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# Electrical conductivity in granular media and Branly's coherer: A simple experiment

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We show how a simple laboratory experiment can be used to exhibit certain electrical transport properties of metallic granular media. At a low critical imposed voltage, a transition from an insulating to a conductive state is observed. This transition comes from an electro-thermal coupling in the vicinity of the microcontacts between grains where microwelding occurs. The apparatus used allows us to obtain an implicit determination of the microcontact temperature, which is analogous to a resistive thermometer. The measurements also illustrate a 100-year-old problem, the explanation of Branly's coherer effect - a radio wave detector made of metal fillings used for the first wireless radio transmission.

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## I. INTRODUCTION

The coherer effect or "Branly effect" is an electrical conduction instability which appears in a slightly oxidized metallic powder under a constraint [1]. The initially high powder resistance irreversibly falls by several orders of magnitude as soon as an electromagnetic wave is produced in its vicinity. Discovered in 1890 by E. Branly, this action *at distance* relates to other phenomena, notably when an electrical source is directly connected with the powder such as; i) a transition from an insulating state to a conductive state observed as an electrical source exceeds a threshold voltage, ii) temporal fluctuations and slow relaxations of resistance that occur under certain conditions [2].

Although used for the first wireless radio transmission near 1900, these electrical transport phenomena in metallic granular media are still not well understood. Several possible processes at the contact scale have been invoked without any clear demonstrations; electrical breakdown of the oxide layers on grains [3], modified tunnel effect through the metal - oxide  $\sim$  semiconductor - metal junction [4], attraction of grains by molecular or electrostatic forces [5], local welding of microcontacts by a Joule effect [6, 7] also labelled as "A-fritting" [4]; each being combined with a global process of percolation [3, 5, 6].

Here we set out to understand this transition of conduction by means of a model experiment with a chain of metallic beads [8] to focus on the local properties (contacts between grains) instead of the collective ones (e.g., typical disorder of a granular). This paper also allows students to explore from an historical starting point, the electrical and thermal properties of non-homogeneous media such as granular media, as well as the influence of electromagnetic waves (see a note recently published in *Physics Education* [9]). After a brief review of the history

of the coherer effect in Sec. II, we introduce in Sec. III A the experimental setup used here which can be easily built in a standard physics laboratory. Then, we present the experimental results in Sec. III B, followed by a qualitative and quantitative interpretation of the conduction transition mechanism in Sec. III C and III D respectively. Finally, our conclusions are given in Sec. IV.

## II. A BRIEF HISTORICAL REVIEW

In 1887, a short time after the publication of Maxwell's theory of electromagnetism, experiments performed by H. Hertz clearly demonstrated free space generation and propagation of electromagnetic waves. He noticed that sparks (high frequency electromagnetic waves of the order of 100 MHz) could induce arcing across a wire loop containing a small gap, a few meters away.

This discovery was anticipated by many people; P. S. Munk af Rosenschöld observed in 1835 the permanent increase in the electrical conductivity of a mixture of metal filings resulting from the passage of a discharge current of a Leyden jar (the former capacitor). In 1879, D. E. Hughes observed a similar phenomenon for a loose contact formed of a carbon rod resting in grooves in two carbon blocks, or with a tube filled with metallic granules (which was called microphone since it was first designed to detect acoustic waves) placed in series with a voltaic cell. Hughes appears to have discovered the important fact that such a tube was sensitive to electric sparks at a distance as indicated by its sudden change in conductivity. At the time, the Royal Society was not convinced, and his results were published some 20 years later [10], a long time after the discovery of the hertzian waves. In 1884, T. Calzecchi-Onesti performed experiments on the behaviour of metallic powders under the action of various electromotive forces, and observed a considerable increase of the powder conductivity by successively opening and closing the circuit. Upon shaking the tube containing the powder, the earlier high value of the powder resistance was restored.

