Pulsed laser deposition—UV laser sources and applications

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Abstract Progress in material research and processing industry is fueled by the technique of pulsed laser deposition (PLD).

High energy excimer lasers enable this technique since every material is amenable to their high photon energies. Spectral properties, temporal pulse and laser beam parameters of state of the art excimer lasers will be compared with frequency converted Nd:YAG lasers.

Both quality and longevity of the deposited layers strongly depend on the degree of accuracy achieved in the thin film ablation and subsequent deposition process.

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1 Introduction

High pulse energy excimer lasers with pulse energies between 300 and 1200 mJ/pulse and photon energies between 5 and 7.9 eV lend maximum flexibility to the technique of pulsed laser deposition. On account of the high energy densities accessible with the latest generation of excimer lasers, the entire material spectrum including high band-gap metal oxides such as ZnO is amenable to precise and controlled ablation with subsequent stoichiometric transfer to the substrate. Because the transferred material needs time to smoothly deposit and position itself optimally on the substrate, the repetition rate of the ablation laser is typically on the order of only 10 Hz. These requirements are best met by pulsed lasers with short wavelengths (248 nm is the most common), high pulse energies (100 to 1000 mJ) and homogeneous spatial energy distribution. Thin film quality is very sensitive to shot-to-shot energy density fluctuations, and because deposition time in a lab takes up to one hour, both spatial (beam profile) and temporal (shot-to-shot) energy stability are essential in order to obtain reproducible results. Therefore, in this paper we will give a comparison of state-of-the-art excimer performance date with flash-lamp pumped Nd:YAG laser beam properties.

2 Experimental

2.1 Ceramic preionization

Based on the proven concept of metal–ceramic technology, the preionization of high pulse energy PLD excimer lasers has been optimized in order to obtain large pulse energies in combination with homogeneous discharge conditions. The newly employed and patented ceramic preionization design (see Fig. 1) uniquely combines the efficiency of a discharge driven preionization source, such as spark preionization designs with the smooth and homogeneous volume preionization as provided by, e.g., the corona preionization which is preferably employed for high repetition rate low pulse energy excimer lasers.

2.2 Optimized gas flow

In order to extend the hands-off operation time of high pulse energy excimer lasers, the gas flow architecture inside the laser tube and the internal electrostatic gas purification systems have been redesigned. Laser gas contaminants are efficiently filtered out by careful optimization of the gas flow
via capable electrostatic filter elements. With the sophisticated gas purification system inside the LPXPro and COM-PexPro, laser gas performance remains to a large extent unaffected during long-term operation even at multi-hundred millijoules of laser pulse energy (see Fig. 2).

### 3 Wavelength and beam considerations

#### 3.1 Excimer versus Nd:YAG laser ablation

In view of optical properties excimer laser have pivotal advantages over Nd:YAG lasers in thin film manufacturing. The advantages are predominantly based on superior ablation characteristics and much better energy stability that are available at comparable costs and similar maintenance expenses. Major drawbacks of the Nd:YAG lasers for PLD include, inherently inappropriate Gaussian beam profile instead of a flat-top profile as well as temperature-induced polarization and thermal lensing effect create donut-shaped beam profile and lateral distortions (see Fig. 3).

In addition, poor short-term and long-term pulse-to-pulse stability of typically 10 to 15%, rms is provided by Nd:YAG lasers due to the necessary frequency conversion in order to attain the ultraviolet regime the efficiency of which is severely degrading toward shorter output wavelengths such as 266 nm. Another limiting factor of Nd:YAG lasers in PLD experiments is the fixed repetition rate of 10 or 20 Hz which is not alterable but represents a fixed parameter. An overview of the optical performance parameters of excimer and flash-lamp pumped Nd:YAG lasers is given in the following table.

As a matter of fact, state-of-the art excimer lasers from the COMPexPro series are benchmark UV laser sources for pulsed laser deposition as can be seen from the hard specifications list in Table 2.

### 4 Laser–material interaction

#### 4.1 Material coupling

When laser light interacts with a solid surface, the photon energy is transformed into heat. The temperature of the solid material increases, leading to melting and evaporation of the solid material. Because the temperature in the vapor plume can rise to high values (10,000 K and higher, see [1]), a plasma is formed. Besides atoms, electrons and ions, the material plume also consists of particulates, with dimensions ranging from nm to µm. The smallest particles (∼nm size) are probably formed in the expanding vapor plume, by condensation of vapor atoms. The larger particles (∼µm size) are likely created by direct ejection from the solid target. Moreover, at very high laser irradiance (above $10^{10}$ W/cm$^2$), explosive boiling of the target material beneath the surface layer, as well as mass ejection of large particulates, may occur.

Higher photon energies, or equally shorter wavelengths, lead in general to faster plume heating and smaller particle sizes, hence supporting a preserved material stoichiometry and smoother thin films without unwanted particulates. The shorter the wavelength, the smaller the penetration depth of the laser radiation into the material. For excimer