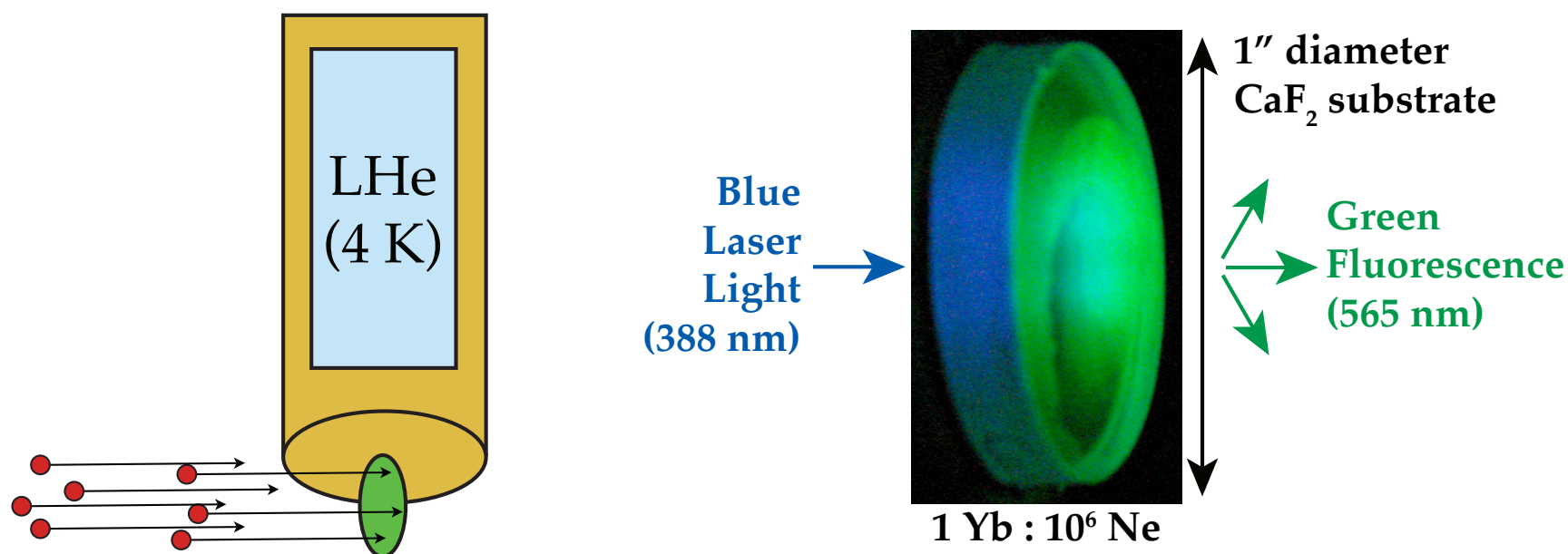


Progress Towards A Single Atom Microscope for Nuclear Astrophysics



spinlab.me

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DAMOP, June 3, 2020 @ 0800: G05.00001

U.S. National Science Foundation CAREER Award #1654610

"It is surprising to find an unstable element in the stars."



Technetium in the Stars

Paul W. Merrill

Mount Wilson and Palomar Observatories

Technetium, the first "artificial" element, was identified in 1937 by Perrier and Segrè in a piece of molybdenum that had been bombarded with neutrons in the cyclotron at Berkeley. Technetium has also been detected among the products of fission of heavy atoms. No completely stable isotope is known; the most nearly stable has a half-life less than a million years.

The spectrum of technetium was thoroughly investigated in 1950 by Meggers and Scribner at the National Bureau of Standards. Their work has made astronomical investigations possible. In 1951 Charlotte E. Moore announced the possible presence of weak lines of ionized technetium in the solar spectrum.

It is surprising to find an unstable element in the stars. Either (1) a stable isotope actually exists although not yet found on earth; or (2) S-type stars somehow produce technetium as they go along; or (3) S-type stars represent a comparatively transient phase of stellar existence.

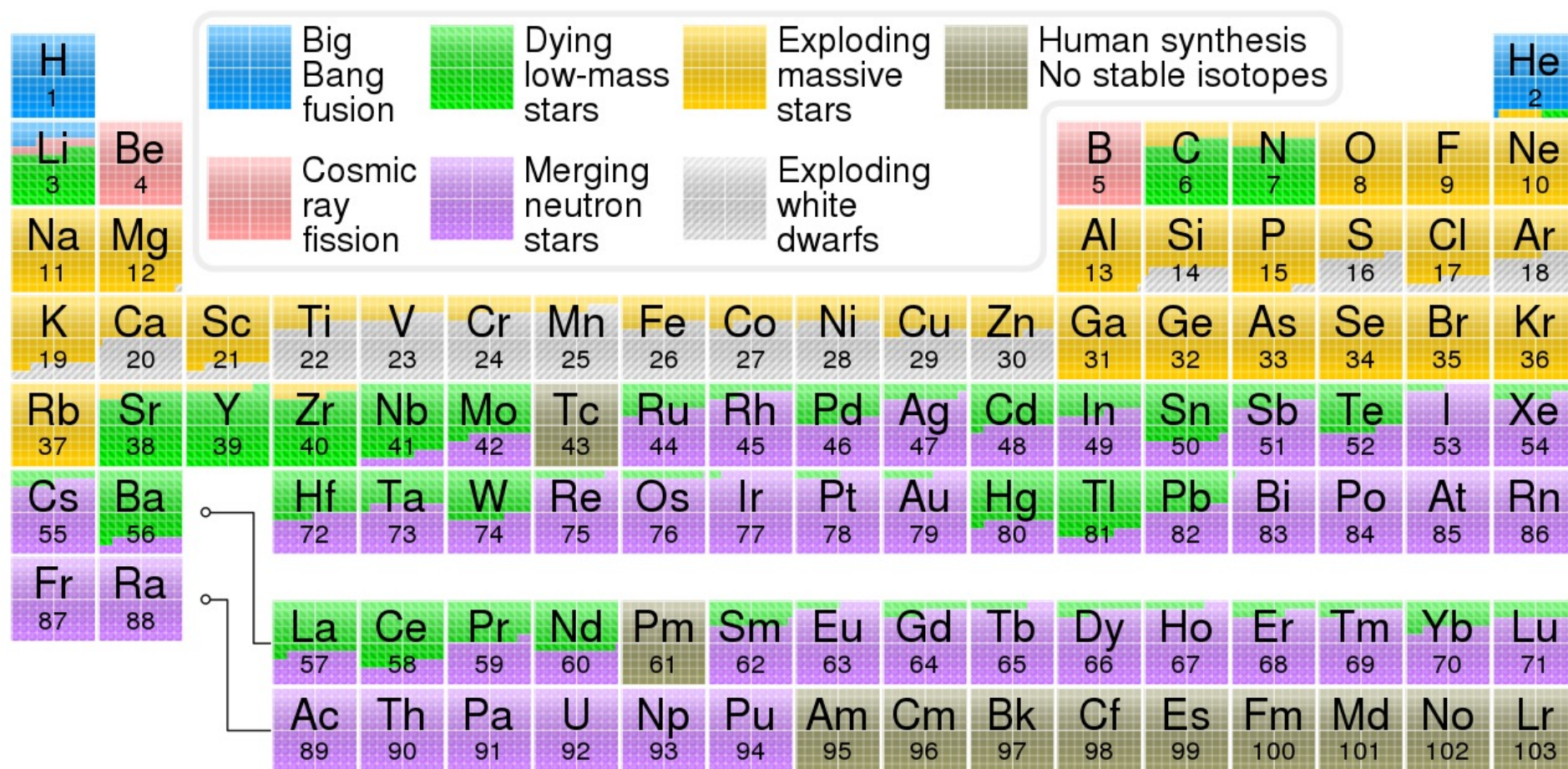
STAR	PLATE	ABSORPTION				
		ZrO	TiO	Ba II	Low-Temp.	Tc I
R And....	Ce 3522	8	3	5	8	4
U Cas.....	Pc 127	7	7	5	6	3
HD 22649..	Pc 192	2	2	5	6	1
R Gem....	Pc 68	5	0	10	7	5
S UMa....	Pc 110	1	0	7	4	1
T Sgr.....	Pc 124	7	0	7	5	3
R Cyg....	Pc 137	10	0	10	5	3
AA Cyg...	Pc 115	8	7	7	8	4
Z Del.....	Pc 112	2	7	3	3	1
χ Cyg.....	Ce 3762	5	20	3	10	3
o Cet.....	{ Ce 4109	1	15	1	7	2
	{ Ce 5925	1	10	2	6	1
R Hya....	Ce 3390	1	15	3	7	1
R Leo....	Pc 40	0	20	1	10	0

Science 115, 484 (1952)

Rev. Mod. Phys. 29, 547 (1957)

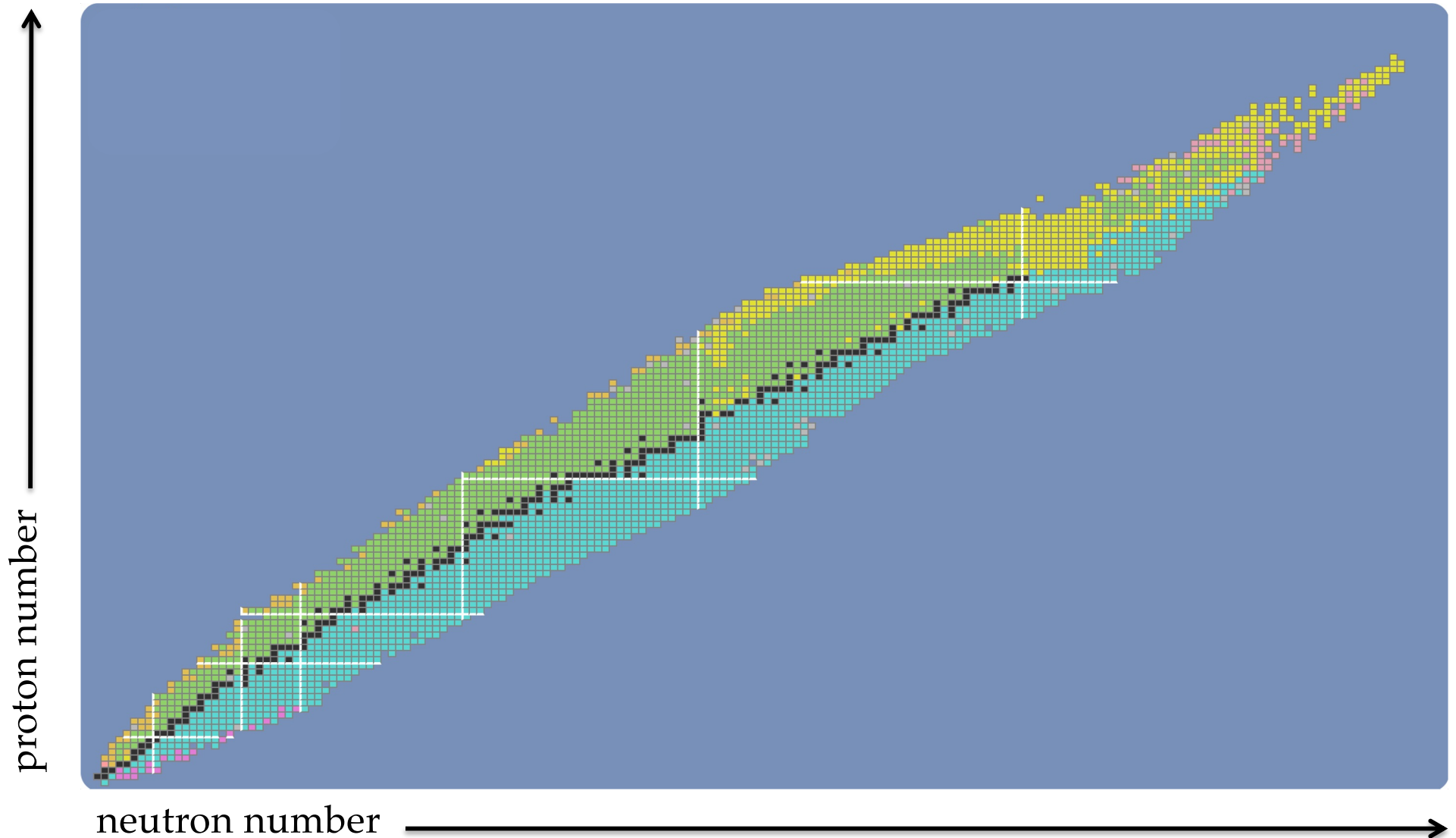
Astrophys. J. 116, 21 (1952)