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The action of nearby electromagnetic waves on metallic powders was observed and extensively studied by E. Branly in 1890 [1]. When metallic filings are loosely arranged between 2 electrodes in a glass or ebonite tube, it has a very high initial resistance of many megohms due to an oxide layer likely present on the particle surfaces. When an electric spark was made at a distance, the resistance was suddenly reduced to several ohms. This conductive state remained until the tube was tapped restoring the resistance to its earlier high value. Since the electron was not known at this time (discovered in 1895-96), Branly called his device a “radio conductor” to recall that “the powder conductivity was settle under the influence of the electric radiations from the spark”; the meaning of the prefix “radio” being at this time “radiant” or “radiation”. He performed other experiments with various powders, lightly or tighly compressed powders, and also found the same effect occurred in the case of two metallic beads in contact, or in the case of two slightly oxidized steel or copper wires laid across each other with light pressure [11]. This loose or “imperfect” contact was found to be extremely sensitive to a distant electric spark.

This discovery caused a considerable stir as soon as O. Lodge in 1894 repeated and extended Hertz’s experiments by changing the wire loop by Branly’s tube, a much more sensitise detector. Lodge improved it into a reliable, reproducible detector, and automated it by associating a mechanical shock to the tube. Lodge called it a “coherer” from the Latin *cohaerere* synonymous with stick together. He said that the fillings were “cohered” under the action of the electromagnetic wave and needed to be “decohered” by a shock. Later, E. Branly and Lodge focused their fundamental research on mechanisms of powder conductivity, and not on practical applications such as wireless communications. However, based on the coherer, and using it as a wave detector, the first wireless telegraphy communications were transmitted in 1895 by G. Marconi, and independently by A. S. Popov. Popov also used the coherer as a lightning recorder to detect the atmospheric electrical discharges at a distance.

Under the action of the voltages that are induce by electromagnetic waves, T. Tommasina advanced in 1898 the opinion that the metallic grains were welded together. According to Lodge, as a consequence of the electric field of the wave, the filings became dipoles and attracted each other by electrostatic forces, inducing small motions of grains to stick together, forming thus conductive chains [12]. A shock should be enough to break these fragile chains, and to restore the resistance to its original value. Branly never believed this hypothesis, and to demonstrate that motion was not necessary, he immersed the particles in paraffin, wax, resin, or used a column of six hard steel balls or disks, few centimeters in diameter. Since the coherer effect persisted, he thought that the properties of the dielectric between the grains played an important role, and was responsible for the “radio-conduction”. In 1900, Guthe et al. performed experi-

ments along the same lines with 2 balls in contact [13]. However, in 1906, the invention by Lee de Forest of the triode, the first vacuum tube known as audion, progressively supplanted the coherer as a receiver, and Branly’s effect sank into oblivion without being fully elucidated.

Several decades later in the beginning of the 1960s, a group in Lille (France) became interested again in this old problem. They suggested a molecular relaxation arising from the removal of applied electrostatic field that held the particles in contact [5]. In the 1970s, numerous works dealt with the conductivity of granular materials for batteries, but not with the transition of electrical conduction [14]. In 1975, a group in Grenoble (France) suggested a mechanism of electrical breakdown of the oxide layer on the grain surfaces, and were interested in the associated  $1/f$  resistance noise [3]. In 1997, the transition of conduction was observed by Vandembroucq et al. by direct visualization (with an infrared camera) of the conduction paths when a very high voltage (more than 500 V) is applied to a monolayer of aluminium beads [6]. More recently, a group in Liège (Belgium) has revisited the action at a distance of sparks [7]. For additionnal information about the coherer history, the reader is invited to see Refs. [15, 16].

### III. ELECTRICAL CONDUCTIVITY OF A CHAIN OF METALLIC BEADS

Understanding the transition of electrical conduction through granular materials is a complicated many body problem which depends on a large number of parameters: global properties concerning the grain assembly (i.e. statistical distribution of shape, size and pressure) and local properties at the contact scale of two grains (i.e. degree of oxidization, surface state, roughness). Among the phenomena proposed to explain the coherer effect, it is easy to show that some have only a secondary contribution. For instance, since coherer effect has been observed by Branly with a single contact between 2 grains [11], percolation can not be the predominant mechanism. Moreover, when two beads in contact are connected in series with a battery, a coherer effect is observed at high enough imposed voltage [13], in a similar way as the action *at distance* of a spark or an electromagnetic wave. In this paper, we deliberately reduce the number of parameters, without loss of generality, by focusing on the electrical transport within a chain of metallic beads directly connected to an electrical source.

