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► **To cite this version:**

Valentin Mazières, Romain Pascaud, Laurent Liard, Simon Dap, Richard Clergereaux, et al.. Time reversal plasmas as a versatile space-time patterning deposition method. 2021 IEEE 16th Nanotechnology Materials and Devices Conference (NMDC), Dec 2021, Vancouver, Canada. pp.1-4, 10.1109/NMDC50713.2021.9677515 . hal-03765762

HAL Id: hal-03765762

<https://hal.science/hal-03765762>

Submitted on 31 Aug 2022

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Time reversal plasmas as a versatile space-time patterning deposition method

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Abstract—The aim of this communication is to discuss the possibility of using the innovative plasma source that we have developed recently, namely the “Space-Time Plasma Steering Source”, as a versatile space-time deposition method. More precisely, we focus on the space-time control capabilities of time reversal plasmas over large and complex 3D surfaces. Simultaneous ignition of several plasmas by time reversal is also experimentally demonstrated. These unprecedented control capabilities of plasmas could lead to the development of an original and versatile method allowing the patterning of complex 2D or 3D structures on large areas.

Keywords—Time Reversal, Space time plasmas, Patterning deposition method

I. INTRODUCTION

In the context of thin film nanocomposite deposition, “fabricating functional structures [...] with arbitrary patterns is central to the development of many technologies” [1]. Thus, one of the challenges faced by these technologies is development of methods allowing an accurate control of the deposition pattern (shape, thickness...) over large areas. Various methods have been considered to attain this goal. They can be divided into two categories: “Mask-requiring methods” - lithography, molding, embossing and microcontact printing [1] - which require fabrication of a new mask for each different design of patterns, thus making them not versatile and “Maskless methods” - dip-pen nanolithography, inkjet printing, laser printing and plasma spray [2] - which consist in local deposition, providing flexibility in the control of the deposition pattern. In addition to the spatial control of the deposition pattern, a control of the process duration may also be of importance, for example for obtaining a better homogeneity of particle distribution in nanocomposites [3].

The aim of this communication is to discuss the possibility of using an innovative plasma source that we have developed recently [4,5,6] as an original versatile space-time patterning deposition method. This source, named “Space-Time Plasma Steering Source”, has been introduced to address the challenge of controlling plasmas in large cavities (overmoded). Contrary to conventional microwave plasma technologies, for which the plasma occupies the entire volume of the plasma vessel, the idea is to rather ignite one or several localized plasmas whose locations in the vessel can be dynamically controlled. To that end, Time Reversal (TR) was used as it allows focusing of the electromagnetic waves [7]. It consists of two steps. First the impulse response

between the desired location (at which we want to ignite a plasma) and an antenna is recorded. Then, this signal is time reversed and transmitted to the antenna, resulting in the back propagation of the waves toward the desired location. A spatio-temporal focusing then occurs at this location, of duration equal to that of the initial impulse and of dimension equal to half the wavelength. If the focusing electric field is high enough, a breakdown occurs at the focusing instant and location. Based on this principle, the locations of plasmas can be dynamically controlled by the shape of the transmitted waveform to the cavity through one emitting antenna [4,5,6].

In our pioneer paper demonstrating the experimental control of plasmas by TR [4], several applications for this innovative source have been mentioned, with among them, “surface treatments, etching, or thin film deposition” [4]. In the present paper, we focus on its possible application for thin film nanocomposite deposition. In section II, the space time capabilities of the Space-Time Plasma Steering Source based on time reversal are presented. Its principle is explained and the first experimental demonstration of this source [4] is briefly presented. Then, its ability to control simultaneous plasmas is experimentally demonstrated. In section III, a discussion is proposed on the possibility to use this original source as a versatile space time pattern deposition method.

II. SPACE-TIME PLASMA STEERING SOURCE BASED ON TIME REVERSAL

A. Principle

The principle of the innovative source we introduced recently is briefly explained in this subsection. It has been introduced to meet the challenge of controlling plasmas in overmoded cavities. Indeed, commonly used microwave plasma sources in cavities are based on the control of the spatial distribution of the field, and thus the plasma, by exciting a stationary resonant mode of the cavity [8]. However, in overmoded cavities (large cavities), it becomes difficult, if not impossible, to control with this principle the spatial distribution of the field [9], and thus of the plasma. The idea of the present source is to dynamically control the plasmas location by changing the waveform of the transmitted signal $s(t)$ to an overmoded cavity. It is then the transient behavior of the waves inside the cavity that dynamically controls the plasmas location, hence the name of “Space-Time Plasma Steering Source”.

