Radioactive noble gas detection
STL-007-21, Year 1 of 2

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Challenge

- We wish to demonstrate the feasibility of atomic hyperfine spectroscopy as a method for detecting and quantifying radioactive xenon, krypton, and argon produced by nuclear activities, such as underground testing, reactor operations, fuel reprocessing, etc.

- Optical atomic hyperfine spectroscopy can detect radioactive atoms before they decay; thus, “dark” (i.e., undecayed) atoms in a sample can be readily probed. A sensitivity enhancement of several orders of magnitude over traditional nuclear spectroscopy becomes possible.

- The hyperfine technique may be able to complement the IAEA’s global monitoring network, and it should be possible to deploy a portable apparatus closer to sensitive areas for more sensitive rapid monitoring.

Innovation

Enhancement from Sampling the "Dark" Population

The exponential decay law relates source activity to source population:

$$\frac{dN}{dt} = -\Gamma N \qquad Activity = \Gamma = ln(2)/T_{1/2}$$

$$\frac{DarkPopulation}{DecaySignal} = \frac{N_0}{N} = \frac{1}{1 - e^{-\Gamma \Delta t}} \approx \frac{T_{1/2}}{\Delta t ln(2)}$$

- For an 8-hour nuclear measurement of ^{85}Kr ($t_{1/2}$ = 10.756 yrs), this formula yields a maximum enhancement (Dark/Decay) of about 10^4. If the dark population can be sampled in minutes instead of hours, then the information is available much sooner.

- For xenon isotopes, the maximum enhancement is not as dramatic, because the 8-hour nuclear measurement time is nearly as long as the half-life (^{135}Xe ~9 hrs). Nevertheless, the decay daughters can also be probed by laser excitation, e.g., ^{85}Rb at 780 nm (from Kr), and ^{133}Cs and ^{135}Cs at 852 nm (from Xe), and they, too, have hyperfine structure.

- Actual enhancement values must account for the efficiencies and branching ratios of both the nuclear and atomic detection methods.

Innovation

Radioactive Xe and Kr Are Red Flags of Nuclear Activity

Table 1. Isotopic abundance and shift relative to ^{132}Xe for the Xe I $6s\ ^2[1/2]^0_1 \rightarrow 6\ ^2[3/2]_2$ transition at 834.682 nm (from Suzuki *et al.*[25]). No isotopic shift values are available for ^{124}Xe and ^{126}Xe.

Mass (amu)	124	126	128	129	130	131	132	134	136
Abundance (%)	0.0096	0.009	1.92	26.4	4.1	21.1	26.9	10.4	8.9
Shift (MHz)	—	—	84.1	96.5	37.1	72.8	0.0	-38.9	-116.9

Smith et al., IEPC-2007-229 (2007).

Isotope	Energy	Half-life	Decay	Daughter	Spin	Nuc Moment
^{131m}Xe	163.930(8)	11.934(21) d	IT	Xe	11/2−	
^{133}Xe	132.9059107(26)	5.2475(5) d	β−	Cs	3/2+	0.813
^{133m}Xe	233.221(18)	2.19(1) d	IT	Xe	11/2−	
^{135}Xe	134.907227(5)	9.14(2) h	β−	Cs	3/2+	0.903

The half-life of ^{85}Kr is 10.756 years, so it is less compelling of a red flag, because a lot has built up in the atmosphere since 1945. However, if a downwind concentration surge is observed somewhere, then it would be a useful indicator (incidentally, so would detection of radioactive ^{41}Ar, $t_{1/2}$ ~ 110 min).

Technical Approach

In atomic physics, fine structure is messy enough, but hyperfine structure takes it to a new level. The spin and deformation of the nucleus introduce new structure to atomic orbitals from the magnetic dipole and electric quadrupole interactions. The effects are small but measurable.

Orbital energies are typically measured in eV (8065.5 cm^{-1}), while hyperfine structure separations are usually measured in MHz (0.000033 cm^{-1})

$$E_{HFS} = \frac{A}{2}C + BD$$

Level	E_i (eV)	A_{129} (MHz)	A_{131} (MHz)	B_{131} (MHz)
$6s\ ^2[1/2]_1^0$	9.56972	−5801.1	1713.7	24.5
$6s\ ^2[3/2]_2$	11.0547	−2892	831.0	26.8

Smith et al., IEPC-2007-229 (2007).

