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Excimer laser technology trends

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Abstract

State-of-the art pulsed UV excimer laser technology has a proven track record of stable performance under three-shift operation conditions in various industrial thin-film applications at stabilized pulse energy levels ranging from 100 to 1000 mJ per pulse. For the last 25 years excimer laser technology has formed the backbone of pulsed laser deposition advances. The available UV average power level nowadays exceeds 1 kW and novel excimer laser beam utilization schemes in combination with sophisticated substrate–target geometries are ready to utilize the full laser upscaling potential and thereby reduce unit costs.

(Some figures may appear in colour only in the online journal)

1. Introduction

In 1987, the year when pulsed laser deposition (PLD) was pioneered by Venkatesan at Bellcore [1], commercial excimer laser technology was still in its infancy. In those days, excimer laser tubes were still made of organic polymers and severely limiting the operational lifetime of the active gas as well as the long-term optical performance of the laser due to outgassing and material ageing. The introduction of the all-metal-ceramic laser tube design in 1994 thus represented an important milestone resulting in a tenfold increase in gas and tube operational lifetime. Since then, insulators and high-voltage feed-through parts are all made of corrosion resistant high-density ceramics and the metal components of the laser tube consist of carbon- and silicon-free alloys preventing the generation of corresponding contaminants.

In 1998 both laser operational lifetime and output beam stability were further improved by replacing the eroding spark discharge by a sliding discharge soft preionization scheme. With rising duty cycles and increasing pulse frequency of excimer lasers, the operational lifetime of the thyratron, in particular, which for every laser pulse switches tens of kilovolts on a nanosecond time scale, became a bottleneck for industrial use. The development of low-voltage semiconductor switching technology combined with magnetic isolators and pulse transformers for achieving the required discharge voltage was hence a major breakthrough. In 2005 the first solid-state pulsed 300 W excimer laser for large-area thin-film annealing was built. The replacement of thyratron switches by its maintenance-free solid-state based counterparts laid the groundwork for the ongoing success of high power excimer lasers on the industrial production floor [2].

Nowadays, all excimer lasers at above 100 W output power are built on maintenance-free solid-state switching technology providing high-energy pulses of 5–35 ns, FWHM pulse width. In the year 2010, novel solid-state switching technology has enabled high power excimer lasers to cross the kilowatt mark for the first time ever. These core innovations together with constant performance and lifetime improvements have rendered excimer lasers today's most cost-effective and dependable pulsed ultraviolet (UV) laser technology.

Today, excimer lasers have become indispensable photonic production tools in various high-tech growth markets as diverse as medical [3], microelectronics [4], flat panel display [5] and automotive industries [6] mainly at the wavelengths 193, 248 and 308 nm.

In the following sections, the technical aspects of excimer lasers relevant for PLD and resulting output energy characteristics are discussed in view of both scientific and an industrial environments.

2. Excimer lasers in PLD thin-film research

2.1. Low pulse count implications

From an excimer laser viewpoint, the typical PLD conditions in thin-film research labs correspond to extremely low duty cycle use. As growing a thin film is mostly performed at low repetition rate between 1 and 10 Hz, usually less than a hundred thousand laser pulses are accumulated per day. Over the course of a year the pulse count in the laboratory environment may add up to 30 million laser pulses. As ablation lasers, compact excimer lasers at pulse energies of 200–1100 mJ are employed mostly at a wavelength of 248 nm supporting a large range of ablation fluences between 0.3 and 30 J cm^{-2}.

