

Complex dynamics in the peeling of an adhesive tape

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Abstract

Fracture is a very complicated phenomenon and its dynamics is not well described up to now by a consistent physical and/or mathematical model. In this paper we synthetically present the main experimental and theoretical results for the peeling of an adhesive tape, i.e. a viscoelastic dissipative system, viewed as a two-dimensional fracture propagation. From recent peeling experiments, using a common adhesive tape, the emergence of hierarchical structures in a broad range of time scales was observed in a definite region of the stick–slip regime, and this is one of the indicators commonly used speaking of complex systems.

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1. Introduction

The fracture process, which is the object of our investigation in this work, is one of the most complicated phenomena of the physical world [1]. Fracture results of the interplay between the creation of new interfaces and the elastic deformation of the bulk material. While the creation of new interfaces is dominated by the properties of elasticity of the surrounding medium and the amount of accumulated strain energy, the constitutive properties of the medium are strongly affected by the fracture propagation. In the full three-dimensional fracture of a brittle material (which is commonly referred to as “rupture”), the medium generally undergoes a progressive process of diffused damaging which then spontaneously concentrates into some region that is gradually crushed into fragments that slide and roll on each other, involving a great deal of different physical phenomena such as friction, adhesion, and plastic deformation [2,3]. Modeling such a complicated mixture of phenomena is almost hopeless even with the spreading power of modern computers [4]. Some simpler context must be chosen where a smaller number of phenomena are considered along with a simplified

geometry. The peeling of an adhesive tape provides an excellent example since the phenomenon is reduced to the propagation of a single coherent fracture front along a predetermined bi-dimensional interface [5]. Moreover, the dissipative nature of the viscoelastic systems has the significant effect of stabilizing the fracture dynamics. Even with these simplifications, the phenomenon remains highly non-linear and the dynamics shows a variety of instabilities and structures that suggest a possible underlying complexity. Furthermore, the peeling of an adhesive tape can be easily set up in experiments that provide very long data series from which it is possible to extract useful information on the non-linear features of the system.

The paper is organized as follows: Section 2 provides some basic knowledge about fracture mechanics in the context of the peeling of an adhesive tape; Section 3 describes the basic phenomenology and previous experiments; Section 4 presents different dynamical models for the stick–slip dynamics; Section 5 illustrates the new experimental results and Section 6 concludes with a discussion of the open problems.

2. Fracture dynamics and the peeling of an adhesive tape

In this section we will focus on the propagation of a plane crack in linear or non-linear elastic solids and

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non-linear viscoelastic systems, giving some general concepts and formulae concerning fracture mechanics, peeling and stick–slip dynamics [6].

The propagation of a crack along a predetermined interface is governed by an energy balance condition determined by Griffith [7]. More precisely, a crack will advance if the strain energy release rate $G = \partial U_M / \partial A$, i.e. the amount of mechanical energy U_M released by the system per unit new crack area A , is larger than the surface energy of the medium. When the dissipative phenomena produced at the crack tip are relevant, the above condition can be restated into an equation relating the strain energy release rate G to the crack tip velocity V

$$G = \Phi(V), \tag{1}$$

where $\Phi(V)$ is an empirical function which has the characteristic sigmoidal shape represented in Fig. 1. Although the qualitative shape of $\Phi(V)$ was explained by Maugis and Barquins [8], a microscopical model for this curve is still lacking.

We refer to “peeling” when a thin film of material is separated from a rigid substrate (Fig. 2). The relation between the fracture mechanics and peeling was established by Kendall [9], who linked the strain energy release rate G to the pull force F applied with a given angle θ to the free end of the film, of initial width b and

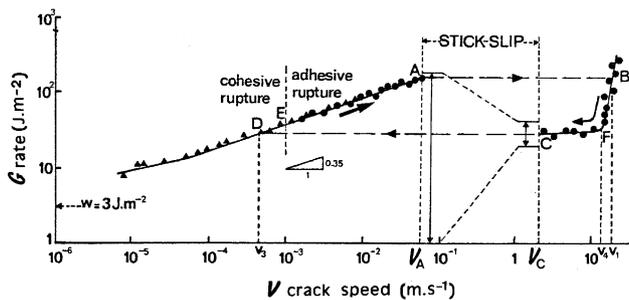


Fig. 1. Empirical $G = \Phi(V)$ curve from [12]. The stick–slip cycles are

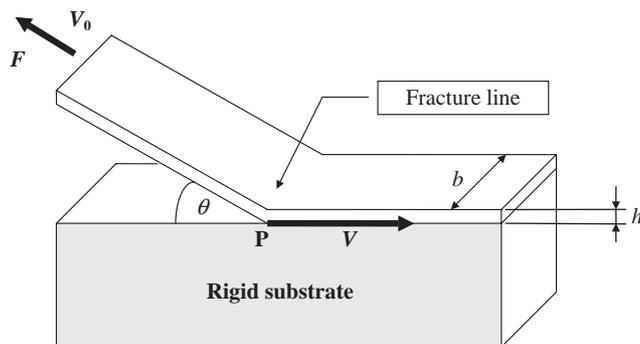


Fig. 2. A film of width b and thickness h is peeled apart from a flat rigid substrate. The force F is applied at an angle θ (peeling angle) relative to the rigid substrate.

