LASER SPECTROSCOPY OF NUCLEAR REACTION PRODUCTS:
RECENT RESULTS AND FUTURE PROSPECTS

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Laser spectroscopic observations of nuclear reaction products produced with intensities of less than 10\textsuperscript{4} atoms/second are now possible with several different methods. We describe the recoil into gas method which has recently been successful. This method is not Doppler-free, but can give reasonable spectra if the resolution requirements of the spectra are not too high. It has the great advantage that it very efficiently uses the atoms, and spectra have been observed with primary production rates of less than 10\textsuperscript{3} atoms/sec. Our recent work has concentrated on developing the recoil into gas method for the refractory element Hf. In order that the atoms could be cycled to produce many fluorescence photons, nitrogen and hydrogen impurity gases were added to the argon buffer gas to quench metastable levels to the ground state. In this way spectra could be obtained with fluxes of 10\textsuperscript{4} atoms/second. Future prospects for trapping radioactive atoms in a magneto-optic trap will be discussed.

1. Introduction

The sensitivity of laser spectroscopy [1] has allowed detailed measurements of exotic nuclei that are very far from stability. The systematic studies that have been done over long chains of isotopes have significantly aided in our understanding of nuclear structure. There are remaining unexplored areas which will require further developments in experimental technique in order to study in detail. These developments proceed along several lines. More intense sources of unstable nuclei are being considered at various facilities, but in addition specific methods to circumvent some of the limitations of the standard collinear apparatus are being studied.

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The new developments of state selective ion and atom detection with the collinear technique have been very well covered by the previous paper of Dr. Lievens, and we wish to focus on an old method which has recently had some success, namely the recoil into gas method. The recent work is now well documented in the literature [2,3], so that we will only outline the method, and then point out some of the specific aspects which are often not communicated well in formal publications.

2. Recoil into gas method

The basic idea of the recoil into gas method is very simple in concept, but requires extreme care in practice. An inert buffer gas is used to stop nuclear reaction products in the vicinity of a laser beam which then induces fluorescence in the atoms. The buffer gas serves to retard the diffusion of the atoms to the walls, so that multiple excitation cycles can increase the fluorescence yield from a single atom. The motivation for using this Doppler limited method is that the atoms can be efficiently deposited in less than $10^{-8}$ s into the laser beam without any consideration of melting point or volatility. However, questions of background suppression, neutralization, and diffusion times must be addressed to successfully observe the fluorescence.

The target arrangement that we used is shown in fig. 1. Because heavy ion fusion evaporation reactions are quite selective, an isotope separator was not necessary, and this allows for efficient use of the limited number of atoms. The fusion products from the reaction are carried out of the target by the momentum of the beam particle. Because of multiple Coulomb interactions in the target, the slow recoil nuclei are scattered to much larger angles than the fast beam particles, and an angular separation can be made. If additional separation is needed, Au foils can be added to spread out the recoil nuclei to larger angles. The thickness of material in the target, gas windows and buffer gas must be carefully adjusted to maximize the concentration of recoil nuclei in the vicinity of the laser beam. Small adjustments in the gas are usually made to optimize the signal. The pressure chosen for the
buffer gas results from a compromise between having a high pressure to maximize the recoil atom concentration, and having a low pressure to minimize the pressure broadening of the atomic transitions. Cooling the buffer gas helps to narrow the Doppler broadening, and the foil thickness and gas pressure are usually adjusted so that the contribution from the pressure broadening is comparable to the Doppler broadening. For Yb in argon at 100 K, this results in a pressure of 10 Torr. The second reason to cool the buffer gas is to increase the diffusion time of the atoms to the walls of the cell. Figure 2 shows the complete arrangement, with the laser beam and detection optics.

