lock-in transition.

Before presenting the theory, we note that its application may not be as direct as mentioned above. The parameter μ is not necessarily a constant for all the relevant physical systems. In particular in the CDW system the periodicity is determined by the Fermi wavevector, which in turn is determined by the electronic chemical potential μ_e. Therefore $\mu \sim \mu_e$ and ρ_s measures the electron density relative to the density in the commensurate situation (Horovitz 1980). If there is an infinite electron reservoir at the energy μ_e then the electron density is ρ_s (μ = constant, T) and is temperature dependent. However in actual systems the 'reservoir' is another band and the total charge is conserved rather than the chemical potential. This point should be considered if a theoretical comparison with actual systems is attempted. In § 6 we consider this point in the case of a fluctuating CDW, i.e. in the 1D fermion problem.

3. Classical limit

The classical ground state is determined by the solutions of the sine–Gordon equation

$$-\ddot{\Psi} + \Psi'' = m^2 \sin \Psi \tag{5}$$

where $\Psi = \beta\varphi$ and $m = m^*$ in this section. The single soliton, or antisoliton, solutions $\Psi = 4 \tan^{-1} \exp(\pm mx)$ determine two important parameters: the single soliton width is $1/m$ and its energy is

$$E_s = 8m/\beta^2. \tag{6}$$

The soliton lattice solution is given by (Frank and van der Merwe 1949)

$$\sin \tfrac{1}{2}(\Psi_s - \pi) = sn(mx/k, k) \tag{7}$$

where sn is the Jacobian elliptic function (Whittaker and Watson 1962) with parameter k, $0 \leqslant k \leqslant 1$. In this solution Ψ increases by 2π as x is increased by $l = 1/\rho_s$, and

$$\rho_s = m/[2kK(k)]. \tag{8}$$

Thus ρ_s is the soliton density and is determined by the parameter k in the range $0 \leqslant \rho_s < \infty$.

The energy per unit length is

$$E_{cl}(\rho_s) = \frac{8m\rho_s}{\beta^2 k}\left[E(k) - \tfrac{1}{2}k'^2 K(k)\right] \tag{9}$$

where $K(k)$ and $E(k)$ are the complete elliptic integrals of the first and second kind and $k'^2 = 1 - k^2$. A low-density expansion gives $k' \to 0$, $\rho_s \to m/(2\ln 4/k')$ and

$$E_{cl}(\rho_s) = \frac{8m}{\beta^2}\rho_s(1 + 4\exp(-m/\rho_s)) + O(\exp(-2m/\rho_s)). \tag{10}$$

The first term represents free solitons, each with energy E_s, and the second term is the long-range interaction which is exponential with a range $1/m$.

The lock-in transition is defined by the function $\rho_s(\mu)$ where $\mu = dE_{cl}/d\rho_s$ is the chemical potential, $\mu/E_s = E(k)/k$. Near the transition $(\mu = E_s)\rho_s \sim \ln^{-1}(\mu - E_s)$, corresponding to a critical exponent of $\bar{\beta} = 0$.

The thermodynamics of the classical grand canonical sine–Gordon system has been studied by Gupta and Sutherland (1976). They show by using the transfer matrix method that the soliton pressure is determined by the eigenvalue of the Mathieu equation $a_0(\nu, Q)$ at the imaginary wavevector $\nu = -i\mu/\pi T$ and parameter $Q = E_s^2/16T^2$. Using equation (102) of Gupta and Sutherland (1976) the soliton density is

$$\rho_s = \frac{T^2}{E_s}\frac{\partial}{\partial\mu}a_0\left(\frac{-i\mu}{\pi T}\frac{E_s^2}{16T^2}\right). \tag{11}$$

From the large ν expansion (Abramowitz and Stegun 1964) we obtain

$$\rho_s = m\frac{2\mu}{\pi^2 E_s}\left(1 - \frac{E_s^4}{8^3(T^2+\mu^2/\pi^2)^2} - \frac{3E_s^8[5(\mu/\pi)^4 + 12(\mu T/\pi)^2 - 11T^4]}{8^7(T^2+\mu^2/\pi^2)^4(4T^2+\mu^2/\pi^2)^2} + \mathrm{O}(E_s/\mu)^{12}\right). \tag{12}$$

This expansion has been used as a high T expansion (Gupta and Sutherland 1976) but it is also a $1/\mu$ expansion for all temperatures.