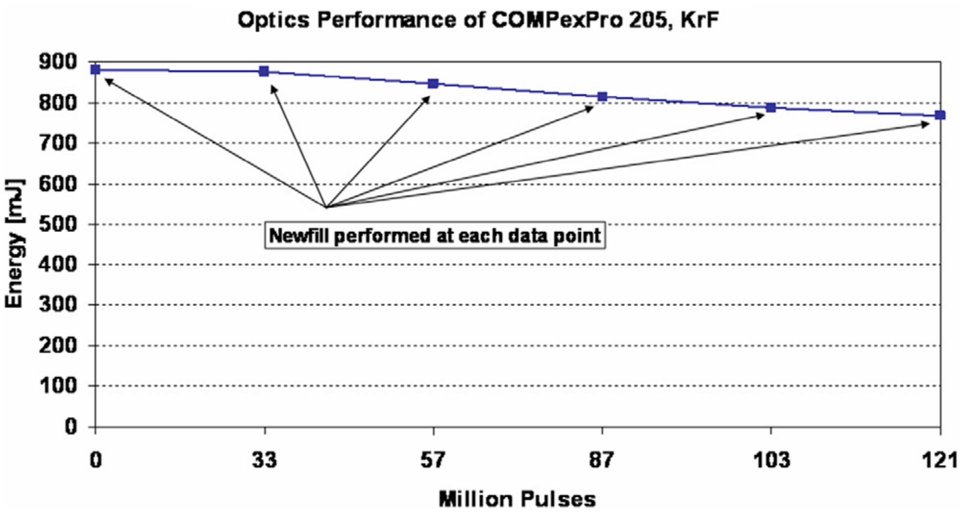

Figure 1. Optics performance over a four-year equivalent pulse count under typical PLD conditions.

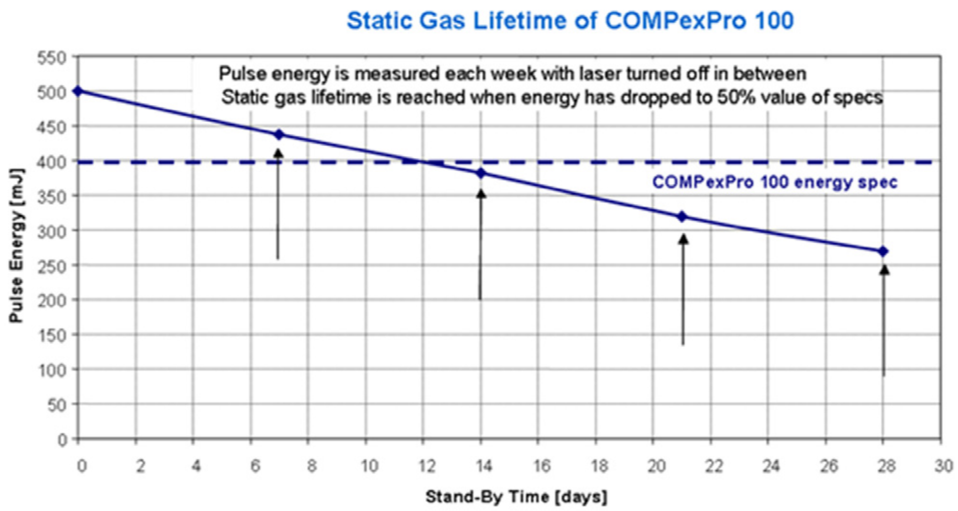

Figure 2. Static gas lifetime of a standard PLD laser filled with 248 nm premix gas.

The low pulse count translates into a lifetime for the laser tube of about 10 years and also into long exchange cycles for the resonator optics of 3–5 years, as indicated in figure 1.

As the excimer laser in a PLD laboratory lingers thus mostly in the off state, the yearly gas consumption is largely determined by the static gas lifetime which is the time interval between consecutive new fills accepting a 50% reduction of the initial output energy before each new fill.

A static gas lifetime of over a month is typically achieved, resulting in about 25 new fills per year which are easily covered e.g. by a 20 l premix gas bottle. In the measurement shown in figure 2, after 4 weeks the reduction of energy is just about 30% with respect to the nominal pulse energy specification value of 400 mJ.

Using premix gas is, in fact, an observable trend in PLD research as the gas installation is much simpler compared with using single gases. Moreover, the halogen concentration in the premix gas bottle is only below 0.2% and is thus very easy to reconcile with individual laboratory safety regulations. As a matter of fact, a modern excimer laser in a PLD laboratory offers lower running costs at superior performance compared with a crystal and flash-lamp-consuming, frequency-converted Nd : YAG system [7].

2.2. Energy control and stabilization

Excimer lasers for PLD must meet high standards with regard to performance and output characteristics. One prerequisite for obtaining reproducible thin films of high quality is that the excimer laser keeps its optical performance stable during layer deposition and growth, i.e. over the course of up to a few hours, independent of the set laser repetition rate and pulse energy.

The basic approach to stabilize the output energy of an excimer laser is to compensate energy variations by adapting the discharge voltage.

Energy stabilization via feedback regulation of the laser discharge voltage, however, induces slight effects on laser pulse length and on the width of the beam profile. As the latter is usually aperture imaged onto the target, changing the discharge voltage indirectly affects the on-target laser fluence as well.

