Comparison of Model and Experimental Data for Resonance Ionization Mass Spectrometry of Uranium: Predicting and Guiding the RIMS Method

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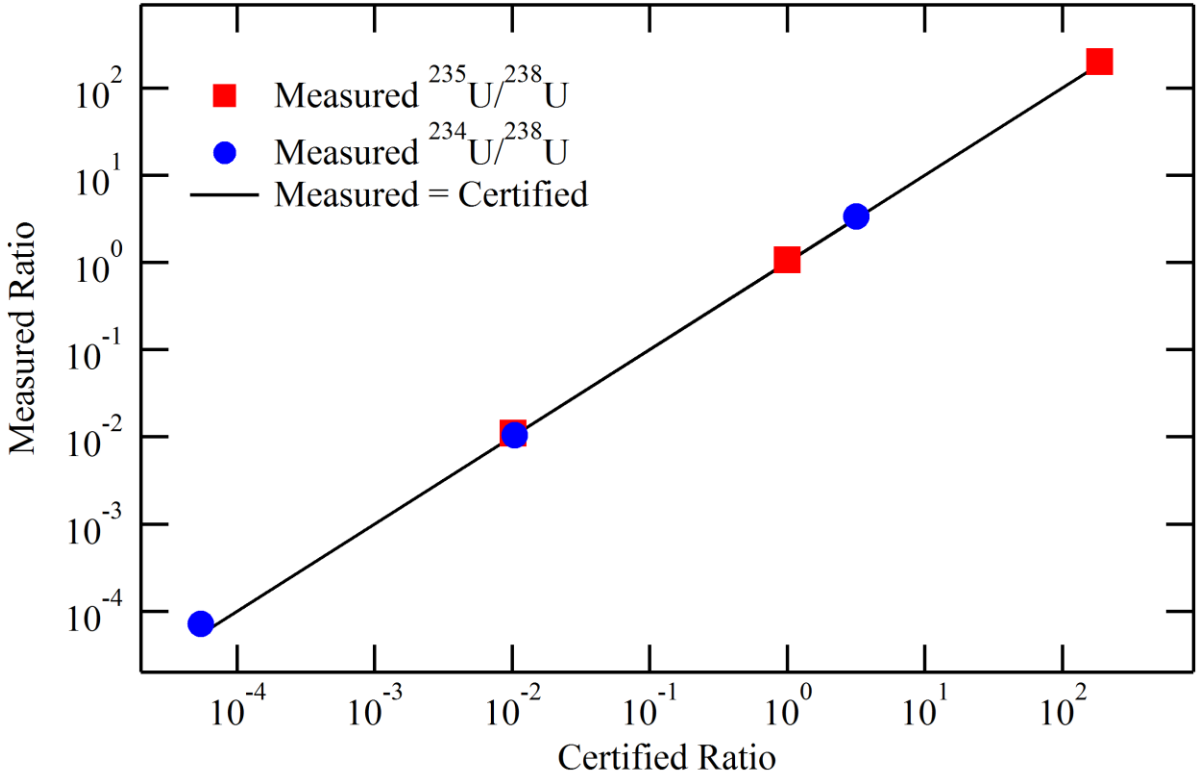
Motivation

Quantifying U isotope ratios using RIMS to <0.5% RSE is feasible.

