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TITLE: **HIGH IONIZATION EFFICIENCY TECHNIQUES FOR CW RIMS**

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High Ionization Efficiency Techniques for CW RIMS

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ABSTRACT: The demand to measure high dynamic range isotope ratios on small samples with RIMS continues to increase. This paper discusses high ionization efficiency methods which can be applied to CW RIMS to potentially achieve several tens of percent ionization efficiencies for certain elements. The primary technique under development to achieve this is an external laser cavity which can generate very high circulating laser powers.

1. INTRODUCTION

The ability to measure large isotope ratios for rare isotopes with small samples continues to increase (Hurst 1986, Miller *et al.* 1986, and Blum *et al.* 1990). The major problem which has limited the accomplishment of this task has been to achieve high overall ionization efficiency with good sample utilization.

Some aspects of this problem are apparent in the choice of excitation and ionization source for the RIMS process. While pulsed lasers have high peak powers and broad tuning ranges, several characteristics limit their general utility: low duty cycle (low efficiency) [largely solved by kHz repetition rates], pulse pile-up difficulties (limited dynamic range), poor stability (poor precision), and laser pulse spectral and temporal concerns (ratio biases). In contrast, CW lasers offer 100% effective duty cycles, easily controlled laser profiles (spectral, spatial and temporal), as well as excellent power stability. The major obstacle which has precluded the utility of CW lasers has been power. While sufficient intensity is available to saturate the resonant transition, efficient promotion of the excited atoms above the ionization potential is difficult.

High dynamic range measurements ($\leq$1:10^5) have been demonstrated previously with CW RIMS , primarily through its high duty cycle and power stability versus pulsed sources (Miller and Nogar 1983, Miller *et al.* 1985). We have also demonstrated that a secondary, non-resonant laser can substantially increase ionization to yield efficiencies of ~0.1% in lutetium (Fearey *et al.*, 1989). However, further increases through more powerful lasers are not available.

Therefore, our primary effort is to significantly increase signal levels through an external cavity technique which has the potential to achieve overall ionization efficiencies of tens of percent for several elements (*e.g.*, Th). Such a technique is anticipated to yield a 100-fold increase in coherent circulating laser power. This paper will focus on the theoretical and experimental aspects of such a system, as well as some of our most recent results.

2. EXPERIMENTAL

Our general CW RIMS experimental setup has been described previously (Fearey *et al.* 1988). Specifically, the external cavity RIMS set-up consist of an Ar^+ laser pumped standing wave dye laser coupled into the external laser cavity which will be constructed around a 90° single sector magnetic mass spectrometer (see Figure 1). The external cavity consists of a 98% reflective input coupler mirror, a ~100% high reflector, a polarization selective element and active-feedback stabilization lock electronics. The stabilization scheme (Hansch and Couillaud 1980) consists of a piezo-electrically driven high reflector mirror whose drive voltage is generated from an error signal derived from the polarization dependent element. The purpose of the stabilization circuits is to maintain a phase matching condition within the cavity such that the external cavity has maximum gain (*i.e.*, the length is $n*\lambda/2$).

