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Narrow-band "noise" and damping in NbSe₃

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Abstract - We report the results of a Narrow-band "Noise" (NBN) study, in NbSe₃ for magnetic fields up to 19 T and temperatures as low as 15 K. The first result of this NBN study is the observation of the decrease of dJ_{CDW}/dv with decreasing temperatures for both transitions. We report also new values for the absolute value dJ_{CDW}/dv for both transitions, and the existence of a curvature of dJ_{CDW}/dv versus frequency, in low frequency range. We have interpreted the decrease of dJ_{CDW}/dv with temperatures in terms of Artemenko's theory which has been used in 1984 to explain the non-linear Hall effect in TaS₃. The total longitudinal current is given by $J_T = J_{CDW}(1-A) + \sigma_1 E$, where the term $-AJ_{CDW}$ is a backward normal current flow due to the moving CDW. From the saturating value of dE/dv obtained in high electric field we have deduced the total damping parameter τ^{-1} which is decreasing with temperature. Using data obtained under application of a magnetic field of 7 T and the Wonneberger's theory for the ac response of the collective mode, we have deduced the thermal variation of the bare phason damping coefficient τ_0^{-1} . We report that for $T < 30\text{K}$, $\tau_0^{-1} \sim AT^{4.5}$ very close to the theoretical predictions of Takada, Wong and Holstein.

Among all low-dimensional conductors which are known to display "sliding" (CDW's) conduction, NbSe₃ is certainly the most widely studied in terms of CDW dynamics [1]. NbSe₃, which undergoes two CDW transitions ($T_1 = 145\text{ K}$; $T_2 = 59\text{ K}$), is the only one to remain semimetallic at all temperatures. A striking phenomenon in CDW systems is the presence of a radio frequency voltage [2], improperly referred to as narrow band "noise" (NBN), when the dc bias exceeds a threshold electric field E_T . The oscillations are the hallmark of the sliding conduction of the CDW, with a fundamental frequency ν_0 shown to be proportional to the excess current density J_{CDW} carried by the CDW. From the ratio $P = J_{CDW}/\nu_0$ the number of condensed carriers can be inferred [3]. NBN has been studied extensively, but up to now no accurate measurements have been reported for low temperatures ($T < 90\text{ K}$ for the first transition, and $T < 25$ for the second transition) and frequencies higher than 30 MHz.

We report on extremely precise NBN experiments made over the range $65\text{ K} < T < 150\text{ K}$ for the first transition and $15\text{ K} < T < 55\text{ K}$ for the second transition.

Making NBN measurements at low temperature (compared with T_1 or T_2) we are faced with the problem of removing sample heating effects, due to the application of high DC electric fields in order to depin the CDW. To reduce sample Joule heating, we have performed four probe resistance measurements by the pulsed technique, with the sample immersed in a cryogenic liquid such as Argon or Nitrogen for the first transition, Neon or Hydrogen for the second transition. Using high quality samples with minimal threshold fields (9 mV/cm at 48 K) we were able for the first time to do Fourier analysis of the current oscillations for samples depinned by a pulsed current. With this technique, pulsed widths of 50 μs to 1 ms have been used with duty cycles of 10^{-4} .

Most of the NBN studies in NbSe₃ so far reveal a linear relation between J_{CDW} and ν_0 over the usual frequency range 0-30 MHz. A few measurements have been made up to 400

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MHz but only at a few temperatures. NbSe₃ samples which exhibit a non linear relation between J_{CDW} and ν_0 have been considered to be poor quality samples. Surprisingly, in orthorhombic or monoclinic TaS₃ [5] and (NbSe₄)_{10/3} [6] curvature can always be found in some specific temperature range. In order to improve our sensitivity, we have plotted the experimental derivative $dJ_{CDW}/d\nu_0$ versus frequency ν_0 . Our results shown in Fig. 1 indicate that even for high quality samples with coherent oscillations, non linearity is present at 45 K for $\nu_0 < 30$ MHz. Unlike TaS₃, the curvature is more prominent at lower temperatures for both transitions. We also have performed NBN measurements on (TaSe₄)₂I and observed curvature, even at 160 K, but in the frequency range 0-3 MHz. We argue, therefore, that the curvature exists for all samples exhibiting NBN, its amplitude is very temperature dependent, but for high enough frequency the linearity between J_{CDW} and ν_0 will be recovered. For NbSe₃ almost all previous measurements made below 120 K and 25 K give an underestimate of the ratio J_{CDW}/ν_0 .

