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JOSEPHSON OSCILLATIONS OF CHARGE DENSITY WAVES

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Abstract

The formation of charge density waves in solids was originally proposed as a possible mechanism for superconductivity by Fröhlich. Although the experimentally discovered materials with charge density waves (CDWs) are found to have finite resistivity as a result of impurity pinning, they nevertheless reveal many interesting features including motion which is analogous to a resistively-shunted Josephson junction of superconductors. The noise spectrum of CDW systems is reviewed with particular emphasis on interactions with normal as well as magnetic impurities. Future prospects for observing an amplitude variation of the noise signals induced by a magnetic field are proposed.

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Some thirty years ago, Fröhlich suggested that a sliding motion of a coherent charge density wave may be an interesting candidate for superconductivity [1]. His results were based on a simple one-dimensional model which favors the formation of a wave (CDW) with charge density of the form

$$\rho(x) = \rho_o + \rho_1 \cos(2k_F x + \phi) \quad , \tag{1}$$

where the wavelength λ is determined by the Fermi wave vector k_F according to the relation $\lambda = 2\pi/k_F$. Accordingly, the wavelength may be incommensurate with the lattice spacing, depending on the filling of the electron energy bands which determines k_F. Since the inertia of the CDW involves the total mass of all the conduction electrons, the wave propagation should in principle be unaffected by scattering from isolated impurities and hence the collective quantum state may exhibit frictionless flow.

To compensate for the electron charge density $\rho(x)$, the ions rearrange themselves to create a lattice distortion which is often referred to as Peierls instability in one-dimensional systems. Hence the electron-phonon coupling plays a central role in creating an energy gap Δ in the electronic system. At first glance this situation is reminiscent of a semiconductor, but the novel feature of a coherent quantum state permits a sliding motion of the CDW without energy loss, while the ion charges merely oscillate about their equilibrium values.

Since one-dimensional systems had not been synthesized, experimental interest in CDW motion waned until recently, when the discovery of new materials such as $NbSe_3$ generated a wealth of remarkable data.

Overhauser stimulated theoretical and experimental interest when he showed [2] that an electron gas may be unstable toward formation of either a charge density wave or a spin density wave (SDW); his calculations demonstrated vividly the influence of reduced dimensionality on the CDW instability and these general results form the basis of much current discussion. Unfortunately, the early attempts to identify CDW states in alkali metals such as potassium were mired in controversy, even though the SDW in chromium was well established by extensive experimentation.

A new wave of interest in CDW modes was generated by the discovery of quasi-one-dimensional compounds [3] which provided a wealth of transport and susceptibility data and rejuvenated theoretical challenge. We shall concentrate our discussion on $NbSe_3$ principally because the discovery of current noise in this material reveals many analogies with the Josephson effect. Furthermore the recent discovery of Devil's staircase structure in the noise spectrum, as well as data on mode locking and chaos provides novel features which may test recent theoretical models with universal features.

Since $NbSe_3$ exhibits a chain structure, the electrons propagate in an environment of reduced dimensionality which favors CDW states. Nevertheless the electronic structure is not one-dimensional in the usual sense. The anisotropy in the conductivity along perpendicular crystal axes varies by roughly a factor of ten, and two distinct charge density waves oriented along different axes were discovered in the resistance R(T) measurements [4] which are reproduced in Figure 1. It is important to emphasize that the R(T) data shown is for a low electric field and presents a dilemna in that the CDW states are found to be not superconducting! Rather, the R(T) data show evidence for two energy gaps in the electron structure with the first one vanishing at a rather high temperature ($T \cong 140k$) and the second one representing an order parameter which vanishes at ($T_2 \cong 59k$).

As the applied electric field is increased beyond a threshold value E_T, a large increase in the DC conductivity of $NbSe_3$ is observed, suggesting that the CDW has been liberated from the pinning influence of impurities. In addition, Fleming and Grimes [5] discovered current noise in $NbSe_3$ which scaled with the CDW order parameter obtained from x-ray diffraction, thus providing convincing evidence for the CDW transport as a noise source.

