Interplay between disorder and quasi-regularity in CDW systems

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Abstract. — We develop here a microscopical model of quasi-regular impurity distribution driven by the CDW appropriate for highly mobile impurities or materials with fictitious CDW transition temperature. Thermal hysteresis effects at the CDW transition reported in the quasi-1D materials $NbSe_3$ and Ta_2NiSe_7 could be interpreted in the framework of the present model. Possible connections with other (alloy, heavy-fermion) systems are also suggested.

1 Introduction.

The displacive motion of the impurities immersed into the one-dimensional system has been studied in detail previously as one kind of quasi-regularity driven by CDW [1]. As a further investigation of this problem we develop here a microscopical model for a different kind of quasi-regular impurity distribution driven by the CDW, consisting of the modulation in the probability of occupied sites. This modulation could be expected in two realistic cases. Firstly, this could happen in the case where impurities could migrate through the crystal, via atomic diffusion processes, towards those sites where the energy of their interaction with the CDW is minimum. Secondly, one may expect such a modulation in the distribution of impurities in connection with the manner in which a certain doped quasi-1D material is synthesised. If this preparation process yields a material whose stable state is a CDW one, one would expect that, in the case where the process is slow enough, impurities would have the possibility to drift towards those sites at which the minimization of the total energy occurs; thence, a modulation in the occupation probability with the same wave vector as the CDW. Such modulated impurity distributions have been previously postulated for KCP [2] and thiourea [3].

We present here a microscopic model which can describe the afore-mentioned modulation in the spatial distribution of impurities. The dependence on impurity concentration of the quantities characterizing the CDW state is obtained. Finally, we discuss the possible connection of our results with the experimental findings in several materials.

2 Model of modulated distribution of impurities.

In the case where the physical conditions permit, impurities could migrate through the crystal to reach those positions which are most favoured energetically in the presence of the Peierls-CDW distortion.

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In the corresponding state of thermodynamic equilibrium, the occupation probability depends on the energy of interaction between impurities and the host system. The energy $E(X_n)$ of interaction between an impurity located at X_n and the CDW has the form:

$$E(X_n) = -\rho_1 U_B \cos(QX_n + \varphi - \varphi_B), \quad (1)$$

where $Q (= 2k_F \bmod G)$ is the CDW wave vector, U_B and φ_B are the amplitude and phase of the Q -Fourier component of electron-impurity scattering potential, respectively, and ρ_1 and φ denote the amplitude and phase of CDW, respectively ($\rho(x) = \rho_0 - \rho_1 \cos(Qx + \varphi)$). Once this interaction energy is known, the thermodynamics requires that the probability of occupying the position X_n is ($\beta^{-1} \equiv k_B T$):

$$w(X_n) \propto \exp[-\beta E(X_n)]. \quad (2)$$

Eq. (2) straightforwardly yields an extra $2k_F$ -potential Δ_i which acts in-phase with the contribution from the Peierls distorted lattice Δ_L on the electron gas. In the continuum limit, Δ_i has the expression:

$$\Delta_i = f_T \frac{x}{d} U_B \frac{d}{dz} \log I_0(z), \quad (3)$$

where x is the fractional impurity concentration, d is the intrachain lattice spacing, $z \equiv \beta \rho_1 U_B$ and $I_0(z)$ is the modified Bessel function with index zero. Obviously, one expects realistically that only a (small) fraction from the total number of impurities is able to effectively reach (through atomic diffusion processes) the sites which are energetically most favoured (cf. Eq. (2)). This effect has been taken into account above by means of the parameter f_T ($\ll 1$), presumably temperature dependent. To get the equation for the CDW order parameter $\Delta (= \Delta_L + \Delta_i$ within the present approach) one has to minimize the thermodynamic potential with the constraint provided by Eq. (3) because ρ_1 depends on Δ . Although the ensuing equation is rather complicated, analytic results can be obtained in the limits of low and high (i. e. near the critical) temperatures if one uses the behaviour of $I_0(z)$ for large and small values of the argument. For low temperatures ($\beta \Delta \gg 1$), Eq. (3) leads to:

$$\Delta_i = f_0 \frac{x}{d} U_B, \quad (4)$$

which shows that in this limit the quantity Δ_i does not depend on Δ . Actually, this model of Δ_i can also be used to describe a converse situation, where a quasi-regular impurity distribution drives the CDW or a system where the distortion has a different (e. g. chemical) origin or the material has a fictitious CDW transition temperature.

Close to the CDW-to-normal transition ($\beta \Delta \ll 1$), one gets from Eq. (3):

$$\Delta_i = f_c \frac{x}{d} \beta \rho_1 U_B^2, \quad (5)$$

revealing that in this region the impurity quasi-regularity is driven by the CDW ($\Delta_i \rightarrow 0$ as $\rho_1 \rightarrow 0$).