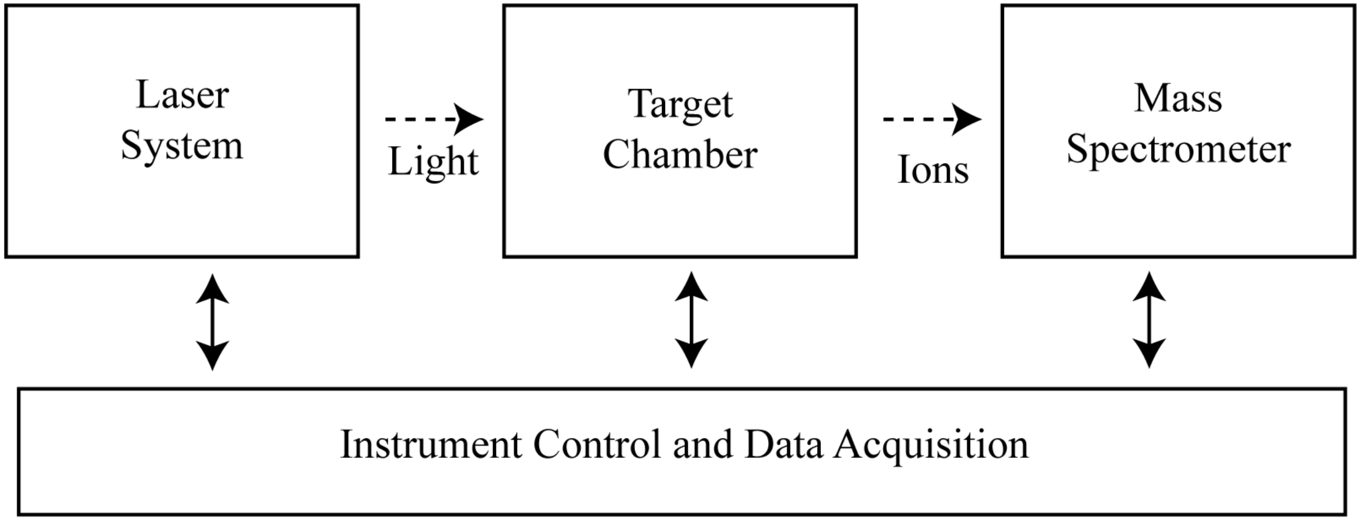


Measurements are highly dependent on the performance of the laser systems.

How good do the laser systems need to be to provide reliable measurements?