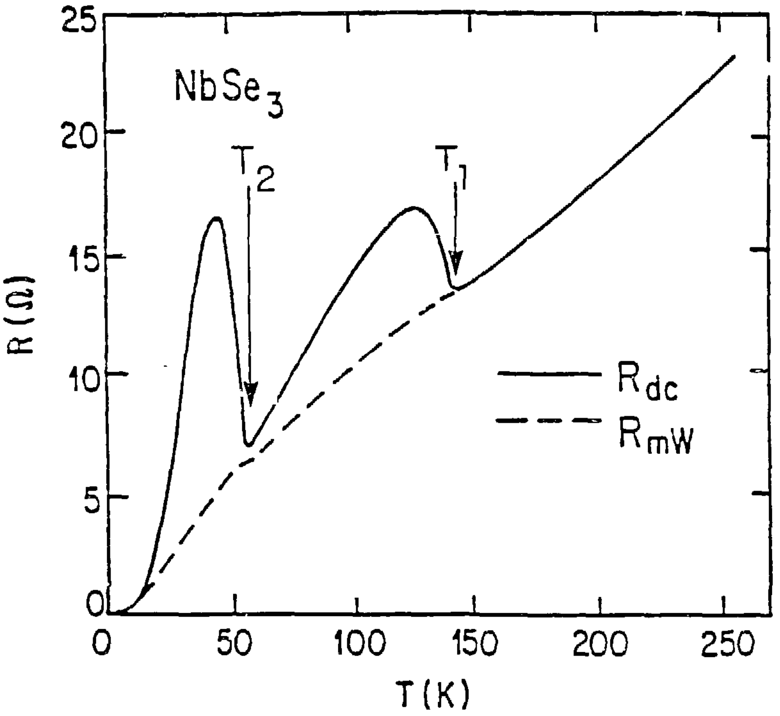


Fig. 1. Resistance data on $NbSe_3$ taken at low electric field so that the CDW is not moving. Evidence for two electronic energy gaps is shown by the drops in R(T) at the transitions T_1 and T_2 (solid curve). Application of microwave fields eliminates the energy gap characteristics as shown by the dotted curve (from Ref. 4).

The surprising damping of the CDW motion has been attributed to impurity pinning whose origin continues to be of wide theoretical interest. A quantum tunneling approach has been developed by Bardeen [6], with quite good success in explaining the highly non-linear field dependence of the conductivity. Classical models have also been proposed, and the microscopic origin of the CDW impurity pinning continues to challenge theorists. Our goal is to review the CDW oscillation phenomena with analogies to the Josephson effect in superconductors. Thus we neglect the discussion of the interesting non-linear response of the CDW to an applied DC electric field and we refer the reader to the review article by Bardeen [6] for further study of this subject as well as the microscopic origin of the impurity pinning potential.

It is instructive to examine the classical equation of motion for a CDW originally proposed by Grüner, Zawadowski and Chaikin [7]. They note that a periodic displacement of the CDW results in the same energy so that the impurity pinning may be represented as a periodic effective potential $V \propto \sin(Qx)$, and then the equation of motion may be written as

$$\frac{d^2x}{dt^2} + \Gamma \frac{dx}{dt} + \frac{\omega_o^2}{Q} \sin(Qx) = \frac{eE}{M} \quad , \tag{2}$$

where $Q = 2k_F$ and Γ is a phenomelogical damping constant. The analogy of Equation (2) to the resistively shunted Josephson junction [RSJ] equation is quite apparent if we simply write the phase $\phi \equiv Qx$ and redefine the corresponding parameters in terms of the current. We shall use Eq. 2 as a padagogic guide in discussing the CDW noise structure even though alternate models have been proposed.

Application of external radiation results in noise structure emanating from the competition of the driving frequency ω_{ac} and the intrinsic CDW frequency ω_f. A moving CDW will have a charge density

$$\rho(x) = \rho_0 \cos\left[2k_F(x - v_D t)\right] \quad , \tag{3}$$

and thus will exhibit a fundamental frequency

$$\omega_f = 2k_F v_D = \frac{2k_F}{ne} J_{CDW} \quad , \tag{4}$$

where v_D is the CDW drift velocity which is simply related to the current J_{CDW}. The proportionality of the observed noise frequency to the CDW current has been extensively verified at various temperatures for $NbSe_3$ [8].