A Chemist's View of the Periodic Table



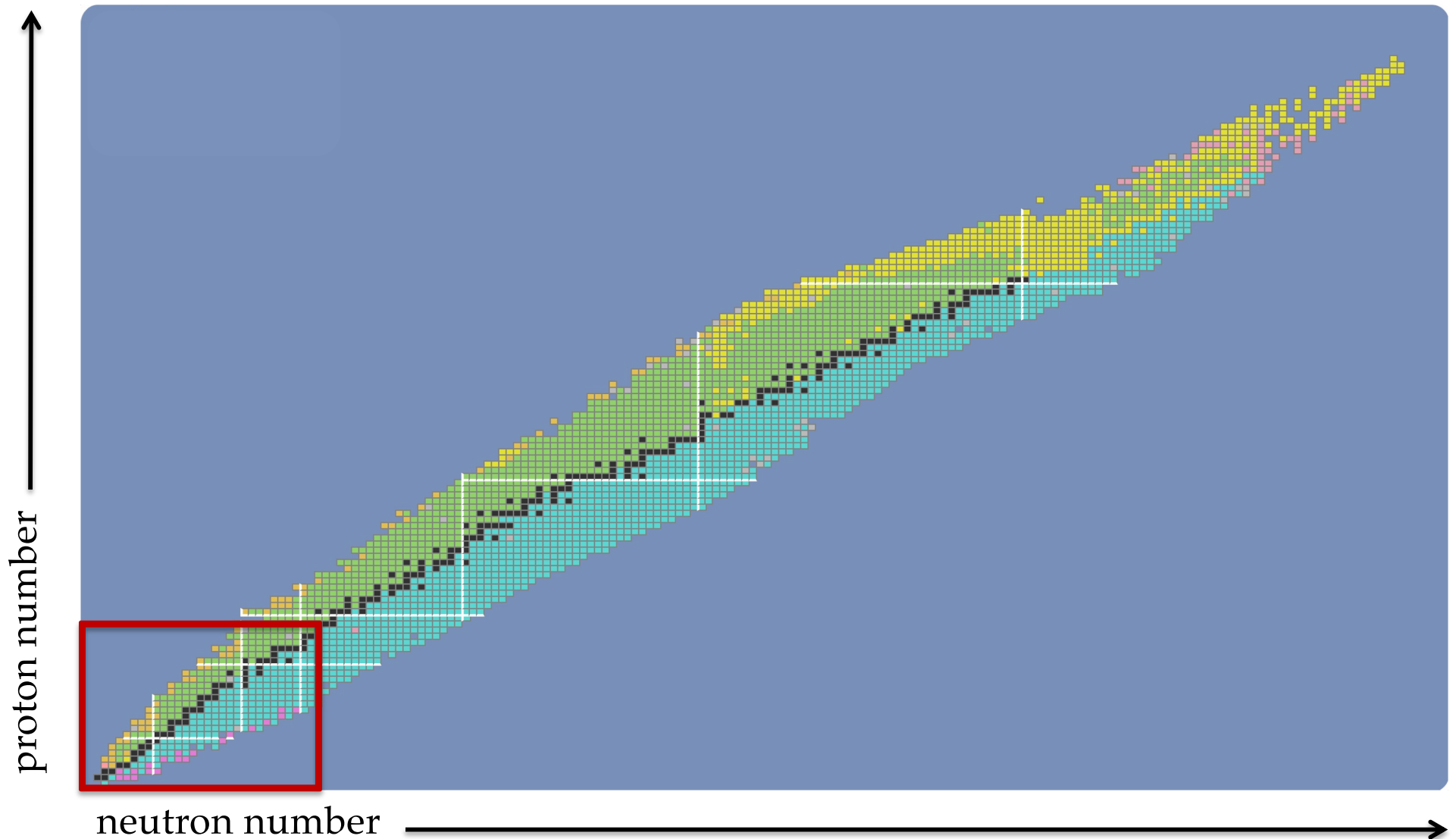
File:Nucleosynthesis periodic table.svg. (2020, March 7). Wikimedia Commons, the free media repository. Retrieved 16:41, May 24, 2020 from https://commons.wikimedia.org/w/index.php?title=File:Nucleosynthesis_periodic_table.svg&oldid=402170545.

A Nuclear Physicist's View of the “Periodic Table”



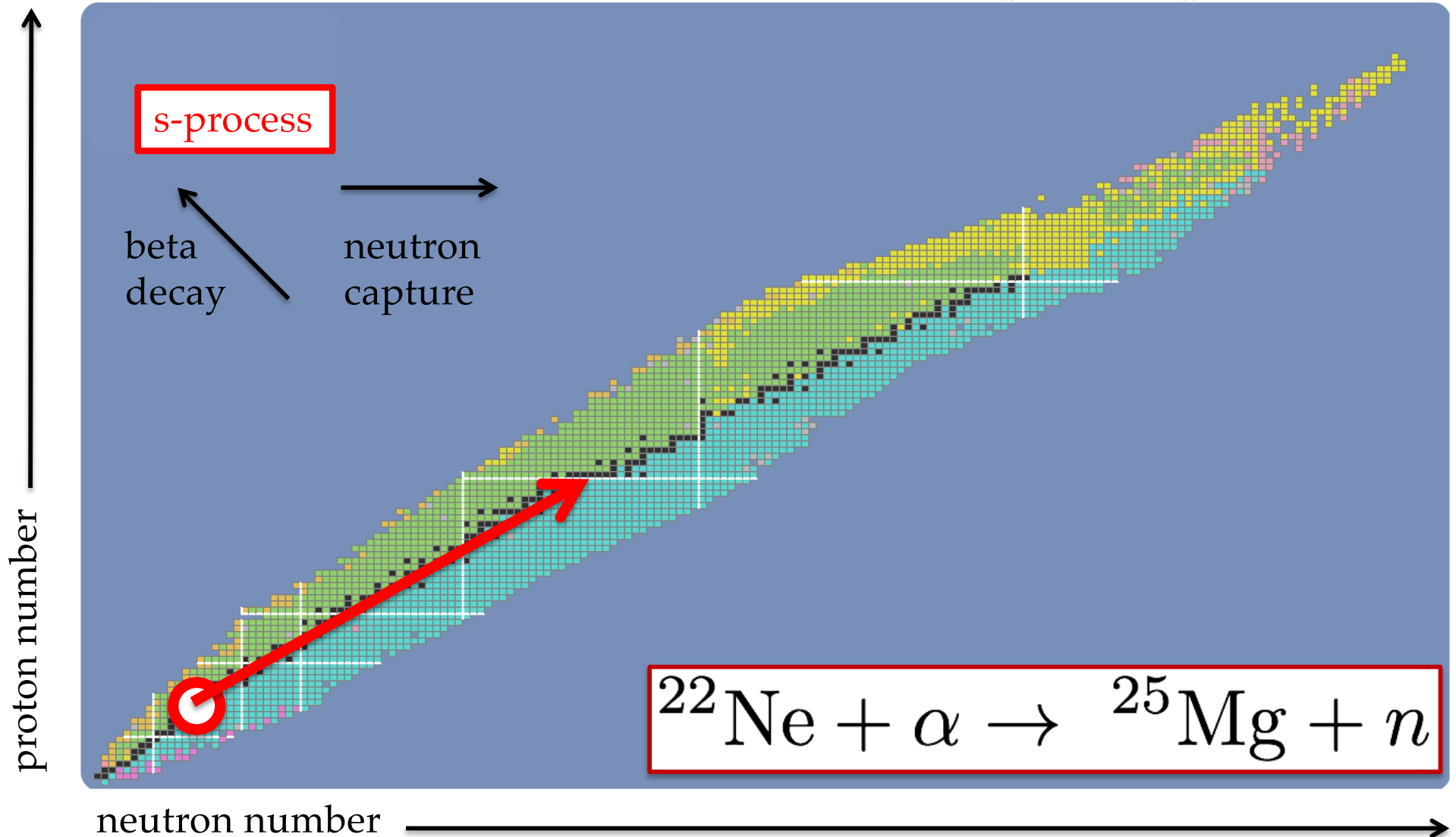
<https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html>

Nuclear Fusion Reactions Up To About Fe-56



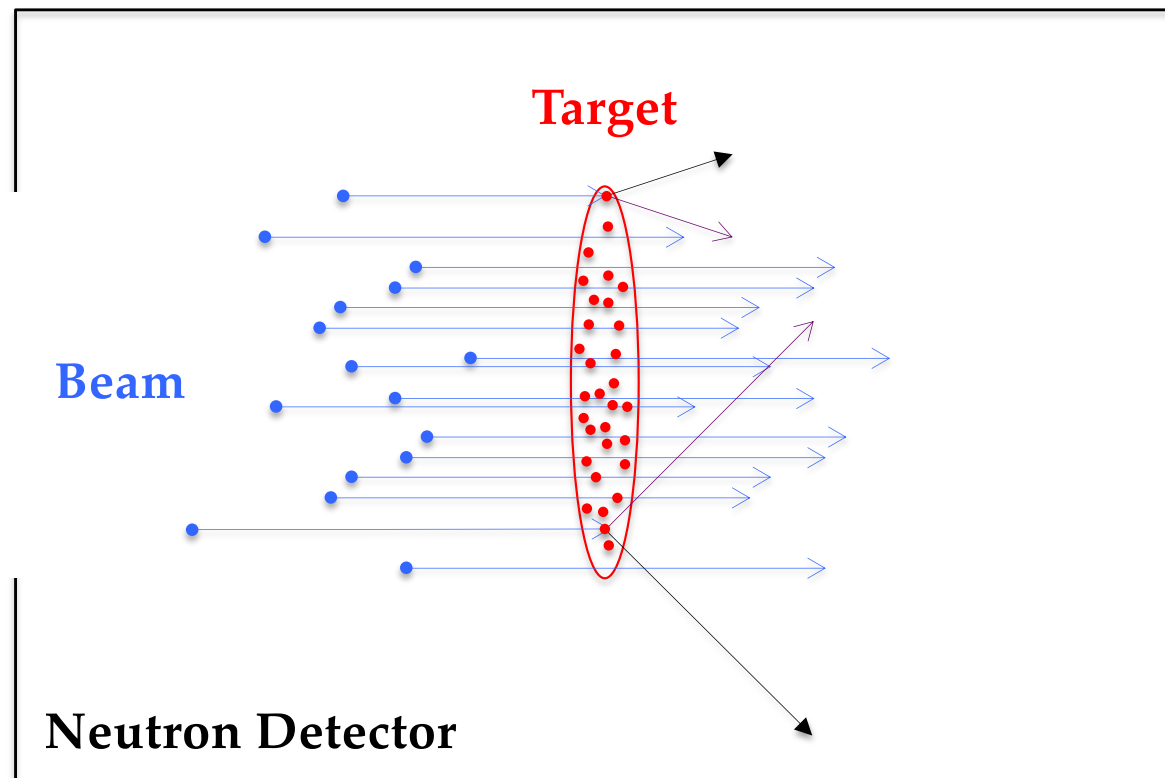
<https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html>

What is the source of neutrons that drives the “slow” neutron capture process?