thickness h , E being the Young modulus (Fig. 2)

$$G = \frac{F}{b} \cdot (1 - \cos \theta) + \left(\frac{F}{b}\right)^2 \cdot \frac{1}{2 \cdot E \cdot h} \tag{2}$$

The first linear term is related to the geometric configuration, and the quadratic one derives from the strain energy of the new peeled film. The Kendall equation is well established since it derives from the conservation of energy and is furthermore confirmed by numerous experiments. For peeling angles $\theta > 30^\circ$ (which is generally verified in our stick–slip experiments) Eq. (2) reduces to

$$G = \frac{F}{b} \cdot (1 - \cos \theta) = \Phi(V) \text{ or } F_0(V) = F \cdot (1 - \cos \theta) \tag{3}$$

where $F_0(V)$ is called the adherence force. As discussed in [10], Eq. (3) is some sort of a state equation, i.e. it describes the relation between the applied force and the crack tip velocity in stationary equilibrium conditions. However, it is expected to work also if the evolution of the variables is not too rapid in relation to some characteristic time which is still not estimated. So its applications in highly dynamical conditions and in the presence of strong non-linearity is very delicate and probably not completely correct in order to describe the instability propagation.

The stick–slip dynamics (or run-arrest) that appears in the peeling of an adhesive tape under certain conditions discussed later, is observed in a variety of phenomena like rock friction, earthquake dynamics, or tearing of rubber, etc. This intermittent motion (or self-sustained oscillations) is created by a mechanism that generates cycles of crack growth (or sliding) instability followed by subsequent arrest. The stability and instability alternation in peeling is produced by the competition between the change in the driving force (or energy release rate) and the change in the crack-growth resistance.

3. The previous main experiments and the empirical results

In general, the experiments on the peeling of an adhesive tape were performed using two different setups. In the first one the peeling is studied when a constant traction velocity V_0 is imposed onto the free end by the action of an electric motor (Fig. 3). In this case, with a fixed geometry, V_0 is the only dynamical control parameter, and the limit between the adhesive tape ribbon and the free tape may be seen as a crack tip propagating with speed V . In a second type of experiment the peeling is studied when a constant applied load is clamped to its extremity and the control parameter is the imposed force.

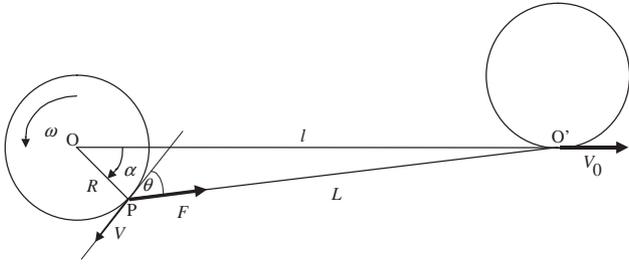


Fig. 3. The film is wound to a reel of radius R rotating with an angular velocity ω . The apparent position of the fracture is indicated by the angle α . V_0 is the traction velocity at point O' at a distance L from the fracture front. θ is the peel angle.

Barquins and Maugis [11,12] performed a series of experiments at constant traction velocity. The observed dynamics exhibits the following behavior: at slow traction velocity the tape is peeled regularly and the dynamics is stationary; at high velocity the dynamics is also regular, but very rapid; in the intermediate range of V_0 a stick–slip phenomenon appears, the peeling of the tape being jerky with emission of a characteristic noise. For increasing values of the traction velocity, the stick–slip motion is at first rather periodic, then it becomes more and more irregular. Moreover, an empirical $G = \Phi(V)$ curve was traced (Fig. 1) showing that the strain-energy release rate varied as a power law of the crack velocity V :

$$\Phi(V) = w + w \cdot a(T) \cdot V^{n_1}$$

with

$n_1 = 0.35$ for the first stable branch, and

$$\Phi(V) = G_C \cdot \left(\frac{V}{V_1}\right)^{n_2}$$

with

$n_2 = 5.5$ for the second (rapid) stable branch.

where $a(T)$ is a parameter depending on the ambient temperature T and w is the Dupré energy of adhesion.