In the non-linear conductivity region ($E \gtrsim E_T$) the CDW may be viewed as sliding down a tilted washboard potential, and the application of an AC microwave field should generate steps in the current-voltage characteristics which are analogous to the Shapiro steps found in superconducting Josephson junctions. In fact this analogy works very well in $NbSe_3$ samples whose I-V curves were measured and analyzed by Zettl and Grüner [9]; their results are shown in Figure 2.

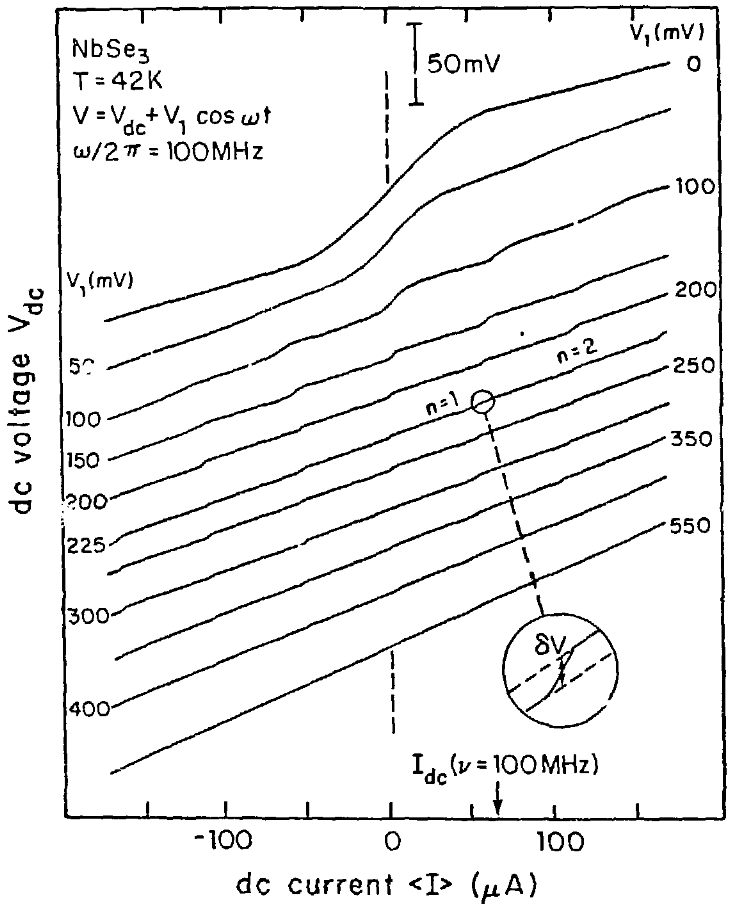


Fig. 2. Current-voltage traces of Ref. 9 for $NbSe_3$ subjected to an rf field at frequency $\omega/2\pi = 100MHz$ and of amplitude V_{ac}. No Shapiro steps are seen for $V_{ac} = 0$, while the maximum step height is achieved for $V_{ac} \cong 100mV$. The step index n is shown.

From Figure 2 we can see the evolution of Shapiro steps with increasing applied field strength V_{ac} : For $V_{ac} = 0$ a smooth I-V curve is observed which shows a well-defined threshold voltage V_T where the conductivity starts to become non-linear; at increasing V_{ac}, well-defined steps appear with height δV at first increasing with V_{ac} and then tapering off to zero. The position of the $n = 1$ step corresponds to a DC current which (at $V_{ac} = 0$) yields an intrinsic oscillation of $f_1 = 100MHz$. The presence of harmonic steps (e.g. $n = z$) and subharmonic steps ($n = \frac{1}{2}$) clearly indicates the interference between intrinsic CDW oscillations and the applied rf radiation. Analysis of the CDW I-V steps shows that the basic equations of the Josephson effect [10] yield a good description of the step heights as a function of the AC voltage amplitude:

thus the RSJ model of Eq. 2 evidently describes the step height oscillations as a function of V_{ac}, and the degree of agreement with the data has been used to argue that 30% to 100% of the $NbSe_3$ sample volume is phase coherent [9].

Despite the advantages of using a model which resembles closely the RSJ equation of motion, it is by no means clear that the classical response of a CDW provides an adequate physical description of the noise spectrum.