#### A. Experimental setup

The experimental setup is sketched in Fig. 1. It consists of a chain of 50 identical AISI 420 stainless-steel beads, each 8 mm in diameter, and  $0.1\mu\text{m}$  in roughness. The beads are surrounded by an insulating framework of polyvinylchloride (PVC). A static force  $F \leq 500$  N is ap-

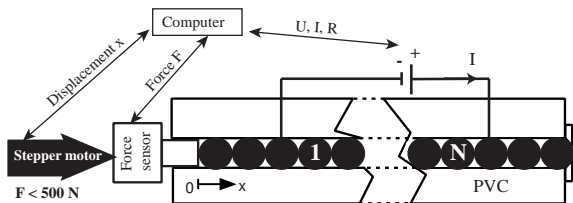


FIG. 1: Schematics of experimental setup.

plied to the chain of beads by means of a stepper motor, and is measured with a static force sensor. The number of motor steps is measured with a counter to determine the total deformation of the chain,  $x$ , necessary to reach this specific force. During a typical experiment, we chose to supply the current ( $10^{-6} \leq I \leq 1$  A) and to simultaneously measure the voltage  $U$ , and thus the resistance  $R = U/I$ . Similar results have been found when repeating experiments with imposing the voltage and measuring  $I$  and  $R$ . The bead number  $N$  between both electrodes is varied from 1 to 41 by moving the “electrode beads” within the chain. Note that the lowest resistance of the whole chain (few  $\Omega$ ) is always found to be much higher than the electrode or the stainless steel bulk material.

## B. Experimental results

The mechanical contact of the bead chain is measured in very good agreement with the non linear Hertz law (given by linear elasticity), that is  $F \propto x^{3/2}$ . This leads to an estimation of the typical ranges of deformation between two beads from 2 to 20  $\mu\text{m}$ , and of the apparent contact radius,  $A$ , from 40 to 200  $\mu\text{m}$ , when  $F$  ranges from 10 to 500 N.

The electrical behavior is much more amazing! Since no particular precautions has been taken for the beads, the bead-bead contact is not metallic and an insulating film (oxide and/or contaminant), few nanometer thick, is probably present. When the applied current to the chain increases, we observe a transition from an insulating to a conductive state as shown in Fig. 2. At low applied current and fixed force, the voltage-current  $U-I$  characteristic is reversible and ohmic (arrow 1) of high and constant resistance,  $R_0$ . This resistance at low current ( $R_0 \simeq 10^4 - 10^7 \Omega$ ) depends in a complex way on the applied force and on the contaminant film properties (resistivity, thickness) at the contact location. The values of  $R_0$  are determined by the slopes of the  $U-I$  trajectories at low increasing current. As  $I$  is increased enough, the resistance strongly decreases and reaches a constant bias  $U_0$  (arrow 2). As soon as this saturation voltage  $U_0$  is reached, the resistance is irreversible upon decreasing the current (arrow 3): this back trajectory is reversible and the resistance reached at low decreasing current,  $R_{0b}$  (of the order of 1 - 10  $\Omega$ ), depends on the maximum current previously applied,  $I_{max}$ , such as a memory effect. The non linear reversible back trajectory is also symmetrical,

when the current applied to the chain is reversed (arrows 4 and 5). When repeating this symmetrical loop up to different values of  $I_{max}$  (solid and dashed arrows), and for various applied forces  $F$ , one can show that the back trajectories depend only on  $I_{max}$ , and follow the same back trajectory when expressed in the variables  $U$  as a function of  $R_{0b}I$  (see inset of Fig. 2). The values of  $R_{0b}$  are determined by the slopes of the  $U-I$  back trajectories at low decreasing current (see Fig. 2).

