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Plasma “anti-assistance” and “self-assistance” to high power impulse magnetron sputtering

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ABSTRACT

A plasma assistance system was investigated with the goal to operate high power impulse magnetron sputtering (HiPIMS) at lower pressure than usual, thereby to enhance the utilization of the ballistic atoms and ions with high kinetic energy in the film growth process. Gas plasma flow from a constricted plasma source was aimed at the magnetron target. Contrary to initial expectations, such plasma assistance turned out to be counterproductive because it led to the extinction of the magnetron discharge. The effect can be explained by gas rarefaction. A better method of reducing the necessary gas pressure is operation at relatively high pulse repetition rates where the afterglow plasma of one pulse assists in the development of the next pulse. Here we show that this method, known from medium-frequency (MF) pulsed sputtering, is also very important at the much lower pulse repetition rates of HiPIMS. A minimum in the possible operational pressure is found in the frequency region between HiPIMS and MF pulsed sputtering.

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I. INTRODUCTION

Magnetron sputtering is a well-known physical vapor deposition technique leading to thin films with properties that can be controlled via the sputtering gas pressure, operational voltage, target-substrate distance, etc. For many thin film or coatings applications it is desirable to have an energetic assist in the deposition process because it can beneficially enhance the density of the film and create a certain film texture.^{1,2}

Energetic atom and ion bombardment of the growing film can be elegantly done with atoms and ions provided by the magnetron discharge.³ Here we can distinguish four groups of particles: (i) sputtered atoms that are ejected from the surface; their energy is described by a Sigmund-Thompson or similar distribution;⁴ (ii) noble gas ions from the magnetron discharge, usually argon, (iii) ions of the reactive gas, if present; those can be positive or negative, where the latter can carry unwanted excessive energy from acceleration in the target sheath,⁵ and (iv) ions of the sputtered material. In common magnetron sputtering, the degree of ionization is very small (of the order of one percent), however, in the emerging technology of high power impulse magnetron sputtering (HiPIMS), the fraction of ionized sputtered material can be large and in some cases approach 100%.⁶⁻⁸ The fundamental difference between atoms and ions in film deposition is that the kinetic energy of the latter at arrival on the substrate surface can be controlled via substrate biasing.^{9,10}

An important parameter in all sputtering systems is the gas pressure because it affects all aspects of sputtering, including the current density at the target as well as the transport of sputtered atoms to the substrate. When it is desired to utilize the ballistic energy from the sputtering process one would try to use a pressure as low as possible

down to the limit of having a stable magnetron discharge. Such lower limit of pressure is typically in the 0.1 Pa (\sim 0.8 mTorr) region, and depends on the magnetic field strength as well as on the kind of gas and target material.¹¹ When the pressure is reduced further, the discharge will be unstable and may extinguish. It was previously shown that by utilizing self-sputtering, one even could approach sputtering in vacuum: for selected materials like copper, sputtering in vacuum was demonstrated for direct current (DC) operation with initial gas flow,^{12,13} for medium-frequency (MF) sputtering with a pulse frequency in the range of 60–90 kHz and duty factor of 80%–90%,¹⁴ and at low pulse repetition rate in high vacuum with the help of a pulsed arc trigger discharge.⁹

Here we report about the idea to not just provide *gas* to the magnetron but *gas plasma* that assists the ignition and maintenance of the magnetron discharge. When plasma from a gas plasma source is directed towards the target, it is thought that such plasma might assist the magnetron discharge and thereby allow HiPIMS technology to operate at lower pressure.

II. EXPERIMENTAL

To test the gas plasma-assist idea, the experimental setup of Fig. 1 was employed. The target material used in this study was niobium, a choice relevant to superconducting coating applications. A 2-inch (5 cm) balanced magnetron (US Inc.) was powered by a HiPIMS magnetron supply (upgraded SPIK2000A by Melec), capable for pulses with a maximum voltage of 1 kV and current 500 A, with variable pulse duration between 5 μ s and 5 ms. The SPIK pulser was charged by a Pinnacle[®] DC charging supply (Advanced Energy).