A new quantum oscillation source of CDW noise has been proposed by Barnes and Zawadowski [11] which invokes the right (R) and left (L) moving components of a CDW as two macroscopic quantum states. The scattering of the electron-hole pairs is treated by analogy to the Josephson calculations and yields a ratio of the CDW current to its natural frequency ω_f as

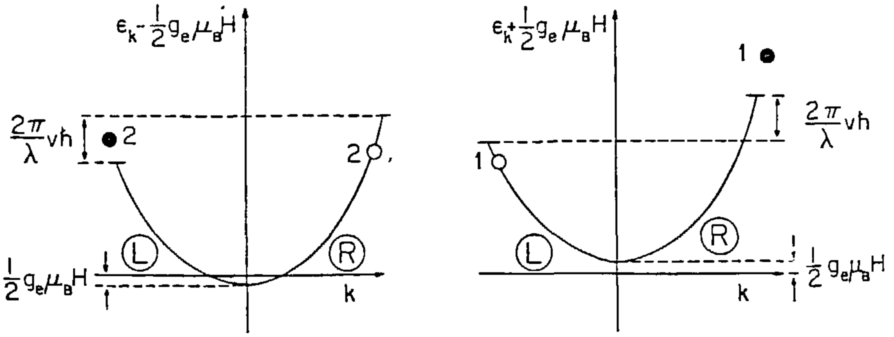
$$\left(\frac{I_{CDW}}{\omega_f}\right)_{B-Z} = e\,\frac{n(T)}{n(T=0)} \quad , \qquad (5)$$

where e is the electron charge and n(T) is the number of electrons in the CDW condensate. By contrast the classical model of Eq. 2 would predict a corresponding ratio $(I_{CDW}/\omega_f) = 2e\,n(T)/n(0)$. Unfortunately, as a consequence of the chain structure of $NbSe_3$ and materials preparation difficulties, n(T) has not been established with sufficient certainty [9] to check these predictions.

The quantum origin of the CDW noise may be probed by other means, including the influence of a magnetic field H. We have recently investigated [12] the response of the CDW current to an external field. Since the CDW has zero magnetization an external field H will couple indirectly to the CDW by means of magnetic impurities, coexisting spin-density waves, or perhaps Fermi surface changes induced by the electron spin splitting.

Our physical approach to the CDW oscillations modified by magnetic impurities and an external field H is illustrated in Figure 3.

Following Barnes and Zawadowski [11], we regard the CDW as a superposition of two macroscopic quantum states characterized by $\pm Q$, where $Q = 2\pi/\lambda$ is the wave number of the quantum state (in the simplest case $Q = 2k_F$). These coherent states consist of electron-hole pairs which are scattered by impurities [13]. The scattering transition takes two electrons from the right-hand side (R) of the Fermi surface to the opposite left-hand side (L), with a corresponding L→R transition of the hole states. The splitting of R and L states is caused by the velocity v of the CDW which imparts a bias to the Fermi surface. Hence a magnetic field provides a test of the above mechanism by splitting the Fermi surface into "up" spin and "down" spin components as shown in Fig. 3.



up spin

down spin

Fig. 3. Dispersion curve of a one-dimensional electron gas in a magnetic field H. When there is a uniform motion of the entire electron gas with velocity v, the right (R) and left (L) Fermi levels are split. Magnetic impurities cause transitions of electron-hole pairs, e.g. states 1→2. In this case, the total momentum changes from $2k_F$ to $-2k_F$ leaving the total spin unchanged.

Performing the computation of the current along the lines of the Josephson method, we find [12] a H-field dependence to the current which is shown in Figure 4. This result allows an independent check on the source of CDW noise since there is not a corresponding classical analogy.

