Charge Density Waves in the Quasi-One-Dimensional Compound

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Charge density wave (CDW) conductors exhibit a rich response to applied electric fields which involves many dynamical degrees of freedom. This complicated dynamics arises from competition between the random pinning of the CDW and the elasticity of the CDW itself. This paper gives an overview of the response of the CDW to an applied electric field.

I.INTRODUCTION

As Peierls [1], pointed out many years ago, a one dimensional metallic system in unstable against a possible deformation of the lattice with the wave vector number $\mathbf{Q}=2\mathbf{k}_F$, $\mathbf{k}_F$ being the Fermi waves number of free electrons. However in 1954, Before the BCS theory, Fröhlich [2] discussed the instability to show that the electrons and the lattice couple together to produce a modulated ground state. According to the relative strength of electron-lattice coupling, the modulated ground state can be a charge density wave modulation in space (CDW) or, if the spin orientation is concerned, a spin density wave (SDW). According to the model proposed by Fröhlich in jelium approximation the CDW slide without friction in an idealised situation. This was suggested by Bardeen [3] as a possible mechanism of high conductivity at low temperature which is observed in some quasi-one-dimensional materials. Lee, Rice and Anderson [4], treated the problem in a mean field approximation and showed that the soft phonon of $\mathbf{Q}=2\mathbf{k}_F$ produces a CDW at zero temperature and also that the excitation consist of the phase mode and amplitude mode. The phase mode has the dispersion $\omega = \mathrm{v}q$ and is important at low temperatures. In fact the phase mode is connected with the transport of charges, which leads to the conductivity $\sigma(\omega)$ to the singularity $(i\omega)^{-1}$ as $\omega \to 0$ corresponding to the free sliding of the CDW. However this is very sensitive to the presence of impurities, which would remove this divergence. In other word the impurities hinder the free motion of the CDW and pin the phase motion.

In order to investigate the pinning effect of impurities on the CDW, Fukuyama [5] made a simplification of the problem by ignoring the amplitude mode; it is then possible to write a semi-phenomenological Hamiltonian of the phase mode including its interaction with impurities and the applied electric field.

In this paper, we give in section 1 a description of the Peierls instability and crystal structure of some materials with CDW transport, in section 2 we report some unusual phenomena which give evidence for the CDW conductivity, in section 3 we present the theoretical model which have been used to simulate this new type of conductivity.

II. CDW GROUND STATE

A. Peierls Instability

The reduction of the phase space from three dimensions (3D) to one dimension (1D) has several important consequences. Both interactions effects and random potentials have a more profound effects in one than higher dimensions and the fluctuations are also more important. Because of the simple Fermi surface in 1D system, the interactions between electrons can be expressed in term of two coupling constants, one for q=0 and one for $\mathbf{Q}=2\mathbf{k}_F$; leading to a simple phase diagrams for the occurrence of the various broken symmetry ground states which arise as a consequence of these interactions. The response of an electron gas to a time independent potential is usually treated within the framework of linear response theory. The rearrangement of the charge density expressed in terms of an induced charge related to the external perturbation trough:

$$\rho^{ind}(\mathbf{r}) = \chi(\mathbf{q})V(\mathbf{r}) \tag{1}$$

$\chi(\mathbf{q})$ is so-called Lindhard response function.

In contrast to a 3D electrons gas, the response function in the 1D diverges at $\mathbf{q}=2\mathbf{k}_F$ [6], this fact has several important consequences. Equation (1) imply that the external perturbation leads to a divergent charge redistribution; this suggests, through self-consistency, that at T=0 the electron gas is unstable with respect to the formation of a varying electron charge or spin density. The divergence of the response function at $\mathbf{q}=2\mathbf{k}_F$ is du to particular topology of the Fermi surface, called perfect nesting [7]. The divergent response function leads to various instabilities at low temperature, and a simple mean field argument gives a finite transition temperature when this happens [6]. The density fluctuations $\rho^{ind}(\mathbf{q})$, dues to the external potential reflect the formation of electron-electron or electron-hole pairs, with the ground state at T=0 being a coherent superposition of the various pairs state. The nature of the ground state depend on the electron-electron and electron-hole interaction, which can be described by a q-dependent interaction potential V($\mathbf{q}$).