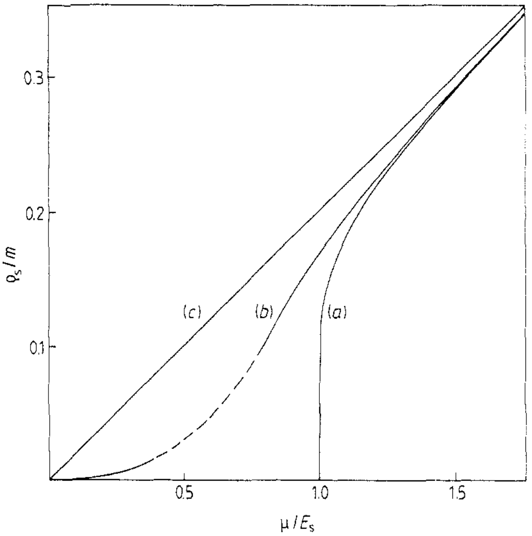

Figure 1. Soliton density in the classical limit. The finite temperature curve is from equations (12) and (13) and the broken section of the curve is an extrapolation between these equations. E_s and $1/m$ are the energy and width of the single soliton solution: (*a*) $T = 0$; (*b*) $T = E_s/8$; (*c*) $T \to \infty$.

For small μ and low temperatures ($T \ll E_s - \mu$) we can use a large Q asymptotic expansion of $a_0(\nu, Q)$ (Goldstein 1929) with the result

$$\rho_s = 8m(E_s/8\pi T)^{1/2}\exp(-E_s/T)\sinh(\mu/T). \tag{13}$$

It is well known that solitons are thermally excited at low temperatures (Currie *et al* 1980). The excitation energies for a soliton or an antisoliton are $E_s - \mu$ and $E_s + \mu$ respectively. The density ρ_s measures the difference between the soliton and antisoliton densities, and equation (13) is indeed proportional to the difference of the appropriate Boltzmann factors.

The results of this section are summarised in figure 1. For $T \neq 0$ there is no sharp transition, as expected in a 1D system. Note also that for $T \to \infty$ the potential $\cos\Psi$ has no effect and ρ_s is maximal. In the context of § 2, the structures do not feel each other in this limit and they have their natural incommensurate periods.

4. Quantum theory: $\beta^2 = 4\pi$

The critical exponent of the quantum lock-in transition is $\bar\beta = \frac{1}{2}$, in sharp contrast with the classical exponent $\bar\beta = 0$ i.e. logarithmic behaviour. The exponent $\bar\beta = \frac{1}{2}$ was first derived for the case $\beta^2 = 4\pi$ (Horovitz 1979), then extended to $4\pi \leqslant \beta^2 < 8\pi$ (Luther *et al* 1979, Pokrovski and Talapov 1979, 1980) and finally shown for all $0 < \beta^2 < 8\pi$ (Haldane 1980, Schultz 1980). Here we present in detail the generalisation of the fermion–boson transformation (Luther and Emery 1974) to the case of variable fermion density for $\beta^2 = 4\pi$, and derive the soliton density $\rho_s(\mu, T)$ as well as the exponent $\bar{\beta} = \frac{1}{2}$.

Consider a system of 'left' and 'right' moving spinless fermions in one space dimension. The dispersion of these fermions is $\pm \tilde{v}_F k$, and the kinetic part of the Hamiltonian can be written in the form (Mattis and Lieb 1965)

$$H_0 = \sum_k \tilde{v}_F k [a_1^+(k)\, a_1(k) - a_2^+(k)\, a_2(k)]$$

$$= \frac{2\pi\tilde{v}_F}{L} \sum_{k>0} [\rho_1(k)\, \rho_1(-k) + \rho_2(-k)\, \rho_2(k)] \tag{14}$$

where $\rho_i(k) = \Sigma_p a_i^+(k+p)\, a_i(p)$ and a_i $(i = 1, 2)$ are fermion annihilation operators for the two branches. Define the boson field,

$$\varphi_1(x) = \sum_{k \neq 0} \frac{2\pi \mathrm{i}}{Lk} \exp(-\tfrac{1}{2}\alpha|k| - \mathrm{i}kx)\, [\rho_1(k) + \rho_2(k)] \tag{15}$$

