Electrical properties of granular matter: From “Branly effect” to intermittency

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Electrical phenomena in granular materials related to an electrical conduction transition (known as the Branly effect) have been interpreted in many different ways but without a clear demonstration. We report the observation of electrical transport through a chain of oxidized metallic beads. A transition from an insulating to a conducting state is observed as the applied current is increased. We show that this transition comes from an electro-thermal coupling in the vicinity of the microcontacts between beads where microwelding occurs. An implicit determination of the microcontact temperature is obtained. In a metallic powder, temporal fluctuations of resistance are additionally observed, and are studied with the tools used in hydrodynamic turbulence. We show that this electric noise displays scale invariance (over 4 decades in time) and intermittency, which trace back to thermal expansion locally creating or destroying electrical contacts. As in the case of hydrodynamic turbulence, the noise properties are well described by a multiplicative cascade model.

1 INTRODUCTION

The coherer or Branly effect is an electrical conduction transition that appears in a slightly oxidized metallic powder under a constraint (Branly 1890). The initial high powder resistance falls irreversibly by several orders of magnitude as soon as an electromagnetic wave is produced in its vicinity. Such a wave detector was used in the first wireless radio transmissions at the end of the 19th century. However, this phenomenon still is not well understood. Several possible processes at the contact scale have been invoked without a clear verification: electrical breakdown of the oxide layers on grains (Kamarinos et al. 1975), the modified tunnel effect through the metal-oxide/semiconductor-metal junction (Holm 2000), the attraction of grains by molecular or electrostatic forces (Gabillard & Raczy 1961), and local welding of microcontacts by a Joule heating effect (Vandembroucq et al. 1997; Dorbolo et al. 2003; Holm 2000). A global process of percolation within the grain assembly also has been invoked (Kamarinos et al. 1975; Gabillard & Raczy 1961; Vandembroucq et al. 1997).

This effect was discovered in 1890 by E. Branly and is related to other phenomena. For instance, when a dc electrical source is directly connected to the granular sample and exceeds a threshold voltage, a conduction transition (the dc Branly effect) is observed (Branly 1902; Guthe & Trowbridge 1900) ; temporal fluctuations and slow relaxations of resistance also occur under certain conditions (Kamarinos et al. 1975; Falcon et al. 2004).

Our goal in this paper is to understand the dc Branly effect by means of an experiment with a chain of metallic beads (Falcon et al. 2004; Falcon & Castaing 2005). Our focus is on the local properties (the contacts between grains). The second goal is to characterize and to explain the origin of the temporal fluctuations of resistance in a metallic powder submitted to a dc voltage in a 3D container (Falcon et al. 2004). In this second part, our focus is on the collective properties of the granular matter.

2 DC BRANLY EFFECT

The experimental setup consists of a chain of 50 identical stainless steel beads, each 8 mm in diameter, and $0.1\,\mu$m in roughness. The beads are surrounded by an insulating medium of polyvinylchloride (PVC). A static force $F \leq 500\,\mathrm{N}$ is applied to the chain of beads. During a typical experiment, we supply a current ($10^{-6}\,\mathrm{A} \leq I \leq 1\,\mathrm{A}$) and simultaneously measure the voltage U, and thus the resistance $R = U/I$. Similar results have been found by repeating the experiment with an applied voltage and measuring I and thus R. The number of beads N between the two electrodes is varied from 1 to 41 by moving the electrode beads within the chain. The lowest resistance of the entire chain (a few ohms) is always found to be much higher than that of the electrode and the stainless steel bulk material. Because no particular precautions were