In an experiment where the peeling was produced by a constant applied load, with the help of a set of different dead loads [13], the first stable region was found to be actually metastable, an unexpected stick–slip regime appears which was related to the inertia of the falling load, and the rapid stable branch was confirmed. The most relevant result was that the average value $\langle V \rangle$ of the measured peeling velocity in the stick–slip regime remains approximately constant increasing the value of the load of one order of magnitude [13].

Obviously, in all these experiments, we must consider some influences due to temperature and relative humidity. Our adhesive tape is substantially composed by polymer melts, made of long flexible molecules that naturally provide the properties of sticky materials: under stress, at long time scales, they have the properties of viscous liquids, and at short time scales they deform

as elastic solids. As it is well known, their properties depend strongly on temperature, especially near the glass transition temperature [14] where the polymer transforms progressively from a viscous material to a brittle solid. However, we want to study the fracture propagation and not the phase transition of the system; if temperature and humidity do not change too drastically, they do not affect the dynamics in a significant way. More precisely, since the stick–slip dynamics is very fast (the slowest cycles have a period of one second), it is not affected by long term environment variations, and even the long series of events are taken in substantially unchanged conditions.

4. The model

As a matter of fact, if one observes finely the macroscopic fracture line, he discovers that it is composed of a huge number of microfractures and microfilaments, but at present a microscopic model for the adhesion force and the crack in a viscoelastic system does not exist. This is the main difficulty in order to give a deep physical interpretation of the investigated phenomenon, i.e. to understand, describe and explain the fracture evolution. So we are constrained at the macroscopic level and generally the authors model the system by means of dynamical equations.

The first model [11] only takes into account the elastic degree of freedom, writing the equations

$$\begin{cases} \dot{G} = -\frac{k}{b} \cdot (V - V_0) \\ G = \Phi(V) \end{cases} \quad \text{with } \theta = \frac{\pi}{2} \quad (4)$$

where k is the stiffness of the free portion of the adhesive tape and b , its width. System (4) explains the stability of the positive-slope branches of the curve $\Phi(V)$, and also the presence of periodic stick–slip cycles when the traction velocity is in the intermediate domain, although it needs the assumption that speed jumps occur when the crack speed crosses the two critical values as shown in Fig. 1.

A common method to model the dynamical systems which have an unstable and/or irregular behavior is increasing the number of degrees of freedom. Following this, a second step was to add the roller inertia [15,13]. If we assume $G = \Phi(V) - \partial U_k / \partial A$, with $U_k = \frac{1}{2} \cdot I \cdot \omega^2$, where I is the moment of inertia and ω , the angular velocity, we can write the equations

$$\begin{cases} \dot{G} = -\frac{k}{b} \cdot (V - V_0) \\ \dot{V} = \frac{b}{m} \cdot [G - \Phi(V)] \end{cases} \quad \text{with } \theta = \frac{\pi}{2} \text{ and } m = \frac{I}{R^2} \quad (5)$$

which represent a two variable model. Choosing the fixed point $[V_0, \Phi(V_0)]$ as the origin and letting $x = V - V_0$ and $F(x) = \Phi(V) - \Phi(V_0)$, Eq. (5) become the

well-known Lienard equation

$$\ddot{x} + \mu \cdot \omega \cdot f(x) \cdot \dot{x} + \omega^2 \cdot x = 0, \quad (6)$$

where $f(x) = \partial F / \partial x$, and μ and ω are constant parameters. This equation is known to have solutions in the form of limit cycles when $f(x) < 0$. The transition from stable stationary dynamics to periodic stick–slip cycles is thus described as a Hopf bi-furcation occurring when the slope of $\Phi(V)$ changes from positive to negative. This two variable model produces results fitting the experimental data only when applied to the initial part of the stick–slip region where the phenomenon is periodic. But when increasing the traction velocity the self-sustained oscillations become more and more irregular, and System (5) is unable to describe and to predict the observed behavior.

Maugis and Barquins [11,12] are quite conscious of this: “the model is more complicated when the variation of the peel angle is taken into account, which gives a third degree of freedom (...) allowing a road to chaos when limit cycles are changed into strange attractors”. Following this suggestion Hong and Yue [16] added a third variable (the peeling angle θ or equivalently the position α (Fig. 3) of the crack tip) obtaining the system¹

$$\begin{aligned} F \cdot (1 - \sin \alpha) &= F_0(V), \\ I \cdot \dot{\omega} &= F \cdot R \cdot \sin \alpha, \\ \dot{F} &= k \cdot [R \cdot \sin \alpha \cdot \dot{\alpha} - (V - V_0)], \\ R \cdot \dot{\alpha} &= V - \omega \cdot R. \end{aligned} \quad (7)$$

Solving these equations numerically the authors affirm that chaotic orbits are present in a narrow range of the traction velocity (they found three positive Lyapunov exponents). The stick–slip would thus be a deterministic chaotic phenomenon and the problem seemed to be closed.