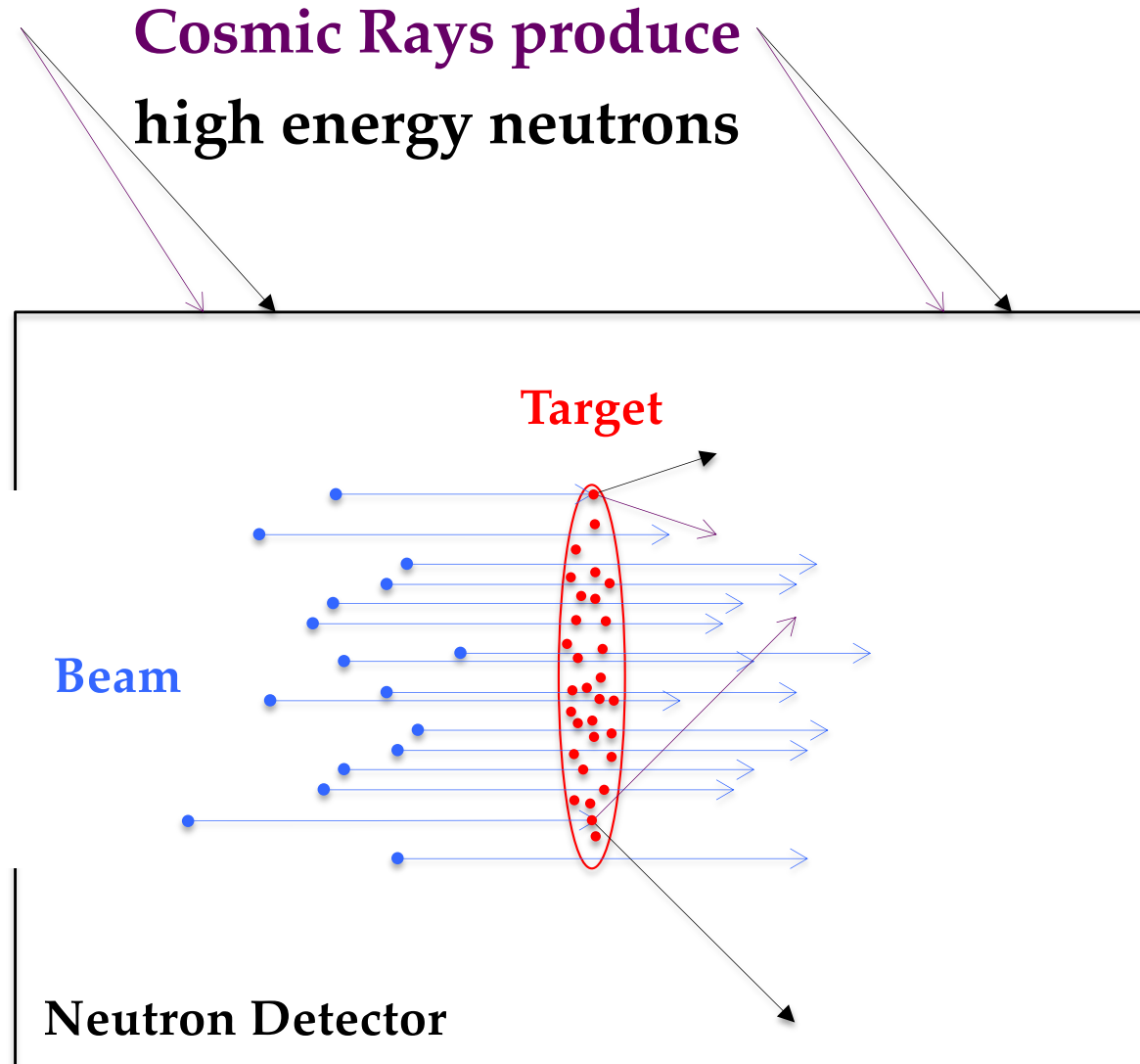


<https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html>

“Traditional Method”: Count the Product Neutrons



Challenge: Cosmic Ray Induced Neutron Background

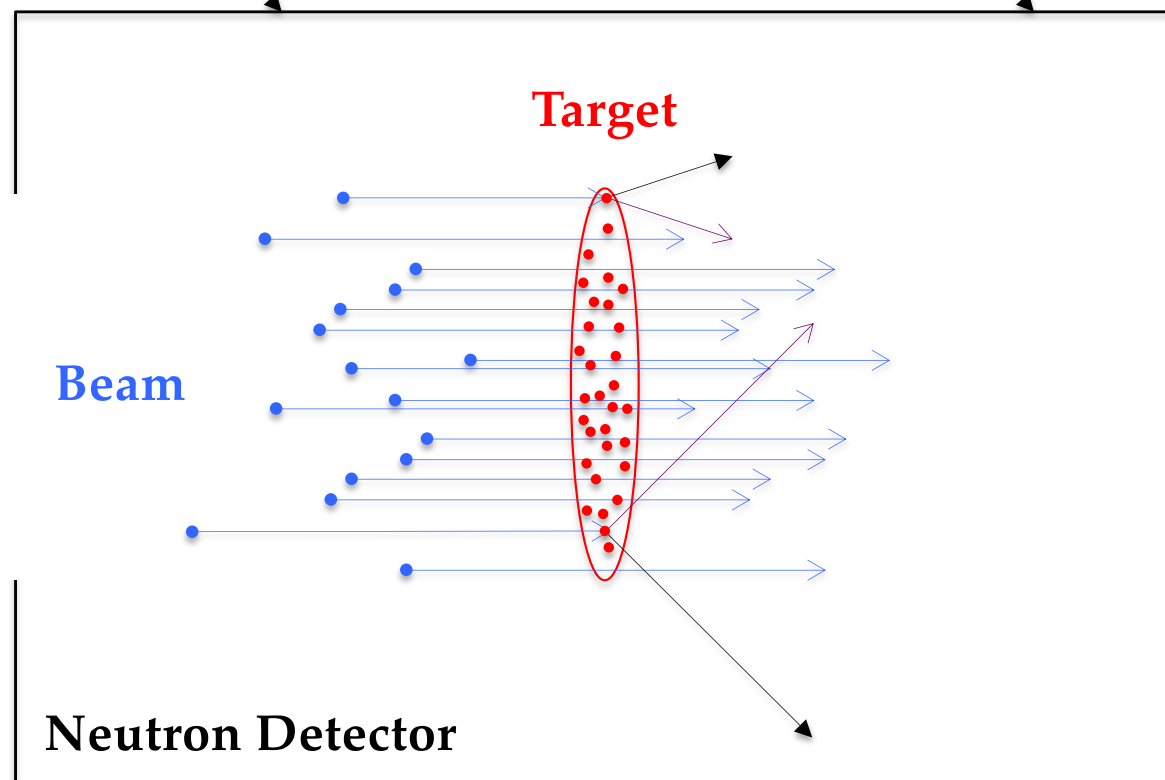


Detecting neutrons is very difficult – you lose all information about the energy of the neutron.

The Beautiful Jaeger 2001 Experiment

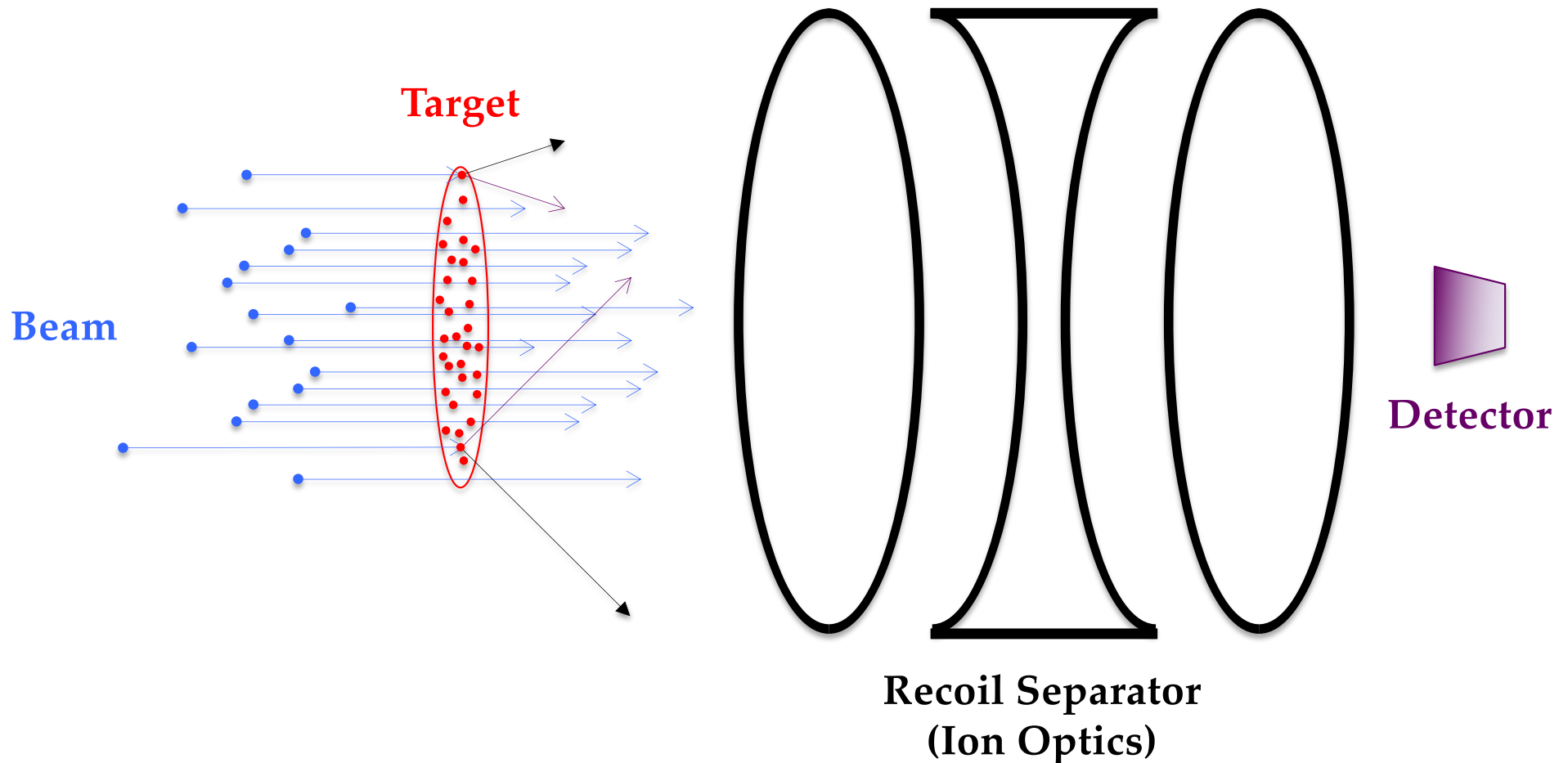
Cosmic Rays produce
high energy neutrons