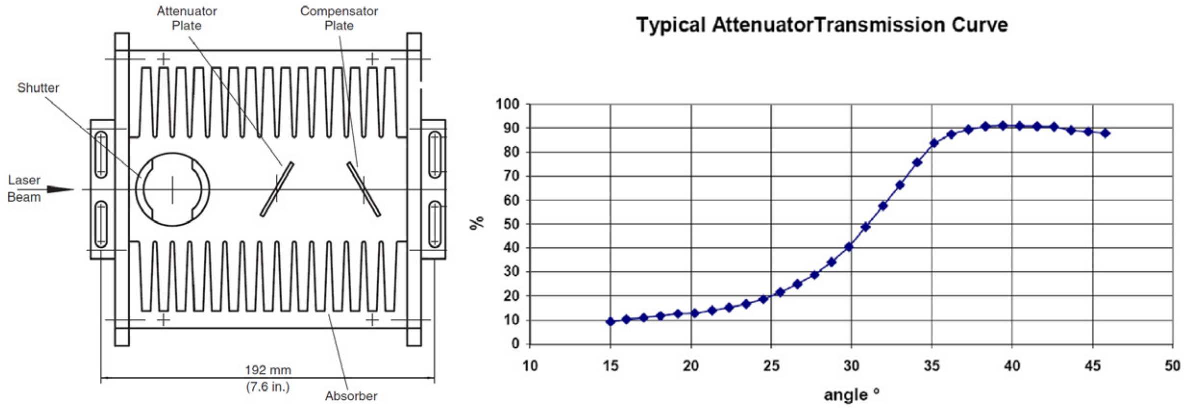

Figure 3. Dual plate attenuator layout (left) and plate angle dependent optical transmission (right).

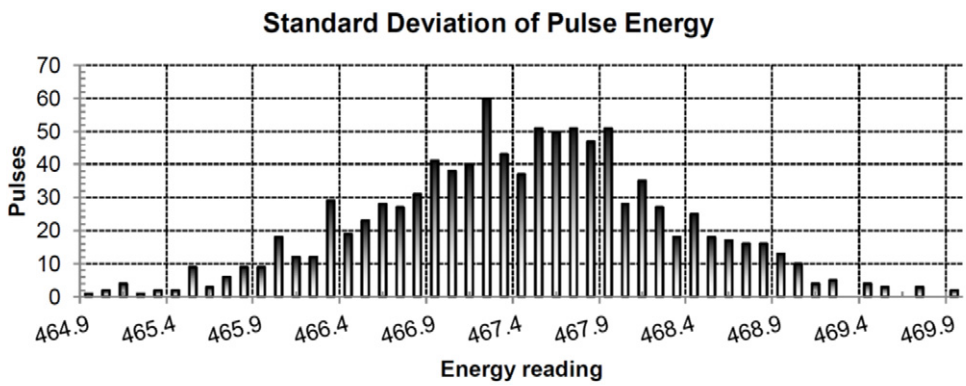

Figure 4. Pulse energy distribution of laser pulses at low pulse frequency and high laser energy.

Hence, the preferred energy stabilization method is via external attenuation using a dual plate attenuator.

Attenuator modules, as depicted in figure 3, left, are either controlled manually or motorized with feedback diode and are increasingly used as a convenient add-on in PLD labs.

Dual plate attenuators work via two counter-rotating substrates with anti-reflective coating and allow seamless energy attenuation and feedback stabilization of the input laser beam from 10 to 90% (see figure 3) while the excimer laser is always running under constant discharge voltage and, hence, also under thermally balanced gas discharge conditions.

2.3. Pulse-to-pulse stability

Pulse-to-pulse stability is an essential requirement for PLD thin-film growth because it largely influences the reproducibility of the functional layer. Excimer lasers as direct UV emitters intrinsically exhibit a much better pulse-to-pulse stability than frequency-converted UV lasers. Mainly as a result of introducing ceramic preionization technology, excimer lasers provide superior pulse-to-pulse stability. Typically, a standard deviation of consecutive laser pulses of 1%, sigma and below is observed over the entire pulse frequency range of even the most compact PLD laser models.

A pulse energy distribution of a thousand laser pulses recorded at low pulse frequency of 5 Hz and maximum discharge voltage is shown in figure 4. The free running laser delivered a mean pulse energy of 467.4 mJ and a standard deviation of 0.2%, sigma.

2.4. High pulse frequency burst mode

Specific thin-film growth techniques such as pulsed interval deposition demand high repetition rate operation at up to 100 Hz in conjunction with bursts of pulses. The number of pulses of each burst typically matches the amount of material required for an atomic layer. Atomic layer formation takes place during the burst pause [8].