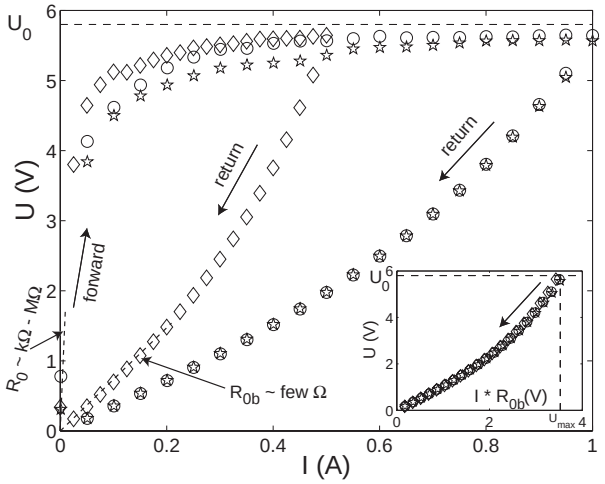

Figure 1. U-I characteristics of a chain of $N = 13$ beads for various forces $F = 56$ N ($\circ$), 100 N ($\star$), 220 N ($\diamondsuit$), and for various current cycles $I = 0 \to 1\text{A} \to 0$ ($\circ$, $\star$), and $I = 0 \to 0.5\text{A} \to 0$ ($\diamondsuit$). The U-I characteristics are nonlinear, hysteretic, and saturate to a low voltage per contact $U_0/(N+1) \simeq 0.4\,\text{V}$. Inset: reversible return trajectories rescaled by $R_{0\text{b}}$.

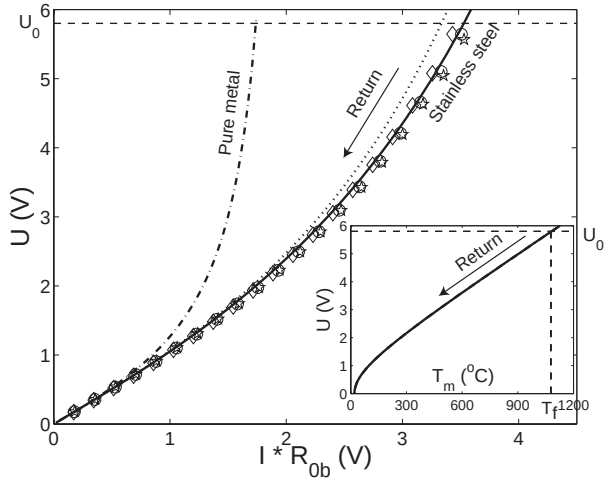

Figure 2. Experimental U-I reverse trajectories of Fig. 1 (symbols) and theoretical curves for an alloy with stainless steel properties [AISI 420 ($-$) or AISI 304 ($\cdots$)], and for a pure metal ($-\cdot$). Inset: theoretical maximum temperature, T_m, reached for one contact as a function of the potential difference, U, across the $N = 13$ stainless steel beads in the chain.

taken for the beads, an insulating film (oxide or contaminant), a few nanometers thick, is likely present at the bead-bead contact.

When the applied current to the chain is increased, we observe a transition from an insulating to a conductive state as shown in Fig. 1. At low applied current and fixed force, the voltage-current U-I characteristic is reversible and ohmic with a high, constant resistance, R_0. This resistance ($R_0 \simeq 10^3$–$10^6\,\Omega$) at low current depends in a complex way on the applied force and on the contaminant film properties (resistivity and thickness) at the contact location. The value of R_0 is determined by the slope of the U-I plot at low current. As I is increased further, the resistance strongly decreases, corresponding to a bias U_0 independent of I. As soon as this saturation voltage U_0 is reached, the U-I characteristic is irreversible if the current is decreased. The resistance reached at low decreasing current, $R_{0\text{b}}$ (the order of 1–$10\,\Omega$), depends on the previously applied maximum current, $I_{\max}$. Note that the nonlinear return trajectory is reversible upon again increasing I, and also is symmetrical when I is reversed (Falcon et al. 2004). For different forces F and values of $I_{\max}$, we show that the return trajectories depend only on $I_{\max}$ and follow the same return trajectory when U is plotted versus $IR_{0\text{b}}$ (see the inset in Fig. 1). The values of $R_{0\text{b}}$ are determined by the slopes of the U-I return trajectories at low and decreasing current (see Fig. 1).

