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Reexamination of the Branly effect

S. Dorbolo, M. Ausloos, and N. Vandewalle
GRASP, Institut de Physique B5, Université de Liège, B-4000 Liège, Belgium

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The electrical resistance of a metallic granular packing has been recorded at room temperature. A nearby burster between which sparks are produced, induces a decrease in the resistance of the granular packing as described in the works of Branly. Our measurements emphasize that the decrease is continuous and the resistance variations behave like a stretched exponential law due to the creation of new electrical paths as in nucleation-growth soldering processes. This behavior has been identified to be a diffusionlike process.

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Among unsolved physical problems, the Branly's coheror is one of the most amazing phenomena. In 1891, Branly discovered that a spark can modify the electrical state of a distant packing of metallic filings [1]. The electrical resistance of a granular packing which is initially high falls down several orders of magnitude as soon as an electromagnetic wave is produced in its vicinity. This was the starting point of radiocommunication. This principle was used by Marconi thereafter: a tube filled with filings of nickel and silver is able to intercept a transatlantic radio signal [2].

Even if scientists understand how an electronic circuit can produce and receive electromagnetic waves, the Branly's coheror remains an enigma as far as its surprising high sensitivity to mechanical and electromagnetic perturbations is concerned [3].

Recent studies have shown that the electrical properties of a metallic granular packing are complex. Indeed, current-voltage i-V curves are nonlinear and exhibit memory effects [4–6]. The origin of such behaviors has to be found in the relationship between the geometrical, chemical, electrical, and mechanical structures of the contact network constituted by the grain junctions.

The macrostructure of granular materials is characterized by arches that distribute the weight of the packing towards the edges of the vessel. As demonstrated by Hertz [7], the electrical resistance R of grain contacts is a power law of the applied pressure on it. A granular packing can be, therefore, reduced to a skeleton constituted by the lines of forces [8–10]. The quality of the electrical contacts can be widely distributed. Conjugated with the distribution of resistances, the nature of the contacts plays a key role since it clearly drives the nonlinearity and the memory effects behavior in i-V curves.

From the microscopic point of view, Branly's effect is better observed when the grains are slightly oxidized. That means that an oxide layer covers the grains to form a metal-oxide-metal junction. This insulator layer constitutes a barrier to the current flow. Such phenomena are well described in solid state physics lectures: such thin barriers lead to exponential relations between the voltage and the current as in diodes [6,11]. It is easily recognized that such a network with tremendous range for the resistances is essentially a percolation problem [12–14]. Taking into account the structure of the network and the nature of the contacts, discontinuities in i-V curves may be explained [6].

The contacts are subject to damage caused by the current flow. Indeed, jumps, first discovered by Calzecchi-Onesti [15], occur when the current reaches a certain value; the resistance then falls several orders of magnitude. This process is very similar to breakdowns in semiconducting devices and to Zener breakdown [16]. However, solderings between grains may occur before the Calzecchi-Onesti transition [17]. These solderings are irreversible processes and explain the hysteretic behavior found in i-V curves of granular packing.

In the present paper, we propose to revisit the Branly effect in view of those recent studies. Using a very sensitive voltmeter and a very stable current source, the electrical resistance of a packing has been recorded as a function of time while electromagnetic waves hit the system. This allows for a fine analysis of the Branly effect and to extract a generalized Avrami exponent [18] that allows us to deduce the type of process occurring in the system.

The experimental setup is very similar to the one used by Branly more than a century ago. About 14 000 polydisperse lead beads are arranged in a parallelepipedic vessel ($50\times 50\times 40$ mm^3). The mean diameter is 2.35 mm and the polydispersity is 2%. Energy dispersive x-ray measurements show that the surface is composed of a nanometric layer of lead oxide. The density of such packing is about 0.75 as is naturally a three-dimensional close packing configuration. Electrical current leads are composed of two lead rectangular plates. They are placed on two opposite faces of the vessel and connected to a Keithley K2400 current source. A Rhumkorff coil is used to produce sparks. The burster is located at a distance between 100 and 1000 mm from the granular packing. The distance between electrodes of the burster is 5 cm and the voltage in the primary coil is 8 V. The geometrical setup of the experiment is displayed in Fig. 1.