State-of-the-art excimer lasers feature integrated burst generation with adjustable burst patterns as well as overshoot compensation via self-learning burst stabilization algorithms. Thus ensuring under such relatively unfavourable laser operation conditions an energy stability of below 1%, sigma. The result is that each burst has the preset energy from the first to the last pulse of each burst, as shown in figure 5.

Thin-film PLD research has been steadily evolving over the last two and half decades. From an excimer laser perspective, stable output is to be provided over a large laser parameter range. Compact excimer lasers for PLD research deliver stable UV output energy over a wide range of control parameters and wavelengths employed by a growing PLD research community constantly shifting the frontiers of thin-film development.

Whereas PLD research is characterized by a low yearly pulse count and various laser conditions, industrial thin-film

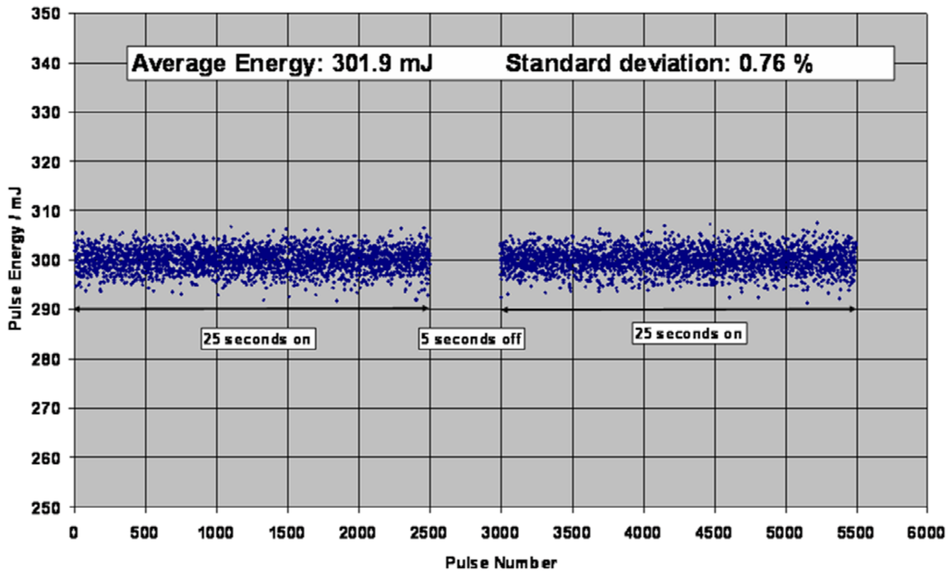

Figure 5. Consecutive high pulse frequency burst at 100 Hz with 5 s burst pause.

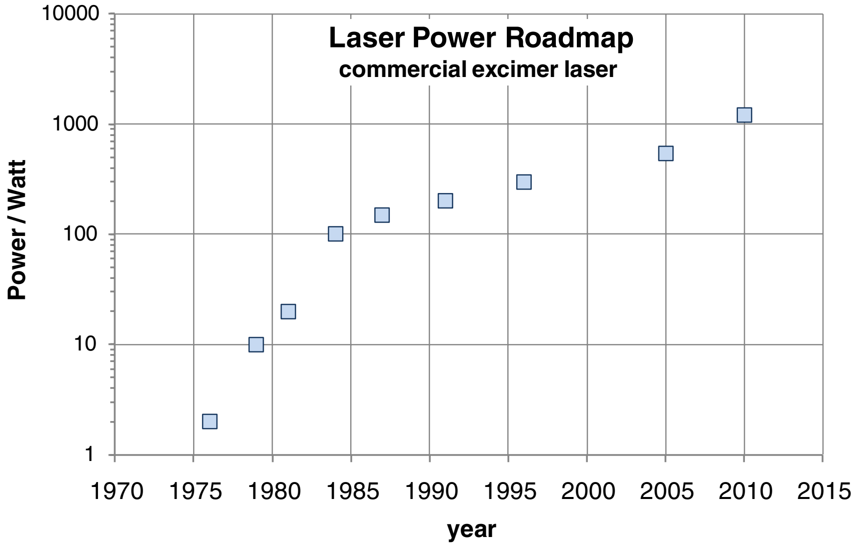

Figure 6. Output power evolution of 308 nm excimer laser systems.

manufacturing via PLD demands exactly the opposite from the excimer laser. The following section will look into the PLD excimer laser requirements for large-scale production.