The average power delivered to the magnetron was limited to a maximum of 1 kW to prevent overheating and damage of the magnetron. This limit, set at the Pinnacle supply, has certain implications assuming a selected fixed pulse length. At high pulse repetition rates, the limit of average power is readily reached and the voltage will drop below the setpoint, and at low pulse repetition rates, while the voltage will stay at its setpoint, the power will not reach the maximum allowable average setting of 1 kW. We point out that the term *pulse repetition rate* (unit: pulse per second, pps) is commonly used with low duty cycle processes (like HiPIMS technology), whereas the terms *pulse repetition frequency* or *pulse frequency* (unit: Hz) are used at higher frequencies, i.e. when the train of pulses is closer to a wave shape. In this contribution, considering both HiPIMS and MF sputtering, we use those terms synonymously.

The assisting plasma to the magnetron was provided by a miniature Constricted Plasma Source¹⁵ (CPS), which is essentially an enhanced glow discharge that can operate with very low pressure outside the plasma source, in extreme cases down to the 10^{-3} Pa (10^{-5} Torr) region. The most stable operation of such source, showing maximum plasma flow, is when the pressure outside the source is in the 10^{-1} Pa (10^{-3} Torr) region. The CPS was powered with a regulated direct current (DC) mode-switched high voltage power supply (Glassman). The plasma density of the gas plasma (Ar) is about 10^{16} m⁻³ and falls off with increasing distance from the source constriction (nozzle), as schematically indicated in Fig 1(b). The distance between the nozzle of the CPS and the center of the magnetron target was 7 cm.

The chamber (stainless steel, 1 m inner diameter, 25 cm inner height) was cryogenically pumped to a base pressure of $3 \cdot 10^{-5}$ Pa and filled with research grade

argon with an adjustable flow rate of up to 100 sccm by a calibrated mass flow controller (MKS). As shown in Fig. 1(a), the gas injection point was through the CPS nozzle near the magnetron target. The pumping speed could be controlled via a variable gate valve (VAT), but in the interest of minimal gas contamination most experiments were done at full pumping speed of 1500 l/s (specification for nitrogen). The magnetron was positioned in the center and the VAT gate valve was off-center mounted on the floor of the chamber. The base pressure was measured with an ion gauge, while the process pressure was monitored with a capacitance manometer (MKS Baratron[®]) at a port between magnetron and pump (Fig. 1(b)). Therefore the local pressure near the magnetron was slightly higher than indicated. The difference, however, should be small due to the large size and gas conductance of the chamber.

III. RESULTS

In a first round of experiments, the gas was injected into the process chamber through the gas plasma source without activating its plasma generation. Stable HiPIMS operation was readily obtained when the gas pressure was relatively high, e.g. one to several Pascal. Figure 2 shows an example of a typical HiPIMS pulse obtained at a repetition rate of 100 pulses per second; here the pulse duration was 80 μ s, which is somewhat longer than what most researchers chose. We can clearly see that the current is still rising and has not yet found its steady-state at the end of the pulse. Longer pulses lead to even higher currents but also increased likelihood for arcing. Shorter pulses provide greater stability but reduce the amount of metal in the plasma because metal self-

sputtering evolves and increases during each pulse.^{16,17} The issue of pulse duration, repetition rate, and pressure will be considered later in greater detail.

In a next step, the plasma source was switched on; it operated for example with a glow burning voltage of 300 V and a current of 270 mA. We found, to our initial surprise, that instead of assisting the magnetron, the magnetron discharge ceased immediately when the CPS was switched on even before we had a chance to lower the pressure! This effect was very reproducible: under pressure conditions when the HiPIMS discharge worked reliably without the CPS, it immediately stopped when the CPS was switched on, and it resumed operation when the power to the CPS was stopped. Clearly, the original idea did not work, we rather deal with an “anti-assistance” to the magnetron discharge. Therefore, it seemed advantageous to optimize the regime of stable operation without external gas plasma assistance and rather focus on the HiPIMS parameters pulse length and repetition rate in order to determine the lowest possible pressure of stable operation.

A profound change in the pulse shape can be noticed when the pulse repetition rate is changed, which is especially relevant for short pulses. Fig. 3 shows the current pulse evolution for almost unchanged voltage pulses of 20 μs with the pulse repetition frequency as a parameter. We can clearly see that the current reaches higher values earlier as the pulse repetition frequency is increased. The peak current is increased as the frequency is increased and therefore the average power increases, too, and eventually will reach the limit of 1 kW set at the charging supply.

From a large number of such curves one can pick the maximum current value (at the end of the pulse) and construct the (peak) current – (constant) voltage characteristics

as shown in Figure 4. The “self-assistance” of the previous pulse is evident by the “accelerated” rise and higher amplitude of the current as the frequency is increased (Figs. 3 and 4).

