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Short Communication

Wavelet Analysis of the Charge Density Wave Dynamics in the Molecular Conductor (Perylene)₂Pt(mnt)₂
(mnt = maleonitriledithiolate)J. Dumas ⁽¹⁾, N. Thirion ⁽²⁾, M. Almeida ⁽³⁾, E.B. Lopes ⁽³⁾, M.J. Matos ⁽³⁾ and R.T. Henriques ⁽³⁾⁽¹⁾ Laboratoire d'Etudes des Propriétés Electroniques des Solides (*), CNRS, B.P. 166, 38042 Grenoble Cedex 9, France⁽²⁾ Centre d'Etudes des Phénomènes Aléatoires et Géophysiques (CEPHAG) (**), ENSIEG, B.P. 46, 38402 Saint-Martin d'Hères, France⁽³⁾ Departamento de Química, ITN, 2686 Sacavem Codex, Portugal

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Abstract. — A new method based on the wavelet analysis of the voltage oscillations generated above threshold electric field for depinning of the charge density wave (CDW) in the molecular conductor (Perylene)₂Pt(mnt)₂ (mnt = maleonitriledithiolate) has been developed. This analysis method permits a better time resolution of the frequencies and phases involved in the CDW motion than that allowed by conventional Fourier analysis. The results are discussed in relation with current models for CDW deformations.

1. Introduction

Charge density wave (CDW) dynamics in quasi one-dimensional conductors has been the subject of intensive studies for the past fifteen years [1,2]. In the CDW state, the conduction electron density is given by $\rho(x) = A \cos(Qx + \phi)$ where A is the CDW amplitude, $Q = 2k_f$ is the CDW wavenumber, and ϕ the phase. Depending on the band filling, the periodicity of the CDW can be commensurate or incommensurate with the underlying lattice. Below a small finite threshold electric field E_t , the CDW is pinned to the lattice because of its interactions with randomly distributed impurities or lattice defects or due to commensurability effects. Above E_t , remarkable non-linear conductivity due to the motion of the CDW with respect to the lattice is observed. At constant current, in the non-linear regime, quasi-periodic voltage oscillations, commonly referred to as narrow band noise (NBN), accompany the current

(*) associated with Université Joseph Fourier, Grenoble

(**) (URA CNRS 346)

flow. The frequency of these oscillations is proportional to the excess current density carried by the CDW. The onset of CDW sliding is also marked by the so-called broad band noise. In the Fukuyama-Lee-Rice (F-L-R) model [3, 4], where only the phase of the CDW is taken into account, pinning results from a competition between the elastic energy of the CDW and preferred CDW phase at impurity sites. Other models involve structural CDW defects, such as phase dislocations [5], neglected in the F-L-R model. The analogy between depinning and onset of plastic deformations of a crystal has also been considered [6]. The origin of the voltage oscillations is still the subject of some controversy. Since the interaction energy of a CDW with impurities or with the lattice is unchanged by a translation through one CDW wavelength, the impurity pinning potential is expected to be periodic. The CDW motion in this potential should give rise to periodic oscillations [7]. Voltage oscillations might also result from periodic formation of phase vortices required for CDW to normal carrier conversion near contacts or at strong pinning centers [5].

These oscillations can be observed in the transient CDW response to current pulses. At constant current, the oscillations can also be studied by Fourier analysis. However, a spectrum analyser involves time averaging, whereas the amplitude, phase and frequency of the oscillations may fluctuate in time. The wavelet analysis presented here is a relatively recent method which allows time averages to be drastically reduced to a minimum and information to be obtained on short-lived events or intermittency. It is especially well-adapted for analysis of the narrow band noise due to the CDW motion.