The motivation for returning to the recoil into gas method, which had been tried earlier by several groups with little success, has been for several reasons. First, the signal available from the standard collinear method is usually about one photon per atom, and this is further reduced by a factor of 100 to about 1/100 count per atom because of quantum efficiency and solid angle. State selective atom detection has removed this second reduction factor. If an atom has a transition which can be repetitively cycled with a time $t_{\text{cycle}}$ and the atom can be observed for a time $t_{\text{observation}}$, then the number of counts can be increased by the ratio $t_{\text{observation}}/t_{\text{cycle}}$. The observation time for the recoil into gas method is limited by the diffusion time of the implanted atom to the wall of the cell, and this time is typically 0.5 sec. Since atomic cycling times are usually between 0.01-1 $\mu$sec, large signal enhancements can be obtained. However, atoms often have "trap" states or optical pumping which limit the number of cycles before the atom is no longer optically active.

Another potential advantage of recoiling the atoms into gas is that the time from the nuclear reaction until the atoms are in the laser beam can be as short as $10^{-8}$ seconds. This allows for the possibility of detecting laser fluorescence or optical pumping of short lived nuclear isomers. However, the neutralization time is not known well under most circumstances.

![Fig. 2. Schematic view of experimental apparatus.](image-url)
3. Experimental results

Fig. 3. shows the results obtained for the $N = 82$ isotope, $^{152}\text{Yb}$, and other heavier isotopes. The standard interpretation of isotope shifts attributes the shift to a combination of the size of the proton distribution and the deformation of the distribution. Observation of the $N = 82$ isotope, which should be very close to spherical because of the closed neutron shell, was important to be able to separate the two effects.

![Fluorescence spectra of very light radioactive Yb isotopes obtained with the recoil into gas method. Spectra were obtained with the order of $10^3$ atoms/sec created in the target. (Note that the usual rate is quoted in a secondary beam.)](image)

The element Yb was chosen for the first studies because of its simple atomic spectrum, but the real motivation for developing the recoil into gas method was to be able to measure isotope shifts of the refractory elements such as hafnium which cannot be quickly released from available ion sources. Our first attempts to observe hafnium fluorescence spectra were not successful, even though we tried many different laser transitions. The atomic spectrum of hafnium, and indeed of all of the refractory elements is very complex, with many levels below 18,000 cm$^{-1}$. A closed excitation-decay system cannot be found with available lasers, and there always
exist decay branches which can trap the atom in a non-fluorescing state. Excitation
to the $^3D_2$ 16163 cm$^{-1}$ level with 618 nm laser light, with detection of the 724 nm
decay branch to the $^3F_3$ state at 2356 cm$^{-1}$ allowed observation of a very weak
resonance. The strength of the resonance was increased by three orders of magnitude
by the addition of mTorr amounts of H$_2$ and N$_2$ gases to the argon buffer gas. These
molecular gases have energy levels which are near-resonant with excited states of
the hafnium atom, and allow for efficient depopulation of the "trap" states back to
the ground state. The stable isotopes of Hf were accelerated to energies of 30 MeV
in our tandem Van de Graaf and implanted into the gas cell, and isotope shifts and
hyperfine structure were measured in order to calibrate the atomic parameters of the
transition used. Fig. 4. shows fluorescence spectra obtained with the even stable
isotopes. With the optimal gas mixture, the sensitivity of the Hf system is about a
factor of 10 less than Yb, mainly because of the fragmentation of the atomic

![Figure 4](image-url)

Fig. 4. Fluorescence spectra of stable even isotopes of Hf.
transition strength over many levels. Experiments with radioactive isotopes will therefore require more intensity than is available at the Stony Brook Superconducting LINAC.

4. New prospects

A new activity in our laboratory is focused on the physics which can be studied with trapped radioactive atoms. Once a sample of radioactive atoms can be routinely put into an atom trap and cooled to microkelvin temperatures, then many experimental investigations become possible:

Parity non-conservation: The beautiful experiments on parity non-conservation in the Cs atom of Wieman et al. [4] could be extended to radioactive Cs isotopes in order to decrease the sensitivity of the measurements to the unavoidable uncertainties in the atomic calculations [5], or they could be performed on the francium atom, where the size of the parity non-conserving signal would be a factor of 15 larger. Before this latter experiment could be meaningful, however, a systematic study of the francium atomic properties must be made, in order to test carefully the atomic calculations.