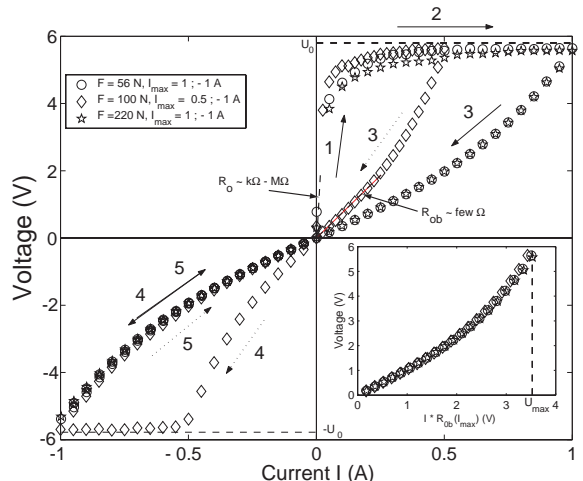


FIG. 2: Symmetrical characteristics  $U-I$  of a chain of  $N = 13$  beads, for various current cycles in the range  $-I_{max} \leq I \leq +I_{max}$ , and for various forces  $F$ . Inset shows the reversible back trajectories rescaled by  $R_{0b}$ .  $U_{max} \equiv R_{0b} * I_{max} \simeq 3.5$  V. (See text for details).

This decrease of the resistance by several orders of magnitude (from  $R_0$  to  $R_{0b}$ ) has similar properties as that of the coherer effect with powders [1] or with a single contact [11, 13]. Note that after each cycle in the current, the applied force is reduced to zero, and we roll the beads along the chain axis to have new and fresh contact between beads for the next cycle. With this methodology, the resistance drop (the coherer effect or Branly effect) and the saturation voltage are always observed and are very reproducible.

This saturation voltage  $U_0$  is independent of the applied force, but depends on the number of beads,  $N$ , between the electrodes. When varying  $N$  from 1 to 41, the saturation voltage per contact  $U_{0/c} \equiv U_0/(N + 1)$  is found constant and on the order of 0.4 V per contact. However, this saturation voltage slightly depends on the bead materials ( $U_{0/c} \simeq 0.4$  V for stainless steel beads;  $U_{0/c} \simeq 0.2$  V for bronze beads; 0.3 V for brass beads), but is of the same order of magnitude [8].

## C. Qualitative interpretation

Assume a mechanical contact between two metallic spheres covered by a thin contaminant film ( $\sim$  few nm).

The interface generally consists of a dilute set of microcontacts due to the roughness of the bead surface at a specific scale [4]. The mean radius,  $a$ , of these microcontacts is of the order of magnitude of the bead roughness  $\sim 0.1 \mu\text{m}$ , which is much smaller than the apparent Hertz contact radius  $A \sim 100 \mu\text{m}$ . Figure 3 schematically shows the building of the electrical contact by transformation of this poorly conductive film. At low applied current, the high value of the contact resistance ( $\text{k}\Omega - \text{M}\Omega$ ) probably comes from a complex conduction path found by the electrons through the film within a very small size ( $\ll 0.1 \mu\text{m}$ ) of each microcontact (see lightly grey zones in Fig. 3). The electron flow then damages the film, and leads to a “conductive channel”: the crowding of the current lines within these microcontacts generates a thermal gradient in their vicinity, if significant Joule heat is produced. The mean radius of microcontacts then strongly increases by several orders of magnitude (e.g., from  $a_i \ll 0.1 \mu\text{m}$  to  $a_f \sim 10 \mu\text{m}$ ), and thus enhances their conduction (see Fig. 3). This corresponds to a nonlinear behaviour (arrow 1 until 2 in Fig. 2). At high enough current, this electro-thermal process can reach the local welding of the microcontacts (arrow 2 in Fig. 2); the film is thus “piercing” in a few areas where purely metallic contacts (few  $\Omega$ ) are created (see black zones in Fig. 3). [Note that the current-conductive channels (bridges) are rather a mixture of metal with the film material rather than a pure metal. It is probable that the coherer action results in only one bridge – the contact resistance is lowered so much that puncturing at other points is prevented]. The  $U-I$  characteristic is then reversible when decreasing and then increasing  $I$  (arrow 3 in Fig. 2). The reason is that the microcontacts have been welded, and therefore their final size  $a_f$  does not vary any more with  $I < I_{max}$ . The  $U-I$  back trajectory then depends only on the temperature reached in the metallic bridge through its parameters (electrical and thermal conductivities), and no longer on its size as previously.