The first of these state develop in response to the interaction for which the total momentum q=0; this is called the particle or Cooper channel. The resulting states are the well known (singlet or triplet) superconducting states of metals. The last with a finite total momentum of pairs, develop as a consequence of the divergence of the fluctuations at $\mathbf{q} = 2\mathbf{k}_F$; this is the particle-hole channel, usually called the Peierls channel. These states develops a periodic variation of the charge or spin density, they are called the charge density wave (CDW) and the spin density wave (SDW) ground states. The period $\lambda = \dfrac{\pi}{k_F}$ associated with the spatial variation of the charge density leads to a gap in the single particle excitation spectrum at the Fermi level. For an arbitrary band filling the period is incommensurate with the underlying lattice.

Below the Peierls temperature transition, the electronic charge density modulation is defined as:

$$\rho(\mathbf{r}) = \rho_c + \rho_0 \cos(\mathbf{Qr} + \varphi) \qquad (2)$$

ρ_c is the uniform charge density, ρ_0 is the amplitude of the modulation and $\mathbf{Q} = 2\mathbf{k}_F$ the wave vector. The phase φ describe the location of the CDW relative to the lattice. The local electron charge density is partially neutralised by a concomitant displacement of each ion to a new equilibrium position, the displacement of the n^{th} ion initially at nr_0, being:

$$u_n = u_0 \sin(n\mathbf{Qr} + \varphi) \qquad (3)$$

In the Fröhlich model, the energy gap reduce the elastic scattering of individual electrons because there is no state available for relaxing energy. The motion is therefore without friction dissipation and the compound becomes superconducting. The Fröhlich mode is a direct consequence of the translation invariance, the CDW energy becomes phase independent. In real system, as shown by Lee, Rice and Anderson [4], this translation invariance is broken because the phase is pinned to the lattice. The pinning can be proved by, impurities and / or defects, commensurability between the CDW wavelength and the lattice, or by coulomb interaction between adjacent chains. An applied electric field can supply the CDW with an energy higher than the pinning one and above a threshold electric field, the CDW can slide and carry a current, but damping prevents the supeconducting phenomenon. This extra conductivity has been first observed in 1976 by Monceau in the $NbSe_3$ compound [8] and since this time, an intense experimental and theoretical activity has been devoted to the understanding of the properties of this collective mode. See for example charge density waves in solids ed L. P. Gor'kov and G. Gruner, North Hollan

B. Materials and CDW transition

A large number of organic and inorganic solids have crystal structure in which the fundamental structural units form linear chains. While most of these materials are insulators or semiconductors; several groups have partially filled electron bands, and consequently display metallic behaviour at high temperature. The overlap of the electronic wave functions in various crystallographic directions leads to strongly anisotropic, so-called quasi-one- dimensional electron band. This is the prerequisite for the development of the Peierls instability.

The existence of a CDW ground state in quasi-one dimensional and two dimensional compound appears to a rule rather the exception, and to date a large number of system which show transitions to CDW phase have been discovered. The first system determined to exhibit dynamic CDW properties was the transition metal trichalcogenide $NbSe_3$, first synthesised in 1975 [9]. $NbSe_3$ was for several years the only known system with CDW transport. Later studies on the related materials TaS_3 [10] and $NbSe_3$ [11] indicated transport properties for these trichalcogenides similar to these observed in $NbSe_3$. Recently two new groups of materials have been shown to display collective CDW conduction: halogen transition metal tetrachalcogenides [12] and bronzes [13]. The halogen group contains the iodine compounds ($TaSe_4$) and $NbSe_3$ while in the second group the blue bronze $K_{0.3}MoO_3$ is the most prominent example.

III. EXPERIMENTAL EVIDENCE FOR THE CDW TRANSPORT

One of the most fundamental phenomena of CDW dynamics is non-linear conduction of collective charge transport by means of CDW sliding below the Peierls transition. At low electric fields, the CDW is prevented from moving by pinning to impurities and normal conductivity characterises the single particle excitations of the system. The applied field must overcomes the pinning force before the CDW began to slide, thus, the CDW is deppined from the underlying lattice above a relatively low threshold field E_T of about 100mV/cm figure. 1 Well above the threshold field, where CDW conduction dominates the single particle current J_{CDW}, the current density and the excess conductivity σ_{CDW} follow empirical power laws as a function of the applied electric field equation (4) et (5).

$$J_{CDW}(E) = J_0\left(\frac{E}{E_T} - 1\right)^{\alpha} \qquad (4)$$

$$\sigma_{CDW}(E) = \sigma_0 \frac{E_T}{E}\left(\frac{E}{E_T} - 1\right)^{\alpha} \qquad (5)$$

The exponent α is temperature dependent [14].
The existence of a finite threshold field for the deppining of the CDW, is a direct consequence of the competition between the CDW elasticity and the pinning of the CDW by defects and / or impurities in the CDW underlattice.