where $1/\alpha$ is a momentum cutoff. Note that $\varphi_1'(x)/2\pi$ is the fermion density operator, except for the $k = 0$ mode. In order to treat a non-zero average fermion density we introduce the field

$$\varphi(x) = \varphi_1(x) + \bar{q}x \tag{16}$$

and measure the fermion density ρ_e from a wavevector which is $\frac{1}{2}\bar{q}$ less than the Fermi wavevector k_F of the original system, i.e. $\rho_e = \bar{q}/2\pi$. The Hamiltonian in terms of $\varphi(x)$ is

$$H_0 = \frac{1}{8\pi\tilde{v}_F} \int \mathrm{d}x [\dot{\varphi}^2(x, t) + \tilde{v}_F^2 \varphi'^2(x, t)] \tag{17}$$

and a term $L\tilde{v}_F\bar{q}^2/8\pi$ was added, so that the energy is measured relative to a level which is $\frac{1}{2}\tilde{v}_F\bar{q}$ lower than the original Fermi level. This density dependent term is important for the generalisation of Luttinger models (Haldane 1979).

Consider now the Hamiltonian

$$H = H_0 + \Delta \sum_k [a_1^+(k)\, a_2(k - 2k_F - \bar{q}) + \text{HC}). \tag{18}$$

Using the fermion representation (Luther and Peschel 1975)

$$L^{-1/2} \sum_k \exp(\mathrm{i}kx)\, a_i(k) \equiv \Psi_i(x)$$

$$= (2\pi\alpha)^{-1/2} \exp[\pm(\mathrm{i}k_F x - \sum_k \frac{2\pi}{Lk} \exp(-\tfrac{1}{2}\alpha|k| - \mathrm{i}kx)\, \rho_i(k))] \tag{19}$$

where +, − correspond to $i = 1, 2$, we obtain

$$H = H_0 + \frac{\Delta}{\pi\alpha} \int \cos\varphi(x)\, \mathrm{d}x. \tag{20}$$

By rescaling $\varphi \to (4\pi)^{1/2}\varphi$ and $x \to \tilde{v}_F x$ we obtain the sine–Gordon Hamiltonian (1) with $m^{*2} = 4\Delta\tilde{v}_F/\alpha$ and coupling constant $\beta^2 = 4\pi$.

The transformation between the fermion and boson Hamiltonians has been done with a constant number of fermions. Also $\varphi_1(x)$ has no $k = 0$ component and so the soliton density in the field $\varphi(x)$ is (from equation (16))

$$\rho_s = \bar{q}/2\pi = \rho_e. \tag{21}$$

Equation (18) is easily diagonalised, and gives free fermions with dispersion $E_k = \pm(\Delta^2 + \tilde{v}_F^2 k^2)^{1/2}$. The condition (21) means that $\bar{q}/2\pi$ states above the gap are occupied (at $T = 0$) and each of these states corresponds to one soliton. A single soliton corresponds to one state above the gap and its energy is Δ.

The chemical potentials of the fermions and solitons are equal (from equation (21)) and therefore we can now pass to the grand canonical formalism and evaluate $\rho_s(\mu, T)$ based on the fermion system with a chemical potential μ.

The Bogoliubov transformation on equation (18) leads to fermions with operators $\alpha_i(k)$ and dispersion $\pm\text{sgn}\, k\, E_k$. Define the operators

$$\tilde{\alpha}_1(k) = \begin{bmatrix} \alpha_1(k) & k > 0 \\ \alpha_2(k) & k < 0 \end{bmatrix}$$

$$\tilde{\alpha}_2(k) = \begin{bmatrix} \alpha_2^+(k) & k > 0 \\ \alpha_1^+(k) & k < 0 \end{bmatrix} \tag{22}$$

and the Hamiltonian becomes

$$H(\mu) = \sum_k (E_k - \mu)\, \tilde{\alpha}_1^+(k)\, \tilde{\alpha}_1(k) + \sum_k (E_k + \mu)\, \tilde{\alpha}_2^+ \alpha_2. \tag{23}$$

