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OBSERVATION OF SHAPIRO STEPS IN THE CHARGE-DENSITY-WAVE STATE OF $NbSe_3$

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We report the observation of ac-induced Shapiro-like steps in the dc I-V characteristics of $NbSe_3$. Analysis of the steps in terms of a classical model of charge-density-wave (CDW) transport, mathematically equivalent to the Stewart-McCumber model of Josephson tunnel junctions, indicates a highly coherent sample response, and that coupling phenomena well known in the Josephson literature also occur for CDW transport.

It is by now well established that the field[1,2] and frequency[3,4] dependent conductivity in $NbSe_3$ and TaS_3 is associated with the collective response of the charge-density-wave (CDW) condensate. The narrow-band noise, with fundamental noise frequency proportional to the excess current in the nonlinear conductivity region,[5,6] provides direct evidence for moving CDW's as originally proposed by Fröhlich.[7] Direct measurements[8] of the current oscillations suggest a highly coherent response, with phase coherence extending across the whole sample.

A tunneling model proposed by Bardeen[9] accounts for the field and frequency dependent response,[10] while the current oscillations[8] are accounted for by assuming a sliding of the CDW over the peaks and valleys of the pinning potential. A phenomenological classical model,[11] which treats the CDW as a rigid object of mass m and charge e moving in a periodic potential, leads to an equation of motion

$$\frac{d^2x}{dt^2} + \frac{1}{\tau}\frac{dx}{dt} + \frac{\omega_0^2}{Q}\sin Qx = \frac{eE}{m} \tag{1}$$

where $1/\tau = \gamma/m$ with γ the damping constant, $\omega_0^2 = k/m$ with k the restoring force constant, $Q = 2\pi/\lambda$ where λ is the wavelength of the CDW, and E is the driving field. Although the classical model does not account for microscopic details of the pinning mechanism, it describes the qualitative features of the nonlinear conductivity when E exceeds a threshold field E_T, and also the frequency dependent response. Both the tunneling and classical models also account for various ac-dc coupling experiments, such as dc conductivity induced by applied ac fields.[12,13] We believe, therefore, that both models should also describe phenomena associated with the intrinsic oscillations produced by the moving CDW.

In this report we shall investigate the dc current-voltage (I-V) characteristics of $NbSe_3$ in the presence of an externally applied ac field. The basis of our discussion and analysis of experimental data will be Eq. (1). We stress, however, that a similar analysis should be possible within the framework of the tunneling model.

The substitution $\theta = Qx$ allows Eq. (1) to be reduced to the dimensionless form

$$\frac{d^2\theta}{dt^2} + \Gamma\frac{d\theta}{dt} + \sin\theta = \frac{E}{E_T} \tag{2}$$

where $\Gamma = 1/\omega_0\tau$, $E_T = (\lambda/2\pi)(m\omega_0^2/e)$, and time is measured in units of ω_0^{-1}. Equation (2) is formally identical to the Stewart-McCumber equation describing a shunted Josephson junction,[14]

$$\frac{d^2\phi}{dt^2} + G\frac{d\phi}{dt} + \sin\phi = \frac{I}{I_J} \tag{3}$$

where ϕ is the phase difference across the junction, I is the current through the junction, and $G = (RC\omega_J)^{-1}$ where R and C are the resistance and capacitance of the junction and $\omega_J = 2eI_J/C\hbar$. I_J is the dc Josephson critical current, and time is measured in units of ω_J^{-1}. This formal correspondence has been recognized by various groups.[15] The current oscillations for CDW transport[5,8] then correspond to the ac Josephson effect.

One of the early evidences for Josephson effects was the observation of so-called Shapiro steps -- a driving current of the form $I = I_{dc} + I_1\cos\omega t$ will produce steps in the dc I-V characteristics of the junction whenever the junction voltage $\langle V\rangle$ equals $n\hbar\omega/2e$, where n is an integer. These steps are a direct consequence of the ac Josephson effect, and have been observed experimentally in Josephson junctions[16] and superconducting microbridges.[17] By explicitly solving Eq. (3) in the high frequency limit ($\omega \gg 2eI_JR/\hbar$), computer simulations and analytic approximations show[18,19] that the height of the n^{th} step is given approximately by

$$\delta I \approx I_J(\omega{=}0)\ 2\left|J_n[I_1/\omega G I_J(\omega{=}0)]\right| \tag{4}$$

where J_n is the Bessel function of order n.