The decrease of the resistance by several orders of magnitude (from R_0 to $R_{0\text{b}}$) is similar to that of the coherer effect with powders (Branly 1890) and with a single contact (Branly 1902; Guthe & Trowbridge 1900). Note that after each cycle of the current, the applied force is reduced to zero, and we roll the beads along the chain axis to form new contacts for the next cycle. The saturation voltage U_0 is independent of F, but depends on the number of beads, N, between the electrodes. The saturation voltage per contact $U_c \equiv U_0/(N+1)$ is found to be constant when N is varied from 1 to 41 and is on the order of 0.4 V per contact. The value of this saturation voltage depends on the bead material (Falcon et al. 2004).

From this simple experiment, we have shown recently (Falcon et al. 2004; Falcon & Castaing 2005) that the transition triggered by the saturation voltage arises from an electro-thermal coupling in the vicinity of the microcontacts between each bead. The current flowing through these spots generates local heating. This current-induced temperature rise (up to $1050°$C) results in the microwelding of contacts (even for a voltage as low as 0.4 V). Since the temperature, T, being limited to the melting or softening one, further increase of the current just increases the microcontact areas. This self-regulated mechanism in temperature is also self-regulating in voltage. Indeed, the thermal conductivity, $\lambda(T)$, and the electrical resistivity, $\rho(T)$, of the bead material are linked through the Wiedemann-Franz law, $\lambda\rho = LT$, where $L = \pi^2k^2/(3e^2) = 2.45 \times 10^{-8}\,\text{V}^2/\text{K}^2$ is the Lorentz constant, k is the Boltzmann constant, and e is the electron charge. If we combine this equation and the thermal equilibrium one, we can express the universal relation between the maximum temperature reached, T_m, at the microcontact, and the applied voltage, U, as

$$T_m^2 - T_0^2 = \frac{U^2}{4L}, \qquad (1)$$

T_0 being the room temperature (the bead bulk one). A voltage near 0.4 V across a contact leads, from Eq. (1), to a contact temperature near $1050°$C (for a bulk temperature $20°$C), exceeding thus the melting point of most conducting materials. Equation (1) shows that the maximum temperature T_m reached at the contact is independent of the materials in contact because both the electrical resistivity, $\rho(T)$, and the

thermal conductivity, $\lambda(T)$, are due to the conduction electrons, which leads to the temperature dependence given by the Wiedemann-Franz law.

Based on the temperature dependence of the resistivity, an analytical expression for the nonlinear U-I reverse trajectory is derived (Falcon & Castaing 2005), and is found to be in good agreement with the data (see Fig. 2). As shown in the inset of Fig. 2, the theory also allows for the determination of the microcontact temperature, T_m, through the return trajectory with no adjustable parameters. The measurement of the saturation voltage per contact allows us to obtain a determination of the melting temperature, T_f, of the bead material (see inset of Fig. 2). We could attempt to directly visualize this process with a microscope or infrared camera. But for this purpose a very powerful electrical source must be applied, far in excess of that necessary to produce true coherer phenomena (Vandembroucq et al. 1997).

3 NOISE AND INTERMITTENCY

One gram of copper powder (100 μm in diameter) is confined in a Plexiglas cylinder, 10 mm in diameter, capped with two brass electrodes. The powder height is 5 mm corresponding to roughly 500,000 particles. A mechanical pressure up to 200 kg/cm^2 is applied on the powder. We let the system relax for one day before performing electrical measurements. A dc voltage, U, is then applied to the powder, and we simultaneously measure the current I across the powder, and thus its resistance $R = U/I$. Two wires can be embedded inside the powder to check that the resistance is not controlled by the electrode-powder interface. Before each experimental run, the container is refilled with a new sample of powder. This procedure ensures better reproducibility than simply relaxing the confining pressure and shaking the container.