Experiments have been performed in several conditions at room temperature: the injected current i through the beads, and the distance between the spark generator and the vessel have been tuned. In Fig. 2, a typical variation of the resistance R normalized by the initial resistance R_0 is shown versus time for an injected current $=0.1$ mA and a distance between sparks and vessel about 100 mm. During the first 20 sec, the resistance R_n is at its maximum, and decreases very slowly with time (logarithmically). This should be attributed to a slow aging of the contacts with time. This behavior can

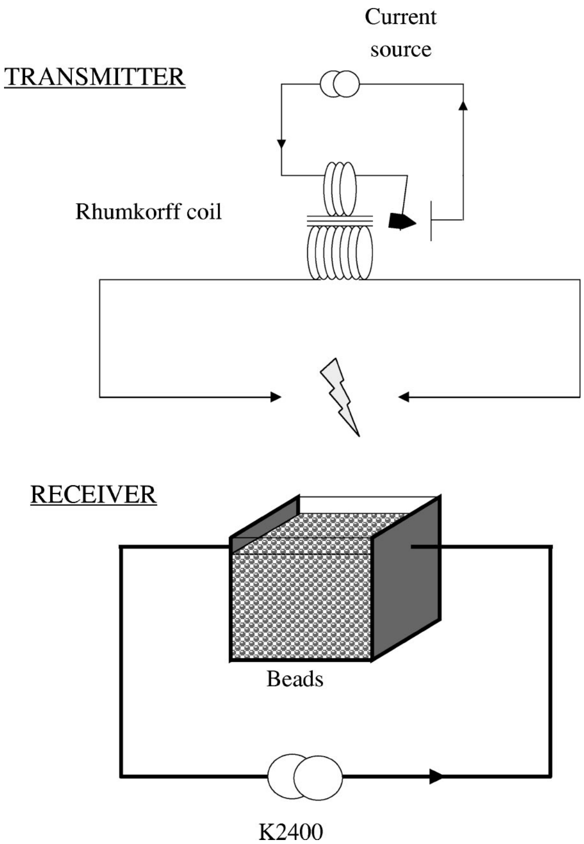

FIG. 1. Sketch of our revisited Branly experiment. A transmitter composed of a Rhumkorff coil produces sparks between the electrodes of a burster. This electromagnetic signal modifies the electrical resistance of a granular metallic packing that is connected to a current source. The resistance (Fig. 2) is recorded by a computer as a function of time (not represented here).

be understood along the lines of nucleation and growth processes, with *nondiffusional* character [19].

Sparks are then produced at 2 Hz frequency for the next 20 sec. The normalized resistance quickly decreases towards a saturating value around 0.82. The continuous behavior is unexpected since the transition was believed to be sharp [1]. The Rhumkorff coil is then not fed anymore and sparks stop.

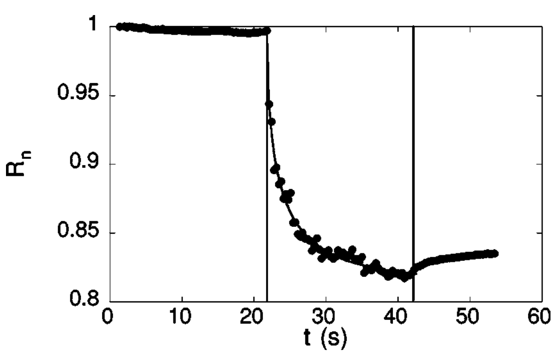

FIG. 2. Evolution of the normalized resistance R_n vs time. The vertical lines indicate the start and the end of the sparks production. The curve is a fit using Eq. (1).

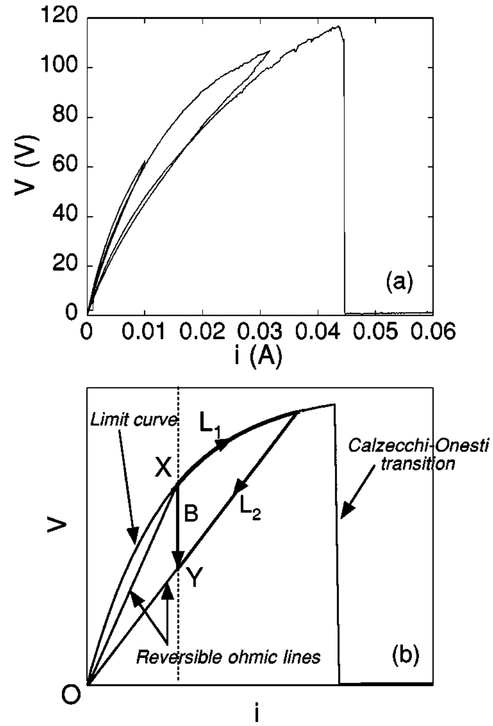

FIG. 3. (a) i-V curves obtained for our packing. The sequence of injected current is described in the text. (b) Sketch of the trajectory in the i-V diagram during sparks (arrow B). The arrows L indicate the path to follow to reach the same point by varying the current intensity.