Rather than giving a time-averaged spectrum, the wavelet analysis decomposes a signal into wavelet components which depend on scale and time [8-10]. We have chosen Morlet's analyzing wavelet [10] which represents a reasonable compromise between time and frequency resolution:

$$\Psi(t) = \exp[ict - t^2/2]$$

where $c = \pi(2/\ln 2)^{1/2}$. With this value of the parameter c , the admissibility criterion for the wavelet is satisfied [9, 10]. $\Psi(t)$ is a complex function with an amplitude and a phase which represents a sinusoidal oscillation convoluted with a Gaussian, as illustrated in Figure 1. The time and frequency resolution of the wavelet obeys a Heisenberg-Gabor relation $\Delta t \Delta f = 1/2\pi$. The wavelet transform of a signal $f(t)$ can be written, for the continuous case, as

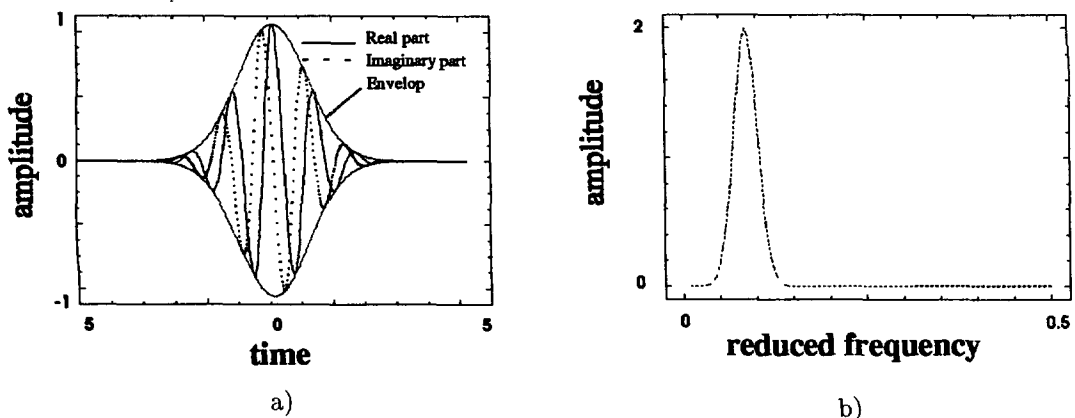


Fig. 1. — (a) Analysing wavelet amplitude as a function of time in arbitrary units; (b) Fourier transform of the analysing wavelet in arbitrary units.

$$W(a, t) = a^{-1/2} \int f(\tau) \Psi^*\left(\frac{\tau - t}{a}\right) d\tau$$

where Ψ^* is the complex conjugate of Ψ , a is the scale parameter $a \equiv f_0/f$, where f_0 is the central frequency of the analyzing wavelet. $W(a, t)$ depends on scale and time. If the parameters a and t are changed independently, the distribution of each component of $f(\tau)$ is obtained in the (a, t) plane, that is, in the time frequency plane. Thus, the wavelet transform allows a time-frequency analysis.

Such an analysis has been performed in the case of the quasi one-dimensional conductor (Perylene)₂Pt(mnt)₂, which shows a Peierls transition at $T_p = 8$ K towards a commensurate CDW and where remarkable voltage oscillations with large amplitude are easily detected in real time by pulse techniques at 4.2 K. The CDW dynamics of the Pt compound will be reported elsewhere [11]. We have recently reported the observation of CDW non-linear transport in the molecular compound (Perylene)₂Au(mnt)₂, isostructural of the Pt compound [12]. Sample and contact preparation procedures are given in reference [12].

2. Experimental Results

The non-linear I-V characteristics and quasi-periodic voltage oscillations were observed at $T = 4.2$ K using the current pulse technique [11]. The computer-controlled arrangement consists of a HP 214B pulse generator, Tektronix AM502 amplifiers and a Tektronix 2230 digital storage oscilloscope. A non-linear I-V characteristic is shown in Figure 2(a). The onset of non-linearity corresponds to a threshold field $E_t = 9$ Vcm⁻¹. Quasi-periodic voltage oscillations with strong amplitude superimposed on the transient voltage response to rectangular current pulses and generated above E_t are shown in Figure 2(b). A delayed onset of the oscillations with respect to the leading edge of the pulse is observed. The delay time decreases rapidly when the current pulse amplitude is increased.