At fixed force and small applied voltage, the powder resistance is very large of value R_0 (few megohms), due to the oxide layer on grains. When U is increased enough, the voltage-current U-I characteristic becomes nonlinear, and undergoes for a critical tension, U_c, a transition from an insulating to a conductive state (few ohms). The main difference with the 1D experiment in Sect. 1 is that the critical tension depends on the applied force, such that the transition threshold always corresponds to the same Joule dissipated power $P_c \equiv U_c^2/R_0 \simeq 10^{-4}$W whatever the applied stress (Falcon et al. 2004). This suggests that the transition comes from a thermal instability. This is consistent with the 1D experiment, but in a more complex way. Since the contact distribution in a powder is very inhomogeneous, one would also expect an inhomogeneous difference potential distribution between each contacts. Thus, the number of contacts carrying the current (the current path) should depend on the initial contact configuration, that is on R_0.

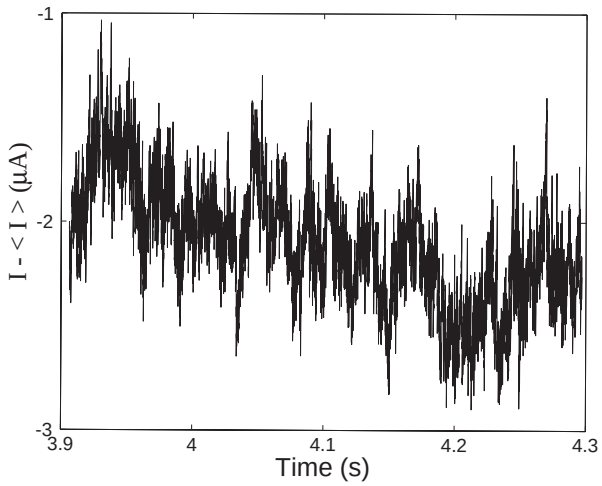

Figure 3. Typical fluctuations of current, $I(t)$, as a function of time (quasi-stationary current noise). The powder resistance is of the order of $k\Omega$ and the applied tension is few Volt.

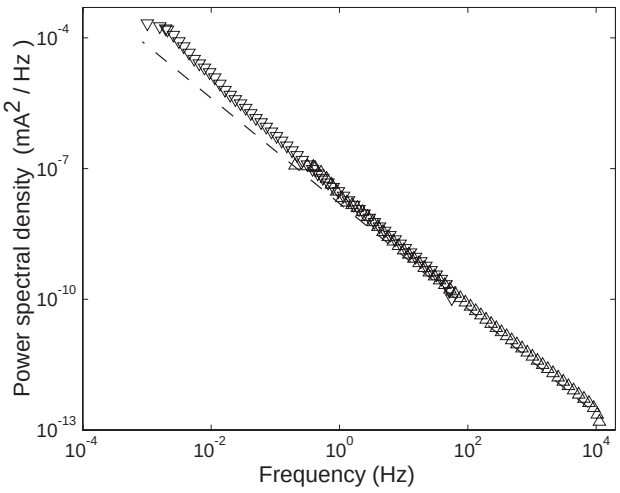

Figure 4. Power spectral density, $S(f)$, of current fluctuations sampled at low ($\bigtriangledown$) and ($\bigtriangleup$) high frequencies. Dashed line corresponds to $S(f) \sim f^{-1.3}$.

Let us focus on the temporal evolution of the current through the powder. According to the value of the dissipated power $P \equiv U^2/R_0$, two cases occur: for $P > P_c$, the current fluctuates rapidly as a noise, whereas for $P < P_c$, its evolution is slow as a relaxation. Only the first case is discussed below. An example of quasi-stationary current noise in the powder is shown on Fig. 3 with stochastic fluctuations. Both increasing and decreasing events occur for the current. This is contradictory to what would result from a cascading electrical breakdown for the oxide layers, as previously proposed (Kamarinos et al. 1975).