3. Excimer lasers in industrial PLD manufacturing

3.1. Evolution of laser power and stability

Technically, the industrial success of PLD in a production environment essentially hinges on upscaling the deposition rate without sacrificing overall thin-film quality and homogeneity. For a given PLD system architecture which is capable of exploiting the full laser power, throughput linearly scales with the average output power of the excimer laser used for target ablation. 308 nm has become the preferred excimer laser wavelength in industrial PLD applications, e.g. for coated conductor manufacturing. The reason is that the maintenance intervals of the optical components at this wavelength are longer than for 248 nm photons and consequently lower running costs are associated with 308 nm high power excimer laser manufacturing.

The UV output power levels achieved with commercial 308 nm excimer laser technology have been constantly increasing over the course of the last three decades by two orders of magnitude, as can be seen in figure 6.

The power roadmap has been largely driven by the thin-film silicon annealing market. Since 2010, it extends up to 1.2 kW of average output power obtained from the latest 308 nm high power excimer laser flagship model used in three-shift volume manufacturing of high resolution display backplanes. At the same time, the dynamic gas lifetimes, i.e. the maximum amount of pulses which can be achieved per gas fill at a given stabilized pulse energy and also the pulse-to-pulse stability has been significantly improved over the last 15 years, as illustrated in figure 7.

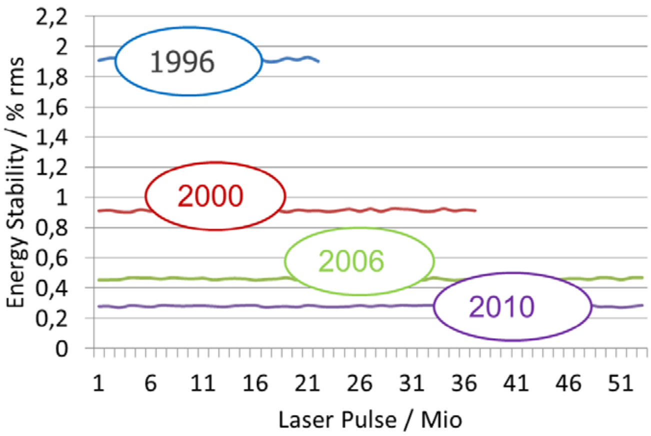

Figure 7. Evolution of the dynamic gas lifetime and output energy stability of 308 nm excimer lasers.

For reliable, high power industrial excimer lasers, the high pressure gas discharge is the only suitable excitation technique. The improvements in pulse-to-pulse stability as well as in laser output power achieved over the last two decades are the combined results of technological advances in laser discharge preionization, discharge circuit design as well as laser gas cooling, ventilation and cleaning.

The technique to produce and control a homogeneous gas discharge is crucial for the performance of a high power excimer laser. Until 1998 preionization pins generated a spark discharge some 10 ns before the main discharge. The sparks produce UV radiation which is sufficient to preionize the laser gas between the electrodes with a homogeneous initial seed density of about 10^8 electrons cm^{-3}. Thereafter the sliding discharge preionization has been introduced. This was superior to conventional preionization schemes because of their ability to generate the same electron density levels with a much higher spatial uniformity. The effect was a twofold improvement of pulse-to-pulse stability and a 20% output power increase from the same laser tube. The threshold values for population inversion in excimer lasers are high due to the short wavelength and the considerable linewidth of the relevant transitions. The typical concentration of the excited active species is 10^{15} cm^{-3}. These can only be obtained by very high pump energy densities of about 10^{-2} J cm^{-3} supplied in a short time interval of some 10^{-8} s as achieved by a high power discharge pulse. The discharge circuit is designed to match closely the dynamic behaviour of the gas discharge, where the ohmic resistance is rapidly changing with time. The discharge is controlled and triggered by a high-voltage switch. This switch must withstand the high voltage and current during the discharge cycle (peak voltage up to 50 kV, peak current at about 100 kA, current rise times of ~30 ns.

In the first excimer lasers, spark gaps were used as trigger switches. These were quickly replaced by thyratrons due to their better reliability and lifetime characteristics. A thyratron tube is filled with H2, provided by heating a hydrogen–metal reservoir (palladium). The hydrogen pressure determines the hold-off voltage of the tube. In the non-conductive state, the grid between the electrodes is negatively biased in order to keep the free electrons, released by a heater close to the cathode. By applying a positive trigger pulse, the electrons can pass the grid and the thyratron switches to the conductive mode. Improved thyratron designs with additional grids helped to achieve a higher hold-off voltage, more reliable triggering and a more homogeneous discharge. Subsequently, larger cathodes for higher achievable forward currents, made the thyratron less vulnerable for reverse currents. The introduction of inductance magnetic switches compressing the discharge current allowed operating a thyratron well below its rise time limit.

