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Charge Density Waves in Solids

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AC CONDUCTIVITY OF THE BLUE BRONZE $K_{0.3}MoO_3$

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Abstract: We have measured the low field ac conductivity of the blue bronze $K_{0.3}MoO_3$ in the charge density wave (CDW) state. For temperatures above 70K and over an extended frequency range, our results are consistent with the model proposed by Cava et. al. Below 70K, however, there appears at high frequencies an additional relaxation mechanism. Associated with this new mode are unusual hysteresis effects and an ac conductivity similar to that found in TaS_3 .

The low field ac conductivity of $K_{0.3}MoO_3$ is currently the subject of much interest. We have performed careful low field ac conductivity measurements in the frequency range 10 Hz to 2.3 GHz. Figure 1 shows the ac conductivity as function of frequency for several samples of $K_{0.3}MoO_3$ at two temperatures. At the higher temperature, 77K, the conductivity is well described by $\sigma(\omega) = i\omega\epsilon(\omega)$, where[1]

(1) $$\epsilon(\omega) = \epsilon_\infty + (\epsilon_0 - \epsilon_\infty)[1 + (i\omega\tau_0)^{1-\alpha}]^{-\beta}$$

In figure 1a, a fit to this expression is indicated by the solid and dashed lines for Re $\sigma(\omega)$ and Im $\sigma(\omega)$ respectively. Note that the characteristic relaxation time, τ_0 , is unusually high. This value of τ_0 corresponds to a characteristic pinning frequency $\omega_1 = 1/\tau_0$, several orders of magnitude lower than characteristic frequencies seen in $NbSe_3$[2] and TaS_3[3].

As the temperature of the material is lowered, ω_1 moves to still lower frequencies. Figure 1b shows the ac conductivity at 42K. As indicated by the low frequency crossing of Re σ and Im σ, the characteristic frequency is now below 1kHz. Because of the

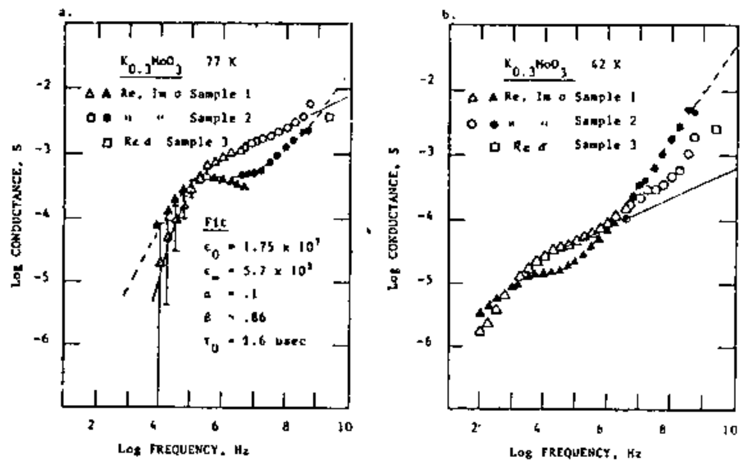

Figure 1: The impedance of $K_{0.3}MoO_3$ at 77K and 42K.

uncertainty in the data below 1kHz, only the high frequency limit of equation 1 is fitted. For $\omega\tau_0 \gg 1$, the real and imaginary components of $\sigma(\omega)$ obey simple power laws:

(2) $\quad \mathrm{Re}\, \sigma(\omega) \sim \omega^{1-\beta(1-\alpha)}$

$\quad \mathrm{Im}\, \sigma(\omega) \sim \omega$

There are clear departures from equation 2 in the 42K data. The real component of $\sigma(\omega)$ goes as $\omega^{.16}$ between 10kHz and 1MHz, but then curves away from this behavior at higher frequencies. Im $\sigma(\omega)$ does not go as $\omega^{1.0}$, but rather as $\omega^{0.9}$ at high frequency. To explain these departures from equation 2, we look at $\sigma_{HF}(\omega) = \sigma_{CDW}(\omega) - \sigma_{LF}(\omega)$, where $\sigma_{LF}(\omega)$ is given by equation 1. When this is done, we find that Re $\sigma_{HF}(\omega)$ and Im$\sigma_{HF}(\omega)$ both obey the same rough power law, $\sigma(\omega) \sim \omega^{0.9}$. This behavior is identical to the low frequency behavior recently observed in the ac conductivity of TaS_3[4].

