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a.c. CONDUCTIVITY OF NEARLY COMMENSURATE CHARGE DENSITY WAVES AND APPLICATION TO $NbSe_3$ AND TaS_3

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The a.c. conductivity of an overdamped one dimensional sine-Gordon system with a low density of kinks is evaluated. This corresponds to a nearly commensurate charge or spin density wave with a kink-lattice representing the deviation from commensurability. The results explain the unusually broad crossover regime of $NbSe_3$ as compared with the similar but more commensurate compound TaS_3. When TaS_3 becomes commensurate ($T \lesssim 130$ K) we predict that its a.c. response increases and its shape slightly sharpens.

THE PHENOMENON of charge density wave (CDW) transport in $NbSe_3$ and TaS_3 is of considerable recent interest [1]. $NbSe_3$ exhibits two CDW transitions at $T_1 = 142$ K and $T_2 = 59$ K with the wavevectors [2] $q_1 = (0, 0.2412, 0)$ and $q_2 = (0.5, 0.2604, 0.5)$ respectively. The conductivity is non-ohmic when the electric field E exceeds a critical value E_c and well-defined frequencies in the noise spectrum appear in a d.c. field. These phenomenon are associated with depinning of the CDW, which is pinned by either impurities [3] or by the lattice [4].

The theory for lattice pinning is based on the values of q_1 (or q_2) being almost commensurate with a lattice reciprocal wavevector. To gain the commensurability energy the CDW is mostly commensurate except for a lattice of kinks (solitons) or discommensurations [5, 6] which carry the excess charge $\Delta q_y/\pi = q_y/\pi - 1/2b$, where b is the lattice constant. Since the charge of a single kink is [6] 1/2 the kink density is $n_k = -0.035/b$ and $n_k = 0.042/b$ for the two CDW's respectively; thus the distance between neighbouring kinks is $28b$ and $24b$ respectively.

Recent data on TaS_3 (in its orthorhombic phase) shows [7] that it is closer to being commensurate with $n_k = 0.02/b$ below 210 K, and becomes commensurate ($n_k = 0$) below 130 K. This system exhibits similar non-ohmicity and frequency generation [7, 8] as $NbSe_3$. TaS_3 is therefore an excellent candidate for studying the effect of commensurability on both linear and non-linear response. In particular the critical field E_c is smaller in the commensurate phase [7], in agreement with the model for lattice pinning [4].

Here we are mainly concerned with the CDW linear response to an a.c. electric field. The qualitative features of the a.c. response have been interpreted by an overdamped oscillator in a periodic potential [9–11]. The oscillator degree of freedom describes the oscillation of rigid CDW, neglecting internal degrees of freedom. This model, however, does not fit the rather broad crossover region of $NbSe_3$ while it fits better the data of TaS_3 [12].

The rigid CDW model corresponds, in the context of lattice pinning, to an Mth order commensurate system, i.e. the ratio of CDW wavelength to the underlying lattice constant in an integer M. The oscillator degree of freedom is a phase variable $\psi(t)$ such that the CDW has the form $\sim \cos[(2\pi x/b + \psi)/M]$. The phase variable is sufficient to describe charged configurations for $M \geqslant 3$ [6].

$NbSe_3$ and TaS_3 are almost $M = 4$ systems. The use of phase variable is then justified with a constraint to describe the deviation from commensurability. This constraint means that ψ is space dependent such that its average gradient reproduces the correct incommensurate wavevector. This space dependence is achieved by a phase-kink lattice [5, 6].

In the present paper we extend the oscillator model to include the internal degrees of freedom. The CDW is then described by a space and time dependent field $\psi(x, t)$. We find that the presence of phase kinks broadens the crossover region of the a.c. response. This is in agreement with data on both $NdSe_3$ and TaS_3, i.e. TaS_3 being closer to commensurability has a lower kink density and therefore fits better the single oscillator model [9–12]. From a fit to the $NbSe_3$ data one can determine the coherence length ξ (or kink width). This

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length is not available from other data since the electron gap is not yet known.