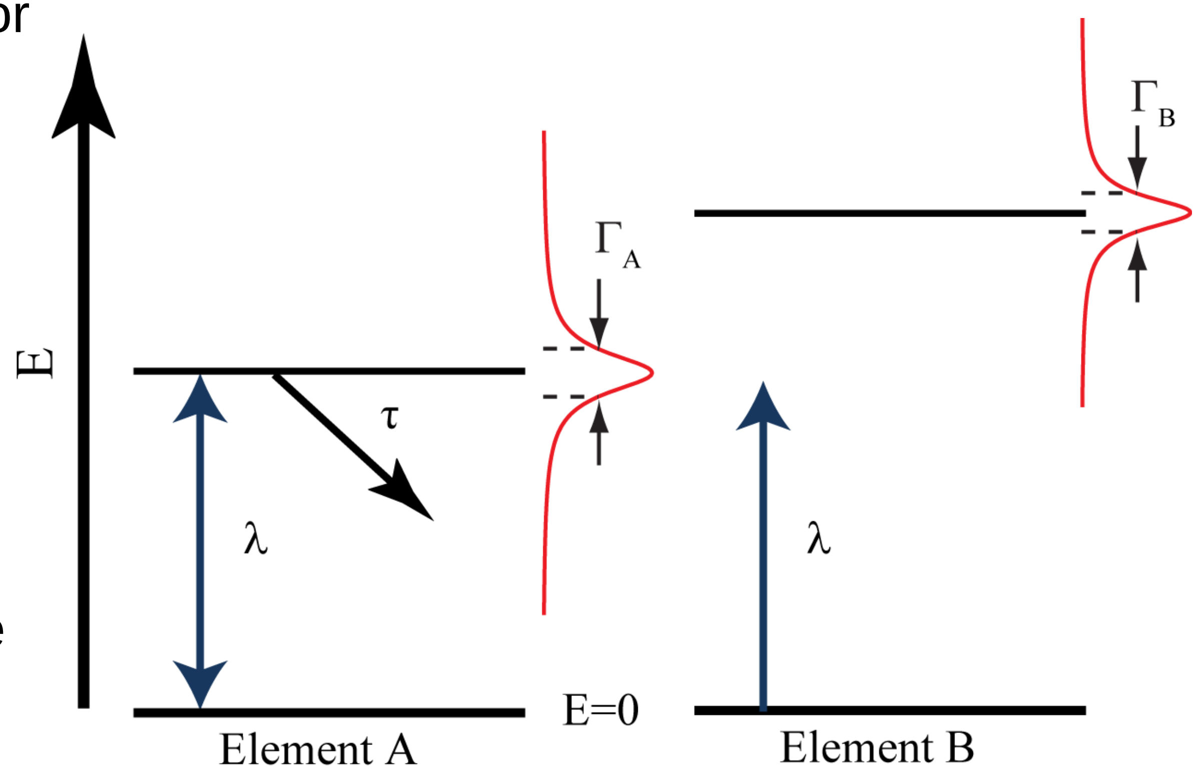
Instrument - CHARISMA

1. Generate neutral atoms in the gas phase.
2. Use resonance excitation to selectively ionize atoms.
3. Extract the ions into mass spectrometer for analysis.



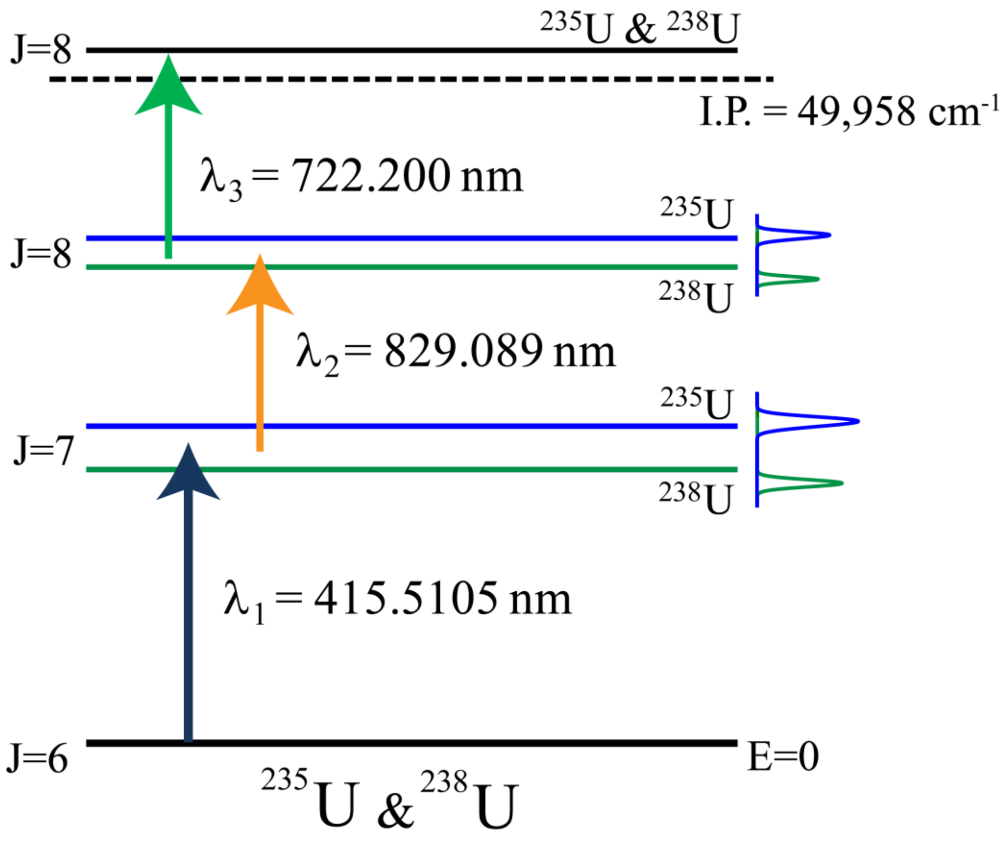
Resonance Ionization in RIMS

- Resonance: probability for finding excited state in energy.
- Stimulated Absorption, Stimulated Emission, Spontaneous Decay
- Differences in energy of excited states between elements.
- Atoms of element B have a very low excitation probability.