The linearity observed in previous measurements is often the consequence of a too small range of frequency (0-30 MHz), an example is given on the insert of Fig. 2.

Fig. 3 shows the result of a temperature dependent study for both transitions [4]. Each data point is actually the saturated value $P = dJ_{CDW}/d\nu_0$ after correction of the heating effects. Studies have been carried out on more than 20 crystals and the overall behaviour is not sample dependent. P_1 (the average value on 14 samples) is equal to 40.2 A MHz⁻¹ cm⁻² [8] and $0.75 P_1 < P_2 < 0.85 P_1$, depending of the sample. The previous results have been evaluated in the regime where P_1 or P_2 do not saturate which explains the scatter of the data published by various authors [1]. With the above value for P_1 we deduce for λ pinning λ_{CDW} a value of 0.93, this is very close to 1, in agreement with the classical model, as already observed in TaS₃ [7]. The central result of our study is the observation of the decrease of $dJ_{CDW}/d\nu_0$ with temperature for both transitions, strongly at variance with various theories.

For the second transition we have performed NBN measurements under application of magnetic fields. In Fig. 4 we plot the saturated value $dJ_{CDW}/d\nu_0$, after correction of heating effects, versus temperature, at $B = 0$ and $B = 7T$.

Magnetic field has only a small effect (4 - 7 %) on the slope P_2 for temperatures below 45 K. For temperatures above 45 K any effect of the magnetic field is lost in our experimental resolution ($\sim 1\%$).

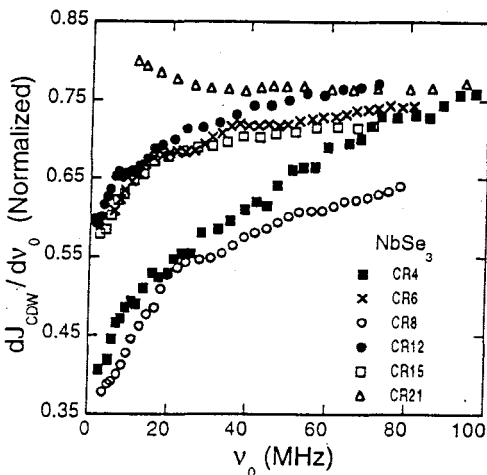


Figure 1

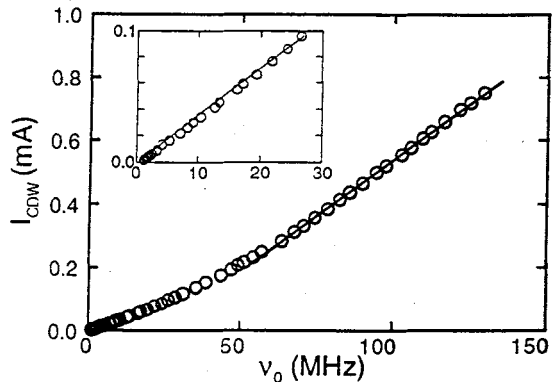


Figure 2

Figure 1 : Experimental slope $dJ_{CDW}/d\nu_0$ (normalized for each sample to the maximum value obtained at the first transition) versus frequency for different samples at $T = 45$ K.

Figure 2 : I_{CDW} versus frequency at $T = 36$ K. The data show that the linearity obtained in low frequency range (see insert) is misleading, as shown by the measurements made in the higher frequency range.