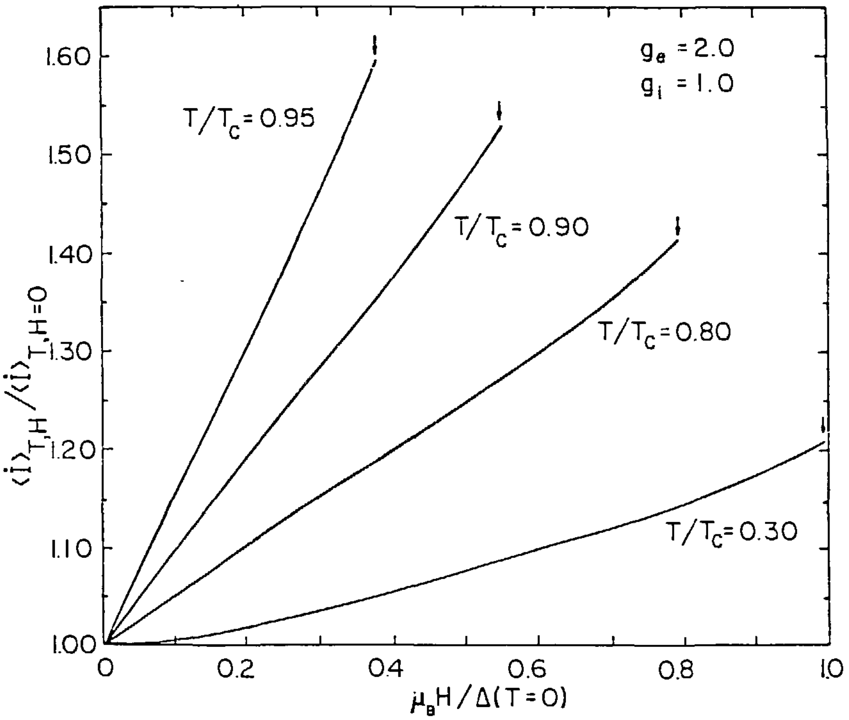


Fig. 4. Magnetic field dependence of the CDW current oscillations at various temperatures as predicted by the quantum scattering theory of the noise [12]. The electron g-factor is taken as 2 while the impurity g-factor is 1. The arrows indicate the values $\mu H = \Delta(T)$.

Since the addition of magnetic impurities to compounds such as $NbSe_3$ may pose undesirable complications in materials preparations, it is interesting to speculate on other mechanisms which may couple a CDW to a magnetic field. One possibility is the presence of a competing spin density wave (SDW) which is favored by the same type of electronic structure which induces CDWs as originally shown by Overhauser [2]. Hence the SDW coupling to an external H-field may in turn influence the CDW order parameter by generating field dependent gaps in the electronic structure.

The pinning of spin density waves and their response to external H fields has recently been calculated [14] and the results suggest a new type of SDW current noise with amplitudes which may be field-dependent in a manner distinct from CDW motion.

Application of large magnetic fields to $NbSe_3$ samples results in a dramatic increase in the resistivity as a function of T [15], even though the CDW transition temperature T_2 (as in Figure 1) remains unaffected by H. At the low electric fields of these measurements the CDW remains pinned by impurities and therefore the resistivity changes suggest modifications in the electronic structure created by the magnetic field. Field-dependent energy gaps and "nesting" regions of the Fermi surface [16] may alter the balance between CDW and SDW instabilities and accordingly influence their associated current oscillations.

Future studies of higher purity $NbSe_3$ samples subjected to various combinations of external fields should help to resolve the controversy regarding the origin of the narrow-band CDW noise. In addition to the classical and quantum oscillations suggested above, the origin of CDW noise has also been attributed to solitons [17] and contact effects [18]. Also the quantum tunneling theory of Bardeen [6] provides a microscopic basis for the pinning potential which is essential in the analysis of the noise spectrum [8].

Finally it is interesting to mention the onset of chaos and studies of the "Devil's staircase" structure in the noise spectrum. These phenomena reach into various branches of physics as well as mathematics. The original ideas stem from mode locking phenomena observed centuries ago in the synchronization of weakly coupled clocks, and recent studies have advanced to Josephson junctions [19, 20]. Theoretical studies by Per Bak and colleagues [21] have proposed a new approach to the onset of chaos which predicts certain universal features in the response of dynamical systems, including the Josephson equation model discussed here. The recent discovery of Devil's staircase structure in the current-voltage characteristics of CDW oscillations [22] yields a fractal dimension quite close to the theoretical prediction [$D \sim 0.87$]. Thus we may expect further stimulating results in this field of endeavor.

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