Magnetic Dipole

$$A = -\left(\frac{\mu_I}{I}\right)\frac{B_J \cdot J}{J \cdot J}$$

$$C = F(F+1) - I(I+1) - J(J+1)$$

Electric Quadrupole

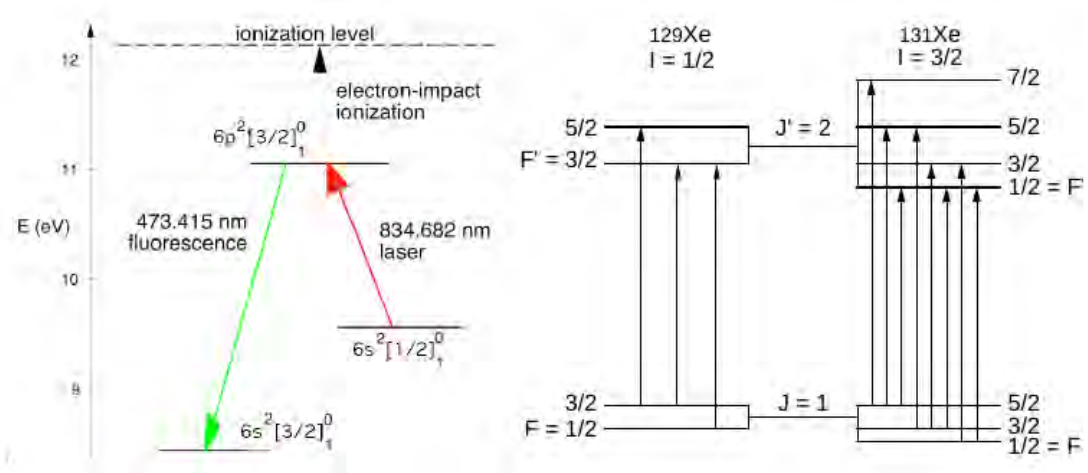
$$B = e^2 q_I Q$$

$$D = \frac{3(I \cdot J)^2 + \frac{3}{2}I \cdot J - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

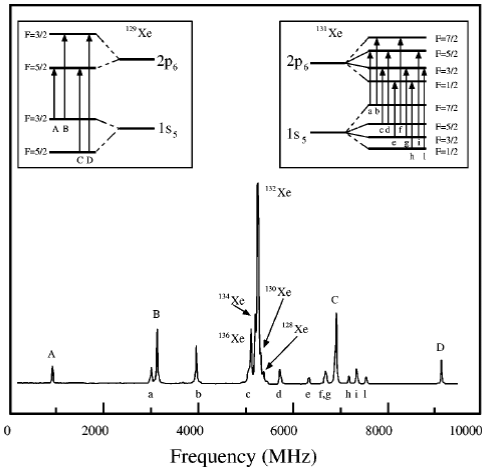
Technical Approach

Excitations from Various 5p → 6s Configurations Are Possible

The $5p^6$ ground state of Xe is too tightly bound (12.1 eV) for direct laser excitation, so electron impact excitation, or electron capture of the ion, is required as a first step; then laser excitation to a higher excited state becomes feasible.



Smith et al., IEPC-2007-229 (2007).



D'Amico et al., Hyperfine Interactions 127, 121 (2000).

840.919 nm : $5p^5(^2P°_{3/2})\ 6s\ ^2[3/2]°_2 \rightarrow 5p^5(^2P°_{3/2})\ 6p\ ^2[3/2]_1$ (Racah notation)

834.682 nm : $5p^5(^2P°_{1/2})\ 6s\ ^2[1/2]°_1 \rightarrow 5p^5(^2P°_{1/2})\ 6p\ ^2[3/2]_2$

828.012 nm: $5p^5(^2P°_{3/2})\ 6s\ ^2[3/2]°_1 \rightarrow 5p^5(^2P°_{3/2})\ 6p\ ^2[1/2]_0$