In the low frequency limit, where the capacitance of the junction is neglected, computer calculations[18,19] have yielded solutions for δI which somewhat resemble Bessel functions.

The analogy between Eq. (3) and Eq. (2) suggests that similar "Shapiro steps" should appear in the dc I-V characteristics of a CDW

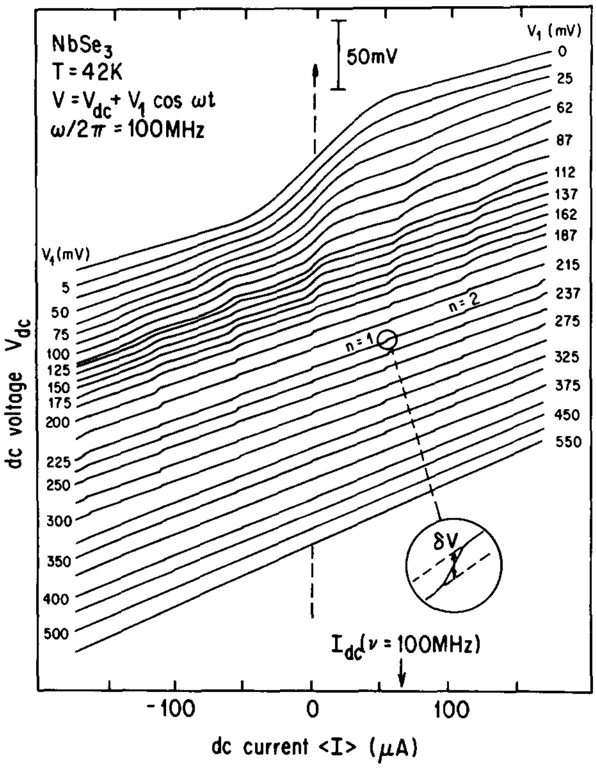

Fig. 1. Dc I-V traces for $NbSe_3$ at 42 K in the presence of an applied rf field at frequency $\omega/2\pi$ = 100 MHz and of amplitude V_1. The step height δV is defined in the Figure. No Shapiro steps are seen for $V_1 = 0$, while the maximum step height is at approximately V_1 = 100 mV. The arrow indicates the dc current which yields a noise frequency ν = 100 MHz. n is the step index (see text).

system driven by an external field $V = V_{dc} + V_1 \cos \omega t$. The analogy also allows further predictions concerning CDW transport to be made, as discussed by Ben-Jacob.[15]

In this communication we report the observation of Shapiro-like steps in the dc I-V characteristics of the CDW systems $NbSe_3$, and we analyze our experiments in terms of expressions worked out originally for Josephson junctions, and adapted for CDW transport. A comparison of our experimental findings with these expressions leads to a highly coherent sample response, in agreement with conclusions drawn from our previous analysis[8] of current oscillation phenomena in $NbSe_3$.

Rf-induced steps in the dc I-V characteristics of $NbSe_3$ were first reported by Monceau et al.,[5] who found sharp peaks in the differential conductance in the nonlinear conductivity region, indicating interference effects between the applied ac frequency and the narrow-band noise frequency. Our experiments on $NbSe_3$ reported here, which were performed on samples with well-defined and uniform geometries, allow us to observe the steps directly on dc I-V plots. We observe interference effects approximately 100 times larger than those reported by Monceau et al.[5]