Preliminary work by Gruner[5] at UCLA suggests that both Re σ and Im σ continue to climb in the low microwave frequency range. Eventually Im σ must turn over at some crossover frequency ω_c. This second characteristic frequency would correspond to the more familiar

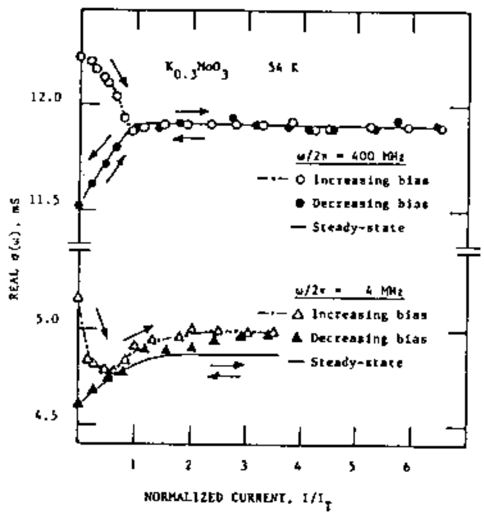

Figure 2: The effect of a dc bias on the high frequency ac conductivity of $K_{0.3}MoO_3$.

pinning frequency encountered in $NbSe_3$ and TaS_3 . It is likely that the high frequency response is present at all temperatures, not just below 70K. At high temperatures (70K - 100K) the effect of the high frequency resonance is to a large extent masked by the low frequency relaxation process, which displays an Arrhenius behavior in both its characteristic frequency and its magnitude.[1]

Figure 2 shows the effect of an applied dc bias on the ac conductivity. The measurements were made according to the following procedure. First the sample was warmed above 54K and then cooled back down. Next the bias was swept out so that $I > I_T$, and then reduced back to I = 0. Subsequent sweeps of the bias were made until the curve of $\sigma(\omega)$ versus I became repeatable. As shown in the top of figure 2, a "steady-state" was reached as soon as I became greater than I_T when $\omega/2\pi$

was 400 MHz. At $\omega/2\pi$ = 4MHz, two or three sweeps of the bias were required to achieve a repeatable curve.

There are several unusual features to the data presented in figure 2. First is the existence of a metastable, enhanced ac conductivity state which can only be reached by temperature cycling. A related temperature hysteresis effect has been observed in the dc conductivity by Tsutsumi et. al.[6] This ac effect, however, can not be explained solely in terms of a changing dc conductivity offset; the magnitude and bias dependence of the ac effect is enhanced at 400 MHz compared to 4 MHz. Second, at 400 MHz the conductivity is independent of bias past $I = I_T$. Increasing the bias past E_T does not increase $\sigma(\omega)$ even though $\sigma(\omega)$ has not saturated at some high-frequency, high-field limit. The scaling relation between $\sigma(\omega)$ and $\sigma(E)$ observed in $NbSe_3$ and TaS_3 can apply to $K_{0.3}MoO_3$ only under limited conditions.

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SUBHARMONIC SHAPIRO STEPS, DEVIL'S STAIRCASE, AND SYNCHRONIZATION IN RF-DRIVEN CDW CONDUCTORS

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Explanation of recent Shapiro steps studies in CDW conductors requires consideration of internal degrees of freedom and of associated finite velocity correlation lengths. The synchronization of different regions of the specimens with increasing rf is demonstrated through coalescence of noise peaks, reduction of fluctuations in noise peaks, and narrowing of steps.

The ac-dc interference phenomenon known in Josephson junctions as Shapiro steps was discovered in CDW conductors by Monceau, Richard, and Renard,[1] who used it to monitor narrow band noise in $NbSe_3$. Later work by Zettl and Grüner[2] stretched the Josephson junction analogy further, and the authors' most recent studies of steps shed new light on the microscopics of CDW motion.[3] Here we outline: (1) the restrictions which can be placed on the CDW equation of motion; and (2) the extent to which CDW response can be synchronized throughout the specimen.

I. Equation of Motion

Perhaps the most intriguing aspect of CDW steps studies so far is the presence of subharmonic steps. "Principal" steps occur whenever the internal (narrow band "noise") frequency f_{int} is near an integer multiple p of the applied rf driving frequency f_{ext}. Subharmonic steps are not restricted by this rule; they occur whenever $pf_{ext} = qf_{int}$ for q an integer not equal to 1. Figure 1 shows Shapiro steps in TaS_3, represented as peaks in the differential resistance dV/dI. This representation emphasizes small, sharp details which would be missed in direct I-V curves. When huge rf is applied, the usual linear region vanishes and steps appear. In addition to the principal steps (p/q = 1/1, 2/1), one can see three small subharmonics (1/2, 1/3, 2/3). These have been found in several TaS_3 specimens, while a few showed fewer subharmonics and others showed more. The differential