The equation of motion for ψ in the presence of damping, commensurability and an expectric field $E\exp(i\omega t)$ is given by [4]

$$\frac{\ddot{\psi}}{\omega_0^2}+\frac{\dot{\psi}}{\omega_c}-\xi^2\psi''+\sin\psi = Ee^{i\omega t}/E_1, \qquad (1)$$

where dot is $\partial/\partial t$ and prime is $\partial/\partial x$; ω_0 is the pinning frequency, ω_c the crossover frequency and E_1 is the depinning field *if* the system were commensurate. (In general $E_c \geqslant E_1$ [4].) The kink density imposes the boundary condition $\int \psi'(x,t)\,dx/L = 2\pi n_k$ where L is the length of the system. The kinks are considered to be two dimensional walls which are deformable along the chain direction x. Thermal fluctuations are not included since these walls are macroscopic objects.

We are interested in situations where the density of kinks is small enough to ignore their mutual interaction, i.e. $\bar{n}_k \equiv n_k\xi \ll 1$. In this limit each kink contributes independently, and by the same amount, to the a.c. response of the system. (If antikinks are present, they contribute to the a.c. current with the same sign as kinks since having opposite charge [6] they oscillate 180° out of phase with respect to the kinks). The current is defined by [4]

$$J = e\rho_{\text{eff}}\langle\dot{\psi}\rangle/(4\pi S), \qquad (2)$$

where ρ_{eff} is the fraction of the total electron density which participates in the CDW transport, S is the area per conducting chain and $\langle\dot{\psi}\rangle$ is the space average of $\partial\psi/\partial t$. We derive the dimensionless conductivity $\bar{\sigma}(\omega) = \langle\dot{\psi}\rangle E_1/[E\omega_c\exp(i\omega t)]$ so that the actual conductivity is

$$\sigma(\omega) = A\bar{\sigma}(\omega); \qquad A = e\rho_{\text{eff}}\omega_c/(4\pi SE_1). \qquad (3)$$

We employ the perturbation theory developed by Fogel *et al.* [13, 14] to expand around the single soliton solution $\psi_s(x) = 4\tan^{-1}[\exp(x/\xi)]$,

$$\psi(x,t) = \psi_s(x)+\chi_T(t)\psi_s'(x)+\chi(x,t), \quad (\chi_T,\chi \ll 1). \qquad (4)$$

Here $\chi_T(t)$ represents the amplitude of the translation mode ($\psi_s'(x)$) which is specifically separated from the remaining response, with the constraint

$$\int \psi_s'(x)\chi(x,t)\,dx = 0. \qquad (5)$$

With this separation, $\chi(x,t)$ contains the kink shape oscillations and the background response [13–16].

Substitution of equation (4) into equation (1) and subsequent linearization in $\chi_T(\tau)$ and $\chi(z,\tau)$ yields

$$(\ddot{\chi}_T\psi_s'+\ddot{\chi})/\Gamma^2+\dot{\chi}_T\psi_s'+\dot{\chi}-\chi''+(\cos\psi_s)\chi = $$
$$= E\,e^{i\Omega\tau}/E_1, \qquad (6)$$

where $\Gamma = \omega_0/\omega_c$, $z = x/\xi$, $\tau = t\omega_c$ and $\Omega = \omega/\omega_c$. The eigenfunctions $f(z)$ satisfying the Schroedinger type equation

$$\left[-\frac{d^2}{dz^2}+\cos\psi_s(z)\right]f(z) = \epsilon^2 f(z), \qquad (7)$$

form a complete set consisting of a single "bound state" (translation mode)

$$f_b(z) = \psi_s'(z), \qquad \epsilon_b^2 = 0, \qquad (8a)$$

and a continuum of extended modes,

$$f_k(z) = \frac{e^{ikz}}{\sqrt{L/\xi}\,\epsilon_k}(k+i\tanh z), \qquad \epsilon_k^2 = 1+k^2. \qquad (8b)$$

Therefore, if $\chi(z,\tau)$ is expanded in continuum modes,

$$\chi = \sum_k \chi_k(\tau)f_k(z), \qquad (9)$$

then it is automatically orthogonal to $f_b = \psi_s'(z)$ as required by equation (5).

We now multiply equation (6) by $\psi_s'(z)$ and integrate over z to obtain

$$\chi_T(\Omega) = \frac{\pi E}{4E_1}\frac{e^{i\Omega\tau}}{i\Omega-(\Omega/\Gamma)^2}. \qquad (10)$$

The space average yields a factor of $2\pi n_k\xi$ and hence the translation mode contribution to the conductivity is

$$\bar{\sigma}_T(\Omega) = \frac{\pi^2}{2}\frac{\bar{n}_k}{1+i\Omega/\Gamma}. \qquad (11)$$

In the strongly overdamped limit ($\Gamma \gg 1$) and in the frequency range of experimental interest [9] ($\Omega \ll \Gamma$)$\bar{\sigma}_T$ is *independent* of frequency, $\bar{\sigma}_T \simeq \frac{1}{2}\pi^2\bar{n}_k$.