Figure 1 shows several dc I-V traces for $NbSe_3$ at T = 42 K, using a two probe mounting configuration. The excitation applied to the sample was of the form $V = V_{dc} + V_1 \cos \omega t$, with $\omega/2\pi$ = 100 MHz. For $V_1 = 0$, a smooth, nonlinear I-V curve is observed. At higher values of V_1, well-defined steps appear in the nonlinear region. The step height δV, as defined in the Figure, in general first increases with increasing V_1, and then decreases. The position of the n = 1 step (identified in the Figure) corresponds to a dc current <I> which, in the absence of external ac, yields an intrinsic oscillation of frequency ν = 100 MHz = $\omega/2\pi$. We also note the presence of harmonic steps corresponding to n = 2 (where ν = 200 MHz). The steps are thus clearly an interference effect between the intrinsic current oscillation and the externally applied rf excitation. The coherent current oscillations were observed directly during the same experimental run, and the fundamental oscillation frequency ν was found to vary as $\nu \sim I_{CDW}$, where I_{CDW} is the excess current carried by the CDW, in agreement with previous studies.[5,6,8]

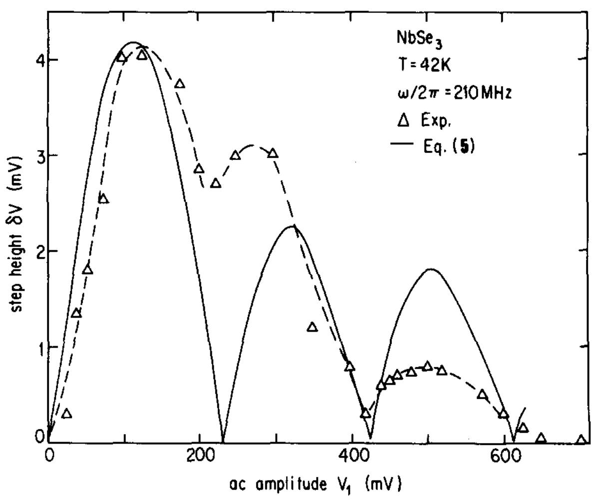

Fig. 2. Step height δV versus amplitude V_1 of the applied rf signal. The rf frequency is 210 MHz and the step index n = 1 (see text). The solid line is Eq. (5), with parameters given in the text. The dashed line is a guide for the eye.

In Fig. 2 we show the detailed form of the n = 1 step height δV as a function of V_1, for a different sample and with $\omega/2\pi$ = 210 MHz. We find δV resembles a decaying oscillatory function, with well-defined maxima and minima.

The step heights, as shown in Fig. 2, are strongly frequency dependent. We have made measurements similar to those shown in Fig. 2 at different values of ω, and we find that the general form of δV versus V_1 remains essentially unchanged between $\omega/2\pi$ = 10 MHz and 200 MHz. The maximum step height attained, however, strongly decreases with decreasing ω. In Fig. 3 we show the maximum step height δV_{max} as a function of ω, for the same sample as used for Fig. 2. The data indicate a saturation of δV_{max} near 200 MHz, although measurements at higher frequencies are needed to confirm this conjecture.

We now analyze these results in terms of the classical model of CDW transport, Eq. (2). In the high frequency limit ($\omega >> \omega_0\tau$), the first (n = 1) step height is given, in direct analogy to Eq. (4), by

$$\delta V = \propto V_T(\omega{=}0)\ 2\left|J_1[V_1\omega_o^{\,2}\tau/\omega V_T(\omega{=}0)]\right| \quad . \quad (5)$$

The parameter $\propto$ represents the volume fraction of the sample which responds collectively to the external field. Equation (5) predicts that the maximum step height, δV_{max}, is independent of frequency. The strong frequency dependence of δV_{max} shown in Fig. 3 then indicates that Eq. (5) applies only to data above approximately $\omega/2\pi \sim 200$ MHz. In Fig. 2 (which is for $\omega/2\pi = 210$ MHz), we have plotted Eq. (5) with chosen parameters $\omega_0{}^2\tau$ = 503 MHz and $\propto = 0.17$. V_T is fixed by experimental conditions at 24 mV. The positions of the maxima and minima of the Bessel function of Eq. (5) are in remarkably good agreement with the experimental data. The value of the cross-over frequency $\omega_0{}^2\tau/2\pi$ deduced from this fit is 80 MHz, in reasonable agreement with the value of 45 MHz obtained from frequency-dependent conductivity studies.[10] Equation (5), however, underestimates δV near the second peak in Fig. 2 and overestimates δV near the third peak, although we remark that the approximate Bessel function solution is quite sensitive to the chosen form of the pinning potential, and that a slightly different choice of pinning potential in Eq. (2) would distort the amplitudes of the peaks in the solution of δV. The value $\propto = 0.17$ indicates that a large fraction of the sample is responding coherently to the external perturbation. Analysis of similar data for another sample yielded an even higher value of $\propto = 0.60$, which corresponds to 60% of the sample volume being phase coherent. The high coherence is in agreement with other studies[8,20] of the current oscillations and switching phenomena of $NbSe_3$.