RIMS of Uranium

Uranium Ionization Scheme:

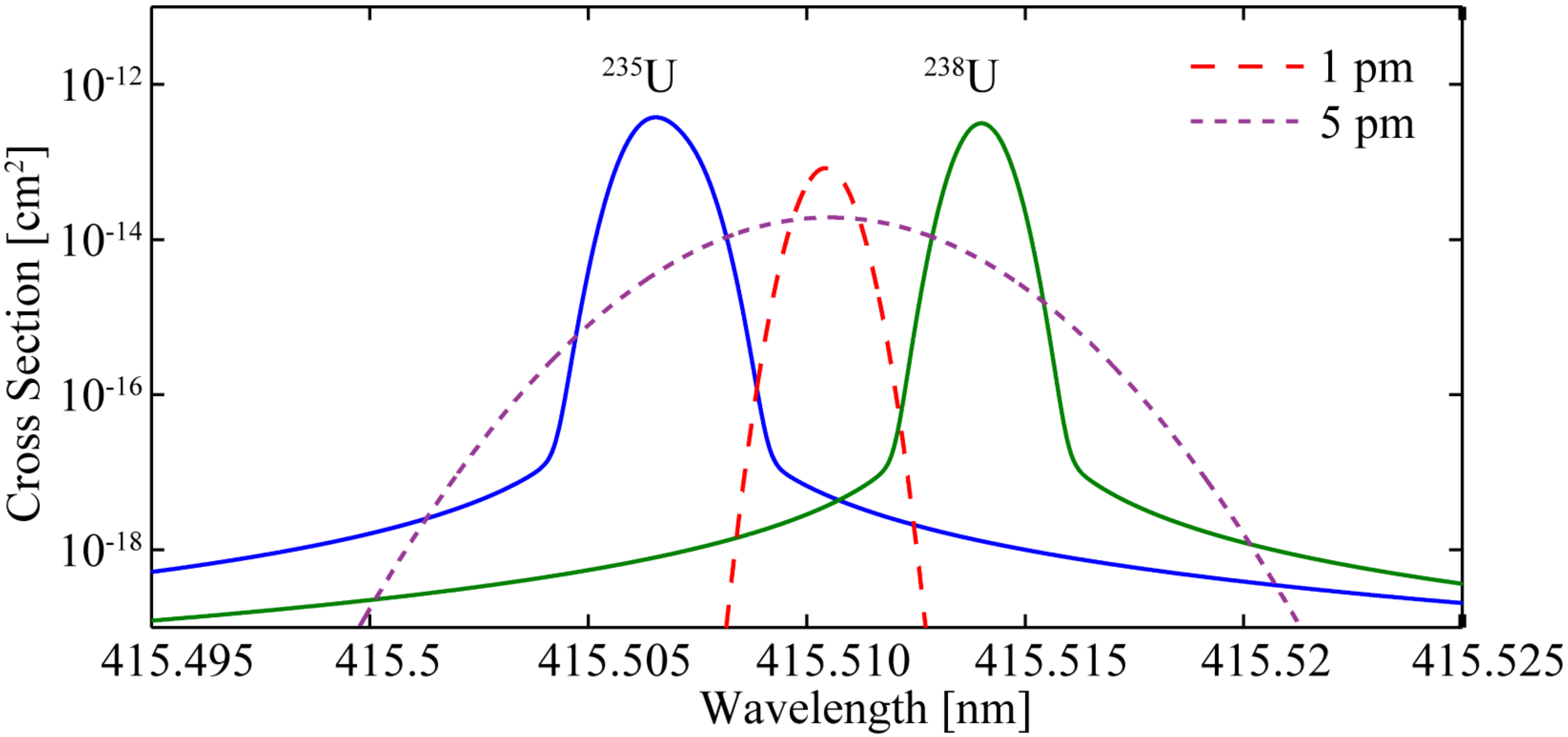


Differences Between Isotopes:

- Small Shifts in Energy
- Angular Momentum Differences

Excitation Using Broad Bandwidth Lasers

Atomic Resonances of ^{235}U and ^{238}U for First Transition



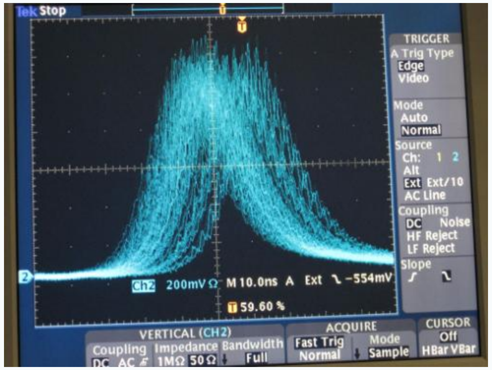
Broad bandwidth lasers can excite both resonances simultaneously.

Controlling Laser Performance

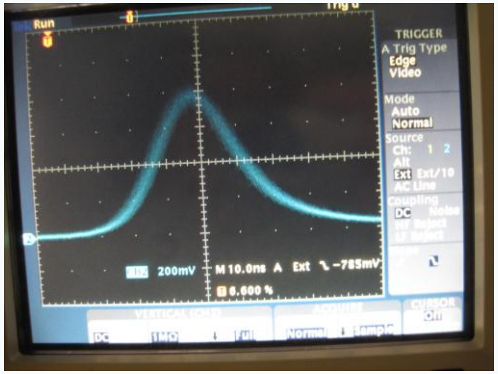
Real lasers vary in:

- Wavelength
- Position
- Power
- Time

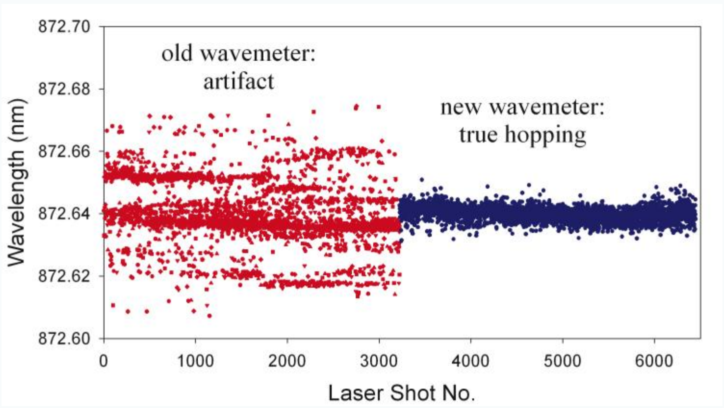
Time Variation from Pulse to Pulse:



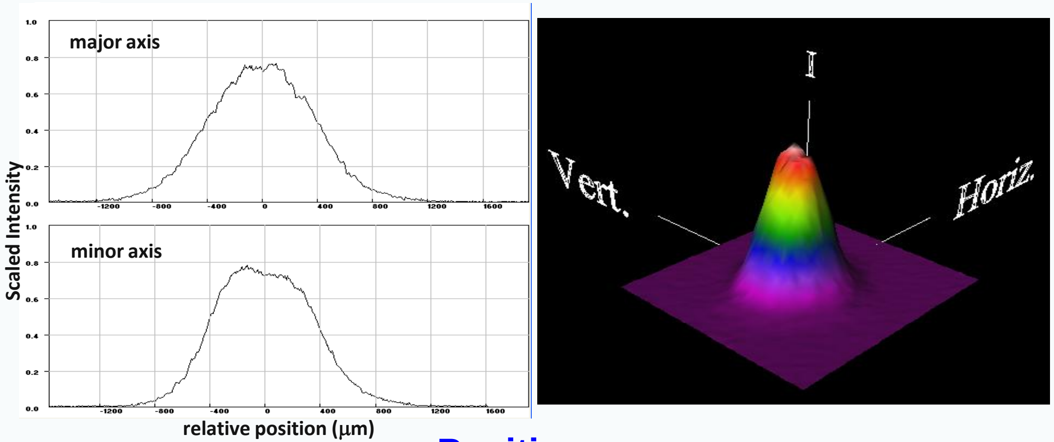
Old laser: 14 ns FWHM



New laser: 4 ns FWHM



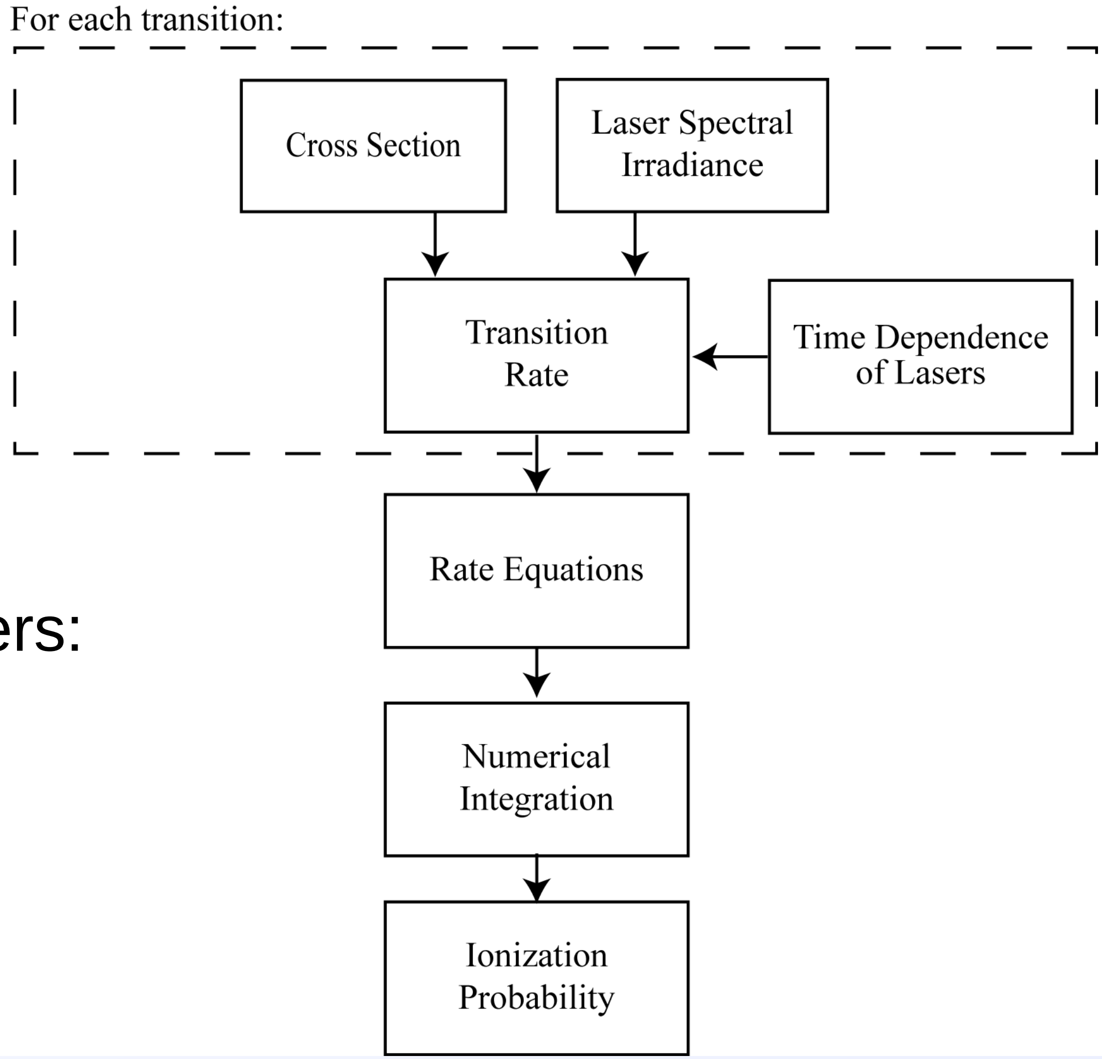
Wavelength Variation



Position

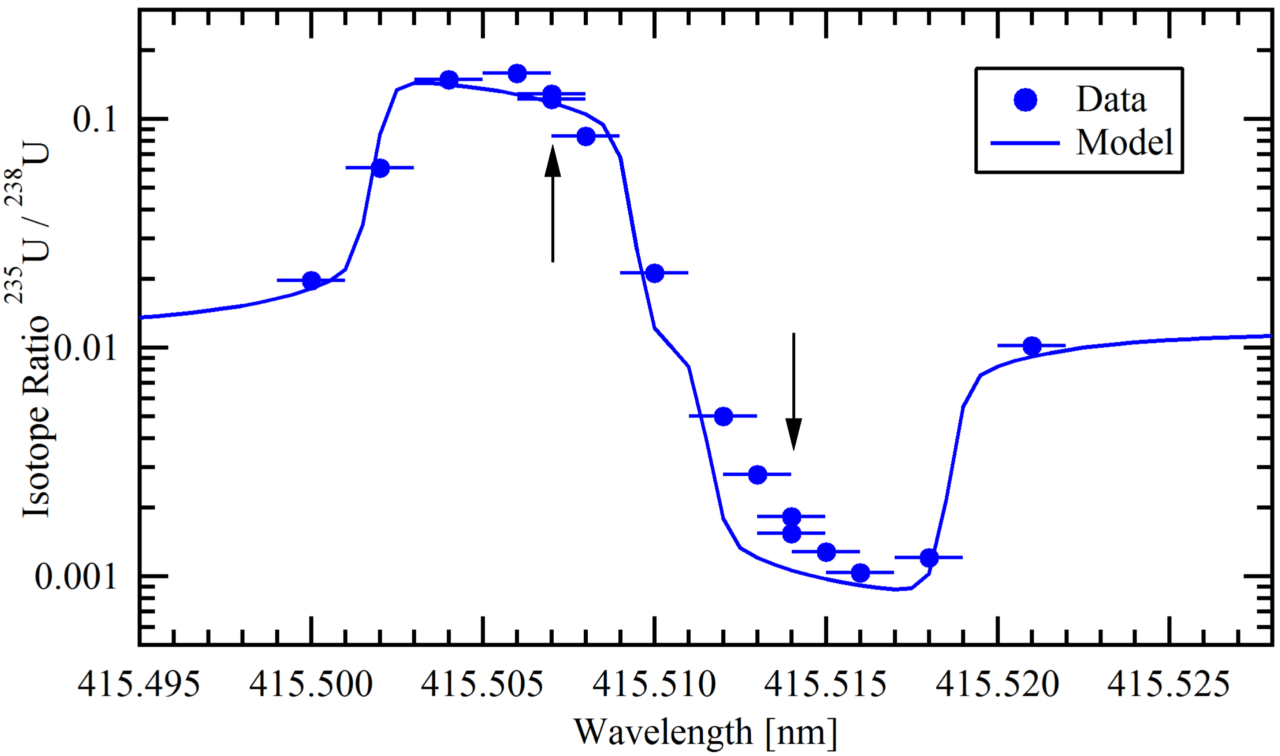
Model: Atomic Resonances and Laser Behavior

- Rate Equations:
  - Change in populations of excited states
- Atomic Cross Sections:
  - Lifetimes
  - Angular Momentum
  - Empirical Corrections
- Average Laser Parameters:
  - Wavelength
  - Bandwidth
  - Time Dependence
  - Intensity
  - Spatial Dependence