At this point, we should come back to the original question and address which conditions would allow us to operate in a stable manner at the lowest pressure possible. Since we have seen the rather dramatic influence of the pulse repetition rate, or “self-assistance,” we explore the lowest possible pressure in a wide range of pulse repetition frequencies, and in fact cover the whole range from single pulse operation (~ 1 pulse per second) up to medium frequency pulsed sputtering. Of course, as mentioned before, for a fixed short pulse length, the system will operate at very low average power at very low pulse frequencies, and in the other extreme, at medium frequencies of many kHz, the limitation of the average power will kick in to protect the magnetron from overheating, i.e. we operate at the maximum allowable power (1 kW) and a voltage less than the setpoint (1 kV in this series of experiments). Figure 5 shows the lowest possible operational pressure for a fixed pulse length of 30 μs ; it was measured by starting the discharge at a higher pressure of 0.15 Pa and gradually lowering it until the discharge stopped. For frequencies exceeding 1 kHz we observed an initial voltage less than 1 kV caused by the limiting setpoint of average power. By gradually reducing the pressure, the current is gradually reduced, too, and thus more voltage was “allowable” within the limit of average power, and in fact, at extinction, the voltage was always at the setpoint of 1 kV. This procedure of determining the lowest possible pressure resulted in a minimum in the frequency region between HiPIMS and MF pulsed sputtering.

IV. DISCUSSION

The initially puzzling observation of “anti-assistance” by an external gas plasma can be readily explained if we think in terms of gas *density* instead of pressure. Our large processing chamber, with a volume of about 0.25 m³, ensures that the process is isobaric (apart from transients). Using the ideal gas equation for isobaric conditions, $p_{gas} = n_{gas}kT_{gas} = const.$, where n_{gas} is the gas (particle number) density, p_{gas} is the gas pressure, T_{gas} is the gas temperature, and k is the Boltzmann constant, we see that any increase in the gas temperature leads to a reduction of the gas density. The plasma source will not just provide gas plasma, it will also heat and hence dilute the gas in front of the magnetron target. A certain minimum gas density is required for the interaction of energetic secondary electrons with the gas, producing ions, which in turn sustain the plasma by generating replacement secondary electrons. The magnetron discharge itself causes a similar gas dilution effect, which is well-known as gas *rarefaction*, characterized by a substantial reduction of the local gas density mainly due to elastic collisions of the sputtered particles (kinetic energies of several eV) and background gas.^{3,18-22} Rarefaction is less important at low gas pressure,²³ but significant when the flux of sputtered atoms is high, especially for materials with high sputter yield.^{19,21,22} Rarefaction increases with increasing discharge current, and it is clear that the effect is very important for the high currents typical for the HiPIMS discharge. Gas plasma assistance to the magnetron discharge leads to an undesired amplification of the gas rarefaction effect, which can cause the complete extinction of the magnetron discharge.

The experiments showed that operating the HiPIMS discharge relies on “self-assistance,” which is associated to the presence of charged or “activated” particles from

the previous pulse. The assistance to plasma development by the afterglow from the previous pulse is well documented for MF pulsed sputtering, in the frequency range 10-350 kHz.^{14,24-26} It is not trivial, however, that such effect can also be found for the much lower pulse frequencies typical for HiPIMS.

One reason for the lack of the observation is simply that when MF pulsed discharges were studied, like the 100 kHz discharge by Welzel et al.,²⁵ or Bryant et al.,²⁶ slower processes with long time constants cannot be seen due to the onset of the next pulse. Consistently, those authors report about characteristic times in the microsecond range.

Bäcker et al.,²⁴ studying pulsed radio-frequency (RF) magnetron plasmas with 10 ms on and 10 ms off time, found two different groups of time constants for electron temperature and density, where the longest decay times for density was about 500 μ s. More recently, the same group investigated pulsed dc magnetron discharges using a time-resolved Langmuir probe. They found again two groups of electron decay times, the longer times being 250 μ s to 600 μ s far from the target.²⁷ Also using time-resolved Langmuir probes, Seo and coworkers²⁸ found a variety of time constants which were generally shorter than 100 μ s; they associated the variations with the different groups of electrons in the dynamic electron energy distribution function. The longer of the characteristic times support the finding that HiPIMS discharges can be “self-assisted,” however we observed the effect of “self-assistance” to be much longer than what is generally reported.