Veto Detector



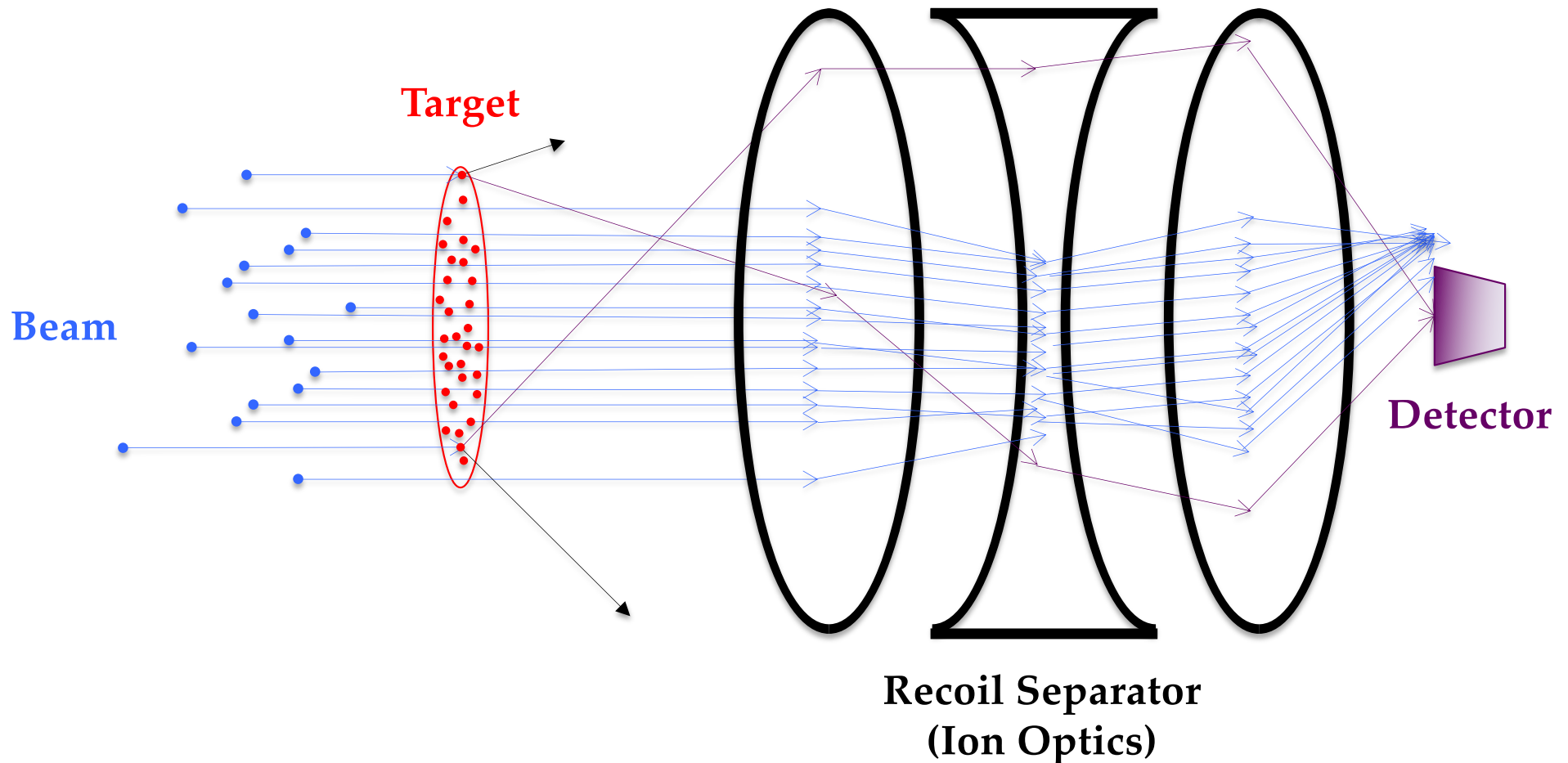
Veto detector that registers a “hit” simultaneously as the neutron detector helps rule out background neutrons...great but not perfect.

“Modern Technique”: Count the Product ^{25}Mg Atoms



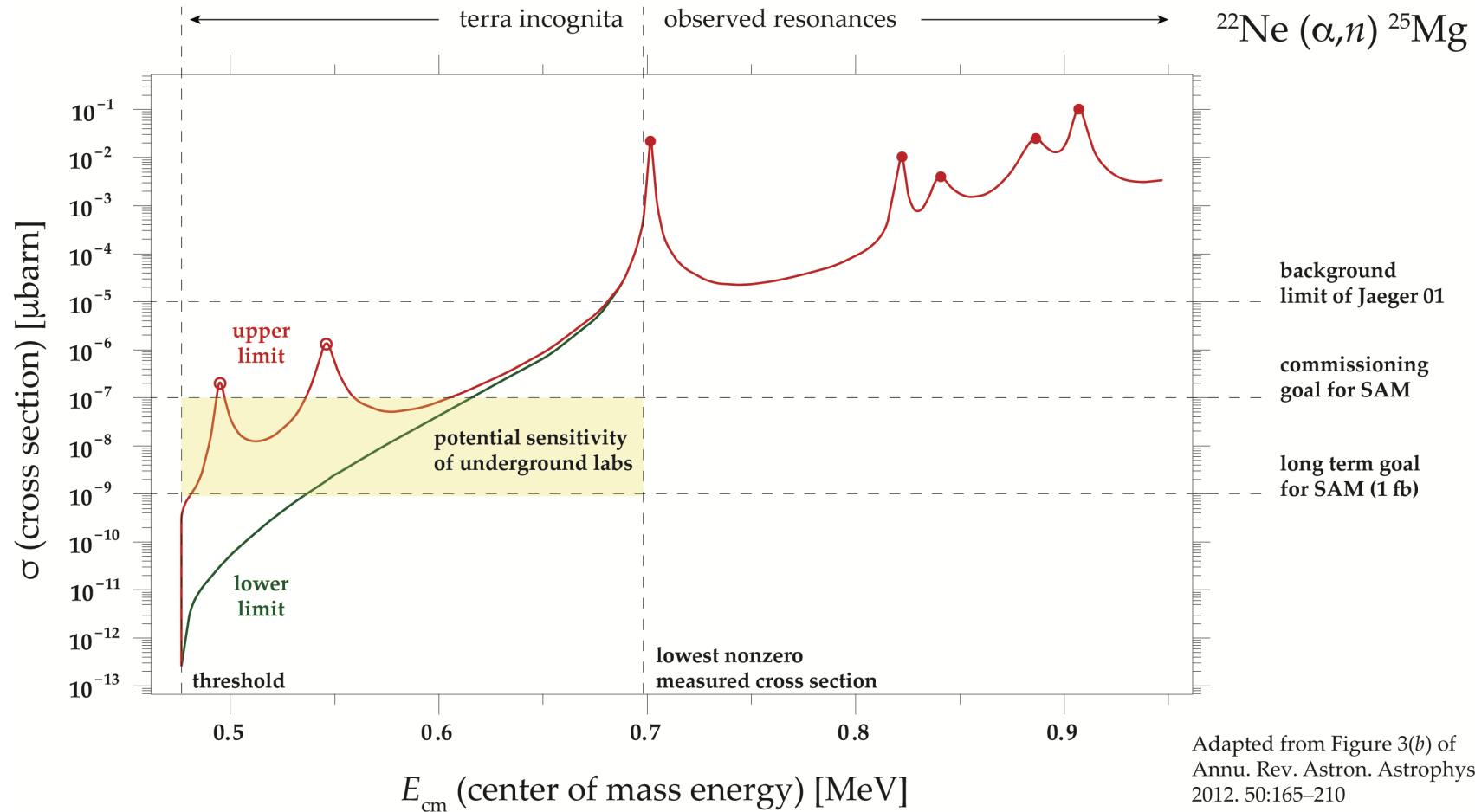
Electromagnetic recoil separator collects all of the products, separates them from the beam, and *counts everything that makes it through*.

Recoil Separators Are Awesome But Not Perfect!



The best recoil separators only let 1 beam atom through out of 100,000,000,000,000,000 or 10^{17} beam rejection efficiency, which is not quite good enough!

Towards Sub-Picobarn Cross Section Sensitivity



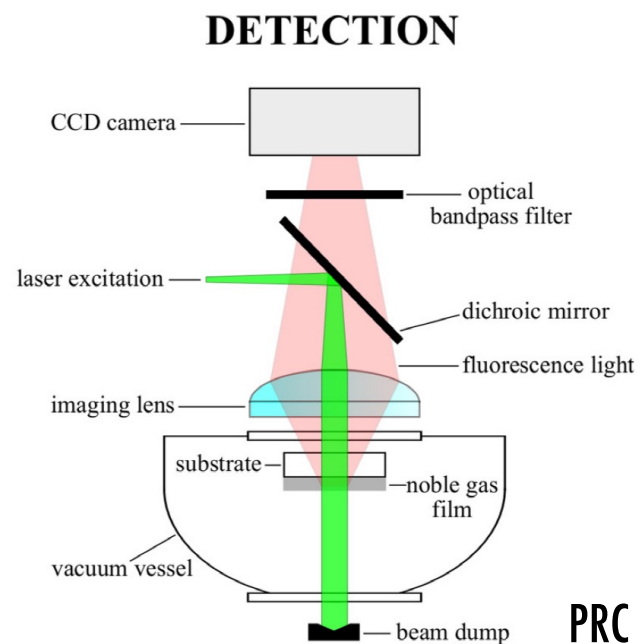
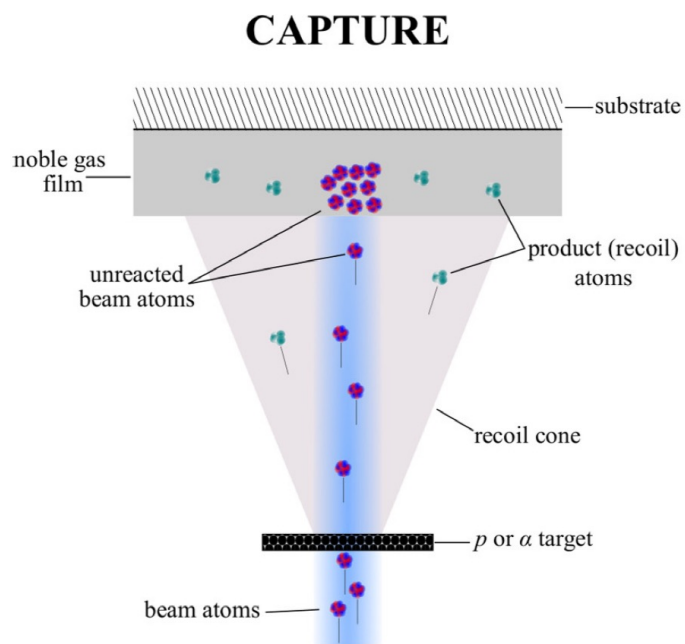
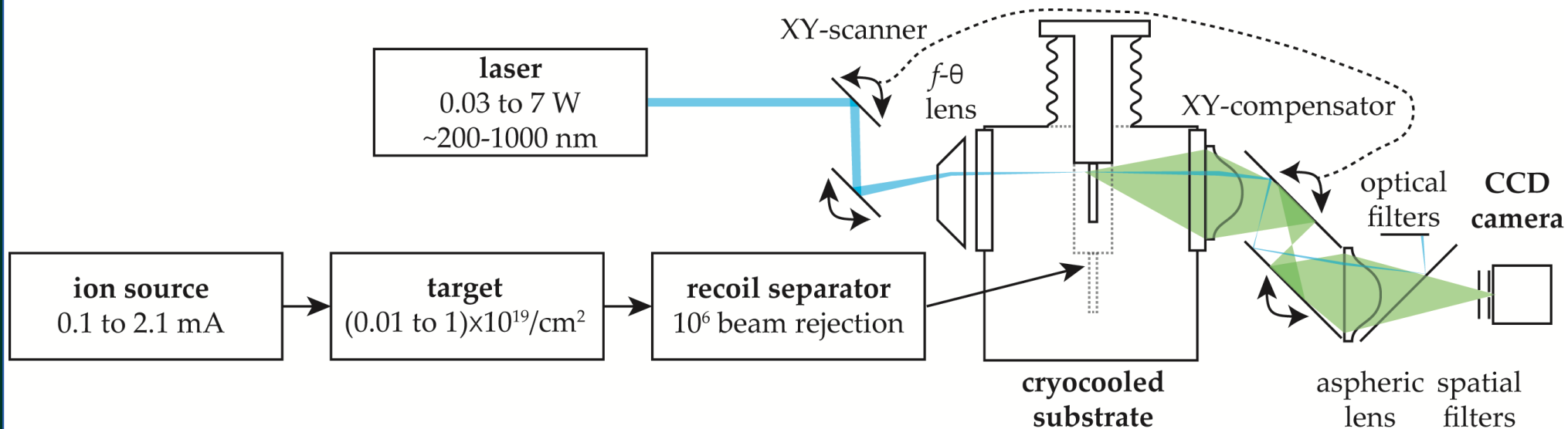
Underground labs are expected to have a factor of 100 or less background.