The current fluctuations are recorded with a low and a high sampling frequency (respectively $f_l = 128$ Hz and $f_h = 25.6$ kHz) during long times (respectively 65 min and 20 s). Each type of data acquisition was repeated 20 times to extract averaged quantities, due to quasi-stationary feature of the signal. The log-log power spectral density of the current fluctuations is displayed on Fig. 4, and shows a power law over more than 4 decades in frequency (1 Hz $< f <$ 10 kHz). This scale invariance means that no typical frequency is relevant to this problem. It is cut at

low frequency by the typical diffusive time of a thermal perturbation within the typical size sample estimates corresponding to 1 - 10 s. This order of magnitude agreement supports the hypothesis of a thermally driven phenomenon.

The probability density functions (PDF) of current increments, $\delta I_\tau(t) = I(t+\tau) - I(t)$, are measured for various time lags τ. These PDF are symmetric and their shapes change with the time scale τ – this is a direct signature of intermittency (Falcon et al. 2004). Moreover, the mean-squared deviation of $\ln|\delta I_\tau|$ decrease logarithmically with the time scale, as well as its two-time correlation, and the slopes are equals (Falcon et al. 2004). The agreement between them and the linearity in $\ln\tau$ strongly supports a multiplicative cascade description (through the time scales) for the intermittency. Thus, as in the case of hydrodynamic turbulence, large scale events condition those at a smaller scale. In turbulence, this is the energy that cascades from the large scales toward small ones, before being dissipated. Here, the cascading quantity is still unknown.

We can now propose a physical picture of what is taking place. As shown above, the driving parameter of the conduction transition is the total dissipated power. This suggests either local heating, able to change the electrical properties of contacts (several hundreds of degrees are needed in this case, as in the 1D experiment in Sect. 1), or thermal expansion, which locally creates or destroys contacts. The power involved is of the order of 10^{-4}W. When divided by the number of contacts, it becomes so small that it cannot generate a temperature of several hundreds of degrees. Moreover, the influence of the large time scales on the small ones, as shown by the observed long–time correlations, cannot be taken into account by such a local process. Therefore, thermal expansion seems to be the mechanism driving the intermittent noise.

In this spirit, we attempt to show the correspondence between the formal multiplicative cascade and electrical conduction in our powder. Since the contact distribution in a powder is very inhomogeneous, one would also expect an inhomogeneous current distribution. Thus, very small Joule heating should create inhomogeneous increasing stresses in the powder. Very small thermal expansion can result in dramatic changes in the current paths, thus in the distribution of this Joule heating, and so on. Such events can occur at any scale, ranging from the size of the sample and the grain size. The large scale events should influence the small ones, as suggested by our study. Note that such a very small thermal expansion of a single particle has been shown to produce a change in the force chains, and thus in the sound propagation within the medium (Liu 1994).

4 CONCLUSIONS

By means of an experiment with a chain of metallic beads, we show, for the first time, that the mechanism of the electrical conduction transition (dc Branly effect) results from the local heating of the microcontacts between each bead where microwelding occurs (Falcon et al. 2004; Falcon & Castaing 2005). The increase in temperature reaches $1050°$C for an applied voltage as low as 0.4 V. The electrical conduction transition is connected to the local properties of the contact between two grains. It is a first step towards more realistic granular media such as a 2D network of ordered beads (including the disorder of the contacts), and a metallic powder sample (including the disorder of position). When a dc voltage is applied to a metallic powder, the temporal evolution of the current is then very noisy under certain conditions. We show that this electric noise has interesting properties of scale invariance (over 4 decades in time) and of intermittency which come from thermal expansions locally creating or destroying the electrical contacts (Falcon et al. 2004). These expansions can occur at any scale, ranging from the size of the sample to the grain size. The large scale events condition those at a smaller scale, as in the case of hydrodynamic turbulence. This phenomenon of self-similarity traces back to the collective effects of the granular matter.

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