A typical wavelet analysis of the transient voltage response, performed at CEPHAG/ EN-SIEG in Grenoble is shown in Figures 3 and 4 with a logarithmic scale for the frequencies. Figure 3 (a) reveals three frequencies as a function of time, each with a finite spectral width, for a current pulse amplitude above threshold for depinning. The fundamental frequency $F_1 \sim 75$ kHz is accompanied by harmonics F_2, F_3, F_4 which appear in pink, red, orange and yellow, respectively, in Figure 3(a). A direct Fourier analysis of the voltage oscillations reveal

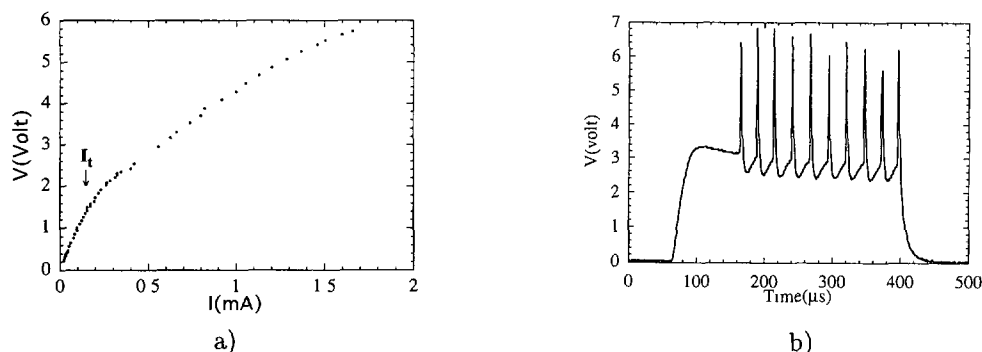


Fig. 2. — (a) I-V curve obtained by current pulse technique showing a non-linearity at $I_t = 0.18$ mA; pulse width 350 μ s; $T = 4.2$ K; (b) Transient voltage response to current pulses with an amplitude $I = 0.496$ μ A.