#### D. Quantitative interpretation

Let us now check quantitatively the interpretation in Sect. III C. Assume a microcontact between two clean metallic conductors (thermally insulated at uniform temperature  $T_0$ , with no contaminant or tarnish film on their surfaces). Such a clean microcontact is generally called a “spot”. If an electrical current flowing through this spot is enough to produce Joule heating (assumed totally drained off by thermal conduction in the conductors), then a steady-state temperature distribution is quickly reached ( $\sim \mu\text{s}$ ) in the contact vicinity. The maximum temperature reached,  $T_m$ , is located at the contact, and is linked to the applied voltage,  $U$ , by [8]

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

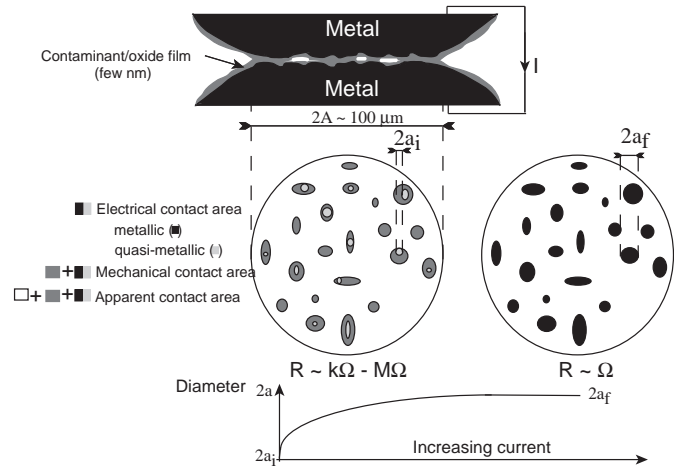


FIG. 3: Schematic view of the electrical contact building through microcontacts by transformation of the poorly conductive contaminant/oxide film. At low current  $I$ , the electrical contact is mostly driven by a complex conduction mechanism through this film via conductive channels (of areas increasing with  $I$ ). At high enough  $I$ , an electro-thermal coupling generates a welding of the microcontacts leading to efficient conductive metallic bridges (of constant areas).

where  $L = \pi^2 k^2 / (3e^2) = 2.45 \times 10^{-8} \text{ V}^2/\text{K}^2$  is the Lorentz constant,  $k$  the Boltzmann’s constant, and  $e$  the electron charge. Stemming from the thermal equilibrium and Ohm’s law, this relationship shows that the maximum temperature  $T_m$  reached at the contact is independent of the contact geometry and of the materials in contact! This is a consequence that both the electrical resistivity,  $\rho(T)$ , and thermal conductivity,  $\lambda(T)$ , are related to the conduction electrons, leading thus that their respective dependence on the temperature,  $T$ , follows the Wiedemann-Franz law [4]

$$\lambda\rho = LT, \quad (2)$$

A voltage near  $0.4 \text{ V}$  across a contact thus leads from Eq. (1) to a contact temperature near  $1050^\circ\text{C}$  for a bulk temperature  $T_0 = 20^\circ\text{C}$ . This means that  $U \simeq 0.3 - 0.4 \text{ V}$  leads to contact temperatures that exceed the softening or/and the melting point of most conductive materials. Efficient conductive metallic bridges are therefore created by microwelding. Beyond the quantitative agreement with the experimental saturation voltage  $U_{0/c}$  (see Sect. III B and Fig. 2), Eq. (1) also explains why  $U_{0/c}$  is the relevant parameter in the experiments in Sect. III B, and not the magnitude of the current. In addition, when  $U$  approaches  $U_{0/c}$  on Fig. 2, the local heating of microcontacts is enough, from Eq. (1), to soft them. Then, their contact areas increase thus leading to a decrease of local resistances, and thus stabilizing the voltage, the contact temperatures and the contact areas, since the current is fixed. The phenomenon is therefore self-regulated in voltage and temperature.