Recoils separators would need 10^{19} - 10^{20} beam rejection ratios.

$(1 \text{ pb}) (10^{17} / \text{cm}^2) (150 \mu\text{A}) = 5/\text{day}$

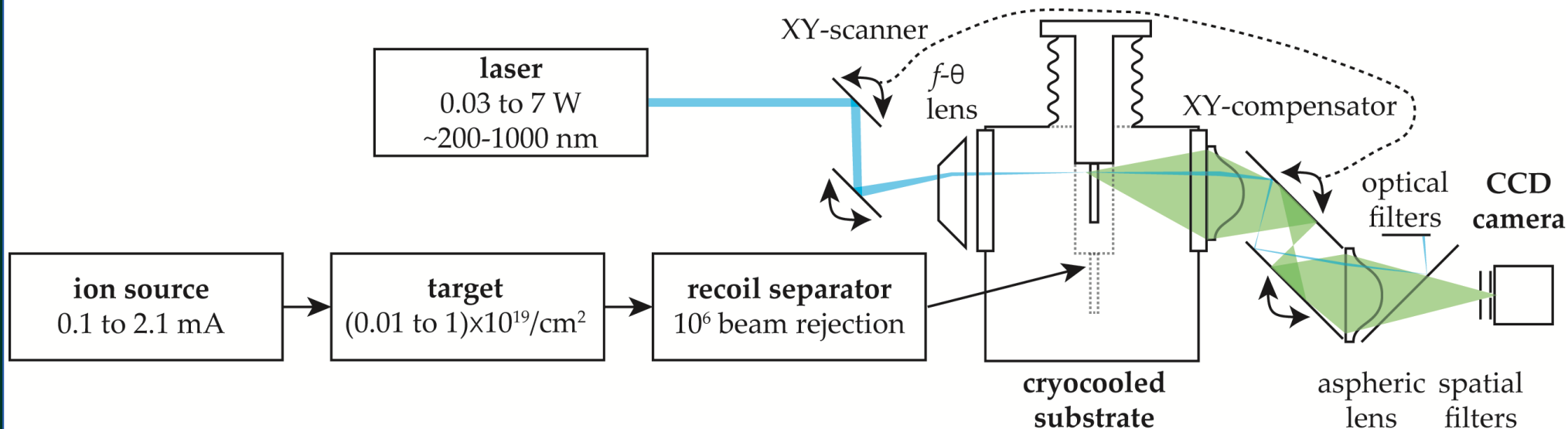
$(1 \text{ fb}) (10^{19} / \text{cm}^2) (2.1 \text{ mA}) = 7/\text{day}$

Single Atom Microscope Concept



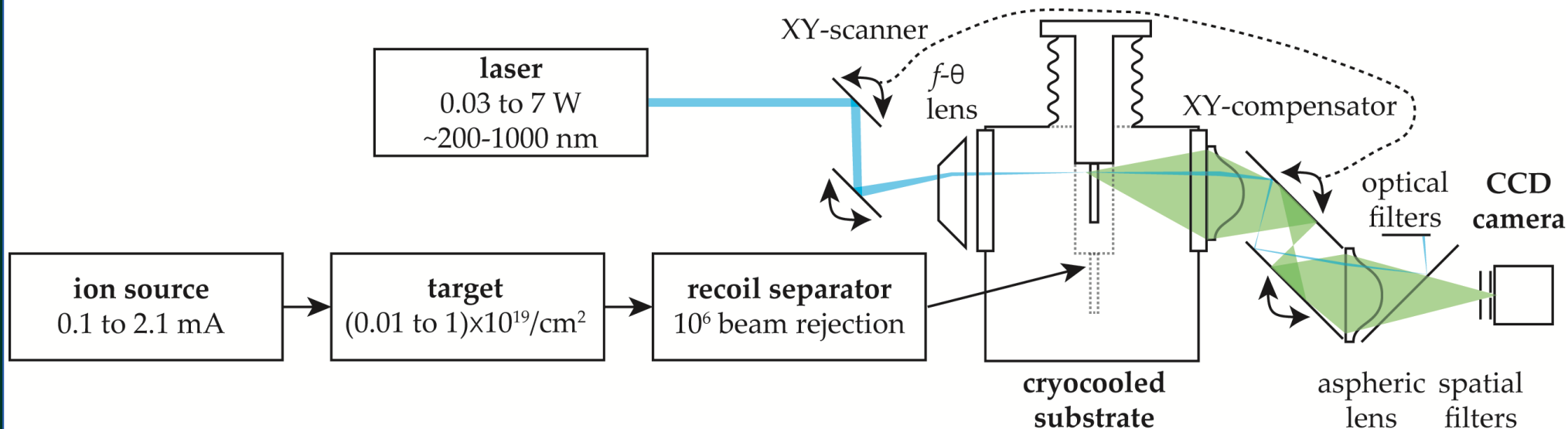
PRC 99, 065805 (2019)

Capture Every Product Atom



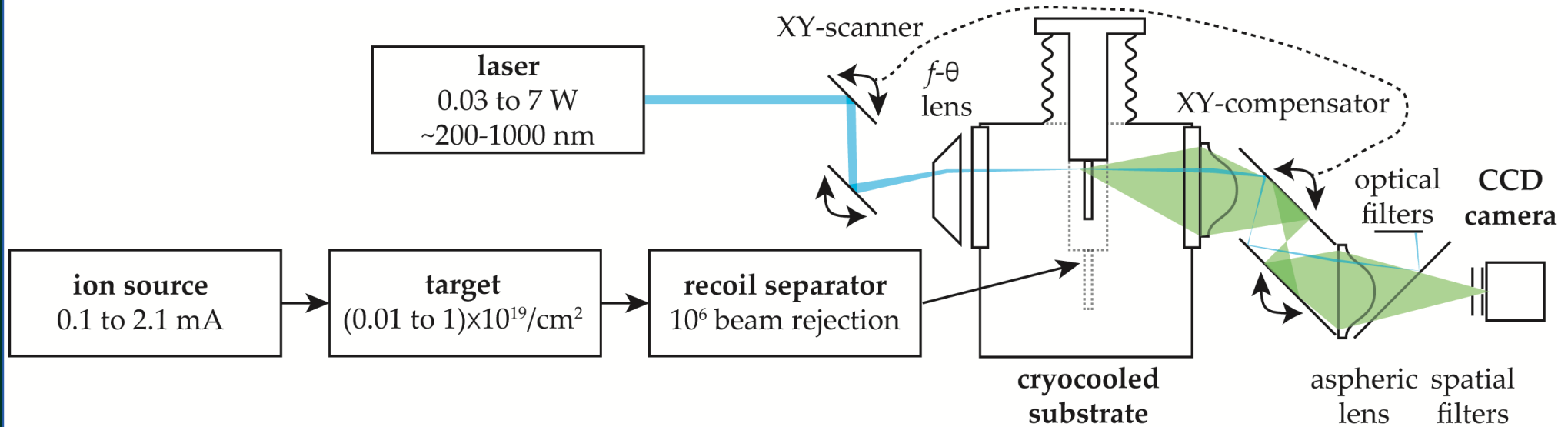
- Efficient: cryogenic NG film captures everything (both products and beam)

Count Only Product Atoms

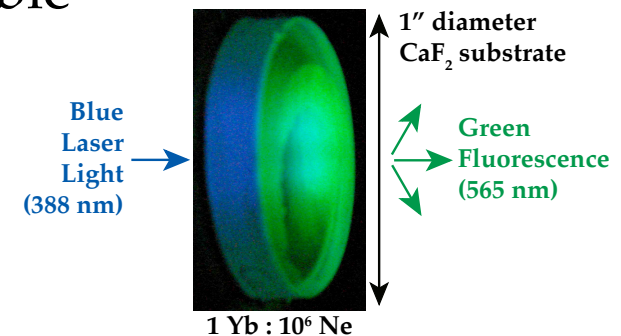


- Efficient: cryogenic NG film captures everything (both products and beam)
- Selective: product atoms identified by localized resonant laser excitation

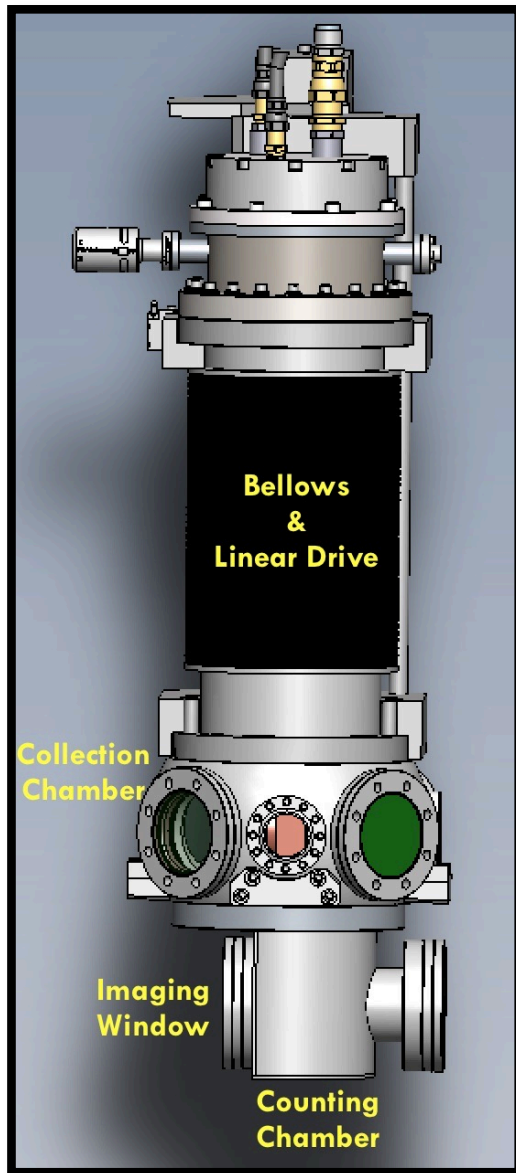
Challenge: Single Atom Sensitivity



- Efficient: cryogenic NG film captures everything (both products and beam)
- Selective: product atoms identified by localized resonant laser excitation
- Sensitive: large shift (few nm to 100's of nm) between **excitation spectrum** and **emission spectrum** coupled with spatial & optical filtering makes optical single atom detection feasible
- **Recoil separator is needed to:**
 - minimize heat load on NG film from beam
 - discriminate between isotopes