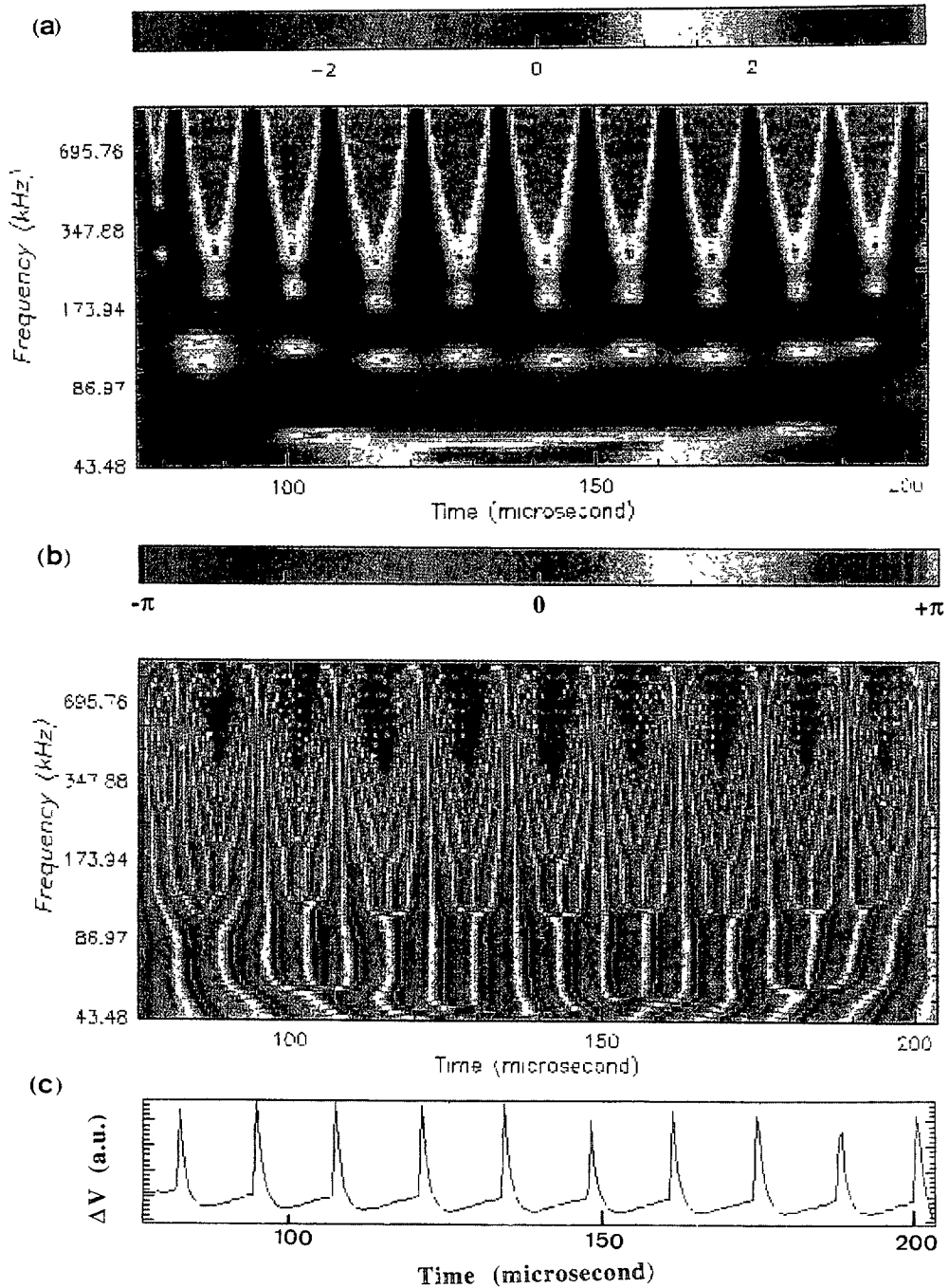


Fig. 3. — (a) Frequency (log scale) as a function of time for $I = 0.496 \mu\text{A}$. The intensity of the wavelet component increases when the color changes from black to pink. The color palette is in a logarithmic scale; (b) Phase as a function of time. The vertical axis is the frequency axis in log scale. The phase changes from $-\pi$ to $+\pi$ when the color changes from black to pink; (c) Transient voltage response as a function of time (arbitrary units).