The soliton density is now the difference in the density of the $\tilde{\alpha}_i$ fermions

$$\rho_s = \int_{-\infty}^{\infty} \frac{\mathrm{d}k}{2\pi} \{[\exp[(E_k - \mu)/I] + 1]^{-1} - [\exp[(E_k + \mu)/I] + 1]^{-1}\}. \tag{24}$$

At $T = 0$

$$\rho_s = \frac{1}{\pi \tilde{v}_F} (\mu^2 - \Delta^2)^{1/2}\, \text{sgn}\, \mu\, \theta(|\mu| - \Delta). \tag{25}$$

Equation (25) shows the lock-in transition which occurs at $\mu = \Delta$ with a critical exponent $\bar{\beta} = \frac{1}{2}$. This exponent is also related to the effective long-range interaction between solitons. For a given soliton density μ is determined from equation (25) and the energy is (at $T = 0$)

$$E(\rho_s) = \Delta\rho_s[1 + \pi^2 \tilde{v}_F^2 \rho_s^2 / 6\Delta^2 + O(\rho_s^4)]. \tag{26}$$

Therefore the effective interaction between solitons behaves like ρ_s^2 for low soliton density. This interaction is much stronger here than the exponential interaction in the classical case.

For $T \neq 0$ the lock-in transition is smeared as in the classical case (figure 1) while at $T \to \infty \rho_s \to \mu/\pi\tilde{v}_F$ and the $\cos\varphi$ interaction has no effect. For small μ and low $T(T \ll \Delta - \mu)$ the result (24) is similar to equation (13) corresponding to thermal excitation of solitons and antisolitons.

5. Charge transfer in 1D fermion systems

In this section we consider the electronic charge ρ_e in a 1D conductor, and show that it is temperature dependent when an Umklapp scattering is considered. This effect may be observed experimentally and is an important application for the results in the previous sections.

The model is identical to that used by Emery *et al* (1976) or Gutfreund and Klemm (1976) except that here we do not restrict the electron band to be half filled. The electrons interact through long- and short-range interactions with strengths V and U respectively. A second-order Umklapp process with strength $W_\perp$ involves a reciprocal lattice wave-vector G and scatters two electrons from one side of the Fermi surface to the other side. The scattering requires a momentum of $4k_\mathrm{F}$, and we consider the effects of the Umklapp process when $\bar{q} = 4k_\mathrm{F} - G \neq 0$.

Define a boson field, similar to equation (15),

$$\varphi_1(x) = \frac{\mathrm{i}}{\sqrt{2}} \sum_{k\neq 0} \frac{2\pi}{Lk} \exp(-\tfrac{1}{2}\alpha|k| - \mathrm{i}kx)\,[\rho_1(k) + \rho_2(k)] \tag{27}$$

but here $\rho_i(k) = 2^{-1/2}\Sigma_{p,s} a^+_{i,s}(k+p)\, a_{i,s}(p)$ and s is the spin index. The electron density (except for the $k = 0$ mode) is $\varphi_1'(x)/\pi$. It is interesting to note that the same result holds in the ordered phase, where $\varphi_1(x)$ is the phase of a complex order parameter (Horovitz and Krumhansl 1978).

In order to describe an electron density of $\rho_\mathrm{e} = \bar{q}/2\pi$ we define the field

$$\varphi(x) = 2\varphi_1(x) + \bar{q}x. \tag{28}$$

The Hamiltonian for the charge density part (equation (7) of Emery *et al* 1976) can be written in the form

$$H = \frac{1}{16\pi v_\mathrm{F}} \int \mathrm{d}x[(1 - W_\parallel/2\pi v_\mathrm{F})^{-1}\dot{\varphi}^2(x,t)$$

$$+ (1 + W_\parallel/2\pi v_\mathrm{F})v_\mathrm{F}^2\varphi'^2(x,t) + (8v_\mathrm{F}/\pi\alpha^2)W_\perp \cos\varphi(x,t) \tag{29}$$

where $W_\parallel = 2V - U$. A constant $v_\mathrm{F}\bar{q}^2(1 + W_\parallel/2\pi v_\mathrm{F})/16\pi$ was added here, so that the energy is measured relative to the level of the half filled band as in equation (17) (Haldane 1979).