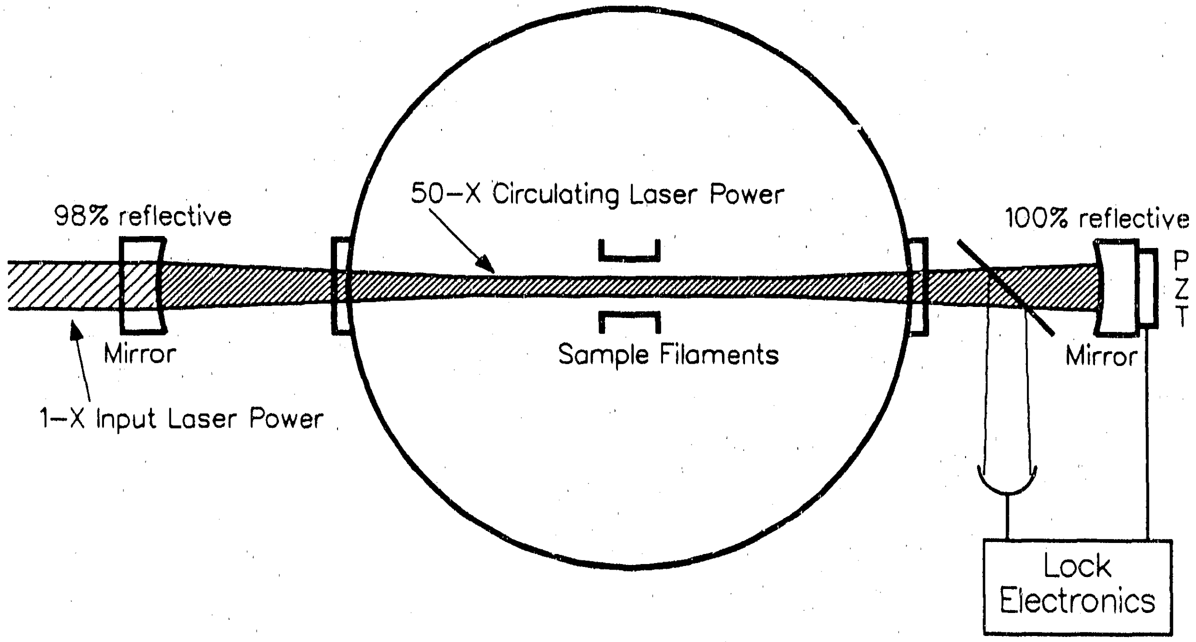

Fig. 1. Mass spectrometer source and external laser cavity configuration to be used in RIMS experiments. With mirror reflectivities shown, a 50-fold circulating laser power increase over the input laser power is calculated.

3. BASIC PRINCIPALS

Although the basic theory in the properties of external laser cavities has been thoroughly discussed before (Hansch and Couillaud 1980 and Helmcke *et al.* 1977), it is perhaps useful to briefly discuss some of the important basic principals and properties involved. The essential gain function can be described as follows: if the reflectivity of the input coupler is R, and the round-trip attenuation is V, the enhancement factor, E, of the input power by the cavity is given by: $E = (1 - R)/(1 - sqrt(RV))^2$. Upon examination, the maximum enhancement occurs when the input coupler reflectivity exactly matches the round-trip attenuation (*i.e.*, $R = V$, such that $E = (1-\mathrm{R})^{-1}$).

In addition, several other factors must be controlled and optimized. These include the external cavity length, mode matching to the cavity, and variable beam waists (Corney 1977). Importantly, the capability to vary beam waists allows the optimization of the cavity gain with the atom reservoir for maximum efficiency.

4. RESULTS

The experimental apparatus is in the construction and testing stage. The present external laser cavity (in its simplest form) consists of two spherical mirrors (100% high reflector and a 98% reflective input coupler). With optimal conditions and different reflectivity mirrors, a ≥100-fold increase in circulating power can be achieved (Berquist 1988).

We have recently completed prototype testing of the cavity in an unstabilized configuration (*i.e.*, without feedback electronics). Under these conditions, we have demonstrated extremely promising values of an average gain factor of ~20 (corresponding to 4 watts of circulating laser power) and a peak (instantaneous) enhancement of ~45-fold (9 watts). In an active feedback stabilization set-up, we expect to be able to match or exceed the peak enhancement factor, approaching the theoretical value of ~50-fold increase in input laser power. In addition, the importance of mode matching and variable external cavity beam waists has been verified.

The actively-stabilized cavity configuration is presently in testing. It has been shown in our bench-top experiments that the PZT motion is intimately involved in the gain (as expected). Our first-stage electronics have been shown to be performing essentially as expected. The more comprehensive second-stage electronics are presently in fabrication. An actively phase-locked cavity should be operational in the immediate future.

5. DISCUSSION

The potential for a two order of magnitude (or more) gain in the circulating laser power and the corresponding increase in ionization efficiency make the external laser cavity approach to RIMS extremely attractive. Figure 2 shows theoretical modeling calculations (similar to Miller and Nogar 1983) of the relative RIMS signal enhancement (ionization efficiency) under various conditions. The plots show the dependence of relative ionization efficiency as a function of laser beam diameter. The lower curve shows this dependence for a single dye laser with 200 mW of power. The other three curves indicate conditions where a 50-fold gain external cavity is utilized for both/either the dye laser and/or argon ion laser.

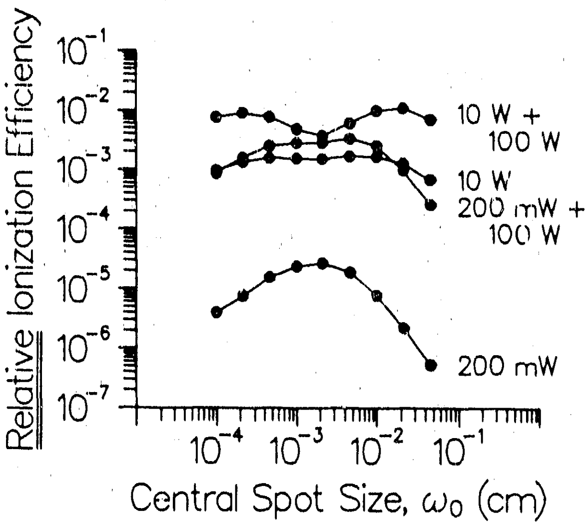

Fig. 2. Detailed calculation of relative ionization efficiencies as a function of beam diameter with differing dye and argon ion laser powers.