Prototype SAM



Pulsed Tube Cryocooler

- <10 micron amplitude vibrations
- 1.3 W cooling power

UHV compatible vertical linear drive

- up to 300 mm in travel
- <10 micron position repeatability

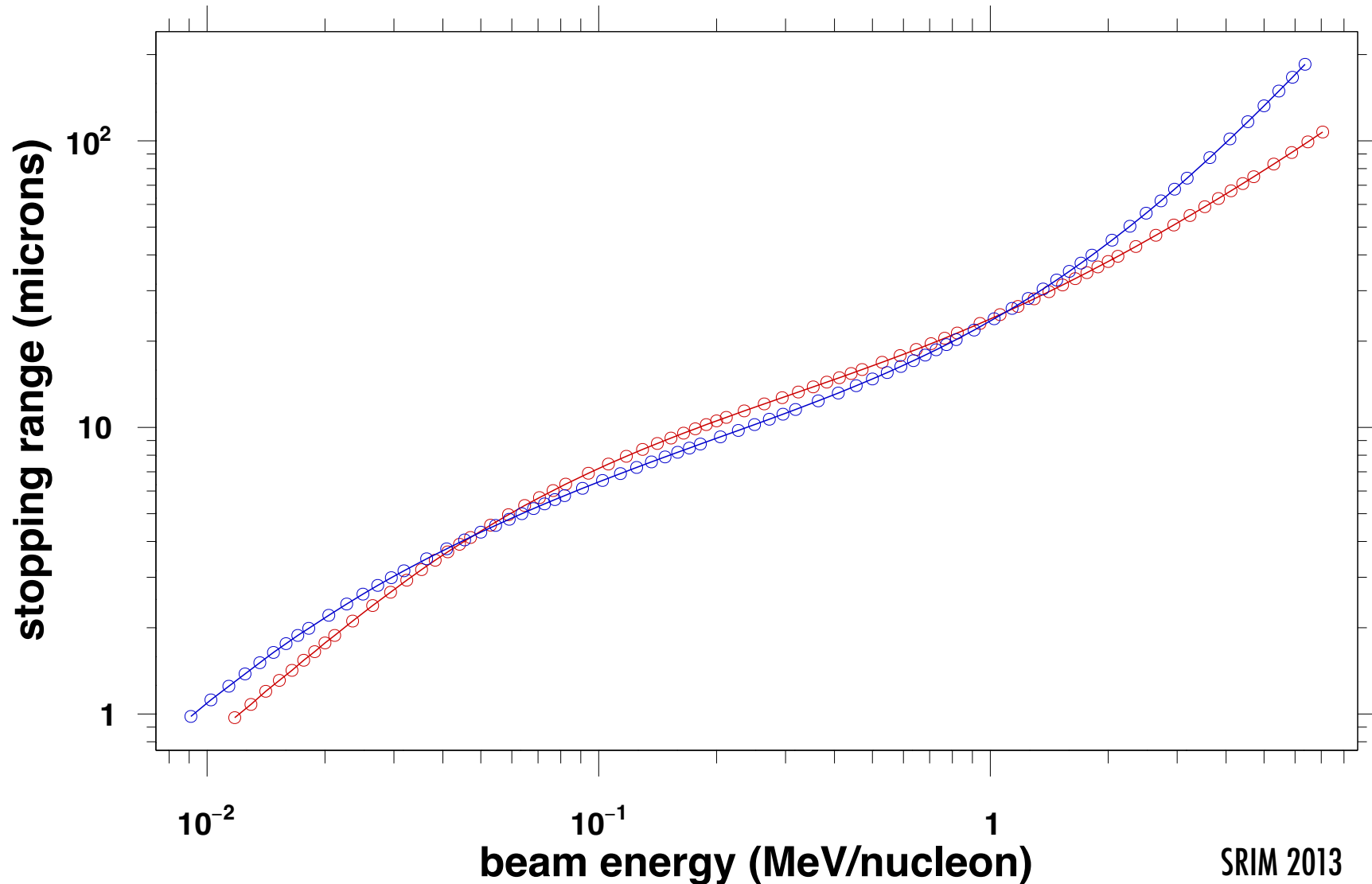
2.5" clear aperture

- 2% light collection w/ single aspheric lens
- DUV Fused Silica

We need 100 μm thick films to fully stop ions.

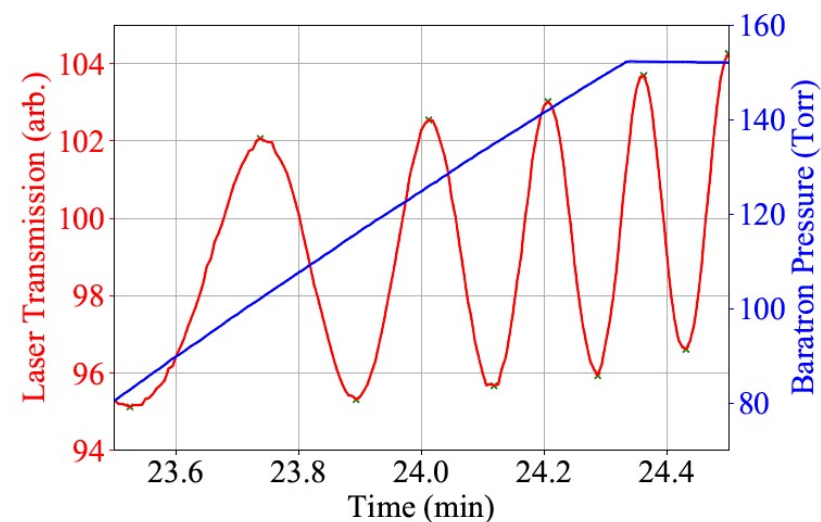
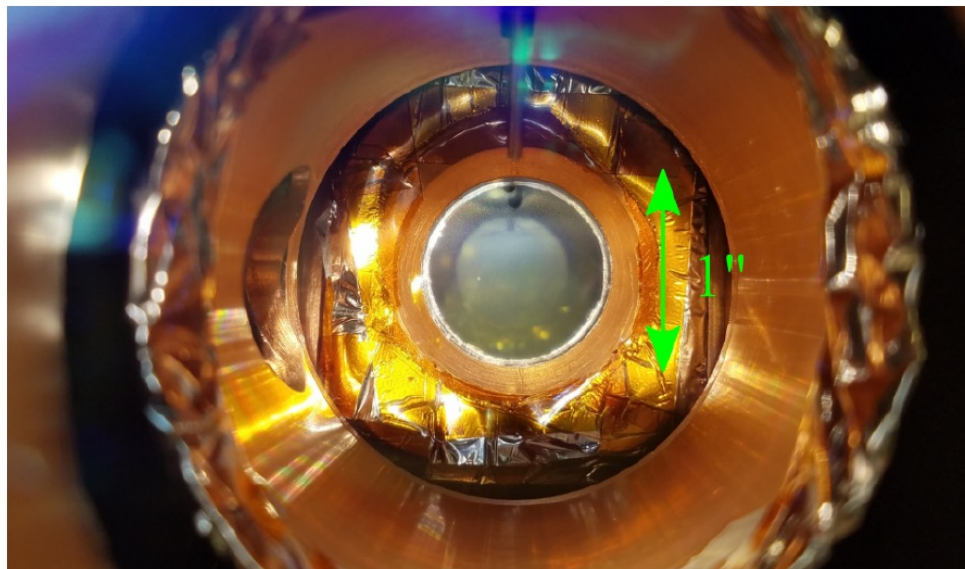
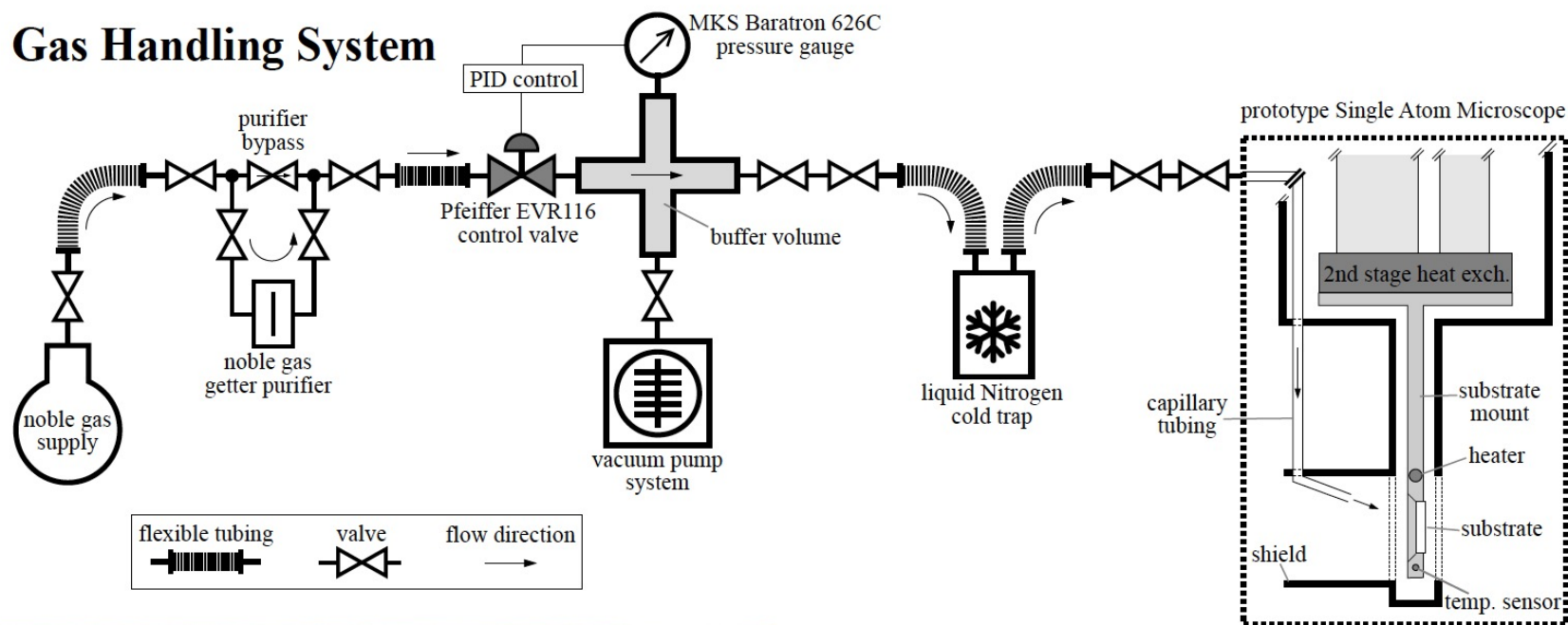
Ne^{+10} in solid Ne

Kr^{+36} in solid Kr



SRIM 2013

Growing Noble Gas Thin Films



There is an art to growing transparent films.