By rescaling $\varphi \rightarrow \beta\varphi$ and $x \rightarrow x v_\mathrm{F}[1 - (W_\parallel/2\pi v_\mathrm{F})^2]^{1/2} \equiv cx$ we obtain the sine–Gordon Hamiltonian (1) with $m^{*2} = 4v_\mathrm{F}W_\perp(1 - W_\parallel/2\pi v_\mathrm{F})/\pi\alpha^2$ and

$$\beta^2 = 8\pi\left(\frac{1 - W_\parallel/2\pi v_\mathrm{F}}{1 + W_\parallel/2\pi v_\mathrm{F}}\right)^{1/2}. \tag{30}$$

The transformation to equation (29) is done with a constant number of electrons so that $\int \varphi_1'(x)\,\mathrm{d}x = 0$. From equation (28) the soliton density is $\rho_\mathrm{s} = \bar{q}/2\pi = \rho_\mathrm{e}$ and therefore the electron and soliton chemical potentials are equal. As in § 4, we can now pass to the grand canonical formalism but here we evaluate $\rho_\mathrm{e}(\mu, T)$ from the function $\rho_\mathrm{s}(\mu, T)$.

Assume that the charge is known for $W_\perp = 0$, or equivalently for $T \rightarrow \infty$ as $\bar{q}_0/2\pi$. In this limit $\rho_\mathrm{s} = \mu\beta^2/4\pi^2 c$ so that the corresponding chemical potential is

$$\mu_0 = \tfrac{1}{4}\bar{q}_0 v_\mathrm{F}(1 + W_\parallel/2\pi v_\mathrm{F}). \tag{31}$$

This is the electron chemical potential for $W_\perp = 0$ measured from the half filled band level. If this chemical potential is fixed, i.e. if there is an infinite electron reservoir at the Fermi level, then the electron density ρ_e is temperature dependent and is given by $\rho_\mathrm{e}(T) = \rho_\mathrm{s}(\mu_0, T)$. However, in a realistic system the Fermi level is determined by an energy balance between two or more bands such that the total number of electrons in these bands is conserved. Thus, $\rho_e(T)$ can be determined if one has a detailed knowledge on all the relevant bands.

As a specific example we consider a charge transfer salt of the TTF-TCNQ type (Khanna *et al* 1977) where the charge transfer from the TTF chains to the TCNQ chains is determined by the crossing of two bands. We assume that both types of chains are described by a 1D interacting electron gas with a charge density Hamiltonian of the form (29). The TCNQ band has Fermi velocity $v_F^a > 0$ and charge density ρ_e^a while the TTF band has $v_F^b < 0$ and charge density ρ_e^b. The total charge in both bands is two electrons per molecule, and since we measure charge from the half filled band level (one electron per molecule) charge conservation implies $\rho_e^a + \rho_e^b = 0$.

In order to measure the electron chemical potential μ_e from the same level in both bands we shift it to the level where the bands cross when $W_\perp = 0$, i.e. $\mu^{a,b} = \mu_e + \mu_0^{a,b}$ where the parameters in equation (31) depend on the chain index. The chemical potential and charge transfer are now determined by solving the equation

$$\rho_e^a(T) = \rho_s^a(\mu_e + \mu_0^a, T) = -\rho_s^b(\mu_e + \mu_0^b, T) \tag{32}$$

where $\rho_s^{a,b}(\mu, T)$ are the soliton density functions with parameters v_F, $W_\|$ and $W_\perp$ appropriate for the chains *a* or *b*. If these parameters happen to be equal on both chains, then the solution of (32) is simple. Since $\bar{q}_0 \equiv \bar{q}_0^a = -\bar{q}_0^b$ we obtain $\rho_s(\mu_e + \mu_0, T) = \rho_s(-\mu_e + \mu_0, T)$ and therefore $\mu_e = 0$ and the charge transfer is $\rho_s(\mu_0, T)$.

An interesting consequence of equation (32) is that in the absence of Umklapp terms ($W_\perp^{a,b} = 0$) the solution is $\mu_e = 0$ and $\rho_e^a = \bar{q}_0/2\pi$ is temperature independent. Thus the interactions V, U cannot change the chemical potential or the degree of charge transfer. This conclusion seems to be a consequence of the assumed electron–hole symmetry, i.e. the linear electron dispersion, and it may not be valid for systems with non-linear dispersion.