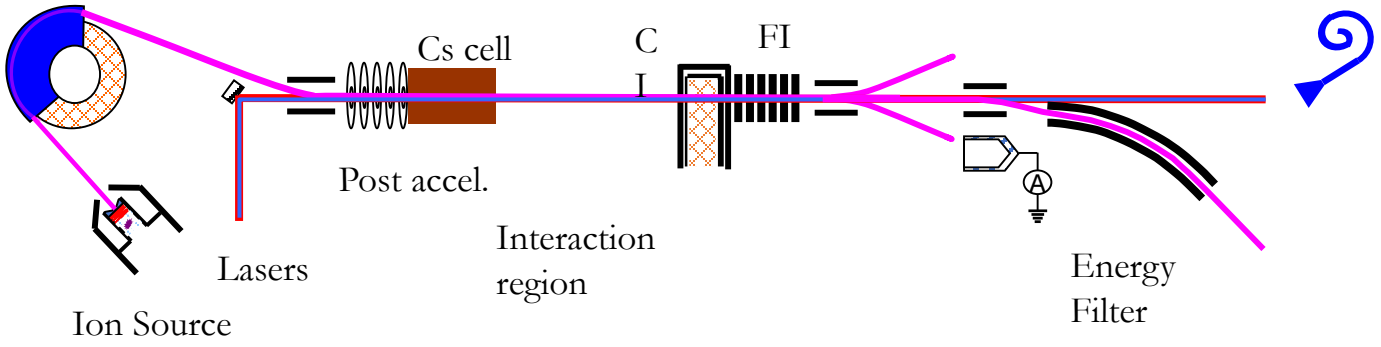
823.163 nm : $5p^5(^2P°_{3/2})\ 6s\ ^2[3/2]°_2 \rightarrow 5p^5(^2P°_{3/2})\ 6p\ ^2[3/2]_2$

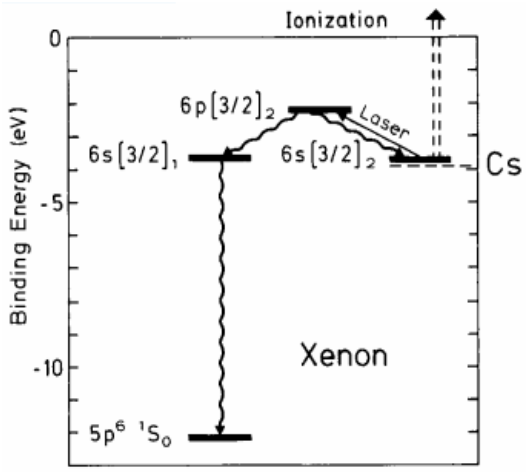
820.634 nm : $5p^5(^2P°_{1/2})\ 6s\ ^2[1/2]°_0 \rightarrow 5p^5(^2P°_{1/2})\ 6p\ ^2[3/2]_1$

etc.

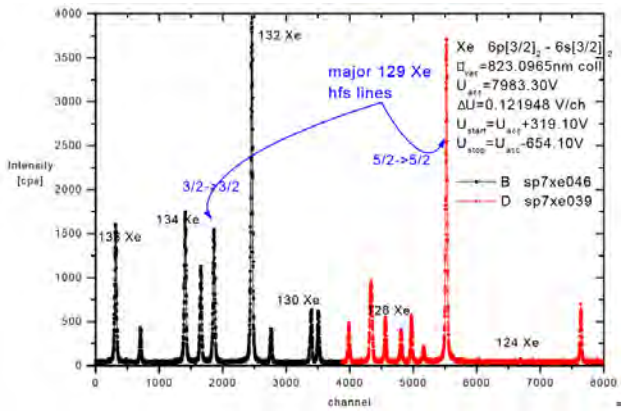
Technical Approach

Concept of Fast Ion Beam Laser Spectroscopy Experiment





Collisional ionization due to charge transfer from the Cs ground state



Hyperfine structure of Xe recorded by the particle detector

Technical Approach

- Numerous excited states can be laser probed for hyperfine structure, and the detection concept we envision is to drive a transition from a tightly bound state to a higher state that is more susceptible to impact or field ionization.

- Fluorescence and absorption measurements are other possibilities, but signal strengths are expected to be too small from real-world gas samples to be viable.

- To gain a better understanding of the various experimental options, we have begun ab initio calculations of some of these possibilities.