One plausible consideration is to consider the metastable levels of atoms and ions, which can be very long lived and contribute to the slow decay, such as the metastable $1s_3$

and $1s_5$ states of neutral argon.²⁹ Considering optical transition probabilities of excited levels in general (see for example the Atomic Spectra Database of NIST³⁰), we find that the transition times vary by about 10 orders of magnitude for different excited levels, with the fastest being a few nanoseconds, and the slowest ($1s_5[np^5(n+1)s, ^3P_2]$ for argon) longer than 38 seconds.³¹ As Katori and co-workers³¹ pointed out, those metastable states are much more likely to be de-excited by collisions than by radiative decay. Recombination of electrons and ions contribute to the generation of new metastable atoms. For example, Nafarizal and coworkers³² observed an increase in the density of metastable atoms 1.5 ms *after* the magnetron discharge pulse was terminated.

In light of those data one can state that afterglow time constants span a wide range depending on the kind of material and process considered.^{33,34} For pure (gas-less) self-sputtering of copper, however, the time between pulses must not be too short: the discharge extinguishes in the absence of argon for frequencies less than 70 kHz.¹⁴

As stated before, the lifetimes of long-living levels are limited by collisions, and therefore one needs to consider the collisional processes of HiPIMS discharges. The density of charged particles decays by volume recombination and diffusion. Volume recombination is known to be only effective at high density when both the energy and momentum conservation can be satisfied via three-particle collisional recombination. Diffusion of plasma away from the dense region is generally ambipolar, thus determined by the less mobile particles. These are usually the heavier ones (here argon and metal). Diffusion away from the target region is additionally hampered by the flux reversing gas rarefaction. Those processes are rather complicated and require further measurements

and modeling. Most models of gas heating in magnetrons are limited to the steady-state situation^{19-22,35,36} and do not take into account the pulsed return flux.

After having discussed plasma afterglow and dynamic rarefaction, it is time to look at the final result of this study displayed in Fig. 5. It shows the lowest possible pressure at which the discharge operates as a function of the pulse repetition frequency. A minimum was found between the regions typical for HiPIMS (very high current pulses, relatively low repetition rate) and MF pulsed sputtering (moderate current and frequencies here up to 20 kHz). This minimum is the result of (at least) two counteracting processes, and again the interplay of dynamic rarefaction (formation and collapse of a low density zone) and plasma decay come to mind.

The rise of the high-frequency branch of Fig. 5 with increasing frequency can be attributed to the lower local density near the target when the next pulse is provided. In other words, the effects of rarefaction are seen stronger by the next pulse when the time lag to the previous time was shorter. The increase towards high frequency levels off at about 0.105 Pa. The last data point (at 20 kHz) is a situation where the duty cycle exceeds 50% (30 μ s on, 20 μ s off), and thus DC conditions are approached because rarefaction under our conditions takes typically 50-100 μ s to be established.⁸ The time between pulses (20 μ s) is much shorter than afterglow time constants, such as the characteristic time of ambipolar diffusion, and therefore self-assistance by the previous pulse is effective.

Now, considering the low-frequency branch, plasma decay will reduce the plasma density and the associated self-assistance as the time lag to the previous pulse is increased. However, no leveling off is found at very low frequencies although one would

expect that eventually one would reach the single-pulse mode that is independent of frequency because the time between pulses is long enough to completely dissipate any afterglow plasma and to fully restore the original gas density in front of the target. The reason for the lack of those data points is that for the fixed pulse length of 30 μs in the test, no discharge can be started, at any pressure, when the repetition rate drops below 8 pulses per second. This by itself is a clear indication for the role of self-assistance by the previous pulse. The notable finding is that the effect of such self-assistance is much longer lasting than expected or known. Processes with times greater than 100 ms must exist, and we speculate that contributions include the full restoration of the density as it was before rarefaction, and the formation of long-living metastable atoms. Future work is needed to clarify the contribution of those and other processes.