IV. NUMERICAL SIMULATION OF THE CDW DYNAMICS

We investigate by numerical simulation to what extent a classical treatment of the Fukuyama-Lee-Rice model (FLR) [15] on 1D incommensurate CDW in the weak pinning regime can reproduce the peculiar transport properties of such quasi-one-dimensional conductors.

A. Model of the deformable phase

The FLR model describe the CDW as a classical deformable medium pinned by random impurities. The behaviour of the CDW is described in terms of its variable $\phi(x,t)$, while its amplitude is kept constant. The phenomenological hamiltonien for phase pinning model of a 1D incommensurate CDW can be written as [4-15]

$$H = \frac{1}{2}\int k\left(\frac{d\phi}{dx}\right)^2 dx - e\frac{E}{\pi}\int \phi(x,t)dx + \sum_j \rho(x,t)V(x-r_j)dx \tag{6}$$

The first term in equation (6) represents the elasticity of the CDW with stiffness k. The second term describes the polarisation energy of the CDW in an uniform applied field E. The third term corresponds to the interaction energy with impurities located at random positions rj and acting only at these positions. The dynamic behaviour of the CDW system is specified by the overdamped equation of motion [16]

$$F_d = -\frac{\delta H}{\delta \phi} \tag{7}$$

F_d is a damping force [17]

The discretised equation of the CDW motion can be written as [18]

$$\frac{d\phi}{dt} = \nabla^2\phi_j + \frac{1}{2}E(x_{j+1} - x_{j-1}) + \varepsilon.\sin(\theta_j + \phi_j) \tag{8}$$

$\phi_j = \phi(x_j)$ is the discrete second derivative:

$$\nabla^2\phi_j = \left(\frac{(\phi_{j+1} - \phi_j)}{(x_{j+1} - x_j)} - \frac{(\phi_j - \phi_{j-1})}{(x_j - x_{j-1})}\right) \tag{9}$$

$x_j = c_j r_j$, $\theta_j = Qx_j$, $E = \dfrac{\rho_c E}{Qkc_i^2}$ is a dimensionless electric field and $\varepsilon = \dfrac{\rho_0 v_0}{kc_i}$ is a dimensionless interaction strength. The strong pinning case [5] corresponds to $\varepsilon >> 1$ and can be realised by either strong impurity potential or a dilute impurity concentration. The opposite limit $\varepsilon << 1$ corresponds to the weak pinning case where the elastic energy dominates. The current density associated with the sliding CDW is related to the time derivative of the CDW phase by the following equation:

$$J_{CDW} = \frac{e}{\pi}\int \frac{d\phi(x,t)}{dt}dx \tag{10}$$

B. Numerical results

The complicated dynamics of the CDW system arises from the competition between the random distributed impurities and the elasticity of the CDW itself. One important consequence of this competition is the existence of a dynamic transition between the pinned and the unpinned CDW states for a well defined threshold field [19]. The threshold field is dimensionless parameter ε and impurities concentration dependent [18-20].
For a given dimensionless parameter ε, is proportional to c_i^2 in agreement with the CDW pinning theory [21] figure 2. This indicates that impurities in a host CDW lattice affect not only the static properties of the CDW condensate, but the dynamical properties as well. The origin of the finite threshold field E_T for the onset of non-linear conduction in dynamic CDW system is generally believed to originate from impurity pinning of the CDW phase.
Above the threshold field, the onset of the CDW conduction depend on the applied field amplitude and the impurities concentration. The excess current density and the associated conductivity obey respectively a power laws equation (4) and (5) figure 3 a and b.

V. DISCUSION

In the presence of impurities, it is energetically favourable for the CDW to distort, these deformation caused by the impurities lead to a non-linear current-voltage characteristics, where the CDW motion appears to set in at nonzero threshold field. Thus, the central issue in study of the CDW conductors is the nature of the CDW-defects interaction. In real system defects are always present, then their effects are present in substantial numbers even high quality samples. The defect not only pin the CDW phase at a preferred values, but the CDW amplitude is affected as well [22]. Numerical simulation based on the 1D FLR model reproduces a qualitative understanding of the real 3D situation of many interesting transport properties observed experimentally in some quasi-one-dimensional compounds.