The more interesting (i.e. frequency dependent) contribution to $\sigma(\Omega)$ is that due to the continuum of extended modes $f_k(z)$. These modes define the Greens' function for the propagation of $\chi(z,\tau)$ which after a frequency integration yields the continuum part of the conductivity

$$\bar{\sigma}_c(\Omega) = (\xi/L)\sum_K\left|\int f_k(z)\,dz\right|^2 \epsilon_k^2F(k), \qquad (12)$$

where

$$\epsilon_k^2F(k) = \frac{i\Omega}{\epsilon_k^2+i\Omega-(\Omega/\Gamma)^2}. \qquad (13)$$

The background oscillations contribute a term of order L to the sum in equation (12). To separate this term we subtract from equation (12) (using the completeness relation [14] of $\{f_k, f_b\}$)

$$(\xi/L)F(0)\sum_K\left|\int f_k(z)\,dz\right|^2 = F(0)(1-\pi^2/2L). \qquad (14)$$

The non-trivial perioidic boundary conditions on $f_k(x)$

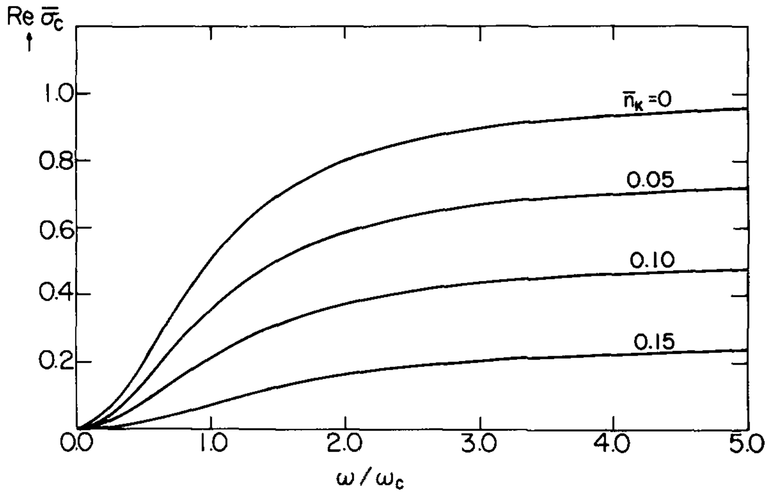

Fig. 1. Real part of the frequency dependent conductivity, excluding the frequency independent conductivity of the kink translation mode. $\bar{\sigma}_c = \bar{\sigma} = \bar{\sigma}_T$. The curves are for various values of the kink density $\bar{n}_k = n_k\xi$.

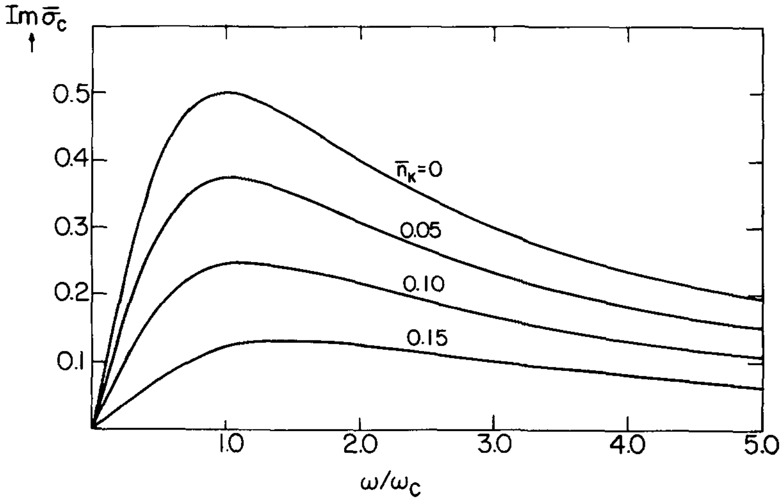

Fig. 2. Same as Fig. 1 for the imaginary part.