However, the issue appears to be more complex. If we study Eq. (7) more finely [17] we find out that the proposed solutions were obtained imposing jumps of the crack velocity V as in the first model discussed above. Using the first equation in System (7) to eliminate α we can rewrite the system in terms of a set of three equations in three variables (F, V, ω) [10]:

$$\begin{aligned} \dot{F} &= -k \cdot \left[\frac{F - F_0(V)}{F} \cdot (V - \omega \cdot R) - (V - V_0) \right], \\ \dot{V} &= \frac{1}{(dF_0/dV)(V)} \\ &\quad \times \left[\dot{F} \cdot \frac{F_0(V)}{F} - F \cdot \sqrt{1 - \left(1 - \frac{F_0(V)}{F}\right)^2} \cdot \frac{V - \omega R}{R} \right], \\ \dot{\omega} &= \frac{R}{I} \cdot [F - F_0(V)] \end{aligned} \quad (8)$$

¹This is actually a slightly modified set of equations due to different reference frame conventions. Moreover, Hong and Yue approximated $\sin \alpha$ with α .

which are valid for $dF_0/dV \neq 0$ and $F \neq 0$, with two singular points at $V = V_C$ and $V = V_A$. The numerical solutions of Eq. (8) admit nice non-periodic cyclic solutions only if they are forced to avoid the singularities, by jumping from one branch to the other as in Fig. 1 when the critical velocities are encountered. Without that assumption, System (8) does not provide any physical solution.

Moreover, we should also point out that a well-defined route to chaos does not exist. And we should also remember that the equation $F \cdot (1 - \sin \alpha) = F_0(V)$ is derived in stationary conditions. Therefore, it is not so natural and obvious to impose it as a constraint in a highly dynamical regime [10]. So we argue that the deterministic chaos in the irregular peeling dynamics seems to be rather artificial and not completely proved as intrinsic to the phenomenon. At this point, we can guess that the philosophy based on increasing the dynamical variables number in order to have a model able to describe the irregular stick–slip regime, is not the more suitable one.

The phenomenon of instability propagation in a viscoelastic medium could be more complicated than a “simple” dynamical system and could have some characteristics proper to a complex system [18,19]. This criticism has stimulated further theoretical and experimental research. The aim was to have a more accurate knowledge of the stick–slip propagation, firstly empirical. The highly non-linear phenomena as the fracture dynamics produce really unpredictable evolutions. In order to extract useful information on the motion from experiments, we must record sufficiently long time series, i.e. sequences of data representing the time evolution of one or more observables. After that, we can use the statistical analysis, or the geometrical reconstruction of the attractors in a suitable phase space, or some other technique to investigate the dynamics. The general philosophy of this approach is to draw out a physical meaning from an empirical signal, bypassing the knowledge of the underlying dynamics and/or the corresponding equations [20,21]. In this optics, we can search the significant points (bi-furcations and so on) and eventually we can detect the emergence of hierarchical structures, one of the most significant complexity indicators [22]. The problem will be to choose the good observable, i.e. the more proper observable to be measured with a sufficient precision and over a convenient long time. For this, we have set up a new experiment and the first provisional results show a stick–slip behavior more complex than we could expect basing strictly on the theory of dynamical systems [23].

5. The new experiment

The new experiment that has been performed aims at a complete description of the phase space of the

stick–slip dynamics and its evolution as a function of the control parameter V_0 .

The experimental assembly is the classic one with constant traction velocity. This condition is enforced with the aid of a very stiff motor that unrolls the tape on a cylindrical core (Fig. 3). We used an adhesive tape 50 mm wide, with a polypropylene backing covered by a rubber adhesive product, commonly utilized to close parcels [23]. Stable peeling is observed for traction velocities smaller than a first critical velocity V_C or larger than a second velocity V_A . In the intermediate range the peeling is jerky and the stick–slip dynamics becomes more and more complicated with rising traction velocity. We focused our attention on this region.

As a first experimental step, we chose to measure the time interval between two subsequent events, i.e. our observable is the period Δt determined by the analysis of the acoustic emissions produced by the stick–slip events. Usually, in physics, and especially in dynamics, the time is not a variable, but rather a parameter. However, in our situation the dynamical equations proposed to describe and explain the stick–slip at least partially failed. The time interval Δt is the indicator that we can measure more precisely in order to evaluate the irregularity of the dynamics. Obviously, the knowledge of the time interval series is not sufficient to model completely the dynamics. For example, we cannot discover if we are in the presence of space–time chaos only by numerical time series of periods, even if these series were chaotic. But at least our philosophy can put into evidence the time structures underlying the phenomenon and give a criterion to discriminate different dynamical regimes, as a function of the control parameter.