TTF-TCNQ has a CDW instability at 54 K, and we are interested here in higher temperatures i.e. the region of a 1D fluctuating CDW. There is indeed experimental evidence (Khanna *et al* 1977, Kagoshima *et al* 1976) from the position of $4k_F$ scattering that the charge transfer changes from 0.55 electrons per molecule above ~200 K to 0.59 below ~160 K. The bands are rather far from being half filled and it is not obvious why the second-order Umklapp process should be important. Let us first demonstrate the application of the present theory and then discuss its limitation and other experiments which may demonstrate $\rho_e(T)$ more clearly.

In figure 2 we show the experimental data along with theoretical curves of the classical (equation (13)) and $\beta^2 = 4\pi$ quantum (equation (24)) cases, assuming that the parameters on the TTF and TCNQ chains are equal. The classical fit gives E_s n. As usual in and $\mu/E_s = 1.2$ while the quantum case gives $E_s = \Delta = 165$ K and $\mu/E_s = 2.0$. Since β determines $W_\|/2\pi v_F$, the result for μ implies a certain bandwidth. The classical fit ($W_\|/2\pi v_F = 1$) yields $v_F k_F \simeq 700$ K while for the quantum case ($W_\|/2\pi v_F = \frac{3}{5}$) $v_F k_F \simeq 250$ K. The expected value is 1000–2000 K so that the classical fit seems more reasonable. This is also consistent with Emery's explanation (1976) of the strength of the $4k_F$ scattering which requires $W_\|/2\pi v_F > \frac{4}{5}$ and $U > 0$. In both fits $W_\perp/2\pi v_F \sim 1$, assuming $\alpha \sim 1/k_F$. This value of $W_\perp$ should not be taken too seriously, since the fermion–boson transformation requires that the limit $\alpha \rightarrow 0$ be taken. As usual in quantum field theory the physical parameter is not the bare-coupling ($W_\perp$) but rather the final mass, i.e. the soliton mass.

Note that the soliton masses in the two fits are very different. Since we consider a system of dense solitons (large incommensurability) the single soliton energy is not relevant. The density ρ_s is determined by the balance between the soliton–soliton interaction and the energy gain for creating an isolated soliton, which is $\mu - E_s$. Thus, the characteristic temperature where ρ_s is changing should be $T \sim \mu - E_s$. For the fits in figure 2, $\mu - E_s$ is 180 K and 165 K—very close numbers in view of the difference in E_s.

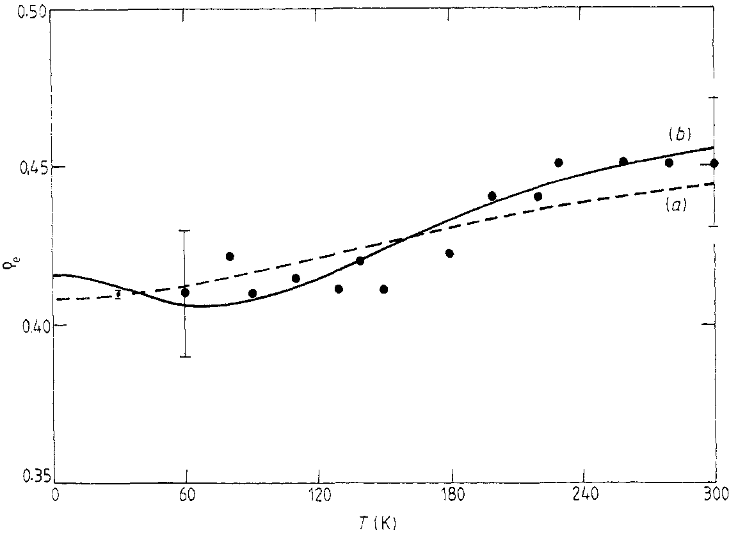

Figure 2. Electron density (electrons per molecule −1) in TTF-TCNQ as a function of temperature. The experimental points and error bars are from Khanna *et al* (1977). Curve (*a*) is the classical fit (equation (12)) ($E_s = 960$ K, $\mu - E_s = 180$ K) and curve (*b*) is the $\beta^2 = 4\pi$ quantum fit (equation (24)) $E_s = 165$ K, $\mu - E_s = 165$ K.