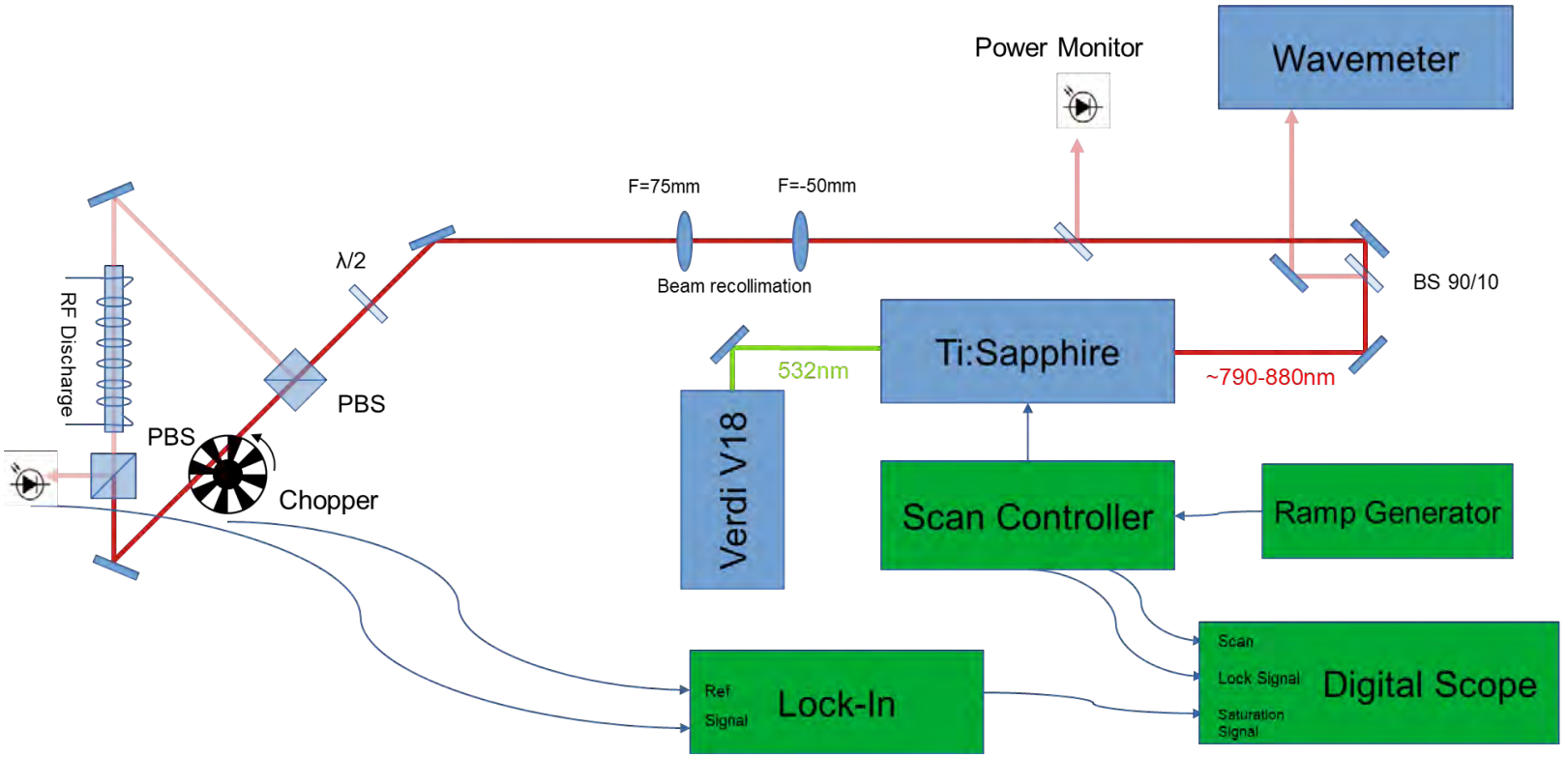
- The relativistic Dirac-Hartree-Fock codes GRASP2018 and AMBiT perform self-consistent field calculations, configuration interaction, and many body perturbation theory estimates of wave functions, energies, lifetimes, transition rates, isotope shifts, and hyperfine structure constants.

- We are using these codes to look for best options of atomic state transitions and to predict line structures. Thus far, we have been limited by computational resources and by divergences of the wave function calculations.

- We have recently (Aug. 2021) obtained access to LLNL's Quartz HPC server. We have compiled GRASP2018 on that platform and have started testing.