Amplification of a single laser leads to a ≥60-fold improvement in the ionization efficiency. That the dependence on laser power is greater than linear, points to increased sample utilization or, equivalently, ionization volume. Figure 3 shows a calculational comparison of the increase in sample utilization between without and with the benefit of an external cavity.

From these calculations, the promising results on our unstabilized external cavity and with our current ionization efficiency of ~1% for Th using simple CW RIMS (described elsewhere in this volume (Fearey *et al.* 1990) — the utilization of the external cavity could potentially lead to overall ionization efficiencies for Th of several <u>tens of percent</u>. This increased efficiency should allow for sub-nanogram isotopic ratio analyses of thorium at the 5:10^6 level ($^{230/232}$Th) which are critical for internal isochron measurements important for geochronology (Fearey *et al.* 1990).

Finally, a second laser, such as an Ar^+ ion laser, can also be used for the ionization step in a 1+1 RIMS scheme (Fearey *et al.* 1989). This second laser can also be amplified via another external cavity which then allows for even further increases in the the volume of atoms addressed and hence, ionization efficiency. Such increases could lead to ionization efficiencies ≥2.5 orders of magnitude (see Fig. 2) greater than single-pass dye configurations. As laser powers continue to increase, saturation conditions will become important; however, this is not necessarily a limitation because the laser beam diameter can be increased. Obviously, there will be some optimum combination of laser power and beam diameter.

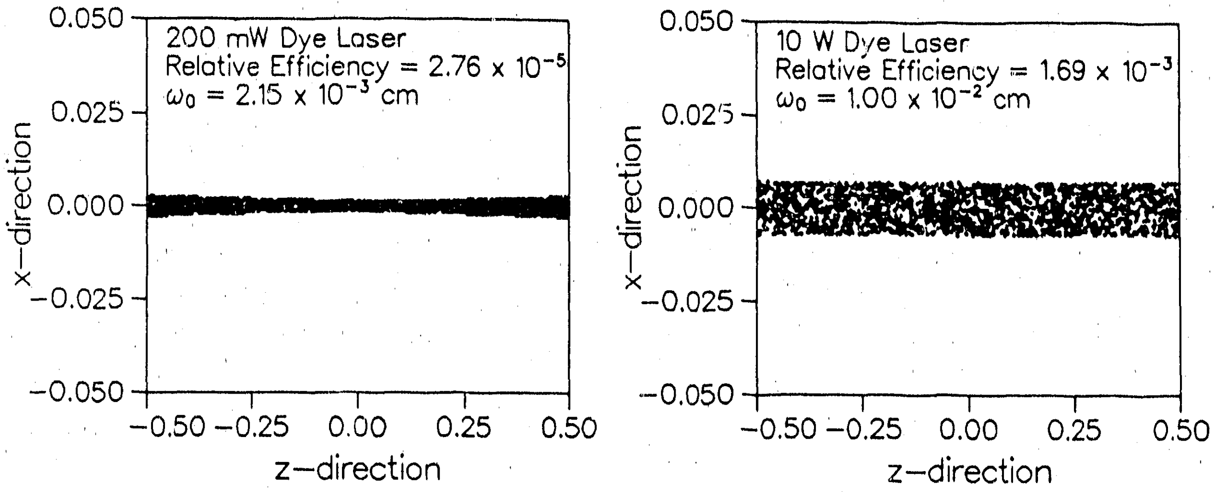

Fig. 3. Ionization calculation showing the comparison in ionization volume both without and with the operation of the external cavity. The x-direction is across the sample filament (cross-section of laser beam) and the z-direction is along the active length of the filament (along the laser beam).

6. SUMMARY

Theoretical calculations and preliminary data have been presented that demonstrate that the utilization of an external laser cavity to boost the circulating laser power leads to substantially increased ionization efficiencies for CW RIMS. An average gain of ~20-fold and an instantaneous gain of ~45-fold have been observed. A gain factor of 100 is expected for our final configuration utilizing a 99% reflective input coupler. Incorporation of the external cavity technique into CW RIMS should lead to ionization efficiencies as high as tens of percent for certain elements. This will largely remove present limitations of CW lasers for RIMS research and analysis.

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