The strong frequency dependence of δV_{max} as shown in Fig. 3 indicates that a low frequency solution is most appropriate below 200 MHz. Neglecting the first term in Fig. 3 yields the standard RSJ (resistively-shunted-junction) model[14] of Josephson junctions. In the CDW case this corresponds to the neglected effects of inertia (overdamped response). The solid line in Fig. 3 is the frequency dependence of δV_{max}, as calculated[14,19] for the RSJ model in the overdamped limit. The value of $\omega_0{}^2\tau/2\pi$ = 47 MHz obtained from this fit is in excellent agreement with the result $\omega_0{}^2\tau/2\pi$ = 45 MHz obtained from the frequency dependence of the low-field ac conductivity,[10] and consistent with that deduced from the analysis of Fig. 2.

We remark that studies[3,10] of the frequency dependent conductivity of $NbSe_3$ have indicated an overdamped response. This does not,

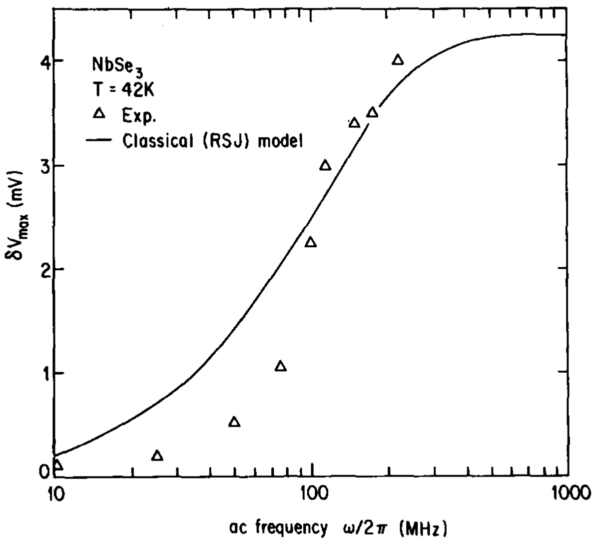

Fig. 3. Maximum step height δV_{max} attained, versus rf frequency. The solid line is the prediction of the RSJ model of Josephson tunnel junctions [Refs. 18, 20]. The data of Figs. 2 and 3 are for the same $NbSe_3$ sample.

however, exclude the importance of an inertial term for the depinned CDW, since the damping parameter may be velocity-dependent. In fact, recent analysis[8] of the coherent current oscillations in $NbSe_3$ indicated that inertial effects may be important well above threshold. In the present discussion and analysis, we find good agreement between experiments and an overdamped Stewart-McCumber model.[14] Whether the inertial term [first term in Eq. (1)] becomes dominant at much higher frequencies and velocities remains to be seen.

We believe that any model which gives both a field and frequency dependent response, along with current oscillations for applied dc fields, will lead to ac + dc coupling behavior. It is in this respect that Eq. (1) can be regarded as a reasonable equation to describe ac-induced dc conductivity and the reverse. The close correspondence between this equation and the equation describing a shunted Josephson tunnel junction[14] demonstrates that various phenomena which are analogous to effects well known in the Josephson literature, also occur in CDW transport. We note that ac induced steps have also been considered recently by Sneddon et al.[21] Only the $V_{ac} \to 0$ limit is calculated, however, and therefore a direct comparison with our experiments is not possible at present.

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