1 inch
diameter
sapphire



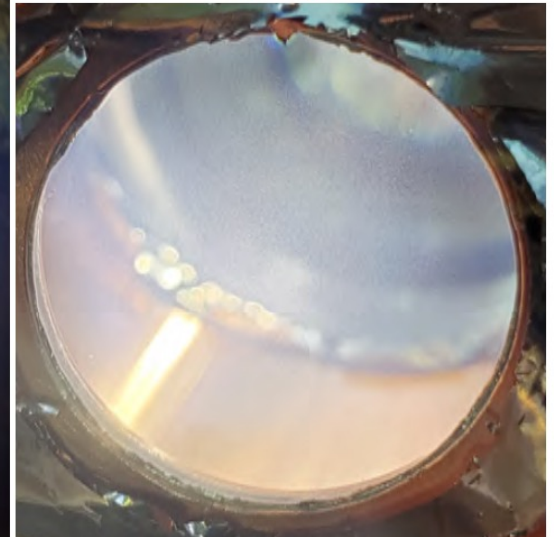
clear



cloudy



hazy



speckled



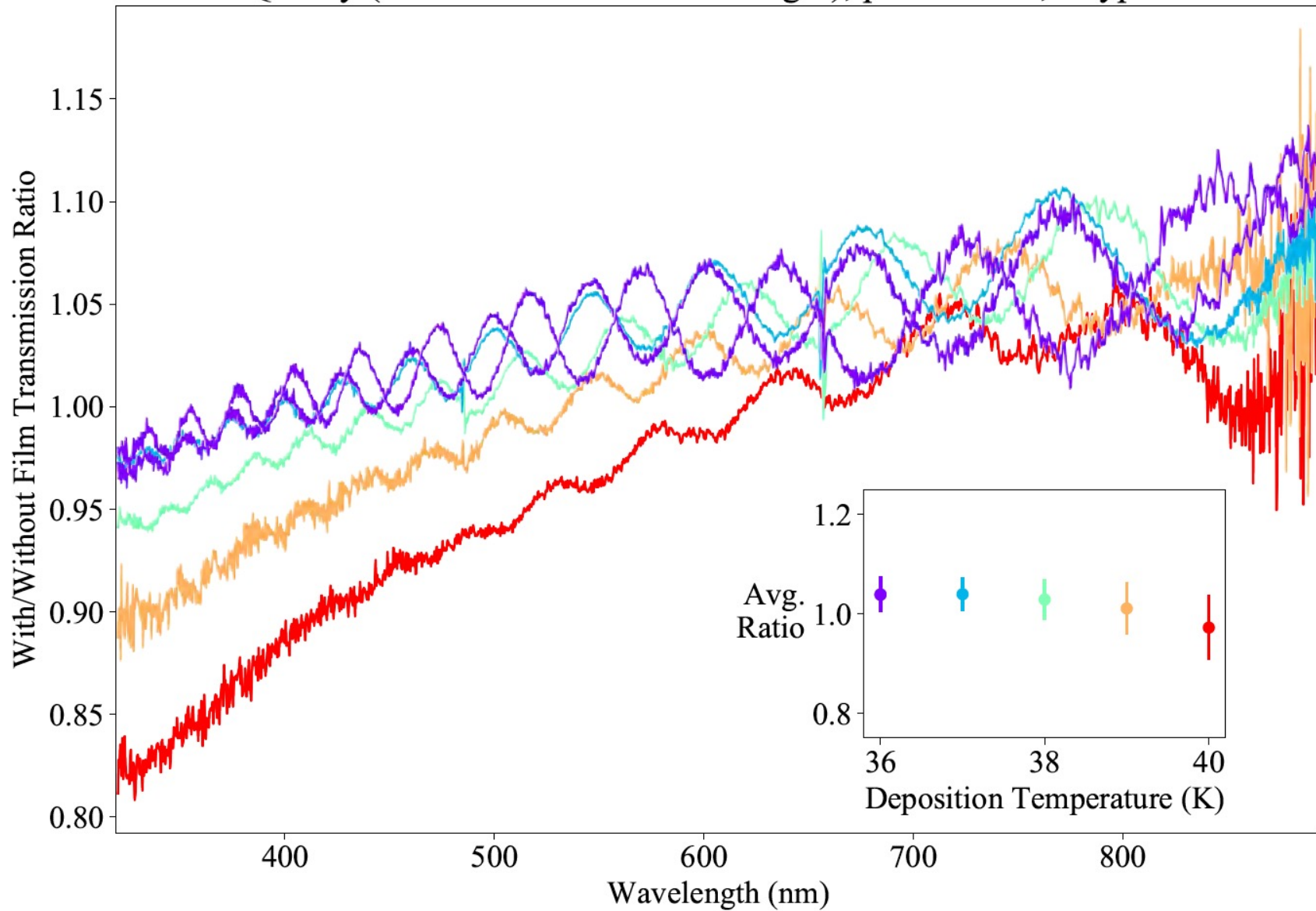
broken



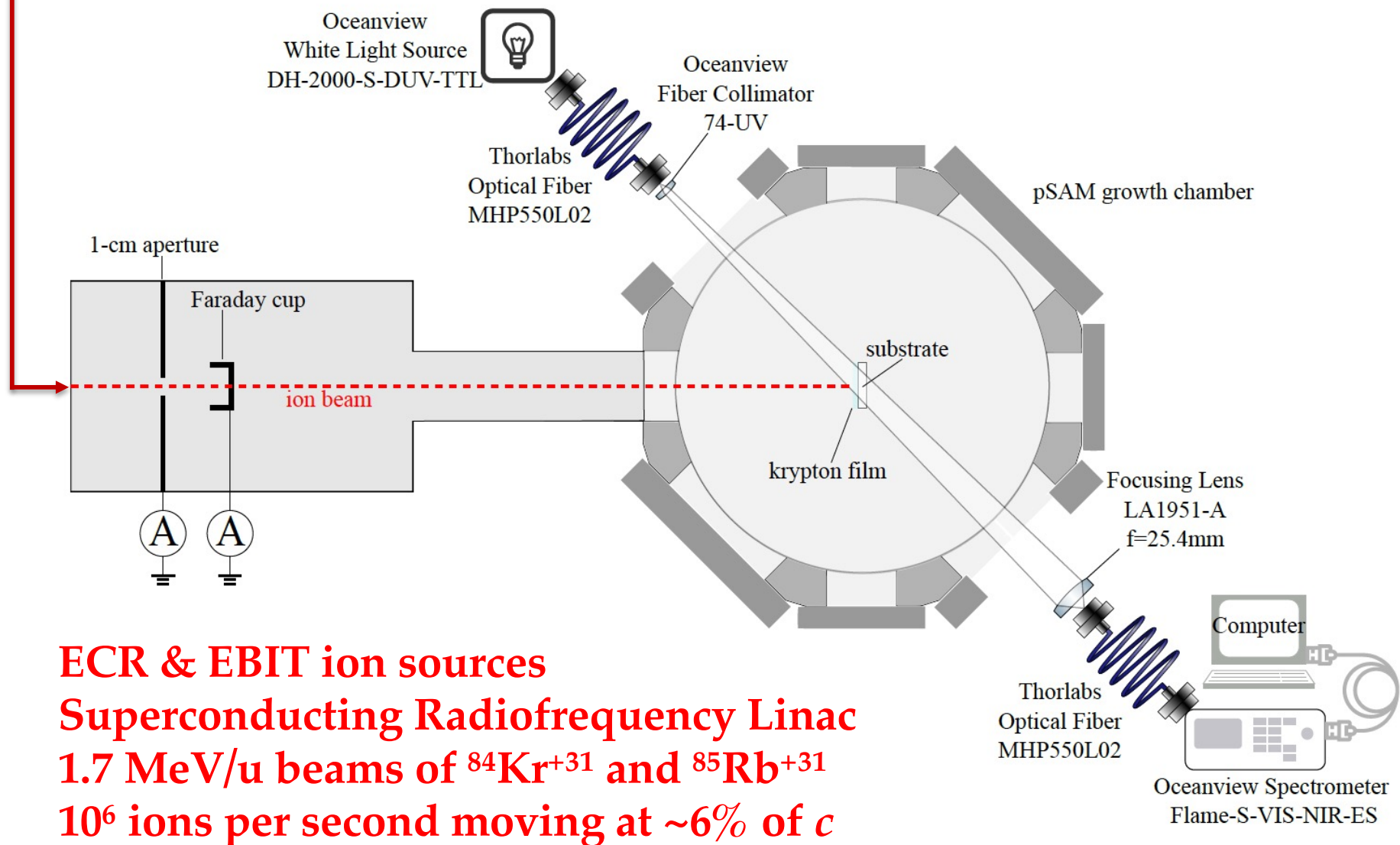
frosty

Film Transparency is temperature dependent.

Film Quality (Transmission vs. Wavelength), pSAM v1.4, Krypton films

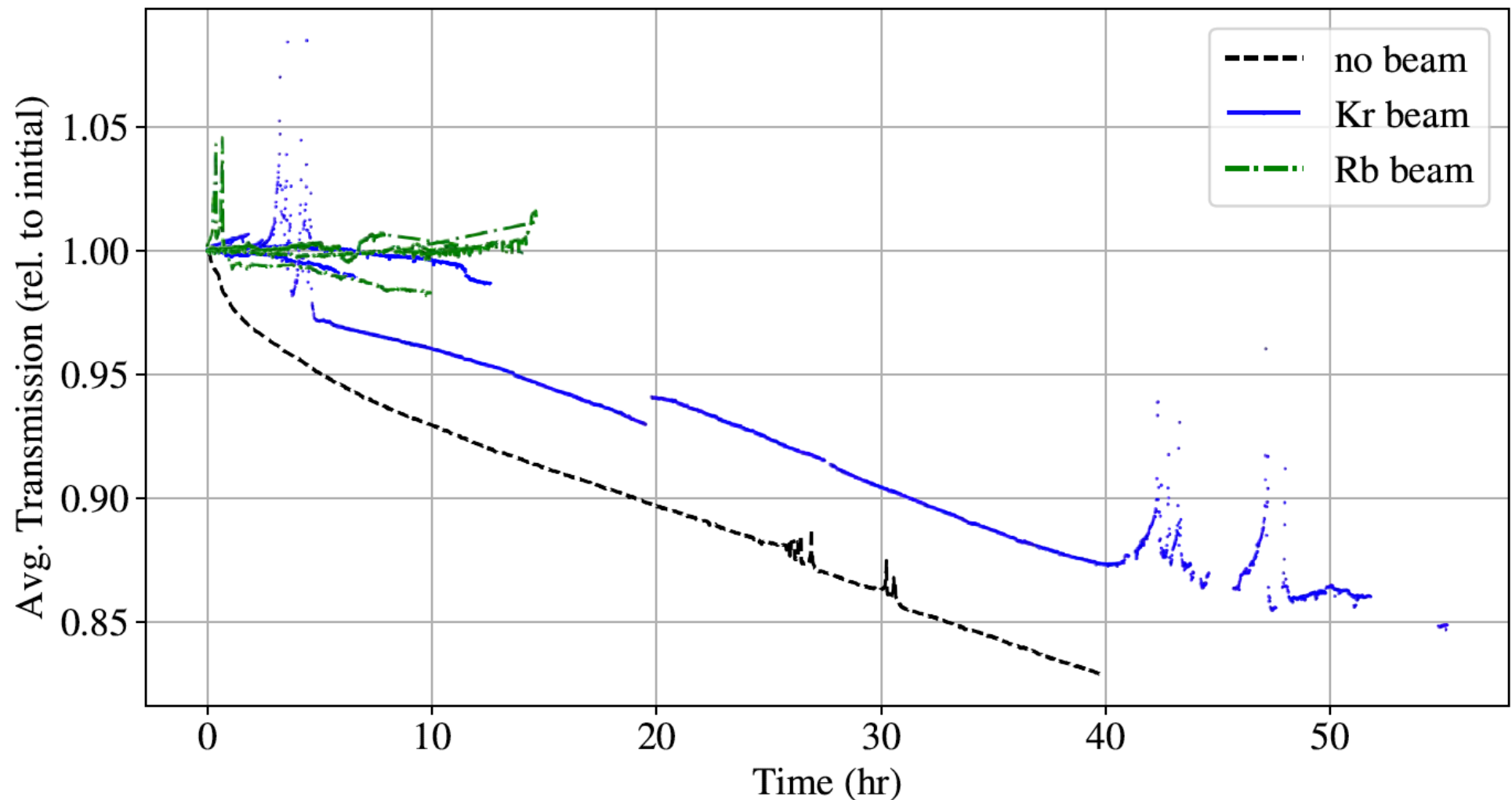


"ReA3" Beamline @ NSCL



- ECR & EBIT ion sources
- Superconducting Radiofrequency Linac
- 1.7 MeV/u beams of $^{84}\text{Kr}^{+31}$ and $^{85}\text{Rb}^{+31}$
- 10^6 ions per second moving at $\sim 6\%$ of c