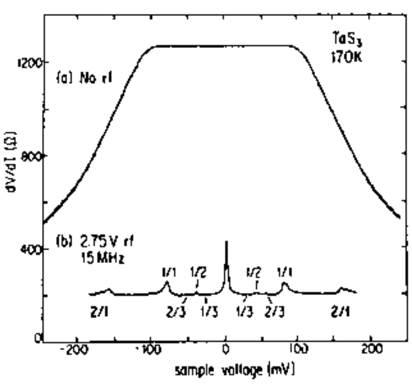

Fig. 1. dV/dI in TaS_3. Peaks in the lower curve correspond to principal and subharmonic Shapiro steps.

resistance at the peaks is much smaller than the linear value, $R_o = 1270\ \Omega$. Figure 2 shows part of a spectacular array of steps in $NbSe_3$. All subharmonic steps for q $\leq$ 14 have been observed, plus some for larger q, for a total of some 80 steps. Not only are there more steps in $NbSe_3$, but dV/dI comes closer to the linear value R_o = 28 Ω.

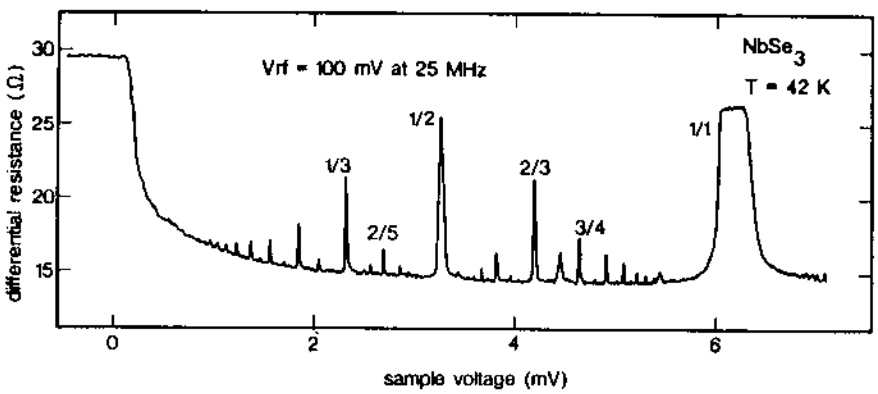

Fig. 2. Segment of a Shapiro step trace in $NbSe_3$.

The steps in $NbSe_3$ exhibit strong similarities to the devil's staircase behavior predicted theoretically[4,5] for the RSJ model of a Josephson junction. The steps generally are shorter and narrower for larger q, and there are so many steps that one begins to suspect they would fill the entire horizontal axis, if only one had adequate experimental resolution. The similarity is, in fact, more than superficial. By taking an expanded version of data like that which appears in figure 2, it is possible to extract a fractal dimension $D = 0.91 \pm 0.03$ which agrees very closely with numerical prediction ($D = 0.87$)[5] and analogue simulation ($D = 0.91 \pm 0.04$).[6] Reference 3 describes the analysis. Any value of $D < 1$ implies that the entire horizontal axis would be occupied by steps, if vanishingly small steps could be included. It is important to note, however, that the same value of D was found for several values of V_{rf} and within different intervals along the x-axis, in conspicuous disagreement with Refs. 5 and 6.

The presence of the subharmonic steps places restrictions on the underlying equation of motion for the CDW. The simplest equation which describes nonlinear and frequency dependent conduction, narrow band noise, and Shapiro steps treats the entire CDW as a single, damped classical particle in a periodic potential.[7] Supplemented by an inertial term it takes the form:

$$m\ddot{x} + \frac{1}{\tau}\dot{x} + \frac{m\omega_o^2}{Q}\sin(Qx) = eE \qquad (1)$$

where E is the applied (dc + rf) electric field. This equation is identical to the

RSJ equation for Josephson junctions, allowing results from the literature to be taken over directly. We therefore expect that the inertial term is essential for Eq. (1) to exhibit subharmonic steps.[4,8] However the observed frequency dependent conductivity is consistent with Eq. (1) only if the inertial term is negligible to at least several hundred MHz.[9] Hence Eq. (1) appears to be inconsistent with Figs. 1 and 2.

What is clearly lacking from Eq. (1) is allowance for internal degrees of freedom of the CDW, which are needed to account for pulse memory effects,[10] long time decays,[11] etc. The extra freedom may be modeled by assigning equations of form (1) with no inertial term to velocity-coherent regions in the sample, then adding coupling terms between the regions.[1] Such systems of coupled first order nonlinear equations can be expected[12] to exhibit mode locking and other behaviors reminiscent of the RSJ equation, and so it is possible that subharmonic steps might result. A distribution of coupling strengths between the regions might be able to explain the lack of strong dependence of D on V_{rf} and on V_{dc}.