[14] yield

$$\int f_k(z)\,\mathrm{d}z = -(\xi/L)^{1/2}\pi(\epsilon_k \sinh \tfrac{1}{2}\pi k)^{-1} \tag{15}$$

with which the remainder of equation (12) is easily calculated. Finally, replacing $1/L$ by n_k (each kink contributes equally) results in

$$\bar{\sigma}_c(\Omega) = (1-2\bar{n}_k)F(0) + 2\bar{n}_k \int_0^\infty \frac{\partial_k F(k)}{\tanh \frac{1}{2}\pi k}\,\mathrm{d}k. \tag{16}$$

In the strong damping limit this reduces to

$$\mathrm{Re}\ \bar{\sigma}_c(\Omega) = \frac{\Omega^2}{1+\Omega^2}(1-2\bar{n}_k) - 4\bar{n}_k\Omega^2 \times \int_0^\infty \frac{3\epsilon_k^4+\Omega^2}{\epsilon_k^4(\epsilon_k^4+\Omega^2)^2}\frac{k\,\mathrm{d}k}{\tanh\frac{1}{2}\pi k}$$

$$\mathrm{Im}\ \bar{\sigma}_c(\Omega) = \frac{\Omega}{1+\Omega^2}(1-2\bar{n}_k) - 8\bar{n}_k\Omega \times \int_0^\infty \frac{\epsilon_k^2}{(\epsilon_k^4+\Omega^2)^2}\frac{k\,\mathrm{d}k}{\tanh\frac{1}{2}\pi k}. \tag{17}$$

Before discussing these results let us present an alternative derivation which exploits the previously calculated kink polarizability [13], defined as

$$\alpha_s(\Omega) = (E\,\mathrm{e}^{i\Omega\tau}/E_1)^{-1}\int \mathrm{d}z\, z\partial_z\chi(z,\tau). \tag{18}$$

Integrating equation (18) by parts and using $\dot{\chi} = i\Omega\chi$ we find the current $\langle\dot{\chi}\rangle$ except for a surface term, i.e.

$$\langle\dot{\chi}\rangle = i\Omega\chi_B(\tau) - i\Omega\bar{n}_k\int \mathrm{d}z\, z\partial_z\chi(z,\tau) \tag{19}$$

where

$$\chi_B(\tau) \equiv \chi(\pm\infty, \tau) = \frac{E\, e^{i\Omega\tau}/E_1}{1 + i\Omega - (\Omega/\Gamma)^2}. \tag{20}$$

The surface term (20) achieves the desired separation of the current equation (19) into a background term and to the kinks' contribution. The continuum part of the conductivity is then

$$\bar{\sigma}_c(\Omega) = \frac{i\Omega}{1 + i\Omega - (\Omega/\Gamma)^2} - i\Omega \bar{n}_k \alpha_s(\Omega). \tag{21}$$

The kink polarizability $\alpha_s(\Omega)$ can be written in the overdamped limit as [13]

$$\alpha_s(\Omega) = \frac{\pi}{2i\Omega} + \frac{2\ln 2}{1 + i\Omega} + \frac{\pi[1 - (i/Q) + \beta(Q/2i)]}{i\Omega Q \sinh(\pi/2)Q} + 4\sum_{n=1}^{\infty} \frac{(-1)^n[\beta(n) + 1 - (1/2n)]}{(4n^2 - 1)(1 - 4n^2 + i\Omega)}, \tag{22}$$

where

$$Q = -\frac{1}{\sqrt{2}}\{(1 + \Omega^2)^{1/2} - 1\}^{1/2} + \frac{1}{\sqrt{2}}\{(1 + \Omega^2)^{1/2} + 1\}^{1/2} \tag{23}$$

and $\beta(x)$ is given by the series [17]

$$\beta(x) \equiv \sum_{k=0}^{\infty} \frac{(-1)^k}{x + k}. \tag{24}$$

For these formula can be evaluated numerically in a straightforward manner and we present results for Re $\bar{\sigma}_c(\Omega)$ and Im $\bar{\sigma}_c(\Omega)$ in Figs. 1 and 2, respectively, for a few values of the kink density $\bar{n}_k$. Note the rather large effect of $\bar{n}_k$ even for values as small as 0.1; our assumption of non-interacting solitons becomes suspect for values of $\bar{n}_k$ much above this. With some straightforward but tedious algebra, it is possible to obtain the limiting forms of $\sigma_c(\Omega)$ for low and high frequencies:

$$\Omega \to 0: \quad \begin{matrix} \text{Re}\ \bar{\sigma}_c(\Omega) \to \Omega^2(1 - 6.300\bar{n}_k) \\ \text{Im}\ \bar{\sigma}_c(\Omega) \to \Omega(1 - 5.674\bar{n}_k) \end{matrix} \tag{23a}$$

$$\bar{\Omega} \gg 1: \quad \begin{matrix} \text{Re}\ \bar{\sigma}_c(\Omega) \to 1 - \tfrac{1}{2}\pi^2\bar{n}_k \\ \text{Im}\ \bar{\sigma}_c(\Omega) \to \Omega^{-1}(1 - 4\bar{n}_k). \end{matrix} \tag{23b}$$