The key element of this source is the shape of the waveform of the transmitted signal $s(t)$. Several techniques

can be envisioned for determining this waveform, allowing the desired space-time behavior of plasmas in the cavity. This technique must allow a concentration of electromagnetic energy at the desired instant and location (at which we want the plasma) and a lower level of energy elsewhere in the cavity. The most obvious technique that meet these criteria is “Time Reversal”, as it allows the focusing of waves inside overmoded cavities. We have shown that this technique allows an accurate control of plasmas on initiators in an overmoded cavity [4,6]. Note that we have also shown that a more elaborate technique allows to enhance the spatial control of plasmas [5], but for simplicity we focus in this paper on “Time Reversal” plasmas.

B. Experimental demonstration of Time Reversal plasmas

The principle of TR was first demonstrated by M. Fink, C. Prada et al. in the 1990's [10]. In practice, two steps are required. The first one consists in sending a short pulse from a source, and recording the signal that has propagated in a more or less complex medium using transducers located around this source. Then, the second step consists in re-emitting by these transducers the information previously recorded, but time reversed. The waves thus back propagate toward the original location of the pulse. In the end, this method allows to focus efficiently microwave energy at given instant and location. The use of a reverberant and ergodic cavity has been shown to allow to reduce considerably the number of transducers needed, down to a single transducer [11]. The principle of the Space-Time Plasma Steering Source based on TR consists in obtaining a focusing field high enough so that a breakdown occurs at the desired instant and location. Following this idea, we have shown that an accurate space time control of plasma is possible on initiators [4]. The results presented in the following of this subsection are a brief synthesis these results.

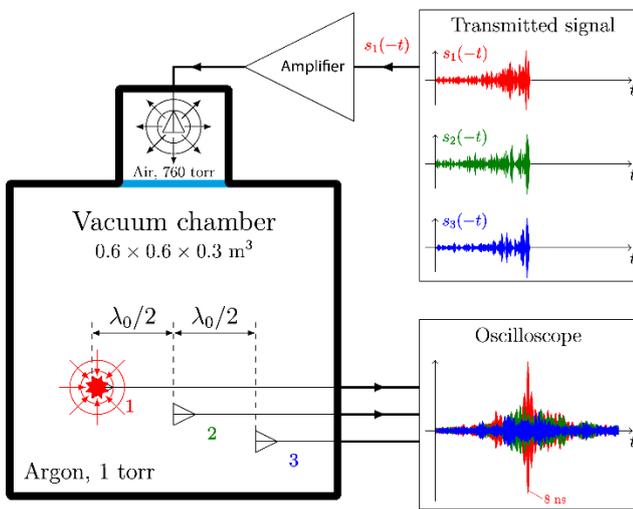


Figure 1: Schematic of a plasma ignition experiment on initiator 1

The experimental setup in which we have shown that TR efficiently allows a space time control of plasmas in an overmoded cavity [4,6] is represented on Figure 1. It consists in a reverberant cavity of $0.6 \times 0.6 \times 0.3 \text{ m}^3$ - that is to say $5\lambda_0 \times 5\lambda_0 \times 2.5\lambda_0$ at our working microwave carrier frequency of about $f_0 = 2.45 \text{ GHz}$ - in which the microwave TR experiments are performed. This cavity includes four antennas: one located in an appendix, at atmospheric pressure

(760 torr) in air, and three others located in the main cavity, at 1 torr in argon. The latter are $\lambda_0/2$ apart from each other, which is the spatial resolution of the TR process (meaning the spatial dimension of the focusing) [7]. These three antennas (which are metallic monopoles), act as emitting antennas during the first phase of the TR process. During the second step, they act as receiving antenna (allowing a measure of the field evolution at their locations) as well as initiators for plasma breakdown, as they produce a local enhancement of the electric field. Hence, they will be referred to as “initiator” in the following of this paper.

The first step of TR consists in recording the impulse responses (from a 8 ns impulse modulated at $f_0 = 2.45 \text{ GHz}$) between initiators 1, 2, or 3 (then acting as emitting antennas) and the high pressure antenna (then acting as receiving antenna). These responses must then be time-reversed ($s_1(-t)$, $s_2(-t)$, and $s_3(-t)$, see Figure 1) and transmitted through the high-pressure transmitting antenna to achieve a focusing (of 8 ns) on initiators 1, 2, or 3. For example, a focusing experiment by TR on initiator 1 is shown in Figure 1.