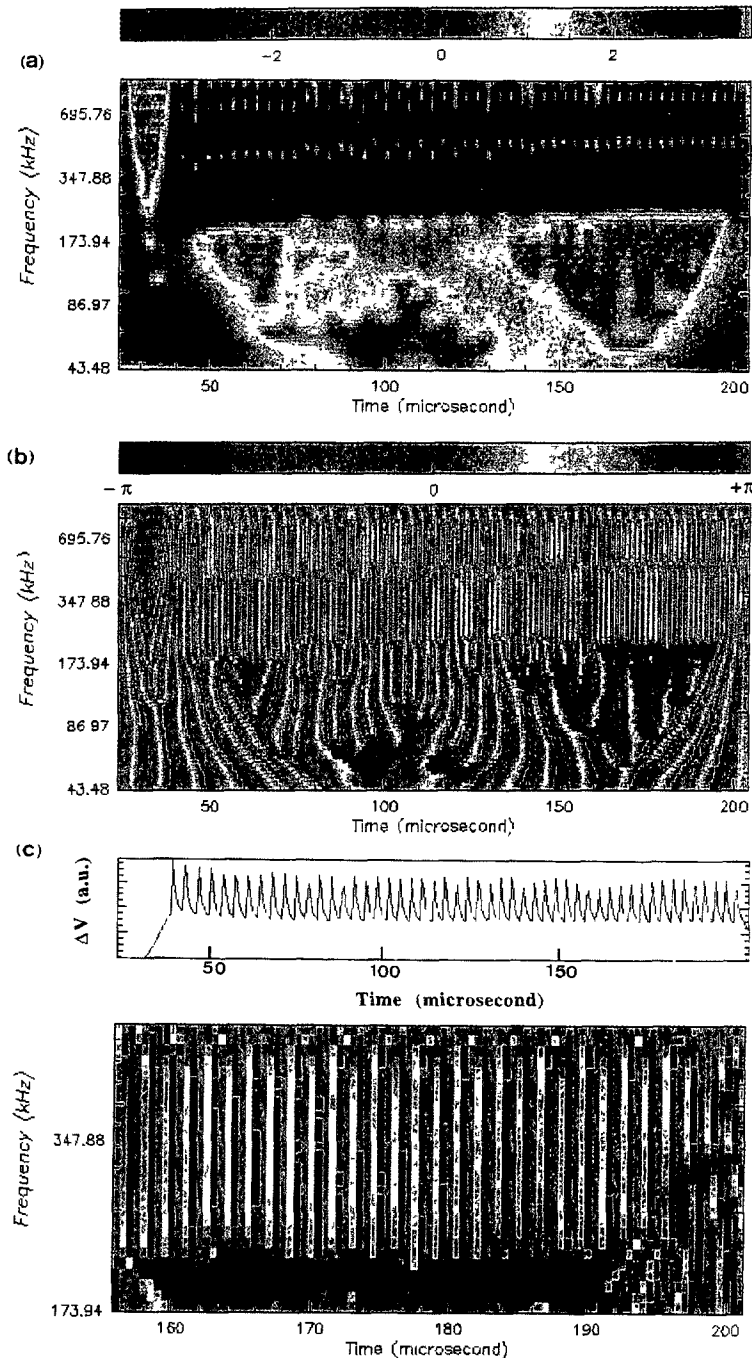


Fig. 4. — (a) Frequency (log scale) as a function of time for $I = 0.896 \mu A$; (b) Phase as a function of time. (c) Transient voltage response as a function of time (arbitrary units); (d) Expanded view showing the phase as a function of time in the time interval 160 – 200 μs .

not only these frequencies, but also higher-order harmonics with decreasing amplitudes. The superimposed stripes, periodic in time, which develop over a broad range of frequencies, correspond to the voltage spikes in the time domain shown in Figure 3(c), which act as Dirac peaks in the wavelet analysis. Figure 3(b) shows the time evolution of the phase corresponding to each frequency F_1 , F_2 , F_3 , F_4 . The vertical scale is still the frequency scale. It can be seen that the phase varies periodically from $-\pi$ (black) to $+\pi$ (pink).

Figure 4 shows the results of the wavelet analysis for a current pulse amplitude well above threshold. Two frequencies, the fundamental and the second harmonics, are observed, each with a finite spectral width, as a function of time. This width shows small fluctuations as a function of time. During a short time interval ($80 \mu\text{s} < t < 140 \mu\text{s}$) an additional low frequency F' , which appears in orange in Figure 4(a), is observed, whose spectral width shows fluctuations. F'

seems to increase during the time interval $80 - 100 \mu\text{s}$, then to decrease in the interval $115 - 140 \mu\text{s}$. The broad structures corresponding to the leading and trailing edges of the pulse current in the real time domain are artefacts. In contrast with the results shown in Figure 3(b), the time evolution of the phase shows a more irregular behavior, as indicated in Figure 4(b). Except for the additional low frequency F' where the corresponding phase varies monotonically, phase jumps are clearly visible, as shown in Figure 4(d), especially near the end of the pulse for the signal with the fundamental frequency F_1 .

3. Discussion

Our wavelet analysis demonstrates for the first time the possibility of detecting time resolved effects involved in the CDW dynamics. It yields a resolution of structures in the time evolution of the frequency and of the phase, not observed by conventional Fourier analysis. Another class of time resolved experiments involves studies of the transient structural response through high resolution X-ray scattering. Information on CDW deformations on a time scale of the order of ms has been obtained by this technique [14,15]. Temporal fluctuations of the NBN have been observed under mode-locking conditions [16].

Various models have been proposed to describe the voltage oscillations. In the simplest one, the CDW moves in a periodic potential which has the periodicity of $\lambda = 2\pi/Q$ along the chains. The CDW motion will be periodic with the fundamental frequency $\nu = Qv$ where v is the drift velocity of the CDW [7]. In an alternative model, probably more realistic, the oscillations originate at contacts or at boundaries between pinned and unpinned regions [5]. The conversion of normal carriers to CDW carriers occurs at CDW phase dislocation lines perpendicular to the chains. The CDW current changes rather abruptly whenever a dislocation line is created at one end or annihilated at the other. Observation of asymmetric voltage spikes, shown in Figure 1(b), seems to support the latter model. Figures 3(b) and 4(b) indicate that the phase fluctuations are periodic just above threshold and show rapid jumps well above threshold. This change of regime may be due to interactions between CDW phase dislocation lines. The frequency F' observed in a short time interval may correspond to a transient depinning followed by a gradual repinning of additional CDW domains.

4. Conclusion

In summary, we have shown that the wavelet analysis provides new data useful for a better understanding of the narrow band noise in CDW materials. This analysis method may open up a way to more systematic studies of dynamical properties and metastable phenomena of different CDW conductors.

Acknowledgments

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