3 Results in the dilute limit.

The full x -dependence of the quantities characterizing the CDW state will be presented elsewhere. Here we shall only give this dependence for the Peierls distortion amplitude A_L at $T = 0$ and the critical temperature T_c in the dilute limit ($\gamma \simeq 1.78$):

$$\frac{T_c}{T_{c,0}} - 1 = \left[\frac{2}{\gamma} \left(\frac{f_c}{\lambda^2} - \frac{\pi^2}{16} \right) - 2 \frac{x_{c,0}}{\lambda \alpha} \right] \frac{x}{x_{c,0}} \equiv S_T x, \quad (6)$$

$$\frac{A_L}{A_{L,0}} - 1 = \left(x_{c,0} + f_0 \frac{1 - \lambda}{\lambda} \frac{v_F}{U_B} - \frac{\pi}{8} - 2 \frac{x_{c,0}}{\lambda \alpha} \right) \frac{x}{x_{c,0}} \equiv S_A x. \quad (7)$$

Here $x_{c,0} = v_F d \Delta_0 / U_B^2$ is the critical concentration in the case of perfectly random impurity distribution and without including the elastic energy renormalization due to impurities [1], λ denotes the electron-phonon coupling strength, α is a dimensionless quantity essentially proportional to the ratio of lattice-lattice and lattice-impurity elastic strengths; the quantities f_0 and f_c denote the fractions of impurities which are quasi-regularly distributed at $T = 0$ and $T = T_c$, respectively.

Depending on the values of parameters entering the model, even in the presently analysed dilute limit, one can get either a decrease or an increase in the quantities T_c and A_L at increasing impurity content. This is illustrated in Fig. 1.

Figure 1: Dependence of the slopes $S_T = d(T_c/T_{c,0})/dx$ and $S_A = d(A_L/A_{L,0})/dx$ on the scattering parameter U_B/v_F and the impurity fractions (f_0 and f_c) (Eqs. (6, 7)). The values of the remaining input parameters are: $\alpha = 1$, $2Wd/v_F = 1$ (W — electron bandwidth) and $\lambda = 0.1$. The positive values of the slopes show that T_c and A_L can increase with x for certain input parameter values.

4 Thermal hysteresis at the CDW transition.

As shown by Eqs. (6) and (7), the quasi-regularity in the impurity distribution rises the values of these quantities as compared to the case of perfect randomness (specified below by the subscript r):

$$\frac{T_c}{T_{c,r}} - 1 = \frac{2 f_c}{\gamma \lambda^2} \frac{x}{x_{c,0}}, \quad (8)$$

$$\frac{A_L}{A_{L,0}} - 1 = f_0 \frac{1 - \lambda v_F}{\lambda} \frac{x}{U_B x_{c,0}}. \quad (9)$$

As discussed previously [4, 1], if one starts with a system in the normal state and cools it down, the CDW onset occurs at the critical temperature of the CDW transition with perfectly random impurity distribution ($T_{c,r}$). Because this value is lower than the critical temperature of the CDW transition in the presence of quasi-regular-impurity distribution T_c (cf. Eq. (8)) which furthermore enhances

the CDW properties (e. g. A_L , cf. Eq. (9)), thermal hysteresis and first-order character at the CDW transition are expected (see [4, 1] for a more detailed discussion). On this basis, an explanation of the different values of the critical temperatures of the lower CDW transition (59K upon heating, 58K upon cooling) and the (large) latent heats in $NbSe_3$ [5] can be suggested.

One may think that the thermal hysteretic behaviour observed in various CDW systems could be related to the relaxation of the CDW phase. Although the present status of both theoretical and experimental investigations cannot rule out an explanation which is different from that presently proposed, data reported in some other systems suggest that the at least in some cases the CDW phase should play but a small part in the thermal hysteretic behaviour. Perhaps most encouraging for the present interpretation is the thermal hysteresis found in the $2k_F$ -scattering intensities (i. e. directly related with a spatial modulated structure) in Ta_2NiSe_7 [6]), along with the failure to detect a non-linear electric conduction which is usually attributed to the depinned CDW phase mode. In addition, one should mention the phase transition of the heavy fermion system UCu_5 which removes parts of the Fermi surface, also displaying a thermal hysteresis [7].

The proposed modulation could also be related with the formation of the long-period superlattice structures from the disordered phase observed in $Ag-Mg$ alloys near Ag_3Mg [8].

Although the afore-mentioned data could be invoked to give support to the hypothesis on impurity quasi-regularity, one should finally point out that all these experiments were not conducted primarily to maximize the effect we discussed here. To get the highest critical temperature T_c , samples in the normal state ($T \gg T_c$) should be cooled down very slowly to temperatures $T \ll T_c$ (such that to allow impurities to follow the Peierls-CDW distortion), then quickly heated (in order to preserve as much impurity quasi-regularity as possible) up to the critical temperature T_c . Similarly, the lowest value of the critical temperature $T_{c,r}$ could be obtained by fast cooling the samples from temperatures $T \gg T_{c,r}$ to the transition region. A more detailed analysis of the various experimental data is deferred to a subsequent publication.

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