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PINNING OF FLUCTUATING CHARGE-DENSITY WAVES IN QUASI-ONE-DIMENSIONAL CONDUCTORS

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It is shown that the contribution of CDW fluctuations to the conductivity of quasi-one-dimensional metals is drastically reduced if the system is commensurate. The effect of commensurability on the single-electron conductivity is much smaller. Thus, commensurability pinning of fluctuation conductivity is a satisfying explanation of recent experimental results on organic conductors.

I. Introduction

Quasi-one-dimensional metals, i.e. systems of weakly coupled metallic chains, show at some critical temperature T_p a phase transition into a charge-density wave (CDW) state [1], as first predicted by Fröhlich [2] and Peierls [3]. One of the most interesting points in the study of these systems is the possibility of dc conductivity by rigid translations of the CDW [2,4]. Below T_p this mechanism is suppressed by impurity pinning or coupling between oppositely charged chains [4,5]. However, above T_p there exist fluctuations into the CDW state, and due to the finite correlation length the pinning mechanisms are expected to be far less effective than in the long-range ordered state below T_p. The existence of a fluctuation contribution to the dc conductivity of quasi-one-dimensional conductors [6,7] has been a controversial subject [8,9]. Recent experiments [10] show a considerable drop of the metallic conductivity of TTF-TCNQ when the CDW becomes commensurate under pressure, i.e. when the wavelength of the CDW is (nearly) three times the lattice constant. This drop has been interpreted as commensurability pinning of fluctuation conductivity, i.e. the fluctuating CDW locks into the lattice. Here we present a theoretical calculation of fluctuation conductivity at commensurability.

Above T_p there is a wide temperature range where fluctuations can be observed by X-ray scattering, for example in TTF-TCNQ (T_p=53K) up to 200K [1,11,12]. Over most of this temperature range (T>60K) adjacent chains are uncorrelated, so that the fluctuations can be treated in a strictly one-dimensional model. Further we limit ourselves here to the lowest order fluctuation effects. The contribution of CDW fluctuations to the conductivity is to lowest order given by the Azlamazov-Larkin (AL) diagram [13-15]. Therefore in the following section we investigate the influence of commensurability on the AL diagram. The fluctuations affect the conductivity also by single-electron scattering [13,14,16]. The influence of commensurability on this mechanism is discussed in the last section.

II. Fluctuation Conductivity in a Commensurate System

In a nearly commensurate situation, the typical wavenumber $2k_F$ of the CDW is given by $2k_F=Q+\delta$, where for the experimentally interesting case of third order commensurability Q is one third of the reciprocal lattice wavenumber and δ denotes deviations from exact commensurability. Due to umklapp processes, an additional term of the form

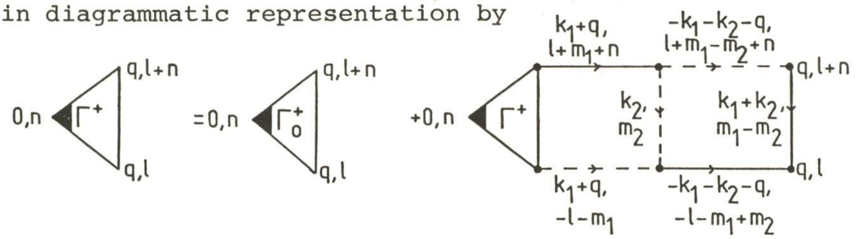
$$G_{com} = -V \int [\phi^3(x)e^{3i\delta x} + \phi^{*3}(x)e^{-3i\delta x}]\, dx \quad , \tag{1}$$

where ϕ is the CDW order parameter, appears in the free energy [17]. This term clearly depends on the phase of the order parameter and thus breaks the translational invariance of an incommensurate (IC) system. Defining a modified order parameter $\psi=\phi\exp(i\delta x)$ an equation of motion for the fluctuation current J can be derived [15]:

$$\frac{dJ}{dt} + \gamma J = \frac{3}{2}iaV \int [\psi^{*3}(x) - \psi^3(x)]\, dx \quad , \tag{2}$$

where $1/\gamma$ is the lifetime of the fluctuations and a is a constant depending on the electronic properties of the system. Equation (2) shows that even for infinite lifetime the current is not a conserved quantity, in contrast to the IC case. Therefore drastic deviations from the IC situation are to be expected.

We now consider the effect of G_{com} on the AL-diagram. First, G_{com} leads to a self-energy renormalization $\omega_o^2 \to \omega_o^2 - 9TV^2/2\omega_o^4\xi$ of the order parameter propagator by third order umklapp processes [15]. ω_o is the minimum of the Kohn anomaly and ξ the correlation length. As long as V is not too large there is only a small change in the conductivity. However, G_{com} leads also to an interaction between the two parallel propagators in the AL-diagram, mediated by another propagator. Arguments similar to those of Holstein [18] show that the most important contribution comes from ladder diagrams, as each rung gives (for certain values of the frequency arguments) a factor $V^2/(\gamma\omega_o)^2$. For long-lived fluctuations this may be very large, so that the whole ladder series has to be summed up. Then one of the two triangular electronic vertices (denoted by Γ_o^+) in the AL-diagram has to be replaced the ladder sum Γ^+. A detailed investigation [19] shows that diagrams with an odd number of rungs are negligible, so that Γ^+ is given in diagrammatic representation by



(3)

where full and dashed lines denote ψ- and ψ^*-propagators, respectively, k_1, k_2, and q are wavenumbers, and l, m_1, m_2, and n are the indices of the corresponding Matsubara frequencies.