Concerning the interpretation of the decrease of P as T falls we favor the idea that the independent two fluid model is not strictly valid. Restrictions to this model have already been published by different authors (Boriak-Overhauser, Lee and Rice [9]). In 1984, Artemenko et al. explained the non linear Hall effect observed in TaS_3 [10], by taking into account the effect of a moving CDW on the distribution of quasiparticles. For NbSe_3 which is still metallic below the Peierls transitions, we must consider a slightly different model with electronic pockets. Unlike semiconducting Peierls materials the imperfect nesting parameter ϵ_0 is not very small compared to Δ . Due to the 2D pockets we must now define $\delta_{1,2}$ ($\delta \ll \Delta$), the energy difference between the Fermi level and the energy in the minimum and the maximum, respectively. To obtain the current, a linearized kinetic equation for a quasiparticle distribution function has been solved. With $T < \Delta$ detailed calculations can be found for semiconducting Peierls compounds in Ref. 11. If we consider the simple symmetric case ($\delta_1 = \delta_2$), where there is no anisotropy in the effective mass, the total longitudinal current is given by :

$$J_T = J_{\text{CDW}}(1 - A F(d)) + \sigma_1 E, \quad (1)$$

where

$$A = (1+2\alpha^{-1})^{-1} \delta / \sqrt{\epsilon_0 \Delta}, \text{ and } F(d) = \ln d^{-1} (1+e^d) + (1+\alpha)^{-1} \text{th } d/2,$$

where $\alpha = v_b/v_f$, $\delta = \delta_1 + \delta_2$, $d = \delta/2T$, σ_1 is the quasiparticle conductivity and v_b and v_f are backward and forward inverse scattering time respectively.

Due to the moving CDW a backward normal current flow, proportional to J_{CDW} , ($J_B = -A F(d) J_{\text{CDW}}$) is added to J_{CDW} and to the contribution of the quasiparticles. The presence of J_B results in non-monotonous temperature dependence of that part of the current which is proportional to the CDW velocity and hence to the NBN frequency. In NbSe_3 , J_B can be large enough to be taken into account, unlike semiconducting Peierls materials where J_B is exponentially small at $T < \Delta$.

The possibility of non-monotonous temperature dependence of J_B for semiconducting CDW material without electronic pockets for high temperature $T \gtrsim \Delta$ was shown by Maki and Huang [12]. The non-monotonous behaviour of J_B , given by equation (1) is less pronounced than the experimentally observed one. A fit to equation (1) has not too much physical signification due to the fact that we neglect the mass anisotropy and temperature dependence of ϵ_0 , δ and α . In the regime when $T < \delta$ we can approximate the back flow current by :

$$J_B/J_{\text{CDW}} = -4n \epsilon_f (\alpha^{-1} + 2^{-1})^{-1} / N\Delta \quad (2)$$

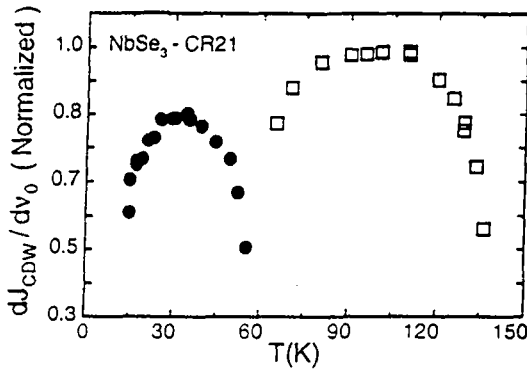


Figure 3

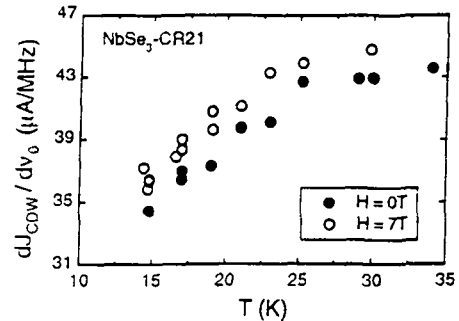


Figure 4

Figure 3 : Temperature dependence of dJ_{CDW}/dv_0 , for both transitions normalized to the maximum value of the first transition.