5.1. Analysis of data

Long records of events have been acquired for many values of the control parameter, i.e. the traction velocity V_0 , spanning the unstable stick–slip regime. Since the sequences may involve thousands of events, some automatic recognition process is necessary for the analysis of the acoustic measurements.

For low values of the traction velocity, the stick–slip events are clearly separated and regular. They appear as abrupt acoustic bursts, followed by a gradual oscillating decay. A simple criterion based on a threshold trigger followed by blind time window is enough.

When the traction velocity grows to higher values, the events become more and more frequent and irregular both in timing and intensity, making their separation problematic. A simple algorithm is no longer sufficient and some more elaborate criterion was developed. For this reason we performed a moving window Fast Fourier Transform on the signal and built a new index

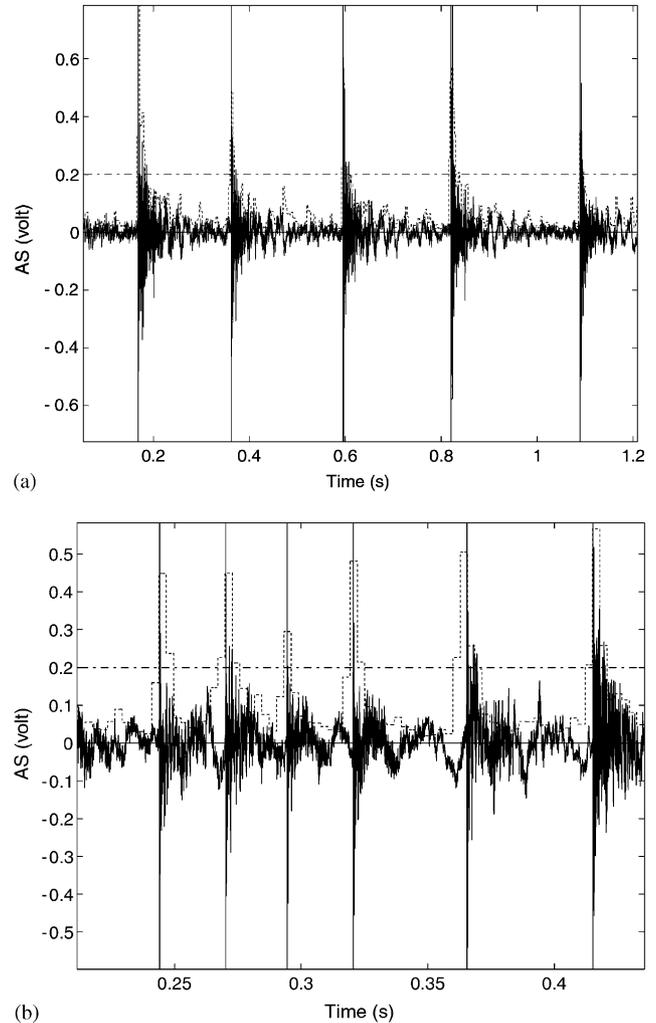


Fig. 4. Acoustic signal (AS, heavy line) recorded in a regular (a) and in an irregular (b) stick–slip region. The dotted signal is the FFT based index used for the automatic recognition of the slip events along with its threshold value (dash dotted).

based on the integration of the high frequency acoustic power. Such an index is plotted in red in Fig. 4 and is evidently well correlated to the events.

This method allowed us to identify the events up to a traction velocity $V_0 = 10 \text{ cm s}^{-1}$ with an efficiency better than 90%. After that, the oscillations in the signal appear to be almost continuous. A possible interpretation is that chaos and/or turbulence is completely installed [24]. But at present we cannot exclude that we have reached the limit of resolution of our apparatus, and that we are not able to distinguish the events.

5.2. The emergence of hierarchical structures

Although the stick–slip cycles are generally not periodic, it is interesting to plot the average period as a function of the traction velocity (Fig. 5). In first approximation the average frequency is proportional to

the traction velocity and a linear fit provides the relation

$$V_m = \frac{1}{\langle T \rangle} = 0.53 \text{ Hz} \cdot \left(\frac{V_0}{2.1 \text{ mm s}^{-1}} \right).$$