The present theory, as applied in figure 2, has a few limitations:

(i) model assumptions—equal parameters on both chains and the linear dispersion;

(ii) thermal expansion of the lattice can also affect the amount of charge transfer (Tiedje *et al* 1977); however $\rho_e(T)$ seems to saturate above 200 K while the thermal expansion coefficient is finite and even increasing strongly above 200 K (Schaffer *et al* 1975);

(iii) the effect is rather small and the experimental error bars are too large to justify a detailed theoretical fit.

Systems with a charge transfer closer to one electron per molecule should be better candidates for the present theory. The effect on $\rho_e(T)$ should be larger and the linear dispersion becomes a better approximation. A good candidate is TTF-TCNQ under pressure, as we discuss in the next section.

6. Discussion

We have studied the soliton density $\rho_s(\mu, T)$ in both classical and quantum theories. The temperature dependence is characterised by three regions: (i) $\mu \ll E_s$, solitons are thermally excited; (ii) $\mu \simeq E_s$, the region of the lock-in transition; the temperature dependence is strong, smearing the $T = 0$ transition; (iii) $\mu \gg E_s$, weak temperature dependence with ρ_s changing slightly around $T \sim \mu - E_s$.

The effects of quantisation are mainly to shift the lock-in transition to the renormalised soliton mass and change the critical exponent $\bar{\beta}$. We obtain $\bar{\beta} = 0$ for the classical case, while $\bar{\beta} = \frac{1}{2}$ for $\beta^2 = 4\pi$.

Most of the theoretical applications of the lock-in transition consider a two- or three-dimensional phase transition within a mean field ($T = 0$) theory. The coefficients of the effective Hamiltonian may depend on temperature and the appropriate order parameter is space dependent only in one given direction. Thus in the CDW problem (McMillan 1976) both μ and E_s depend on temperature and the lock-in temperature T_c is determined from the equation $\mu(T_c) = E_s(T_c)$. The critical exponent of $T - T_c$ is then related to $\bar{\beta}$ through the dependence of μ/E_s on T.

The present theory is more directly relevant to systems with an intrinsic 1D temperature dependence. One possible example is the ω phase anomaly which is observed in Zr–Nb alloys (Horovitz *et al* 1978). Here μ corresponds to the discrepancy in lattice constants of a rod-shaped ω domain and the surrounding BCC matrix. The parameter μ changes with the alloy composition and the lock-in transition appears around 17% Nb. Preliminary measurements on the temperature dependence are consistent with the general features of $\rho_s(\mu, T)$ (D H Bilderback and B W Batterman 1978 private communication).

In § 6 we compared $\rho_s(\mu, T)$ with the temperature-dependent charge transfer in TTF-TCNQ. The observed effect is small, but we expect the Umklapp scattering to be more effective if it is applied to a system with an electron density closer to the commensurate value.

Recent experiments on TTF-TCNQ under pressure (Adrieux *et al* 1979) showed that the CDW becomes commensurate at ~20 kbar below 71 K. This was deduced from resistivity data, while x-ray or neutron scattering data are not yet available. We predict that in some range of pressures around 20 kbar, the charge transfer at room temperature is not commensurate, but it approaches the commensurate value as temperature is reduced, and finally locks-in at the CDW phase transition, i.e. at ~71 K.

It was suggested that TTF-TCNQ near 20 kbar pressure has a $\frac{1}{3}$ filled band (Andrieux *et al* 1979). In that case the theory in § 6 has to be modified to describe a third-order Umklapp process. This process involves the transfer of three electrons across the Fermi sea; this involves at least two chains and therefore a weaker effect than the second-order Umklapp process.

In addition to a shift in the charge transfer, the Umklapp process leads to the apperance of harmonics at $nM2k_F$, i.e. the soliton lattice. In the case of TTF-TCNQ the $4k_F$ scattering is clearly seen, but since it is stronger than the $2k_F$ scattering (at high temperatures) it is probably due to another mechanism (Emery 1976). There is however a weak scattering at $8k_F$ visible in both sets of experimental data (Khanna *et al* 1977, Kagoshima *et al* 1976). All these effects of the Umklapp process are small and within experimental error. Further experiments, e.g. on TTF-TCNQ under pressure, will be very useful in clarifying these points.

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