As a next step thyratrons have been replaced by multi-stage compression techniques and solid–state switches such as IBGTs. A major physical advantage of solid-state switching technology is that it recovers the energy which is reflected by the discharge due to impedance mismatch. This prevents the late return of the energy to the laser electrodes which would lead to poor discharge and excessive electrode wear. Therefore, the introduction of solid-state switching to high power lasers improved long-term beam homogeneity and pulse-to-pulse stability, in particular.

The introduction of highly effective dc resonant charging power supplies followed by highly reproducible switched-mode power supplies with <0.2% voltage repeatability and ~90% overall efficiency contributed to both optical efficiency and output stability improvements as well. As a combined result, the conversion efficiency of high power excimer lasers increased to 4%.

In excimer lasers the excess discharge energy has to be removed efficiently as excess heat. An internal fan causes a forced circulation of the laser gas in order to keep the active medium renewed in the lasing region and to obtain a high flow rate through the gas filter and the heat exchanger. The heat exchanger, which in most designs uses water as cooling medium in a closed or open loop system, needs a sufficient contact area to allow good temperature stability, especially at high pulse repetition rates. Even though the laser gas is continuously cleaned by special gas filters, the lifetime of the active medium is limited due to unavoidable chemical reactions between the laser gas and tube materials or electrode burn-off, causing solid and gaseous impurities. Most of the solid reaction products can be removed by electrostatic dust precipitators, whereas the gaseous metal halide impurities can be largely removed by a cryostatic gas purifier. Some of the cleaned gas is used to flush the windows of the laser tube in order to prevent the deposition of impurities.

A further gas lifetime increasing measure is to add additional halogen gas to the laser gas mixture when the laser output power decreases.

Over the years, advances in tube and electrode materials, gas flow optimization by new fan designs providing gas circulations speeds of up to 50 m s^{-1} and electrostatic dust filter designs have culminated in stable high power tube operation times over 6–10 billion pulses.

3.2. Capitalizing on pulse energy

Sophisticated beam delivery architectures based on laser beam splitting and scanning enable the formation of several parallel plumes. Under the premise that the required excimer laser fluence for each of the illumination spot sizes on the target is

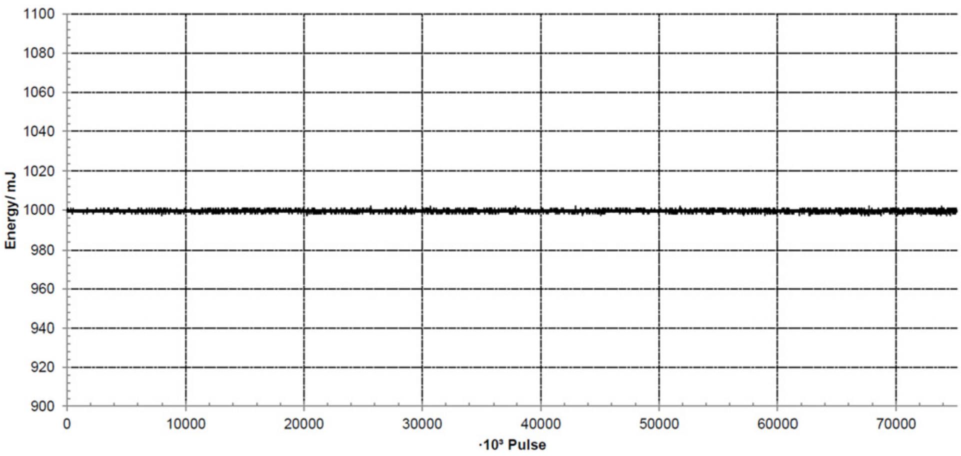

Figure 8. Stabilized energy operation at 1 J pulse energy using a single gas fill over 41 h.

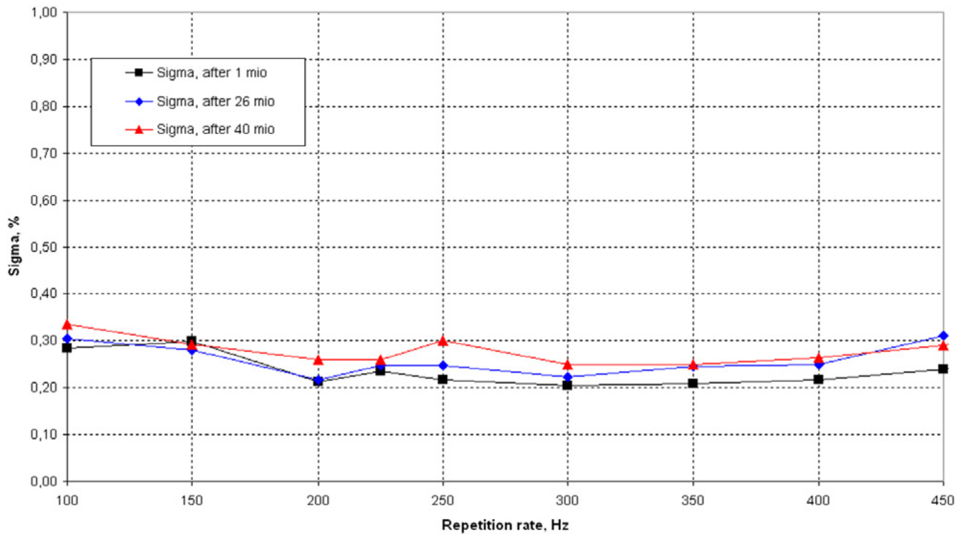

Figure 9. Pulse-to-pulse stability versus pulse count and repetition rate measured within a gas fill run.

achieved, the deposited volume per unit time scales with the number of generated parallel beamlets and material plumes, respectively. In fact, multi-plume deposition is routinely used in coated conductor manufacturing [9]. High-power excimer lasers provide sufficient output energy in order to be distributed over 6 or more laser spots at typically employed fluences of some J cm^{-2}. Figure 8 shows a gas life run of a 308 nm excimer laser stabilized at 1 J pulse energy at 500 Hz pulse frequency over a period of more than 41 h or 75 million laser pulses.

In order for industrial PLD processing not only to be fast but also to achieve high production yield, it is imperative that the pulse-to-pulse energy distribution remains unchanged during every gas life run.

A measurement of the pulse-to-pulse variations over the course of 40 million pulses for a 500 W excimer laser operated at 1 J stabilized output energy at a wavelength of 308 nm is shown in figure 9. Only a marginal increase in the standard deviation is observed, as the gas is constantly refreshed with about 50 on-the-fly micro-injections of halogen gas during every gas life run.

On-the-fly halogen gas injections are unnoticeable in the optical performance of the excimer laser and have become a standard feature in the gas fill management of industrial high-power lasers. They require the use of single gas bottles and extend the dynamic gas lifetime, i.e. the number of achievable pulses from a single gas fill by about a factor of three.

Moreover, active tube temperature control of $\pm 0.2\,^{\circ}$C as well as externally mounted, nitrogen purged resonator optics which are mechanically and thermally decoupled from the laser tube are common features in high power mass production excimer lasers adding to the steadily improving long-term performance characteristics of industrial high power lasers.

3.3. Capitalizing on pulse frequency

In addition to utilizing an excess of laser pulse energy for the generation of multiple beams, i.e. generating multiple material plumes, pulse frequency is the key parameter for production throughput upscaling. On the basis of sophisticated beam scanning designs which avoid pulse overlapping effects during target ablation and ensure homogeneous layer growth, manufacturers can capitalize on the high laser pulse frequency without sacrificing PLD thin-film quality [10].

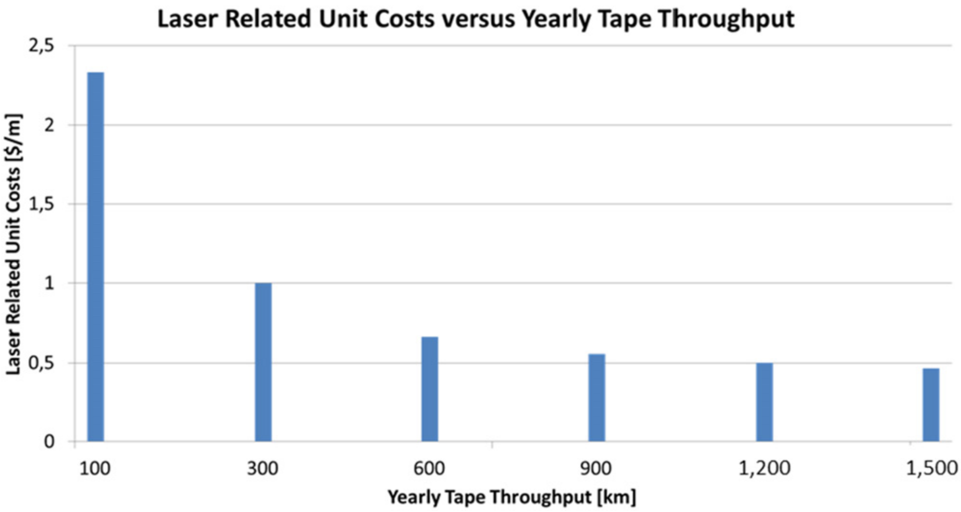

Figure 10. Laser related unit costs versus annual coated conductor production.

