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Wide range investigation of duty cycle and frequency effects on bipolar magnetron sputtering of chromium nitride

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ABSTRACT

Among the different techniques of reactive sputtering, the bipolar and high power impulse magnetron sputtering are growing in interest for the thin films research community. However, the combination of both processes in presence of a reactive atmosphere is extremely complex and the role of the sputtering parameters are key points to control the deposited material properties. In this study, we have investigated the effect of the duty cycle and the pulse frequency on the reactive bipolar sputtering efficiency of chromium in presence of nitrogen. The study has been performed on a wide range of parameters: from 12.5 to 87.5% for the duty cycle, and from 62.5 to 5000 Hz for the frequency. In situ measurements of the magnetron discharge characteristics have been performed (excitation temperature, peak target current and voltage, energy influx at substrate position) in addition to ex situ characterizations of the deposited thin films (structure, microstructure, density, composition, optical and mechanical properties). It appears that the modulation of the duty cycle allows a better control of the mechanical properties due to higher ionization level at the target, while the frequency is better adapted to tune the optical properties that are attributed to a change of texturation and density of the deposited film (confirmed by simulation). All films present a similar microstructure due to the absence of bias applied to the substrate during the deposition process, which leads to a similar energy per atom of deposited species.

1. Introduction

With its widespread industrial applications at the end of the 70's, magnetron sputtering has known several (r)evolutions since the turn of the XXIst century. Among these new evolutions, bipolar sputtering and reactive high power impulse magnetron sputtering (R-HiPIMS) are the most studied topics, as confirmed by a significant increase of newly published works over the last 2 years [1–4]. Reactive bipolar sputtering (R-BP), i.e. the use of two magnetrons with voltage switching from one target to the another as a function of time, permits to avoid the "disappearing anode effect" and electrical arc formation usually observed while the target is poisoned by the reactive atmosphere [5]. Reactive HiPIMS (R-HiPIMS) allows to grow oxide or nitride films at higher deposition rate compared to reactive DC – in compound mode, but the deposition rate is higher in metallic mode using DC sputtering [1] – with a better control of the physical properties [1, 6]. The R-HiPIMS is also characterized by high peak current density (100–3000 W/cm^2) leading to a very high ionization fraction near the target that conducts to self-sputtering of the target (i.e. sputtering of the target, with target ions) and gas rarefaction.

The combination of both bipolar sputtering with two targets and HiPIMS techniques leads to complex deposition mechanisms that have to be investigated, in terms of plasma diagnostics, materials properties and relationships between them. Among the deposition process parameters, the frequency, the duty cycle, the negative substrate bias, the global pressure and the partial pressure of reactive gas are probably the most critical ones. Since the effect of the last three is well reported elsewhere [1, 6–10] and appear to be similar in DC and pulsed sputtering, we restricted ourselves in this work to investigate the effect of the duty cycle and of the frequency. The subsequent questions to be addressed in this work are:

(i) "When" – in terms of ON and OFF pulse duration - starts R-HiPIMS?
(ii) Is the transition between R-HiPIMS and R-BP smooth or sharp?

Previous works investigating the modulation of these two parameters are generally limited to a small variation range, single target, or a single parameter variation [1, 8, 9, 11–13]. Recent work from Bobzin

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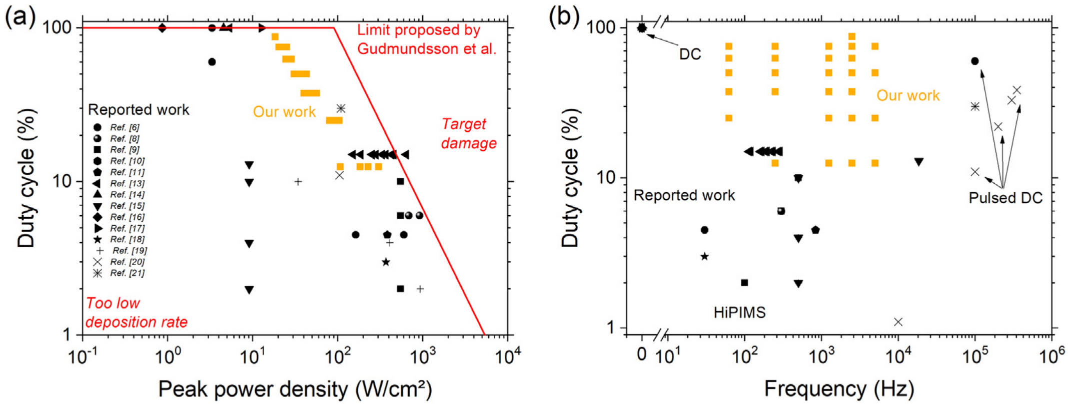