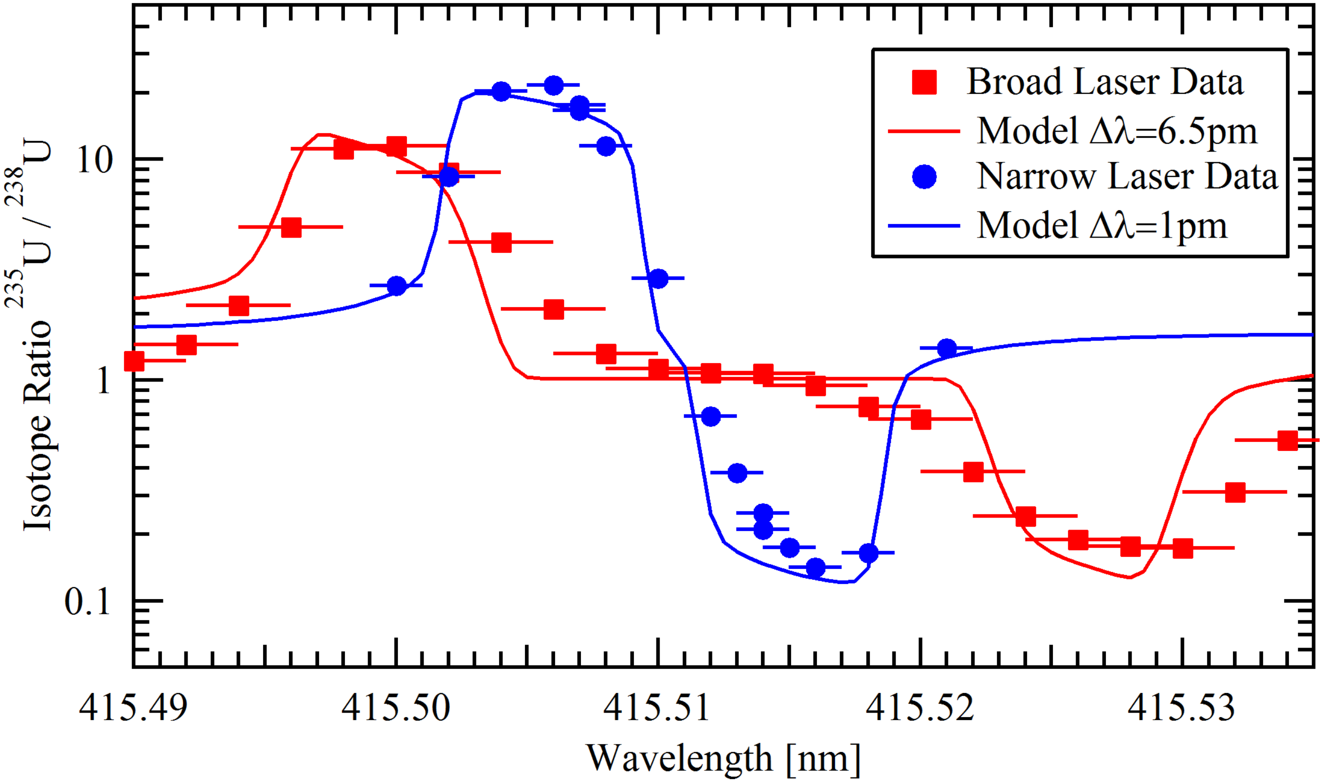
Model Prediction vs. Experimental Data

Variation of Measured Isotope Ratio as a Function of Laser Wavelength in the First Transition



Influence of Laser Bandwidth

Variation of Measured Isotope Ratio as a Function of Laser Wavelength in the First Transition for Two Laser Bandwidths



Conclusions

- Broad bandwidth laser beams are sufficient to saturate the first transition in ^{235}U and ^{238}U simultaneously.
- Variation of laser parameters over experiments does not prevent measurement of the $^{235}U/^{238}U$ isotope ratio.

Future Work:

- 3-D modeling of atom density and laser intensity is required for better description of experimental data.
- Stochastic variation of laser parameters will allow for non-linear effects present in experiments.
- Develop model to predict method performance for other isotopes and elements.