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Article

Supplementary

Control of Lateral Composition Distribution in Partitioned Dual-Beam Pulsed Laser Deposition

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Model for Simulation

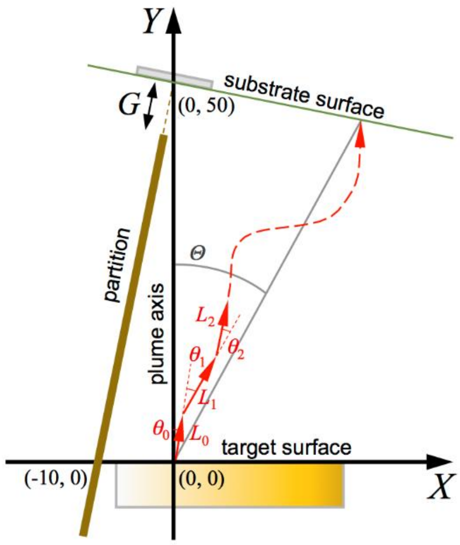

Figure S1: Coordinate system used for the simulation of trajectories of ejected particles from a single target. The point of coordinate (X, Y) = (0, 0) represents the laser spot on the target surface. (0, 50) corresponds to the center of the substrate.

In order to figure out the thickness distribution obtained in the single target ablation experiment of CeO_2 (Section 3.1), the trajectories of particles ablated from a single target to reach the substrate surface under various conditions (pressure p_{O2} and gap G) were simulated by a Monte Carlo method. Figure S1 shows the coordinate system used for the calculation. Expecting qualitative results, we constructed a primitive model to represent the ablation process on the basis of assumptions described below.

1) In the two-dimensional (X, Y) space (Figure S1), the particles ejected from the point (0, 0) on the target surface repeatedly experience elastic collisions with the ambient gas molecules until reaching the substrate surface ($Y = X/5 + 50$).

2) The path length between collisions, L_n, and the change in the direction caused by the collision, θ_n, obey normal distributions [S1]. Here, the subscript *n* indicates the number of collision, except $n = 0$ that denotes the ejection from the target.
3) The path length between the ejecting point (0, 0) to the first collision point, L_0, is 1.5 mm in average [S2]. The standard deviation for L_n is L_n / 5 for all *n* including 0.
4) The distribution of the ejection angle θ_0 is centered on the target normal, with a standard deviation $\sigma_{\theta 0}$ of 11.4° [S3].
5) After the first collision ($n \geq 1$), collisions with ambient gas molecules dominate the movement of the particles. Mean free path in air is adopted as the averaged path length between collisions, L_n ($n \geq 1$). L_n (in mm) is obtained from the pressure p_{O2} (in Torr) using

$$\log L_n = a \log p_{O2} + b \tag{1}$$

where *a* and *b* were taken as -1 and -1.3 [S4].
6) The distribution of θ_n ($n \geq 1$) is with the mean of 0° and the standard deviation $\sigma_{\theta n}$ of 6° [S5].
7) For the particle that falls behind the target surface or that collides with the partition, the calculation is stopped at that moment and is not counted into the results.

The *X* coordinate of each particle at the instant it arrived at the substrate's surface was recorded. For each combination of p_{O2} and *G*, the calculation was done for 6×10^6 ejected particles to obtain the thickness distribution.

References

1 A number of studies have reported that the thickness distribution of conventional PLD processes (without partition) is fitted with $\cos^N \theta$ [S6–S8]. In the present simulation, however, $\cos^N \theta$ is approximated by normal distribution in order to simplify the calculation.

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3 A previous study on the film thickness distribution in ablation from an oxide MO_x (M: Al, Hf, Y) target has revealed that *N* depends on the pressure *p*, the atomic weight *m* of the metallic element M, and the direction of distribution (whether it is in parallel or perpendicular to the longitudinal axis of the laser spot) [S8]. In the case of Ce ($m = 140$ u), in parallel with the longitudinal axis of the laser spot, and *p* of zero, we estimate *N* to be 25 on the basis of the results in ref.[S8]. The normal distribution with $\sigma_{\theta 0}$ of 11.4 gives the same full width at half maximum as $\cos^{25} \theta$.

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5 The $\sigma_{\theta n}$ of 6° is estimated from the maximum scattering angle of a Ce atom (140 u) that elastically collides with a stationary O_2 molecule (32 u), which is calculated to be 13.2°.

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