For very low pulse repetition rates (< 100 Hz), long delay times between the application of voltage and the onset and strong rise of discharge current are well known, especially at low pressure. In fact, the seminal paper on HiPIMS by Kouznetsov and workers³⁷ showed a 50 μs delay in their Fig. 1 for a pulse repetition rate of 50 pulses per second (copper target, applied voltage of 1.5 kV) at the very low pressure of 0.065 Pa (argon). A successful approach to reducing the delay between voltage and current was recently demonstrated for copper and short pulses by having a very low DC current (some mA) discharge at the magnetron, thereby keeping some plasma “alive” between pulses.³⁸

In conclusion, we have shown (i) that external gas plasma assistance to a magnetron discharge does not enable the operation of the magnetron at lower pressure, quite contrary, the gas rarefaction effect can be severe, leading to a complete extinction

of the magnetron discharge; (ii) plasma “self-assistance” by the previous pulse is evident even at pulse repetition rates much lower than previously known; such “self-assistance” reduces the delay time between voltage application and current onset, and it promotes faster development of the current pulse, thereby leading to higher peak currents for short pulses.

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Figure Captions

- Fig. 1 (color online) Experimental setup: (a) sideview of the 1-m diameter cylindrical chamber, with position of the components approximately to scale; the magnetron discharge was assisted by a constricted plasma source (CPS); (b) top view of the discharge region, with the plasma schematically indicated.
- Fig. 2 (color online) Example of a time-dependent, medium-length HiPIMS current pulse, at constant voltage, with a Nb-target at an argon pressure of 0.4 Pa.
- Fig. 3 (color online) Current pulses (bottom) at constant chamber pressure of 0.12 Pa and nominally constant voltage pulses (top) for different pulse repetition rates (2" Nb target, argon gas).
- Fig. 4 (color online) Peak current - constant voltage characteristics for constant pulse length of 20 μs and different pulse repetition rates (2" Nb target, argon gas at 0.12 Pa).
- Fig. 5 (color online) Lower limit of gas pressure needed for stable pulsed magnetron operation with the boundary conditions of 1000 V maximum voltage, 1000 W maximum average power, and 30 μs pulse length (2" Nb target, argon gas).

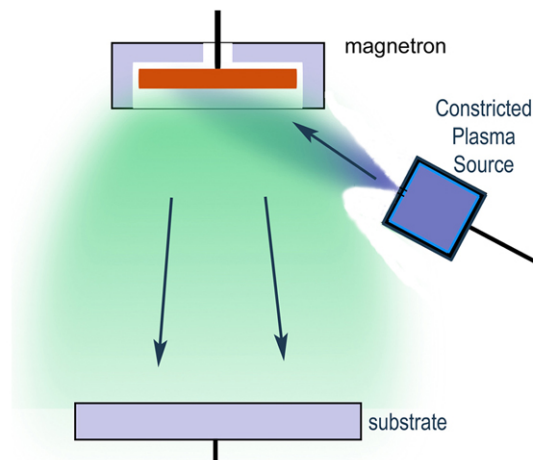
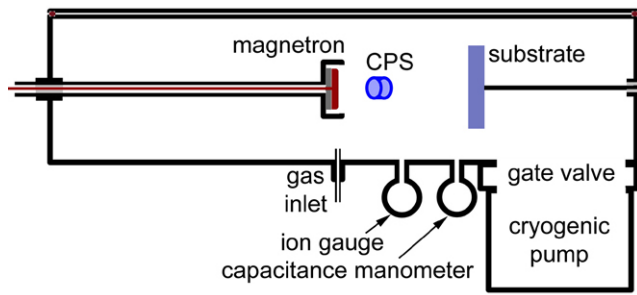