A relaxation of the resistance value is observed: $R_n = R/R_0$ which slowly increases.

The curve in Fig. 2 represents the fit to the experimental data during sparks production. A stretched exponential has been found for the middle region

$$R_n = (1-f) + f\exp(-at^{\alpha}), \tag{1}$$

where f is the decrease amplitude and a is the decay rate. The amplitude f is found to be about 20% and a is typically equal to 1. The exponent α is similar to the Avrami exponent; its value indicates the growing mechanism of the phenomenon versus time [18]. The value of α is found to be 0.5 ± 0.1 (over about 15 measurements).

In order to understand the meaning of the decay, let us first deduce the physical origin of such a decrease. For this purpose, the i-V diagram has to be recorded.

In Fig. 3(a), the measured i-V curve of the lead packing is shown for a sequence of continuously increasing and decreasing injected currents, i.e., several cycles to different maximum currents. This kind of cycle has been also used in order to point out intrinsic memory effect behaviors [17]. The sharp Onesti-Calzecchi transition occurs near 45 mA. For lower currents, a hysteretic behavior is found below a limit curve. After the maximum current of the cycle is reached, if the current decreased and increased back up to this maximum current, the value of the resistance nearly follows Ohm's law. Thereafter, the voltage follows the limit curve. Figure 3(b) is a sketch of Fig. 3(a) and indicates different zones.

Therefore, the i-V trajectory of the representative point of the system during Branly's effect is the following. When the current is injected, the voltage is given by the i-V diagram at point X. During the electromagnetic perturbations, the representative point of the packing moves to point Y along the path indicated by the arrow B. The trajectory is a vertical line in the i-V diagram since the current is kept quasiconstant during the experiment, with small undetected fluctuations through the numerical integration of the Keithley K2400. Transient currents might exist but cannot be observed with our equipment.

Notice that another way to move from X to Y can be achieved by tuning the electrical current i (trajectories L_1 and L_2) [17]. First, the current is increased and the representative point follows the limit curve (trajectory L_1). During this route, the beads knit together due to a strong local increase in temperature inducing, through an irreversible process, a decrease in the differential electrical resistance of the packing. Those new electrical paths shunt the remaining of the heap. When the current is decreased, the electrical resistance of the heap then follows Ohm's law (trajectory L_2). To sum up the L trajectory, the resistance of the system decreases by passing from X to Y as indicated by the difference of the slopes of the OX and OY lines. This is due to the creation of new electrical paths. This is an irreversible process since the representative point of the system in the i-V plane obviously describes a loop. Physically, this behavior can be attributed to microsoldering of the grains by a destruction of the oxide layer due to the local temperature increase [20]. We stress that these solderings occur when the representative point of the system is located on the limit curve.

The fact that the L trajectories allow us to obtain the same situation as the B trajectory shows that the Branly effect introduces irreversible process such as microsoldering of the beads. Moreover, in both (L and B) cases, the system can be reset to its initial resistance by applying a small mechanical tap, that shows the similarity between the states obtained using the L and the B trajectories.

The generalized $\alpha = 1/2$ Avrami exponent of Eq. (1) may be interpreted as follows. Before the spark production, the conductivity of the heap is assumed to be a well-connected cluster of beads, i.e., the ensemble of all the electrical paths. As soon as electromagnetic waves hit the system, microsolderings occur between the beads. This allows the cluster to grow. Since the resistance is an image of the inverse of the size of the electrical path network [8], R_n decreases following Eq. (1). The exponent $\alpha = 1/2$ identifies the mechanism as being similar to a pure diffusion process similar to that in crystal growth [21]. A similitude may be found indeed between crystal growth and the growth of the conducting cluster in this metallic bead heap. Microsolderings occur when the voltage between two grains reaches a critical threshold. Because of the microsoldering, the voltage drops and the electrical stress propagates throughout the heap inducing the growth of the conducting cluster.

This phenomenon is markedly different from that in fuse networks [9,22–25]: for these systems, during breakdown, the junction suffers irreversible damage of a form that supports high conductance between electrodes. On the contrary, in granular materials, contacts are enhanced or created by external perturbation. Bead contacts favor local puncture damage due to Joule heating with subsequent local melting and grain soldering. This leads to an evolving network of better conducting channels.

In summary, contrary to Calzecchi-Onesti transition, which is very sharp and discontinuous [15], we have shown that the Branly effect on resistance decay of a metallic granular packing due to an incident electromagnetic wave is continuous and behaves like a stretched exponential law in time with an exponent 1/2. The physical origin can be explained by the soldering of grains and by the creation of new electrical paths. This process follows the law of diffusion.

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