It is worth noting that for high frequencies, the total conductivity $\bar{\sigma} = \bar{\sigma}_T + \bar{\sigma}_c$ approaches unity, independent of $\bar{n}_k$. This can be seen directly from equation (1) since the $\sin\psi$ term becomes ineffective at high frequencies.

Before discussing $NbSe_3$ and TaS_3, we note that our results are also relevant for highly one dimensional systems where kinks can be thermally activated. In that case n_k is the density of kinks *plus* antikinks and is temperature dependent.

We have not yet attempted a detailed for to the data of $NbSe_3$ since the data is not complete, mainly in the high frequency regime. However, the main feature of the data, namely the unusually broad crossover regime, is consistent with the presence of kinks. As can be seen from equation (17) the kinks introduce a continuous range of crossover frequencies at $\omega = \omega_c(1 + k^2)$ where $0 < k < \infty$. The underlying reason is that the kinks break the translation invariance of the system and then all wavevectors k can couple to the spatially uniform electric fields. The increase of the crossover regime with n_k is also clear from Figs. 1 and 2.

To obtain an estimate of the parameters involved at $T = 42$ K we used $\omega_c \simeq 400$ MHz [1, 9], the high frequency limit of Re $\sigma_c/\sigma_{\text{d.c.}} \simeq 1.1$ ($\sigma_c(\omega) = \sigma(\omega) - \sigma_{\text{d.c.}}$ is the frequency dependent part) and the low frequency limit of Im $\sigma_c/\sigma_{\text{d.c.}} \simeq 0.6\,\omega/\omega_c$ [9, 11]. Using equations (3) and (23) we find $\bar{n}_k \simeq 0.1$, while the overall data [12] is consistent with $\bar{n}_k = 0.1$–0.2. Since $n_k \simeq 0.04/b$ the kink width is $2\xi \simeq 5b - 10b$. Note that the curves in Figs. 1 and 2 when rescaled to the same height become less distinct. Therefore a detailed fit is more difficult when the parameter A [equation (3)] is unknown.

The a.c. conductivity of TaS_3, in contrast with that of $NbSe_3$, fits well the overdampled oscillator result [12]. This is consistent with TaS_3 being closer to commensurability with $n_k = 0.02/b$ [7].

A distribution of ω_c values, representing random impurities, may also explain the broad corssover region in $NbSe_3$ [9]. This, however, is inconsistent with the TaS_3 data, since TaS_3 has more impurities than $NbSe_3$ [8] and yet its crossover region is narrower. Thus the deviation from commensurability seems to be the most relevant parameter which can explain the different AC response of $NbSe_3$ and TaS_3.

The recently discovered commensurate–incommensurate transition in orthorhombic TaS_3 at ~ 130 K [7] provides a remarkable opportunity of studying the effects of commensurability on both nonlinear [4] and linear response. In the commensurate phase ($T \lesssim 130$ K) we predict an increase of both real and imaginary parts of the a.c. response (see Figs. 1 and 2). This is due to the frequency independent contribution of the kink translation mode [equation (11)] which disappears in the commensurate phase. The d.c. conductivity is then reduced while the a.c. response is increased. Other effects however may also modify the intensity of the a.c. response – an increase in the number of conducting chains [7] [reducing S in equation (3)] or a change in the gap which affects E_1. Independent measurements of these parameters (e.g. by crystallography and infra-red absorption) are necessary before our prediction on the intensity of the a.c. response can be tested.

We also predict a slight sharpening of the frequency dependence in the commensurate phase.

Unlike the overall intensity, this feature is uniquely related to the absence of phase kinks in the commensurate phase.

In conclusion, our results account for the unusually broad crossover regime in the a.c. response of $NbSe_3$ as compared with that of TaS_3. Further data on TaS_3 at various temperatures, corresponding to different phase-kink densities, is extremely important for elucidating the role of commensurability in these sliding CDW compounds.

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