Actually, the cycles are only quasi-periodic for low traction velocities, then they become more and more

irregular with increasing speed. The analysis of the time intervals has put into evidence that the dynamics goes through a series of progressive complications when the traction velocity V_0 passes some subsequent critical values. In particular, there is a first low velocity domain in which the cycles are approximately periodic and for which the period is initially about 1 s and then diminishes as the traction velocity is increased (Fig. 6). This domain is followed by the appearance of sparse rapid events which have a period (here simply meaning the time interval between two subsequent events) that is a multiple of the fixed short time interval $t_1 = 0.02 \text{ s}$ (Fig. 7). The events of double or triple period become progressively more frequent and clustered leading to the establishment of an ordered structure with only three possible periods at a traction velocity $V_0 = 3 \text{ cm s}^{-1}$, which will be studied in more detail later. Further increasing the traction velocity, the multiple period events decrease in relative frequency until a new regular regime is observed with substantially periodic events at the short time interval $t_1 = 0.02 \text{ s}$. This time interval does not change with traction velocity up to another critical value where the cycles undergo a second bifurcation with the appearance of sparse events with a duration multiple of a new shorter fixed time interval

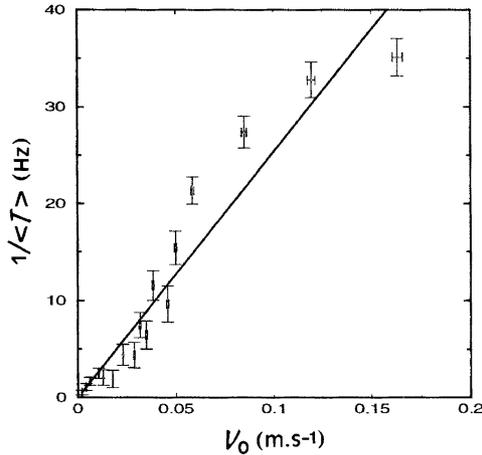


Fig. 5. Average frequency ($1/\langle T \rangle$) of the stick–slip cycles as a function of the traction velocity V_0 .

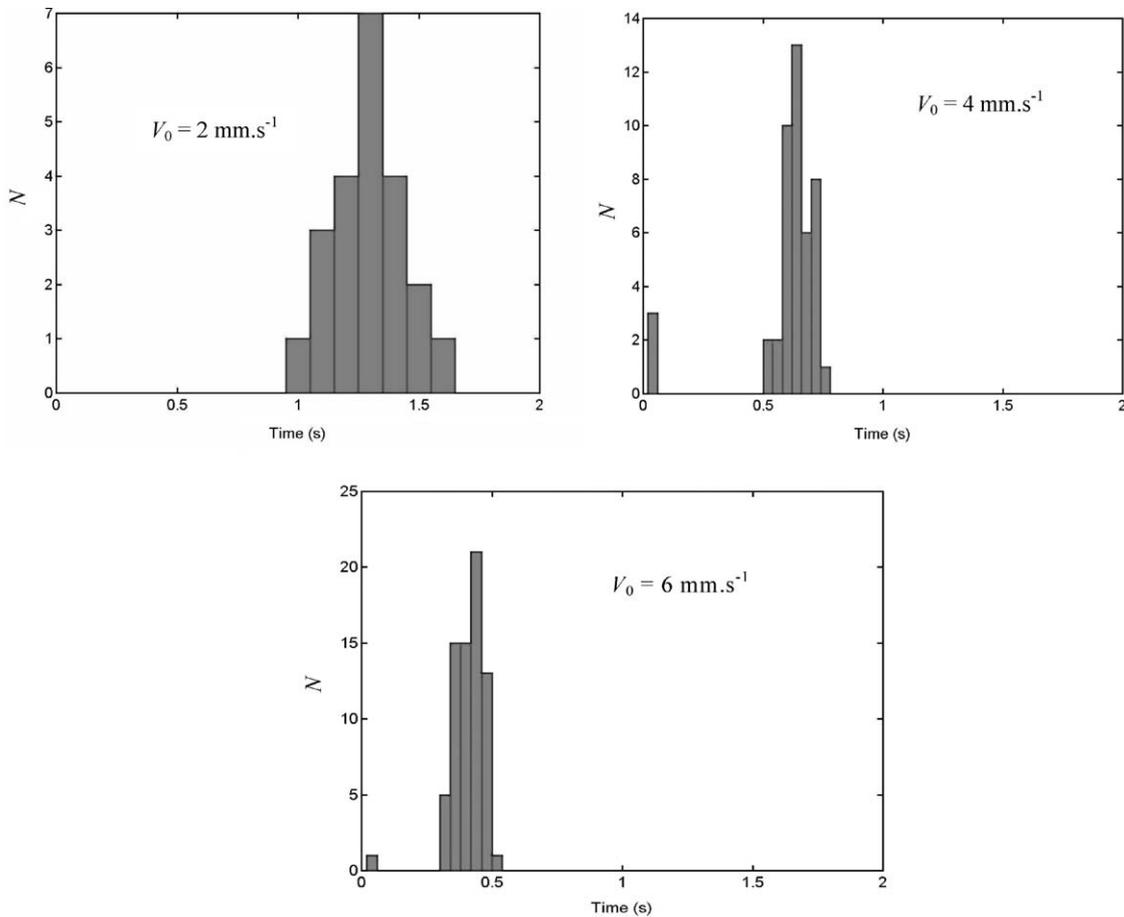


Fig. 6. Distribution of the time intervals for $V_0 = 2, 4$ and 6 mm s^{-1} .