Figure 4 : Temperature dependence of dJ_{CDW}/dv_0 for the second transition with and without an applied magnetic field.

where ε_F is the Fermi energy, n the number of quasiparticles and N the total number of carriers for the second transition. Using the estimated parameters, $\Delta_0 = 350$ K, $\varepsilon_F = 4200$ K, $N = 2 \times 10^{21}$ carriers/cm³ and $\alpha = 1$, We obtain the experimental extrapolation at 4 K ($J_B/J_{CDW} = -0.45$) for $n = 1.9 \times 10^{19}$ which is in agreement with Hall effect measurements ($\ln_0 - p_0 = 7 \times 10^{18}$ carriers/cm³). The effect of the magnetic field on dJ_{CDW}/dv_0 is interpreted in terms of a magnetoresistance effect on J_B rather than an increase of n .

The interaction of the moving condensate with thermally ambient phonons or phasons and with the uncondensed electrons leads to an effective damping parameter τ^{-1} and a viscosity η , with $\eta \propto \tau^{-1}$. Fleming *et al.* [13] have observed an Arrhenius behavior of the excess CDW current I_{CDW} in different materials ($K_{0.3}MoO_3$, TaS_3 , $(TaSe_4)_2I$). These results suggest that the zero-frequency viscosity of the CDW carriers diverges at low temperatures.

A classical starting point for a description of the CDW dynamics has been a phenomenological single-particle description.

For frequencies $\nu < 500$ MHz the inertial term can be neglected compared to the damping term. The electric field is given by :

$$E = E_T + \eta \frac{V_{CDW}}{Ne} \quad (3)$$

where E_T is the threshold electric field, V_{CDW} the CDW velocity, η the effective viscosity with $\eta = m^*N/\tau$. As $V_{CDW} = \lambda_{CDW}\nu$, where ν is the frequency which is obtained from NBN measurements, the consequence of equation (3) would be a linear relation between the electric field and the frequency ν , which is not experimentally shown in small electric field. In high electric field we have experimentally observed a saturation for $dE/d\nu$ which allows us to deduce τ^{-1} :

$$\frac{1}{\tau} = \frac{e}{\lambda_{CDW}m^*} \left. \frac{dE}{d\nu} \right|_{\infty} \quad (4)$$

(The symbol ∞ , for $dE/d\nu$, means that we have taken the saturated values obtained in our experimental electric field limit). Our results will be now analysed in the light of the theory for the ac response of the collective mode in CDW systems which has been elaborated by W. Wonneberger [14]. Extending earlier work of Littlewood [15], Wonneberger's theory is based on an extended Fukuyama-Lee-Rice (FLR) phase approach to the ac conductivity including quasi particle effects. One important advantage of this theory is that it provides a tractable analytical formula for $\sigma(\omega)$. In the weak pinning limit the formula for $\sigma(\omega)$ contains four frequencies : bare phason damping τ_0^{-1} , longitudinal optical phason frequency ω_{LO} with $\omega_{LO} = 4\pi Ne^2/m^*\varepsilon_{\Delta}$, dielectric relaxation frequency of quasi particles ω_r , with $\omega_r = (4\pi/\varepsilon_{\Delta})\sigma_{qp}$, and weak pinning frequency ω_w . The quantity ε_{Δ} is the dielectric constant from the Peierls gap, σ_{qp} is the quasi particle conductivity. Besides ω_w they all can be inferred from experiments.

In the low frequency range or DC limit, Wonneberger has suggested [16] to take for τ^{-1} :

$$\tau^{-1} = \tau_0^{-1} + \left(\frac{1}{16\pi} \frac{\omega_w}{\omega_{LO}} \sqrt{\frac{\varepsilon_{CDW}(0)}{\varepsilon_{\Delta}}} \right) \left(\tau_0^{-1} + \frac{\omega_{LO}^2}{\omega_r} \right) \quad (5)$$