Technical Approach

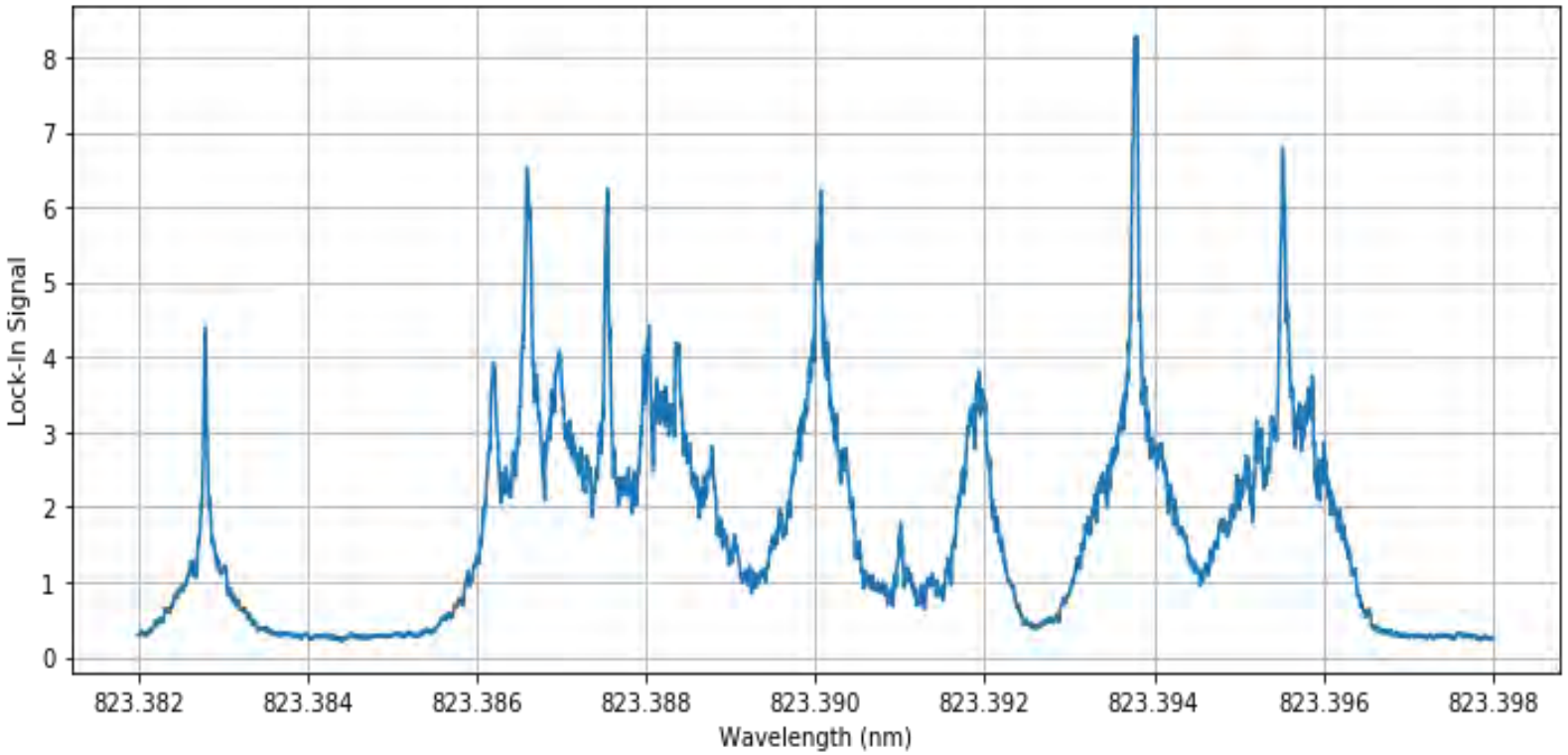
Setup for The Lamb dip measurements of Xe hfs



This discharge cell experiment will not be as clean as the fast beam experiment, but it is a much simpler approach that would be easier to implement in a portable apparatus.

Recent Results from Discharge Cell Experiment

$5p^5\,(^2P°_{3/2})\,6s\,{}^2[3/2]°_2 \rightarrow 5p^5\,(^2P°_{3/2})\,6p\,{}^2[3/2]_2$



Wavelength scale is approximate (from Burleigh wavemeter)

Single lines for ^{128}Xe,^{130}Xe,^{132}Xe,^{134}Xe,^{136}Xe
Three lines for ^{129}Xe
Eight lines for ^{131}Xe

Data from the fast beam apparatus will help sort this out.

Results

- In August 2021, we started getting data from the discharge cell.
- The fast beam experiment should begin at TAMU in Fall 2021.
- Consequently, we plan to bring further discharge cell work to STL.
- Calculations are now being done on the Quartz HPC server at LLNL.
- Those calculations are still being hindered by wave function divergences.
- If we manage to sort out the discharge cell data (including additional calculated radioactive isotopes and isomers), then we can begin to design a portable apparatus.
- We plan to publish our results in a refereed journal, and we will write an invention disclosure for the portable apparatus once it is designed.