ISOTOPICALLY ENRICHED FILMS AND NANOSTRUCTURES
BY ULTRAFAST PULSED LASER DEPOSITION

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I. Project definition:

This project involved a systematic study to apply newly discovered isotopic enrichment effects in laser ablation plumes to the fabrication of isotopically engineered thin films, superlattices, and nanostructures. The approach to this program involved using ultrafast lasers as a method for generating ablated plasmas that have preferentially structured isotopic content in the body of the ablation plasma plumes. In examining these results we have attempted to interpret the observations in terms of a plasma centrifuge process that is driven by the internal electro-magnetic fields of the plasma itself.

The research plan involved studying the following phenomena in regard to the ablation plume and the isotopic mass distribution within it:

1. Test basic equations of steady state centrifugal motion in the ablation plasma.
2. Investigate angular distribution of ions in the ablation plasmas.
3. Examine interactions of plasma ions with self-generated magnetic fields.
4. Investigate ion to neutral ratios in the ablation plasmas.
5. Test concepts of plasma pumping.
6. Fabricate isotopically enriched nanostructures.

Items 1, 2, 3, and 5 were accomplished in considerable detail, while items 4 and 6 were investigated in a preliminary fashion. Additional work is required on these later two items.

II. Results of the Research

Based on the results, as presented in the three annual reports to this project, we are able to present here the following conclusions:

1. It is possible to interpret the angular distributions of the ablated ionic isotopes in terms of a magnetic centrifuge model.
2. Secondary pulses similar to the initial femtosecond pulse, but time delayed by tens of picoseconds, can be used to increase the ionic content of the plume and to also increase the isotopic enrichment of the lighter isotope in the center of the plume.
3. Deposition of isotopically enriched thin films is made complicated by the presence of nanoclusters in the plume. These clusters do not exhibit the isotopic enrichment effect and interfere with the deposition of enriched films.
4. It is possible to greatly reduce the presence of the nanoclusters in the plume by using secondary pulses delayed on the nanosecond time scale.
Selected Results of Research

We compared the isotope enrichment on axis for BN ablation plumes initiated by a single 120-fs pulse having 2.2 kJ/cm² fluence to those initiated and pumped by two pulses of half that energy fluence each and separated by 10 picoseconds. In Fig. 1, the normalized ratio of B¹⁰ to B¹¹ is shown for the two cases, as a function of charge state. The double-pulse case shows an enhancement of enrichment for all charge states. For charge state +2, it is as high as 59% over the single pulse case. Based on a weighted average, an enhancement of 26% is observed for the total ionic enrichment. Further studies of this double-pulse pumping of the plasma plume are in progress.

In Figures 2 and 3 are shown the isotopic mass enrichment and the chemical/mass enrichment of Nickel isotopes and a 50/50 alloy of Nickel and Copper. The scans are performed as a function of angle with respect to the center of the ablation plume. These results are consistent with a self-generated magnetic centrifuge process as described in the second annual report. This is especially true for the chemical/mass data when compared to similar experiments done with mechanically wound magnetic centrifuges. Figures 4 through 6 relate to experimental results obtained on the nanocluster formation observed in these femtosecond ablation plumes. A typical size distribution is presented along with results from double pulse experiments showing how a second femtosecond pulse can break up these clusters. Figure 5 demonstrates that an increase in ion and excited neutrals in the plasma accompany the breakup.
Figure 2: Ni$^{58}$ / Ni$^{60}$ isotope enrichment as a function of charge state and angle in the ablation plume. Charge states $+1$ through $+10$ are shown as gray bars, with the highest enrichment labeled. The total enrichment ratio for all ions is plotted as circles. (The enrichment ratio is normalized to the natural abundances: 68% Ni$^{58}$ / 26% Ni$^{60}$.)

Figure 3: Ni / Cu element separation as a function of charge state and angle. Charge states $+1$ through $+10$ are shown as gray bars, with the highest enrichment labeled. The total enrichment ratio for all ions is plotted as circles. (The enrichment ratio is normalized to the natural abundances: 45% Ni / 55% Cu.)
Figure 4: Size distribution of aggregates after deposition on an Si substrate and fit to a log-normal law.

Figure 5: Evolution of the ion and neutral signals as a function of delay between pulses; perturbation in the grey zone is not described and will be discussed at another time.
Figure 6: Evolution of the aggregate IR optical emission strength as a function of delay between pulses. Maximum decrease rate occurs at ~850 ps. Notice that the cluster signal increases after 1.5 ns whereas the ion and neutral signals are tending toward their non-perturbed zero delay level. It may be that the concentration (i.e. volume density) of clusters goes below a critical level after this time in addition to the plasma becoming too diffuse for efficient interaction to occur at this delay.

Conclusion

Future work in this area should include attempts to understand the cluster formation process and how it can be either eliminated or controlled. We did some work on this using a multi-pulse technique. Experimental work of our own as well as that of others suggests that the clusters are formed by condensation out of the gas phase. Therefore controlling the plasma temperature during the expansion process is one possible way of affecting this phenomenon. Since the isotopic enrichment process in the plume resides in the ions, methods for increasing the ionic content over the neutral or cluster content will help to enhance the process.

Publications:


Conference Proceedings:

