Comment on "Isotope Enrichment in Laser-Ablation Plumes and Commensurately Deposited Thin Films"

In a recent Letter [1], Pronko *et al.* reported discovery of a highly efficient isotope enrichment process directly in laser-ablation plumes generated from ultrafast laser pulses focused on solid targets. We point out that the plasma centrifuge mechanism proposed in this work is not consistent with some of the experimental observations. Taken together with an earlier observation of isotopic enrichment in laser produced plasmas at much lower laser intensities [2], we emphasize the need to look for a new mechanism.

Gupta *et al.* [2] had reported isotopic enhancement in plasma plumes of boron oxide produced by Nd:glass laser pulses of 30 ns duration at an intensity of $\approx 10^{10}$ W cm^{-2}. The ratios of B_{10}/B_{11} in ion signals of an electrostatic analyzer in their experiment were anomalously large, and no definite mechanism could adequately explain the observed behavior. The new observations [1] on isotopic ion ratios in plasma plumes of boron nitride using ultrafast laser pulses (200 fs) at much higher intensities are surprisingly similar to those reported earlier with nanosecond pulses [2].

Pronko *et al.* have supported their observations of isotopic enhancement from spatial variation of B_{10}/B_{11} ratio in a thin film deposited from the plasma plume, and invoked plasma centrifuge mechanism [3] to explain their results. In a plasma centrifuge, radial separation between elements of different masses occurs due to bulk rotation of the plasma in an axial magnetic field [3]. However, data in Ref. [1] does not imply any significant rotation. For typical velocity of boron ions in the range of $(2-3) \times 10^7$ cm/s, time of flight to the deposition site for the ions would be $\approx$300 ns. Using a rotation rate of 3.3×10^5 rad/s stated in their work, only a rotation of 0.1 rad would occur in this time. Even for an order of magnitude larger rotation speed (corresponding to the spatially averaged axial magnetic field of 44 kG), the rotation would be only 1.2 rad. Moreover, this value of self-generated axial magnetic field is itself an overestimate as it is obtained by extrapolating the magnetic field data in Ref. [4] to 3D expansion region up to 7 cm using a power law dependence on distance which is determined only for 1D expansion region.

Next, some experimental observations reported in Ref. [1] are also in disagreement with the implications of isotope enrichment by plasma centrifuge mechanism. The B_{10}/B_{11} ratio in the deposited film showed a maximum value of 0.45 at the center of the plasma plume, which then decreased towards the plume edge. However, the ratio remained the same (0.29) at positions 4 and 5. If the plasma centrifuge mechanism were operative, one would have expected the ratio to be not only monotonically decreasing as one moves towards the plume edge, but more importantly to have become smaller than the natural abundance ratio of 0.25 at the plume edge. Further, the observed B_{10}/B_{11} ratio was highest for $q = +3$ species in comparison to those for $q = +4$ and $+5$ along the target normal. If the plasma plume were rotating, maximum isotopic ratio should have occurred for the highest charge species, which is not the case.

In view of the above, plasma centrifuge mechanism does not appear to be adequate for the observation of isotopic enhancement in Ref. [1]. Nevertheless, the observations are of considerable interest. Since laser-plasma interaction at high intensities using femtosecond laser pulses can be quite different from nanosecond pulse regime in terms of ionization equilibrium, an interplay of ionization and recombination processes occurring during heating and hydrodynamic expansion phases of the plasma plume needs to be examined to understand the reported observations.

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