In a previous paper [15] eq.(3) was solved using a rather crude approximation. In the limit of long-lived fluctuations a more accurate solution has been found [19]. The resulting conductivity is:

$$\sigma_F = \frac{a^2T}{4\pi\gamma\omega_o^2} \int_{-\Lambda}^{\Lambda} \frac{dq}{1+\xi^2q^2} \left\{1+\eta[1+\xi^2(q/2+\delta)^2]^{-3}\right\}^{-1} \tag{4a}$$

with

$$\eta = \frac{(9V^2T)^2}{\pi\omega_o^{12}\xi^2} \frac{\omega_o}{\gamma} \ln\frac{2\omega_o}{\gamma} \quad , \tag{4b}$$

and Λ is a momentum cutoff of the order of $\xi(2T_P)^{-1}$. The analytic evaluation of the integral in eq.(4a) leads to quite complicated expressions which are not reproduced here. Instead we discuss the main results. First, we note that the effect of commensurability is determined by η. Apart from V this factor also depends on the ratio ω_o/γ and may thus be quite large for long-lived fluctuations even for a relatively weak commensurability potential. Physically this means that the fluctuations have to be sufficiently long-lived so as to allow the commensurability force to have an appreciable effect. In Fig.(1) the conductivity resulting from eq.(4a) is shown for different values of η in dependence on $\xi\delta$. This illustration shows that in

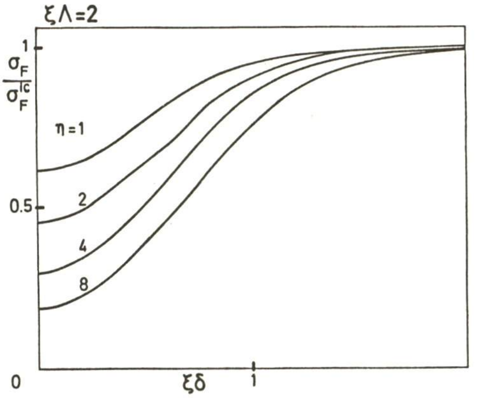


Fig.1 The fluctuation conductivity in units of the value σ_F^{ic} in the incommensurate case

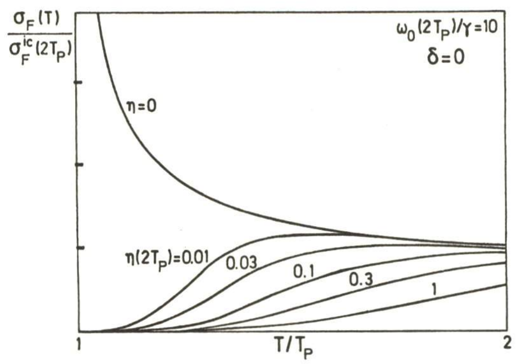


Fig.2 Temperature dependence of σ_F for differnt values of $\eta(2T_P)$, $\eta=0$ is the IC case

an exactly commensurate system ($\delta=0$) σ_F is strongly reduced. For deviations from commensurability σ_F increases and for $\xi\delta>1$, when the CDW can no more lock into the lattice, goes rapidly to its IC value.

A picture of the temperature dependence of σ_F can be obtained assuming $\omega_o^2(T)=\omega_o'^2(T/T_P-1)$. The resulting temperature dependence (Fig.(2)) shows that the conductivity vanishes in the commensurate case for $T\to T_P$, contrary to the IC case ($\eta=0$). We note, however, that our calculation treats fluctuations in lowest order and therefore becomes invalid near T_P. The qualitative behaviour shown in Fig.(2), especially the strong reduction of the fluctuation conductivity with respect to the IC case, is however expected to be valid even near T_P.

III. Discussion

We have shown that the contribution of CDW fluctuations to the dc conductivity of a quasi-one-dimensional metal is strongly reduced by commensurability. Apart from the collective mechanism as described by the AL-diagram fluctuations also scatter electrons individually, thereby decreasing the single electron conductivity. It has been shown [13,14,16] that this effect is proportional to the mean square amplitude of the fluctuations, i.e. to $1/\omega_o$. Therefore commensurability affects the single particle conductivity via the above mentioned self-energy correction of ω_o. This effect is independent of γ, so that for $\gamma<<\omega_o$ the effect of commensurability on the collective part of the conductivity (as represented by the AL-diagram) is much larger than on the single electron contribution.

Quantitative values of the parameters of our theory for TTF-TCNQ are difficult to obtain due to the lack of experimental results on the precursor effects at high pressure. However, an order of magnitude estimate can be obtained using the fact that a term like G_{com} leads to a first order transition with a higher T_P than in the IC case [17]. Using the experimentally observed increase of T_P [10] ($\Delta T_P \simeq 0.1..0.2 T_P$) and the correlation length measured by X-ray scattering at ambient pressure [12], we obtain from Ginzburg-Landau theory [20] $9V^2T/2\omega_o^6\xi \simeq 0.2$ at $T=2T_P$. The lifetime parameter γ is not known experimentally, however, neutron scattering does not indicate overdamping of the soft mode, so that we have $\gamma<\omega_o$. From the above values and assuming $\omega_o/\gamma=5$ we obtain $\eta(2T_P)\simeq 0.7$. This value would lead to a considerable decrease of σ_F, whereas the single electron conductivity is much less affected. We conclude that suppression of the (collective) fluctuation contribution to the conductivity is a reasonable explanation of the experimental results [10]. This picture is supported by the fact that the transverse conductivity of TTF-TCNQ, which is entirely due to single electron processes [21], is nearly unaffected by commensurability [10]. This implies that a considerable part of the longitudinal conductivity of metallic TTF-TCNQ in the incommensurate state is due to CDW fluctuations.

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