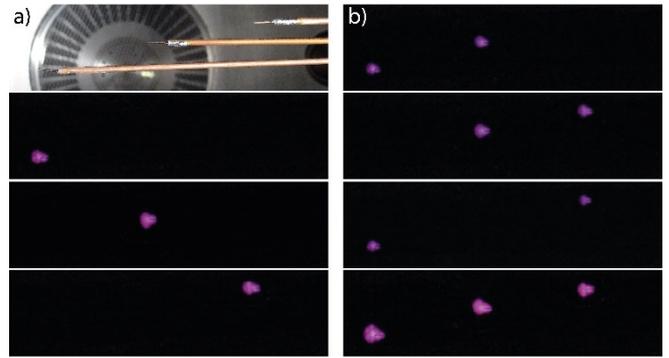


Figure 2: a) Photo of the three initiators 1, 2 and 3 of Figure 1 (top photo) and photos of the plasmas ignited by TR (bottom photos) [4]. b) Photos of the simultaneous TR plasmas.

For plasma ignition by TR, during the second step, the signals are amplified before being transmitted to the antenna (see Figure 1). The high pressure in the appendix allows to avoid parasitic breakdown at the emitting antenna and allows to inject efficiently the microwave power into the low pressure part. Thus, under optimal breakdown conditions for a frequency of 2.45 GHz (argon at 1 torr), focusing by TR on one of the three initiators allows to ignite a local plasma on the corresponding initiator. The 2 kW amplifier used is a pulsed amplifier. Thus the TR focusing peaks are generated periodically, at a period T_{rep} typically equal to $166 \mu\text{s}$ [4].

Photos of the initiators 1, 2 and 3 and the ignited plasmas by TR are shown in Figure 2-a). A plasma discharge can be observed on initiator 1, 2 and 3 for the emission of the signals $s(t) = s_1(-t)$, $s(t) = s_2(-t)$ and $s(t) = s_3(-t)$ respectively [4]. Note that as discussed in [6], the presence of the initiator has certainly a major influence on the plasmas dimension (centimetric) and shape. This is majorly due to the local enhancement of the electric field they produce.

C. Experimental demonstration of simultaneous Time Reversal plasmas

In order to develop a versatile patterning deposition method with high throughput by TR plasmas, it would be

interesting to be able to control simultaneous plasmas. Space-Time Plasma Steering Source based on TR allows the simultaneous control of several plasmas in the cavity. In this case, several TR focusing peaks are obtained simultaneously at the corresponding locations. Based on the principle of superposition, this is achievable by sending the sum of the corresponding time reversed impulse responses to the transmitting antenna (high-pressure antenna of the appendix). For example, the top picture of Figure 2-b) is obtained by sending the signal $s(t) = s_1(-t) + s_2(-t)$ to the transmitting antenna. The two plasmas are ignited at the desired locations, meaning on initiators 1 and 2. The same method allows the ignition of simultaneous plasmas on initiators 2 and 3, and 1 and 3, as shown in Figure 2-b). Sending the sum of the three time reversed impulse responses $s(t) = s_1(-t) + s_2(-t) + s_3(-t)$ leads to the simultaneous ignition of the three corresponding plasmas, as shown in the bottom picture of Figure 2-b). These results show that the position of the simultaneous plasmas is well controlled by the transmitted waveform. This is the first demonstration of simultaneous ignition of several plasmas by TR.

Thus, adequate shape of the signal $s(t)$ sent by an antenna, acquired during the learning phase, allows a space-time control of one or several plasmas in overmoded cavities. This unique property makes this process a very promising candidate for certain applications requiring precise control of the plasma in time and space. A discussion on its use for deposition patterning is proposed in the following section.

It is important to notice that there is certainly a limit on the number of plasmas that can be controlled by TR. This limit depends on the plasma volume compared to the cavity volume and on the plasma frequency (that depends on the plasma density) compared to the wave frequency. Future work is planned on the determination of the limit number of simultaneous plasmas that can be controlled by TR for a given configuration.

III. DISCUSSION ON THE POSSIBILITY OF USING TIME REVERSAL PLASMA A VERSATILE SPACE TIME PATTERNING METHOD

In this section, we discuss the use of the ‘‘Space-Time Plasma Steering Source’’ based on Time Reversal as a possible versatile space time deposition method.

A. Principle

A deposition method based on TR plasmas, allowing the treatment of one or several substrates with complex 3D shapes, would require two steps.

During the first step, impulse responses between the desired locations and an antenna should be evaluated in locations spaced with $\lambda_0/2$ apart from each other, located all around the 3D complex shapes of the substrates. To that end, several solutions can be envisioned. We could for example think of modelling the cavity to obtain these impulse responses in simulation. This has been done recently for time-reversal experiments in 2D cavities [12], but would be for now difficult to implement for 3D cavities. Another and perhaps more promising solution consists in doing a spatial mapping by the means of non-intrusive probe, as follows. First, a non-intrusive probe could be placed at a desired location. The impulse response between this probe and the

antenna should be recorded. Then, the non-intrusive probe should be placed at the next desired location and the corresponding impulse response recorded, and so on.