Our quantitative model only describes the electrical

behavior of a welded contact (i.e. as soon as the saturation voltage is reached, saying at  $I = I_{max}$ ). It describes the reversible  $U-I$  back trajectory (when this contact is cooled by decreasing the current, then eventually reheated by increasing  $I$ ). Here, the contact area is assumed constant since it has been welded, and  $I < I_{max}$ . Let us recall the principles of the derivation for the analytical expression of the nonlinear  $U-I$  back trajectory [8].

The thermal equilibrium means that the heat flux,  $\lambda(T)\mathbf{grad} T$ , across the isothermal surfaces  $T$  is due to the electrical power,  $\varphi(T)I$ , where  $\varphi(T)$  is the potential between the two isothermals on both sides of the contact. Let us introduce the ‘‘cold’’ contact resistance  $R_{0b}$  presented to a current low enough not to cause any appreciable rise in the temperature at the contact [the conductor bulk being at the room temperature  $T_0$ , with an electrical resistivity  $\rho_0 \equiv \rho(T_0)$ ]. The derivation of  $R_{0b}$  involves the same equipotential surfaces during a virtual change between the ‘‘cold’’ state (denoted with a star)  $\varphi^*$ , and the real ‘‘hot’’ state  $\varphi(T)$ . This means that the current density is invariant along an equipotential (which is also an isothermal)  $\mathbf{grad}(\varphi^*)/\rho_0 = \mathbf{grad}(\varphi)/\rho(T)$ . With the thermal equilibrium, this gives

$$\frac{d\varphi^*}{\rho_0} = \frac{d\varphi}{\rho(T)} = \frac{\lambda(T)}{\varphi(T)} dT. \quad (3)$$

Using the Ohm’s law and integrating Eq. (3) between the isothermal surfaces  $T_0$  and  $T_m$  leads to [17]

$$\frac{IR_{0b}}{\rho_0} = 4 \int_{T_0}^{T_m} \frac{\lambda(T)}{\varphi(T)} dT. \quad (4)$$

The temperature dependance for the thermal  $\lambda(T)$  and electrical  $\rho(T)$  conductivities for the material in contact are given by Eq. (2) and

$$\rho(T) \equiv \rho_0 [1 + \alpha(T - T_0)], \quad (5)$$

$\alpha$  being the temperature coefficient of electrical resistivity. Equations (2) and (5) then allow to explicit both  $\lambda(T)$  and  $\varphi(T)$  in Eq. (4). The current  $IR_{0b}$  through the contacts only depends on the temperature  $T_m$  (i.e. on  $U$ )

$$T_m = \sqrt{T_0^2 + \frac{U^2}{4L(N+1)^2}}, \quad (6)$$

and finally reads [8]

$$IR_{0b} = 2(N+1) \frac{\sqrt{L}}{\alpha} \int_0^{\theta_0} \frac{\cos \theta}{[1 + (\alpha T_0)^{-1}] \cos \theta_0 + \cos \theta} d\theta, \quad (7)$$

where  $\theta_0 \equiv \arccos(T_0/T_m)$ . Note that only  $R_{0b}$  depends on the contact geometry, and its value is easily reached experimentally (see Sect. III B)