Coated conductor manufacturers in the pre-production phase utilize about 15–20% of the excimer laser output power available today, reporting PLD throughput rates between 30 and 60 m h^{-1} for depositing the superconducting REBCO layer with a thickness of ∼1 μm on ∼5 mm wide tapes. Accordingly, a throughput of ∼270 m h^{-1} for a 540 W laser used at full power can be extrapolated.

3.4. Coated conductor laser costs projection

Under the assumption of a two-shift operation of 16 h each day and a typical 95% uptime, using the 600 Hz repetition rate, a 540 W high power production excimer laser delivers about 10 billion laser pulses over the course of 5500 h per year. The running costs of a 308 nm high power excimer laser including gas consumption amount to typically 50$ per million laser pulses.

PLD is one of the rate limiting steps in coated conductor manufacturing and tape throughput is driven largely by the available laser power. In order to fully utilize the laser power, it has to be converted into maximum deposition speed and tape feed rate. This is generally achieved via multi-plume formation and parallel tape winding. With ideal laser beam utilization the production capacity limit of a 540 W excimer laser comes close to ∼1500 km per year of high-temperature superconducting tape.

The total 540 W excimer laser costs per metre of tape (both capital costs and running costs over a period of seven years) determined for increasing annual tape production levels which, in a sense, correspond to the experience curve of a laser manufacturer, are given in figure 10. The total 540 W excimer laser costs per metre of tape (both capital costs and running costs over a period of seven years) determined for increasing annual tape production levels which, in a sense, reflect the experience curve of a laser manufacturer, are given in figure 10. As coated conductor tape sells currently with an average price tag of about 50$/m, the projected excimer laser related production costs per metre of tape become as low as 1% of the average per metre sales price.

4. Conclusion and outlook

After 25 years PLD has matured and continues to be a thriving area of material research performed mostly on ∼1 cm^2 size substrates and at less than 10 Hz excimer laser pulse frequency in a growing number of research labs.

Enabled by concurring advances both in excimer laser power and long-term performance and in vacuum deposition technology, off-the-shelf, large-scale pulsed laser deposition solutions are now available opening the path to broader adoption of PLD in large-scale applications. Intelligently capitalizing on multi-hundred watts of average UV power together with evolving setup designs will drive throughput advances in industrial PLD use.

Ultimately, highest coated conductor feed rates and lowest laser related unit costs will be achieved via throughput leveraging multi-plume deposition architectures tapping the full UV power available from state-of-the art excimer lasers.

References

[1] Dijkkamp D, Venkatesan T, Wu X D, Shareen S A, Jiswari N, Min-Lee Y H, McLean W L and Croft M 1987 *Appl. Phys. Lett.* **51** 619–21

[2] Delmdahl R 2010 *Nature Photon.* **4** 286

[3] Krueger R R, Rabinowitz Y S and Binder P S 2010 *J. Refract. Surg.* **26** 749–60

[4] Uhrmann T and Delmdahl R 2013 *Solid State Technol.* **56** 18–20

[5] Paetzel R, Turk B, Brune J, Govorkov S and Simon F 2008 *Phys. Status Solidi* c **10** 3215–20

[6] Herbst L, Lindner H, Heglin M and Hoult T 2004 *Indust. Laser Solutions* **19** 13–16

[7] Delmdahl R and Pätzel R 2008 *Appl. Phys.* A **93** 611–5

[8] Koster G, Rijnders G J H M, Blank D H A and Rogalla H 1999 *Appl. Phys. Lett.* **74** 3729–31

[9] Usoskin A, Kirchhoff L, Knoke J, Prause B, Rutt A, Selskij V and Farrell D E 2007 *IEEE Trans. Appl. Supercond.* **17** 3235–8

[10] Greer J 2006 *Pulsed Laser Deposition of Thin Films: Applications-Led Growth of Functional Materials* ed R Eason (Hoboken, NJ: Wiley-Interscience) pp 191–213