Fig. 1. Results from the present work (yellow squares), as compared to reported studies (black markers) on CrN obtained in pulse reactive sputtering [1, 6, 8–11, 13–21], in terms of duty cycle vs. peak power density (a), and duty cycle vs. frequency (b). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

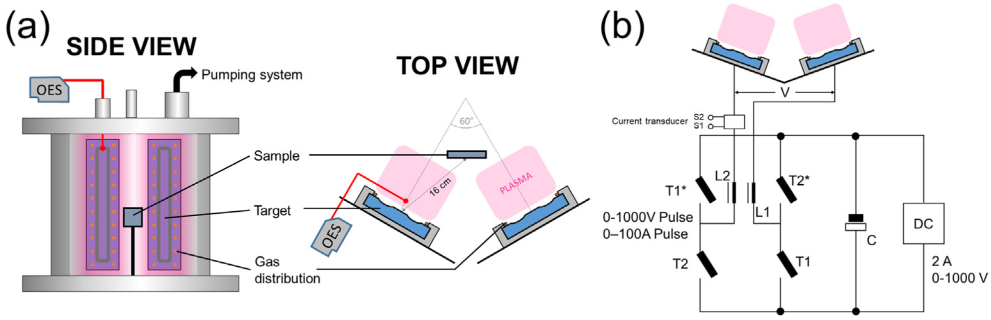

Fig. 2. Schematic side and top views of the location of the sample and of the in situ measurements (a). The orange circles represent the gas inlets. Electric circuit of the bipolar pulse unit (b).

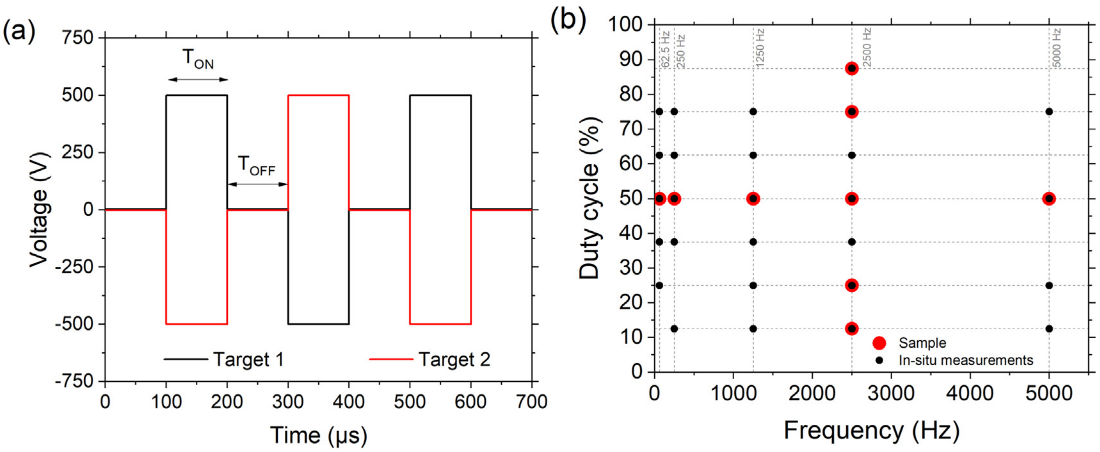

Fig. 3. Schematic variation of each target voltage as a function of time and definition of T_{on} and T_{off} (a), and presentation of the studied conditions and synthesized samples (b).

et al. [2] have carefully investigated the effect of frequency from 300 to 2000 Hz on (Cr, Al)N films, but they used a single target with a constant power and a limited range of duty cycle (0.4 to 8%). In this work, the duty cycle and the frequency were varied from 12.5 to 87.5% and from 62.5 to 5000 Hz, respectively. This allowed us to cover the limits of R-HiPIMS (duty cycle $<$ 15%) to the frontier of pure bipolar sputtering, with a variation of the frequency from mid to high frequency, using a single power supply. The lower limit (12.5% - 62.5 Hz) is the minimum of duty cycle and frequency that can be safely used with by the power supply, while the maximum frequency is arbitrarily chosen to approach high frequency sputtering ($>$ 1 kHz).

In order to facilitate the comparison with previous studies, we report our work in terms of duty cycle as a function of the peak power density (Fig. 1.a) or of the frequency (Fig. 1.b).

As chromium nitride has been widely studied and remains one of the reference systems to compare sputtering mechanisms and films properties, we have carried out our studies on the sputtering of Cr in the presence of Ar and N_2. The effect of the duty cycle and frequency are studied through different in situ plasma diagnostics (i.e. excitation temperature, peak target current & voltage, and energy influx at substrate position) and ex situ measurements of various thin film properties (i.e. structure, microstructure, density, optical & mechanical properties, and chemical composition).