The interest of using a non-intrusive probe is the fact that moving the probe in the cavity does not change the behavior of the waves, so that when it is removed during the second step, sending a time-reversed impulse response by the antenna leads to a focusing at the desired location. This method of scanning the cavity in a non-intrusive manner has been proved to successfully work for microwave TR experiments using electro-optical probes (in a cavity of $6 \times 3.5 \times 2.5 \text{ m}^3$) [13].

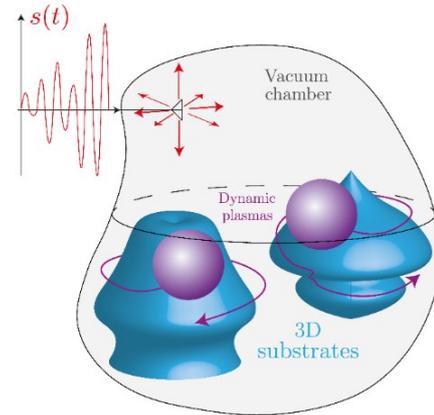


Figure 3: Principle of TR plasmas used as a 3D patterning method

If the focusing field is high enough, breakdown of the gas could then occur at the desired locations without the use of any initiator. If not, pre-ionization of the cavity could be envisioned to reduce the level of this breakdown field [14], pre-ionization that could for example be obtained by an initial TR plasma ignited on an initiator.

During the second step, the transmitted signal $s(t)$ should then be constructed combining the time-reversed impulses responses, according to the desired space time evolution of the deposition process. A schematic representing TR plasmas used as a 3D patterning method of two complex 3D substrates is proposed on Figure 3. Using this method, the two dynamic plasmas could easily be controlled independently of one another, making possible the simultaneous treatments of different spatial 3D shapes.

B. Spatial resolution of the method

The features of this patterning deposition method based on TR plasmas would depend, among other things, on the spatial resolution of the TR process. In a classical case, this resolution is of one-half wavelength [7] and can thus be increased by increasing the wave frequency. Note that subwavelength resolution is also possible in some cases [15].

C. Influence of the substrate properties

The electromagnetic behavior of the substrate has an influence on the method to be followed. Two main behaviors may be distinguished:

Dielectric behavior: in this case, the spatial scanning of the cavity (by means of a non-intrusive probe) should be done once and could be done indifferently before or after placing the substrates inside the cavity, as their presence do not

change the behavior of the waves (as long as they are not too lossy).

Conductive behavior: in this case, the spatial scanning of the cavity has to be done for each configuration of the substrates, and has to be done after putting them inside the cavity, as their presence significantly change the behavior of the waves.

D. Influence of the material properties

The electromagnetic behavior of the material to be deposited may also have an influence on the method efficiency. Indeed, the deposition of conductive materials may not be possible, as their presence could have an influence on the behavior of the waves (but it would depend on how significant is this influence).

E. Influence of the cavity properties

To finish, it is important to notice that TR performances increase with the cavity volume. Indeed, the more the volume of the cavity is large the more the focusing by TR will be important¹ [11]. Furthermore, TR performances also increase with the “chaoticity” of the cavity (in short, a chaotic cavity ensures, statistically, a uniform spatial distribution of the field, avoiding undesired energy concentrations that would compromise an efficient focusing at the desired location). Hence, a deposition method based on TR plasmas would easily deal with a large number of complex 3D substrates. This is a complete break with usual deposition methods, for which the control of the deposition pattern on large area with complex shapes remains major challenge [1,17,18].

F. Note on this discussion

It should be noted that the discussion on the properties of the deposition method based on TR plasmas proposed in this section is a projection of what could be achieved in theory with TR plasmas, based on physical arguments. However, the issue of what materials can be deposited on what substrates of what maximal size has not been experimentally investigated yet. Answering this question requires a more precise and dedicated analysis.

IV. CONCLUSION

These unprecedented capabilities of space-time control of plasmas over large and complex 3D surfaces could offer new possibilities in the context of nanocomposite deposition. Indeed, this method, used in molecular gases, could also be adapted for the deposition of various thin films as well as nanocomposites, *i.e.* of nanostructured or nanofilled coatings. It could lead to the development of an innovative and versatile method allowing the patterning of complex 2D or 3D structures on large areas.

ACKNOWLEDGMENT

The authors would like to acknowledge the DGA/AID for their funding support and the CEA-Gramat for the material support. We are also grateful to K. Makasheva for her valuable comments.

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¹ Note that there is a saturation phenomenon, from which the TR performances become constant with the increase of the cavity volume [16]. However, an increase of the cavity volume certainly leads to a better control of plasmas by TR,

as the proportion of the cavity volume occupied by the plasma decreases with the cavity volume.