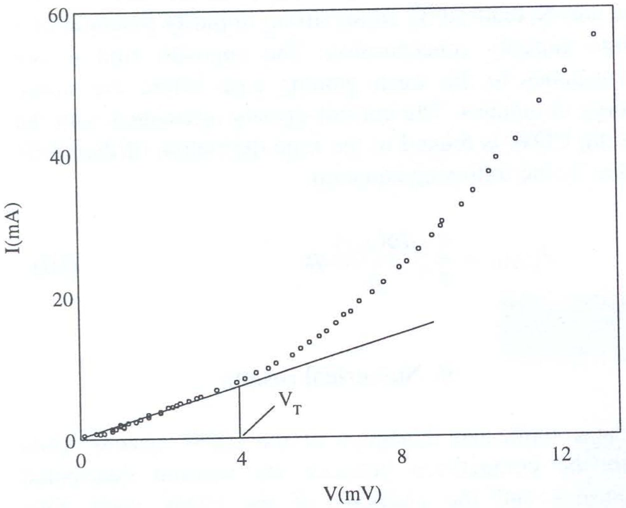

Figure 1. Typical current- voltage characteristic at T=77k on the $K_{0.3}MoO_3$

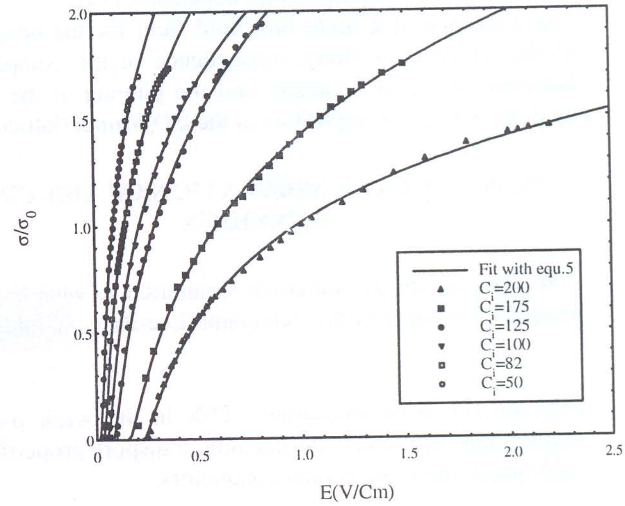

Figure 3-a. Current density versus electric field for various impurity concentrations c_i (c_i in ppm).

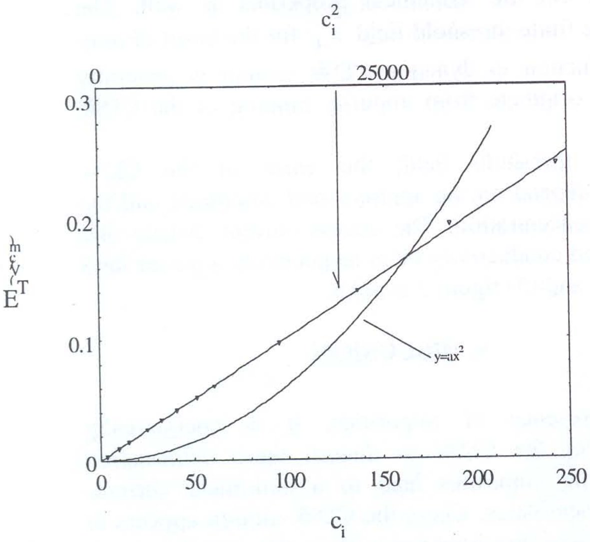

Figure 2. The threshold electric field E_T as function of the impurity concentration c_i (c_i in ppm).

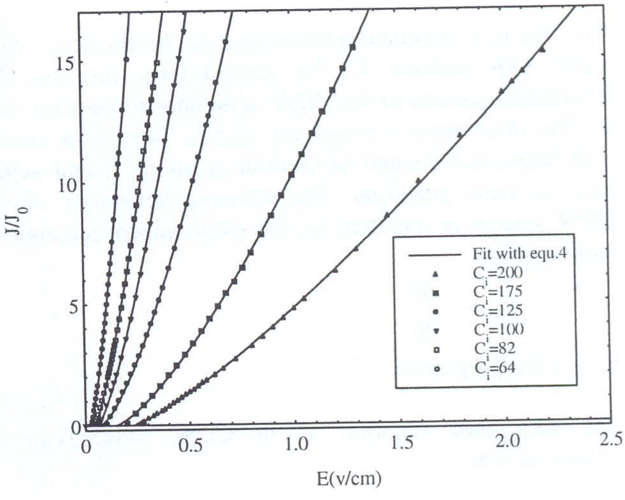

Figure 3-b. Conductivity versus electric field for various impurity concentrations c_i (c_i in ppm).

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