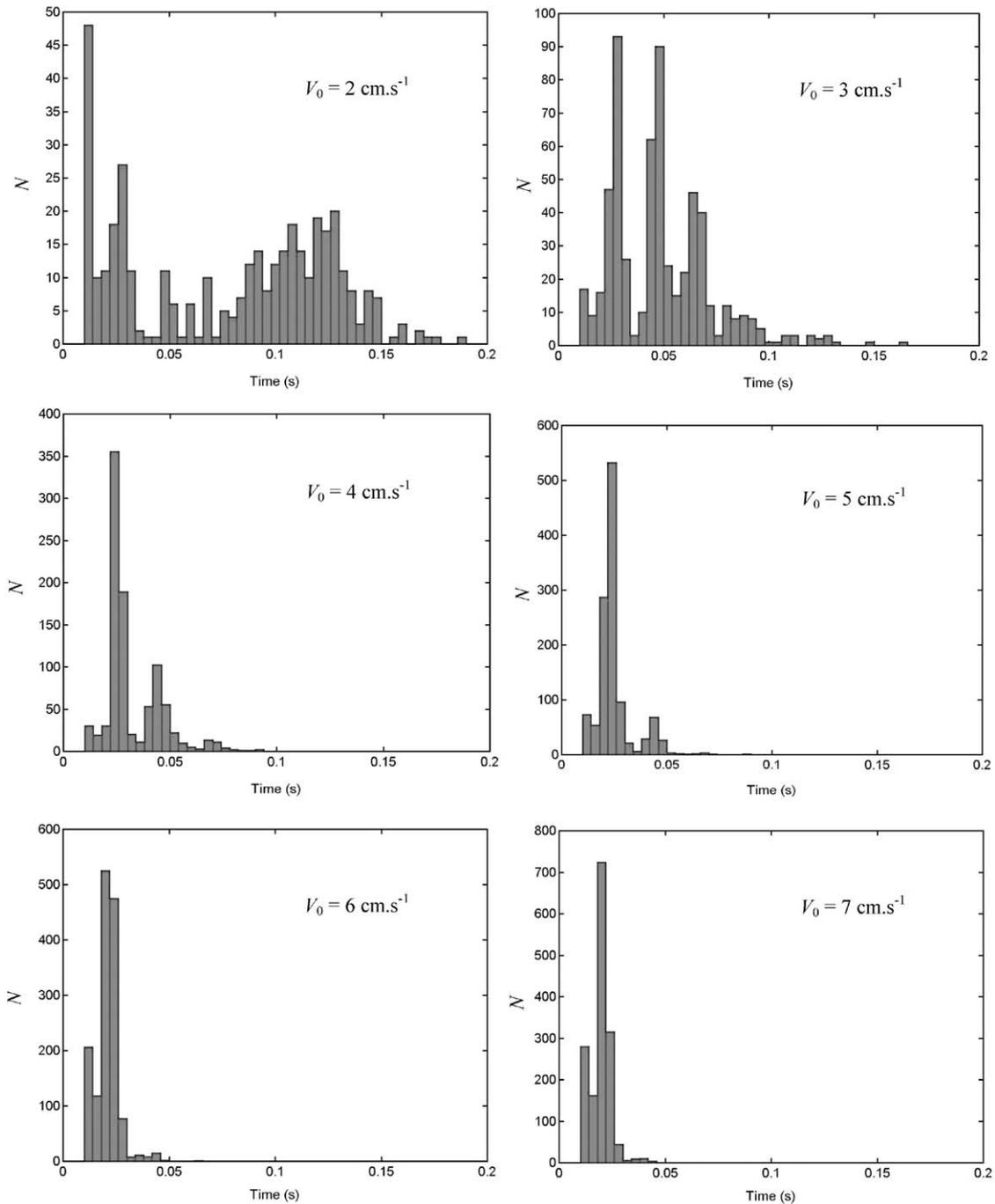


Fig. 7. Distribution of the time intervals for $V_0 = 2, 3, 4, 5, 6$ and 7 cm.s^{-1} .

$t_2 = 0.001 \text{ s}$, which is about 20 times shorter than the previous one. These measurements are still in analysis, due to the increased difficulties of separating such a dense series, but we can anticipate that a new regular structure with fixed multiple periods is again developed. At higher velocities the acoustic signal is mixed to such an extent that we cannot distinguish any event at the present state. However, at a traction velocity of 3 m.s^{-1} the peeling becomes again stable without the crackling

noise and it remains stable until the reach of the maximum traction velocity of the engine 6 m.s^{-1} .