Here again, for pure metals ( $\alpha^{-1} = T_0$ ), Eq. (7) does not depend explicitly on the geometry of the contact,

or on the metal used for the contact! However, in case of alloys in contact, Eq. (7) depends on  $\alpha$  the temperature coefficient of electrical resistivity of the alloy. This additional parameter is related to the defects in the material. The normalized  $U-I$  back trajectory (i.e.,  $U$  as a function of  $IR_{0b}$  of the inset of Fig. 2) are compared in Fig. 4 with the theoretical solutions [Eq. (7)] for the pure metals, and for a stainless steel alloy. A very good agreement is shown between the experimental results and the electro-thermal theory, notably for the alloy case. Qualitatively, the alloy solution has a better curvature than the pure metal one. The agreement is quantitatively excellent (see Fig. 4) when choosing  $\alpha^{-1} = 4T_0$  instead of  $3.46T_0$  (the  $\alpha^{-1}$  value for AISI 304 stainless steel), since the  $\alpha$  value for the bead material (AISI 420 stainless steel) is unknown, but should be close. During this experimental back trajectory, the equilibrium temperature,  $T_m$ , on a microcontact is also deduced from Eq. (6) with no adjustable parameter (see inset of Fig. 4). Therefore, when the saturation voltage is reached ( $U_0 = 5.8$  V),  $T_m$  is close to 1050°C which is enough to soften or to melt the microcontacts between the  $N = 13$  beads of the chain. Our implicit measurement of the temperature is equivalent to the use of a resistive thermometer.

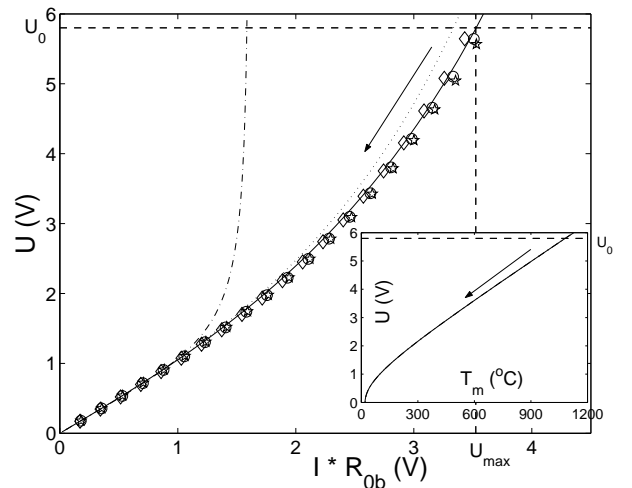


FIG. 4: Comparison between experimental  $U-I$  back trajectories of Fig. 2 (symbols), and theoretical curves from Eq. (7) for the case of an alloy with stainless steel properties [ $\alpha^{-1} = 4T_0$  (—) or  $3.46T_0$  (---)], and for a pure metal [ $\alpha^{-1} = T_0$  (-.-)]. Inset shows the theoretical maximum temperature,  $T_m$  [Eq. (6)], reached in one contact when the chain of  $N = 13$  stainless steel beads is submitted to a voltage  $U$ .

#### IV. CONCLUSIONS

We have reported the observation of the electrical transport within a chain of oxidized metallic beads under applied static force. A transition from an insulating to a conductive state is observed as the applied current

is increased. The  $U$ - $I$  characteristics are nonlinear, hysteretic, and saturate to a low voltage per contact ( $\simeq 0.4$  V). Electrical phenomena in granular materials related to this conduction transition such as the ‘‘Branly effect’’ were previously interpreted in many different ways but without a clear demonstration. Here, with this simple teaching experiment, we have shown that this transition, triggered by the saturation voltage, comes from an electro-thermal coupling in the vicinity of the microcontacts between each bead. The current flowing through these spots generates local heating which leads to an increase of their contact areas, and thus enhances their conduction. This current-induced temperature rise (up to  $1050^{\circ}\text{C}$ ) results in the microwelding of contacts (even for so low voltage as  $0.4$  V). Based on this self-regulated temperature mechanism, an analytical expression for the nonlinear  $U$ - $I$  back trajectory is derived, and is found in very good agreement with the data. It also allows the de-

termination of the microcontact temperature all through this reverse trajectory, with no adjustable parameter, as an analog of a measurement with a resistive thermometer. Moreover, one could attempt to assert that this process may be easily witnessed through direct visualization (microscope, infrared camera). But for that, it is clear that very powerful electrical source must be applied, far in excess of that necessary to produce true coherer phenomena (see for instance Ref. [6]).

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