Neutrino recoil experiments: The atoms in a magneto-optic trap are very well localized, and at a very low temperature, so that the recoil of the atoms from emission of a neutrino might be able to be measured. For example, in a pure electron capture process, the momentum of the atom is equal and opposite to the momentum of the neutrino. Small differences in the momentum could be observed if a neutrino with different mass were sometimes emitted. Very careful experimental design would require to distinguish between other effects such as Auger electron and X-ray emission from the daughter atom.

Decay anisotropy measurements: Atomic traps allow for an extremely high degree of nuclear polarization, which could be utilized to make fundamental measurements of the anisotropy of radiations emitted in nuclear decays.

A magneto-optic trap [6] is currently under construction with the aim to trap radioactive atoms for measurements of the type discussed above. We are implementing the trap with diode lasers for Rb, and we are developing the lasers for Fr. The work of Professor Shimizu presented at this conference has shown that in addition to alkali atoms, rare gas metastables and alkaline earth elements can also be trapped, so that a broad range of nuclear species could be studied.

The process for producing trapped atoms from nuclear reaction products can be broken down into the following steps. A projectile–target combination must be chosen with a nuclear reaction which makes copious amounts of the activity of interest. The choice of target is coupled with the question of diffusion and release from the surface of the alkali atoms of interest. We have chosen a hot (1000 K) target to produce and slow the reaction products, and diffusion out of the target is
increased by the high temperature. A hot tube coated with yttrium surrounding the
target serves to neutralize any ions. The tube serves to produce an atomic beam
which is not collimated. Recent techniques for transverse cooling of neutral atoms
in an atomic beam will be used in order to increase the luminosity of the beam. A
Magneto-Optic Trap (MOT) with a very wide capture range will be used to first
capture the radioactive atoms. This trap is able to cool the atoms to temperatures
of a fraction of a degree kelvin, but that is still too hot for our purposes. A cooling
procedure using the optical molasses techniques should allow reaching micro kelvin
temperatures, with a corresponding increase in the density to that obtained in very
dense atomic beams (optical density higher than 3). However, the presence of the
laser would disturb the atoms for careful measurement. The trapped atoms could be
optically pumped to align the nuclear spins and then transported to a purely magnetic
trap where the measurements could be performed without the presence of the laser
beam. Systematic checks could be implemented with electric and magnetic fields
as well as with polarization to look for the parity non-conservation transitions and
also for other asymmetry measurements.

It is important to stress the power of this method that allows the trapping of
neutral atoms for fractions of minutes at very high densities and with minimal
Doppler broadening. New possibilities of high precision spectroscopy are now becoming
available as a result of the recent advances in neutral atom cooling and trapping.

The goal of the present program is to develop the methods to preserve the
precious population of radioactive atoms from production to measurement. We are
using a simulated reaction product beam of natural Rb isotopes produced in the
sputter ion source of our accelerator and accelerated to reaction product energies
of 30 MeV. In this way intensities of $10^9$/sec can be easily obtained. When the
system is developed, then radioactive Rb isotopes can be produced with intensities
of $10^5$/sec. We have also produced $^{212}$Fr by the reaction $^{205}$Tl($^{12}$C,5n) with 90 MeV
$^{12}$C from the Stony Brook Superconducting LINAC, and identified the reaction
products with alpha spectroscopy. Production rates for $^{212}$Fr of $10^5$/sec are also
achievable with the Stony Brook LINAC, but precision measurements may require
location of the experiment at a more intense source such as the proposed ISOLAB
facility or at other facilities.

References