In order to verify that the observed structures are not generated by artifacts we reanalyzed the most significant datafiles with a standard sound editing software and checked each single event by visual and acoustic inspection. Despite some minor mismatch with the automatic recognition technique, more than 90% of the slip events were confirmed and the structures of

the time interval distribution were shown to be a real effect.

We can resume complication cascade like follows:

- up to 0.7 mm s^{-1} : stable peeling;
- at 0.7 mm s^{-1} beginning of regular stick–slip. Time periods fall with growing traction velocity;
- at 1 cm s^{-1} apparition of first multiplets with $\Delta t \approx 0.02, 0.04, 0.06 \text{ s}$. The structure develops up to 3 cm s^{-1} , then it concentrates on the shortest interval;
- at 6 cm s^{-1} apparition of sub-multiplets with $\Delta t \approx 0.001, 0.002, 0.003 \text{ s}$. The structure develops, but the signal is lost at 10 cm s^{-1} ;
- at 3 m s^{-1} the peeling becomes again stable with a soft hum, up to the maximum traction velocity of the engine 6 m s^{-1} .

5.3. Developments

Since dynamic modeling was proven to be unsatisfactory in describing such a complex phenomenology, the data are now being analyzed with a different approach, that is by a statistical analysis of the series of time intervals, with the aim of investigating the correlation between subsequent cycles and extracting the predictive information hidden in the data [25].

In particular, we analyzed the series of time intervals of the data with $V_0 = 3 \text{ cm s}^{-1}$, for which the intervals only have the three multiple durations (see Fig. 7). In Fig. 8 the same data are represented in the phase space $\{\Delta t(n), \Delta t(n+1)\}$ to put into evidence the establishment of correlations and structures. Preliminary statistical analyses show that the subsequent intervals are not

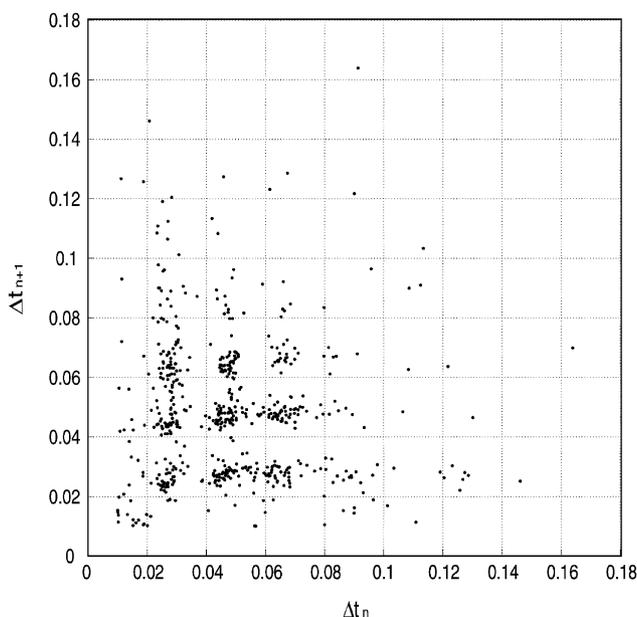


Fig. 8. Correlation between adjacent events of the three level structure ($V_0 = 3 \text{ cm s}^{-1}$) in a phase space $\{\Delta t(n), \Delta t(n+1)\}$.

independent, but they are neither well described by a first-order Markov process. We are presently evaluating the predictive power of the series and the presence of non-linear correlations by an evaluation of the embedding dimension.

6. Conclusions

As we have seen, the main dynamical models constructed to explain the peeling evolution of an adhesive tape can predict correctly only the stationary behaviors and the approximately periodic stick–slip cycles. But when the stick–slip becomes very irregular the proposed models are insufficient even if we increase the number of degrees of freedom.

On the other hand, the results of new experiments show that the stick–slip dynamics is more rich and complicated than a simple bi-furcation's route to chaos. We observe hierarchical structures in a definite traction velocity range that can suggest the emergence of complexity when a fracture is produced and evolves in a viscoelastic system.

It could be possible that the observed complex dynamics leads to time chaos, but a statistical interpretation seems more plausible: for high traction velocities the peeling in the stick–slip regime could not any longer be described by dynamical equations, but it would rather be the result of the interaction of a large number of degrees of freedom cooperating to constitute a self-organized system far from equilibrium.

At last, we want to underline how a common Scotch[®] roller can be so enigmatic and full of mystery to justify the ancient saying “I know that I don't know”. Perhaps this sentence could be the deep meaning of “complexity”.

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