II. Synchronization

The steps are most readily understood as regions of locking between the internal and applied frequencies when $pf_{ext} - qf_{int}$ is sufficiently small. If the locking within such regions is complete, the CDW velocity becomes fixed by f_{ext} and does not respond to changes in the applied dc voltage. Hence dV/dI rises to the linear resistance R_o attributable to uncondensed electrons alone. This situation pertains if the CDW velocity is coherent throughout the specimen. In reality the velocity coherence length must be finite, and f_{int} may vary spatially. If the variation of f_{int} is greater than the width of the region over which locking can occur, locking will be incomplete, and dV/dI will rise to a level less than R_o. Hence the height of dV/dI is expected to correlate with the degree of synchronization across the sample.

Doubling or tripling of narrow band noise peaks is frequently noted in power spectra of $NbSe_3$ current oscillations, graphically illustrating variation of f_{int} within a single specimen. If the splitting is small enough ($\leq$ 100 kHz), the locking effect of large V_{rf} can make the noise peaks coalesce, with corresponding signatures in the steps. An example is shown in fig. 3, for the $p/q = 1/2$ step. Within this step, dV/dI has two plateau levels. Within the lower level, the spectrum contains two peaks (fig. 3a), one of which is locked to $f_{ext}/2$, while the other moves with V_{dc}. Within the higher plateau, the two peaks have combined into a single peak at $f_{ext}/2$ which is independent of V_{dc} (Fig. 3b). Thus the higher dV/dI indicates greater synchronization throughout the sample.

Another common characteristic of narrow band noise in $NbSe_3$ is its fluctuations: the

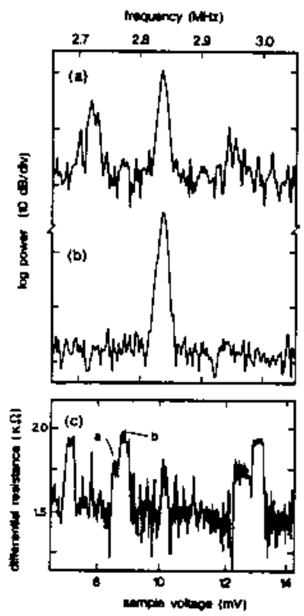

Fig. 3. Demonstration of synchronization in $NbSe_3$ at T = 45 K. (a) and (b) show noise peaks corresponding to labeled plateaus in the p/q = 1/2 step.

amplitude of the noise peak varies widely on a time scale of less than a second. These fluctuations can be reduced with application of large V_{rf}, provided V_{dc} is biased within a step. In samples for which dV/dI = R_o exactly, the time behavior of the fluctuations becomes entirely different, the amplitude becoming stable over the time frame of a minute or more. Figure 4 presents histograms of the noise amplitude, taken in the presence of large V_{rf} over two successive 15 minute periods. One period corresponds to locking on the p/q=1/2 step; for the other f_{ext} was detuned from $2f_{int}$. The 'locked' case exhibits several narrow peaks, each of which were formed by more or less successive measurements of the noise amplitude. The behavior suggests that in the presence of a large rf drive, most fluctuations are unable to disturb the relative phases of the CDW between regions of the sample. The larger fluctuations may allow the CDW to "realign" the phases to a state not much different in energy than before, and the amplitude of the noise takes on the value characteristic of that new state.

Finally, the more anisotropic CDW conductors such as TaS_3 and $(TaSe_4)_2I$ tend to have extremely broad noise spectra,[13] unless they are very pure.[14] This indicates a short velocity correlation length, which probably explains the small, wide steps of Fig. 1. Nevertheless, application of large V_{rf} sharpens the steps (Fig. 5), demonstrating that the phenomena of synchronization are relevant to these materials also.

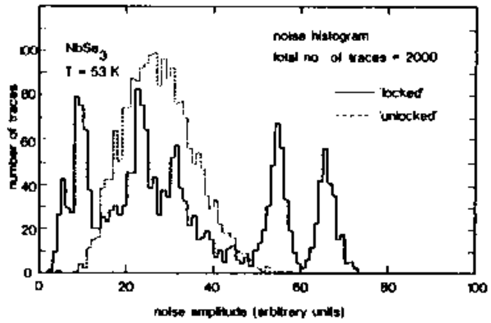

Fig. 4. Histograms of noise amplitude in $NbSe_3$ with with f_{int} tuned to, and detuned from locking criterion f_{ext} = 2 f_{int}. Each histogram contains 2000 points.

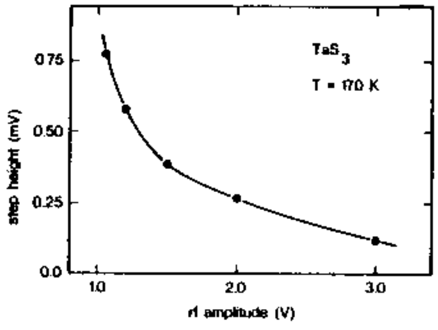

Fig. 5. Rf amplitude dependence of full width at half maximum of the p=1, q=1 step in TaS_3.

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