Fig. 1

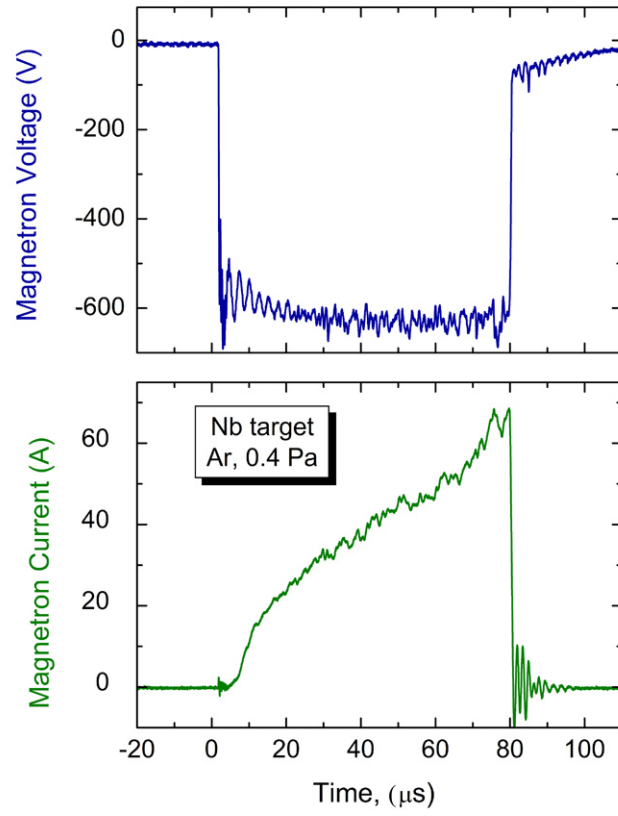


Fig. 2

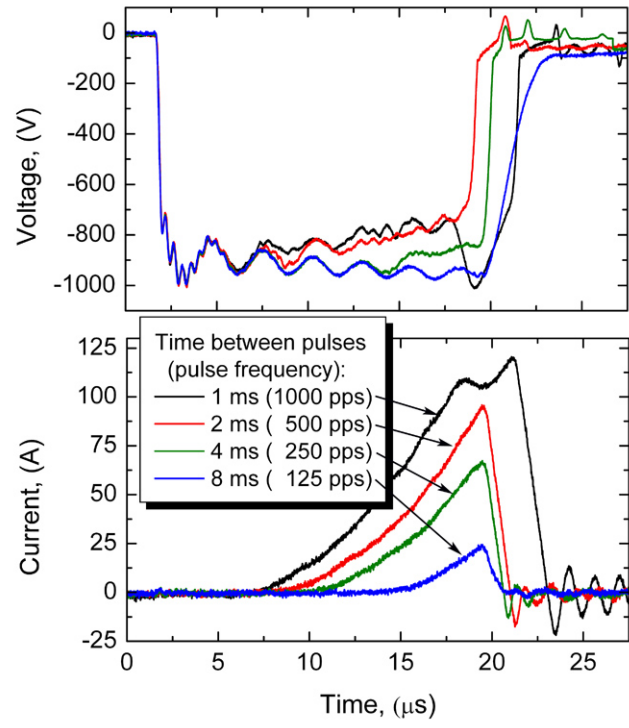


Fig. 3

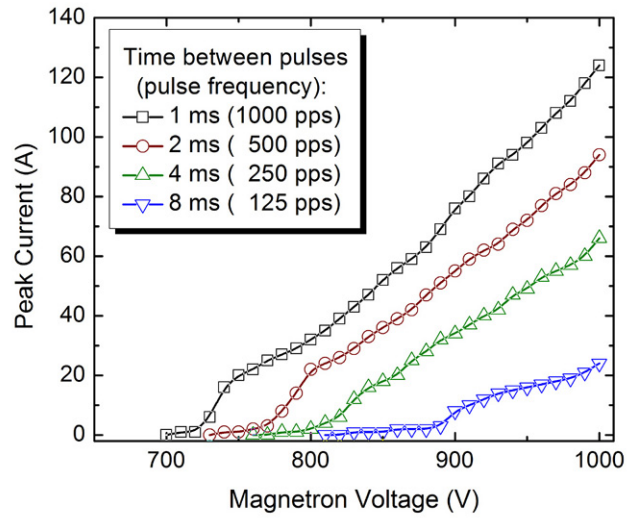


Fig. 4

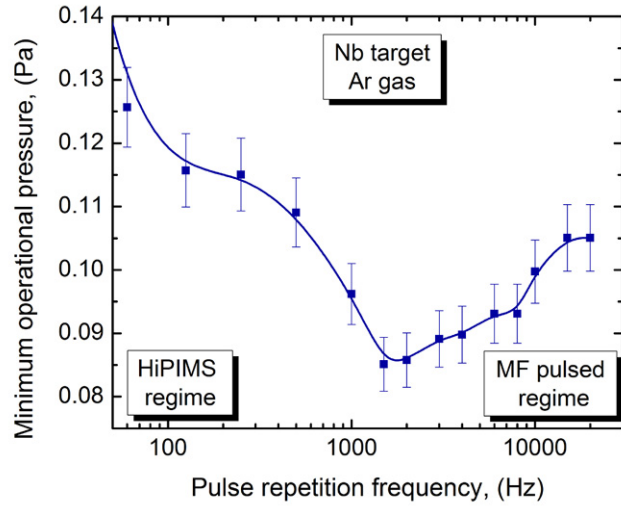


Fig. 5