Using equation (4) for τ^{-1} equation (5) can be rewritten as :

$$\left. \frac{dE}{d\nu} \right|_{\infty} \frac{e}{\lambda_{CDW}} = \frac{Ne^2}{\sigma_M} + B\sqrt{m^*Ne^2} [\sigma_M^{-1} + \sigma_{qp}^{-1}] \quad (6)$$

where B is a constant depending on ω_w and $\varepsilon_{CDW}(0)$

We make the hypothesis that, at least in a first approximation, in the right side of equation (6) σ_{qp}^{-1} is the only term magnetic field dependent. In equation (6) we can introduce $dE/d\nu|_{\infty}$ obtained in measurements without and with magnetic field (7 T), this enables us to calculate $B\sqrt{m^*}$ and then σ_M .

To calculate τ_0^{-1} we must know m^* . The order of magnitude has been given by Reagor *et al.* (17) Using more recent values for N_T and N_0 ($N_T = 3.9 \times 10^{21}$ carriers/cm³, $N_0 = 1.66 \times 10^{21}$ carriers/cm³) [1], we have obtained $m^*/m_e = 63$.

We have assumed below 40 K $N/m^* = N_0/m^*$ with $m^*/m_e \approx 60$, and have deduced the thermal variation of τ_0^{-1} (see Fig. 5). Above 30 K the thermal variation of τ_0^{-1} could be in agreement with $\tau_0^{-1} \propto AT$ deduced by Reagor *et al.* [17], but below 30 K we have deduced $\tau_0^{-1} \propto A_1 T^{4.4}$. Up to now the only precise theoretical prediction for the thermal variation of the bare phason damping was the calculation of Takada, Wong and Holstein, TWH [18].

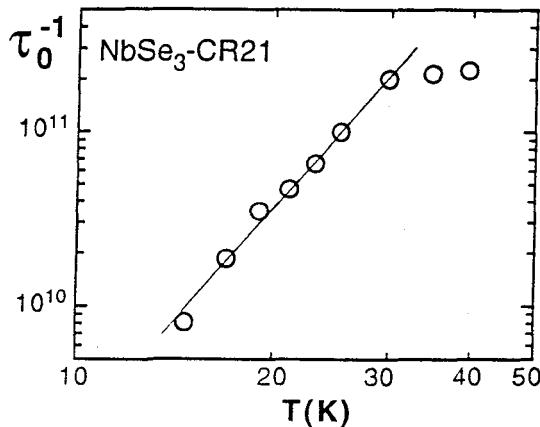


Figure 5 : Temperature dependence of the "bare" phason damping τ_0^{-1} . The solid line is a fit to $\tau_0^{-1} = A_1 T^{4.4}$.

According to these authors the dominant contribution arises from scattering with thermally ambient phasons. Damping due to impurities and normal electrons is neglected. In the low temperature regime $T < \theta_D$ (θ_D : Debye temperature, $\theta_D \approx 100$ -150 K in NbSe₃) TWH have predicted :

$$\tau_0^{-1} = \frac{16\pi^7}{225\mu} \frac{\lambda^2}{\Omega_t^4} T^5$$

where λ is the electron-phonon-coupling constant, $\mu = m^*/m_b$, with m_b the band mass and Ω_t is the transverse-phonon frequency. The T^5 dependence for τ_0^{-1} is roughly observed in the present experiment. Regarding magnitude, using $\lambda = 0.24$, $\mu = 380$ and $\Omega_t = 35$ K the theory yields $\tau_0^{-1} = 3 \times 10^{10}$ Hz at $T = 25$ K a factor of 3 lower than the experimental results. Bearing in mind all the interlocking uncertainties concerning the basic parameters which enter into the theoretical expression for τ_0^{-1} , a disagreement between theory and experiment by a factor of 3 is hardly surprising.

In conclusion of this study, we have shown that for the first time, from NBN measurements up to 400 MHz, we are able, first to measure the total damping constant τ^{-1} , and then using the Wonneberger's theory to deduce the low temperature variation of the bare phason damping constant τ_0^{-1} which seems to verify the prediction of TWH : $\tau_0^{-1} = A_1 T^5$.

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