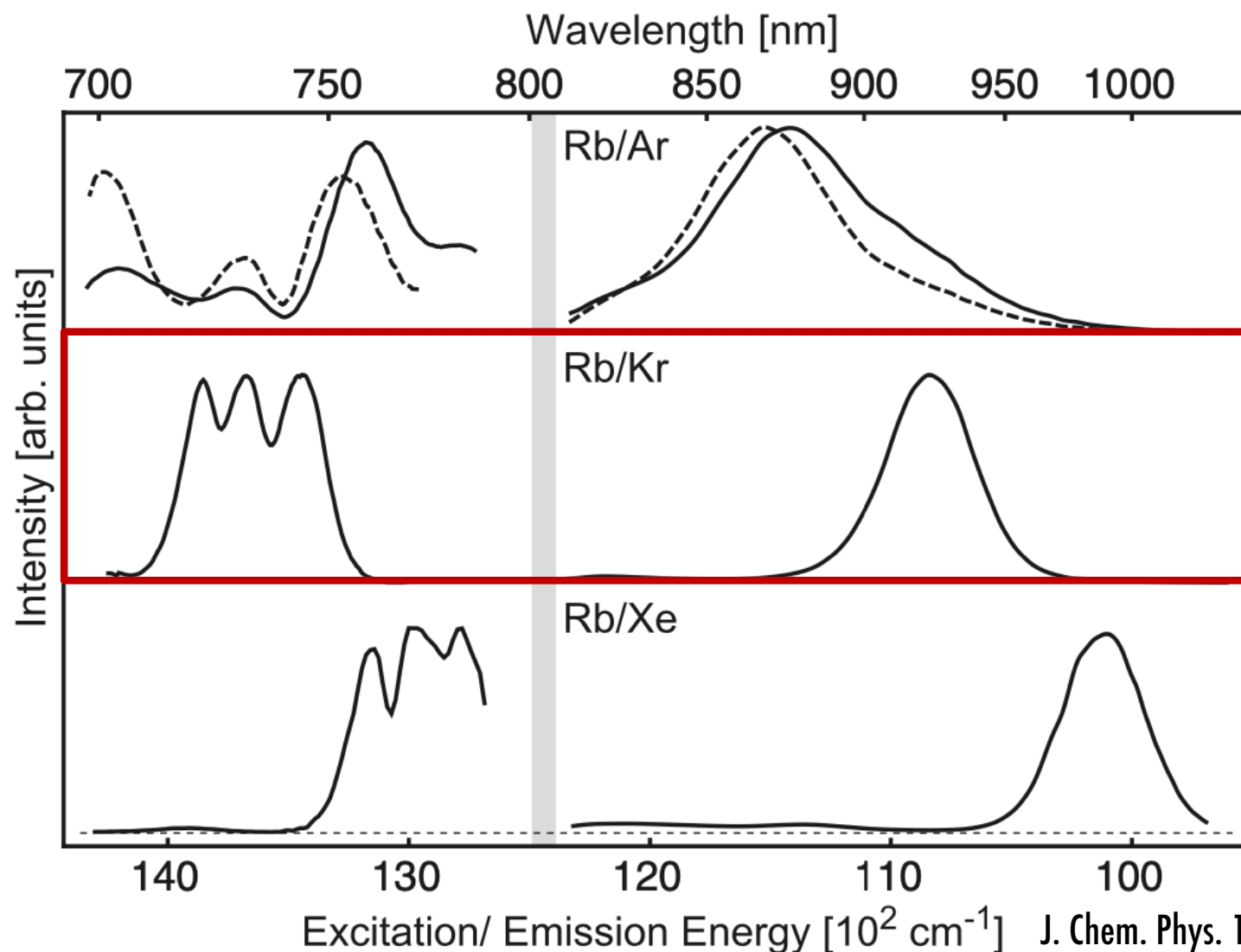
Film transparency is robust against damage.



Start with Alkali Atoms!

Rb in solid Ar/Kr/Xe

Rb in solid Ar



Absorption band (Å)	Emission peak (Å)
---------------------	-------------------

7980 (peak)	8300
-------------	------

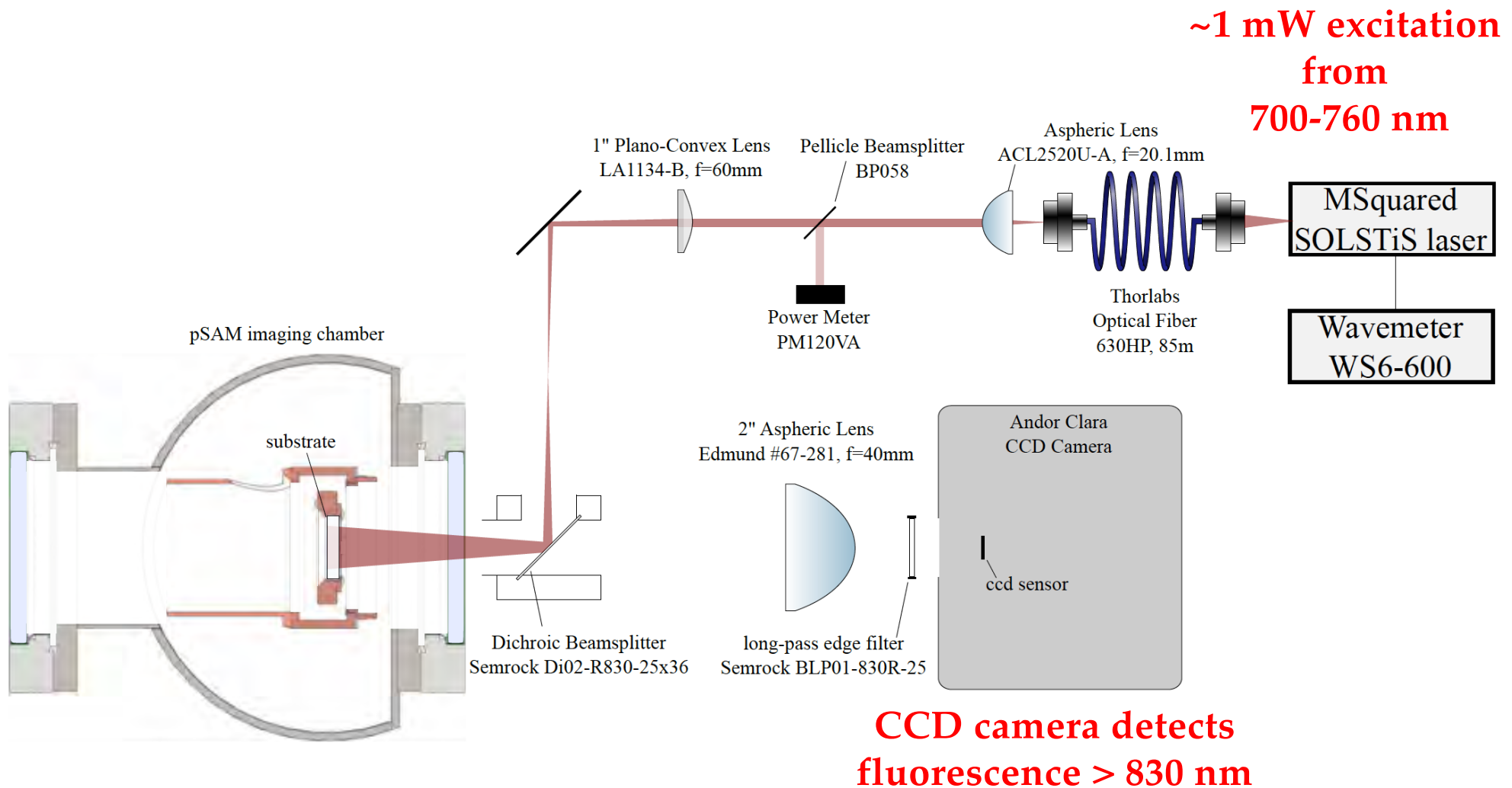
7760 (peak)	8300
-------------	------

7550 (peak)	8300
-------------	------

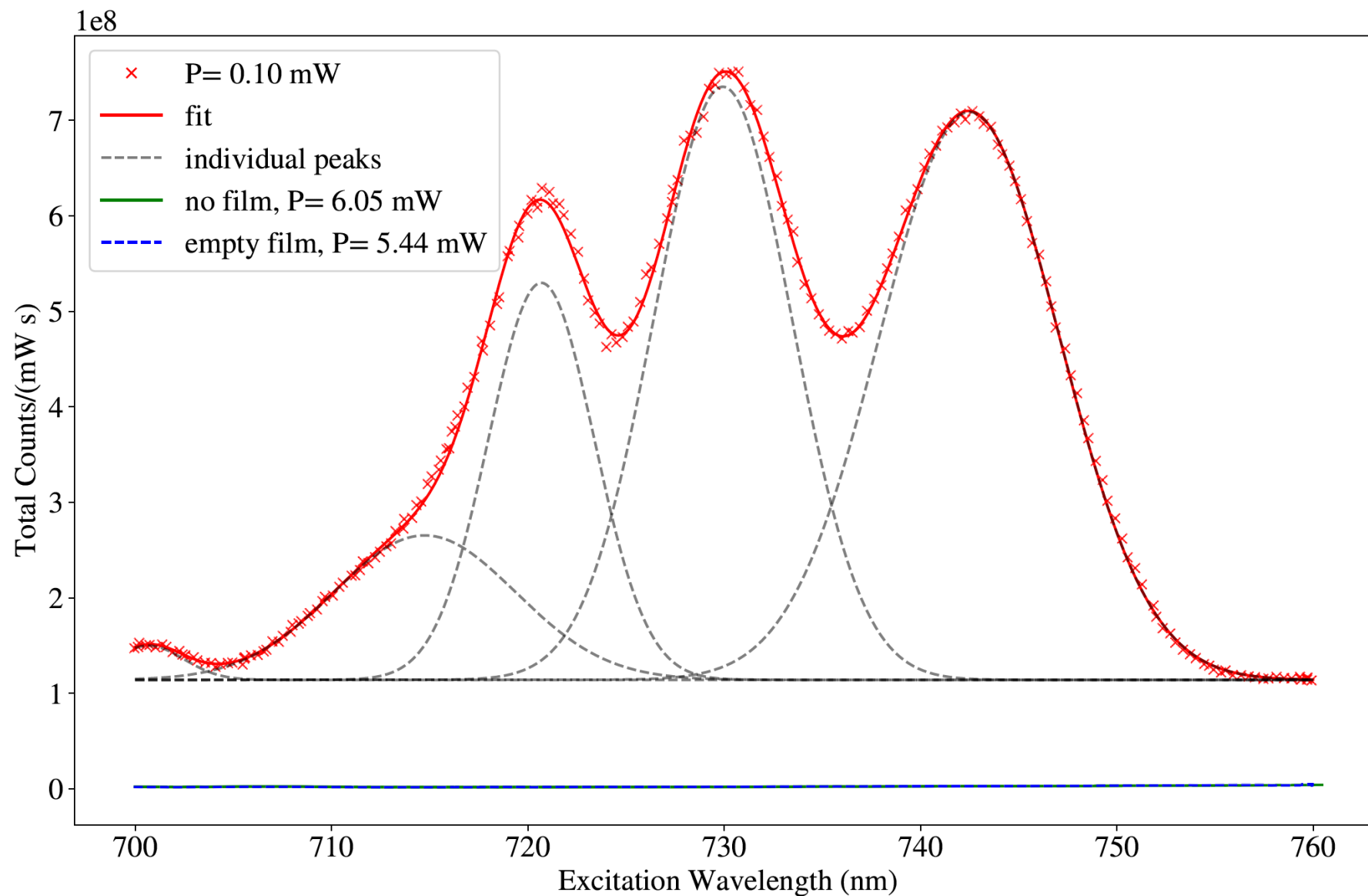
J. Chem. Phys. 78, 592 (1983)

J. Chem. Phys. 137, 014507 (2012)

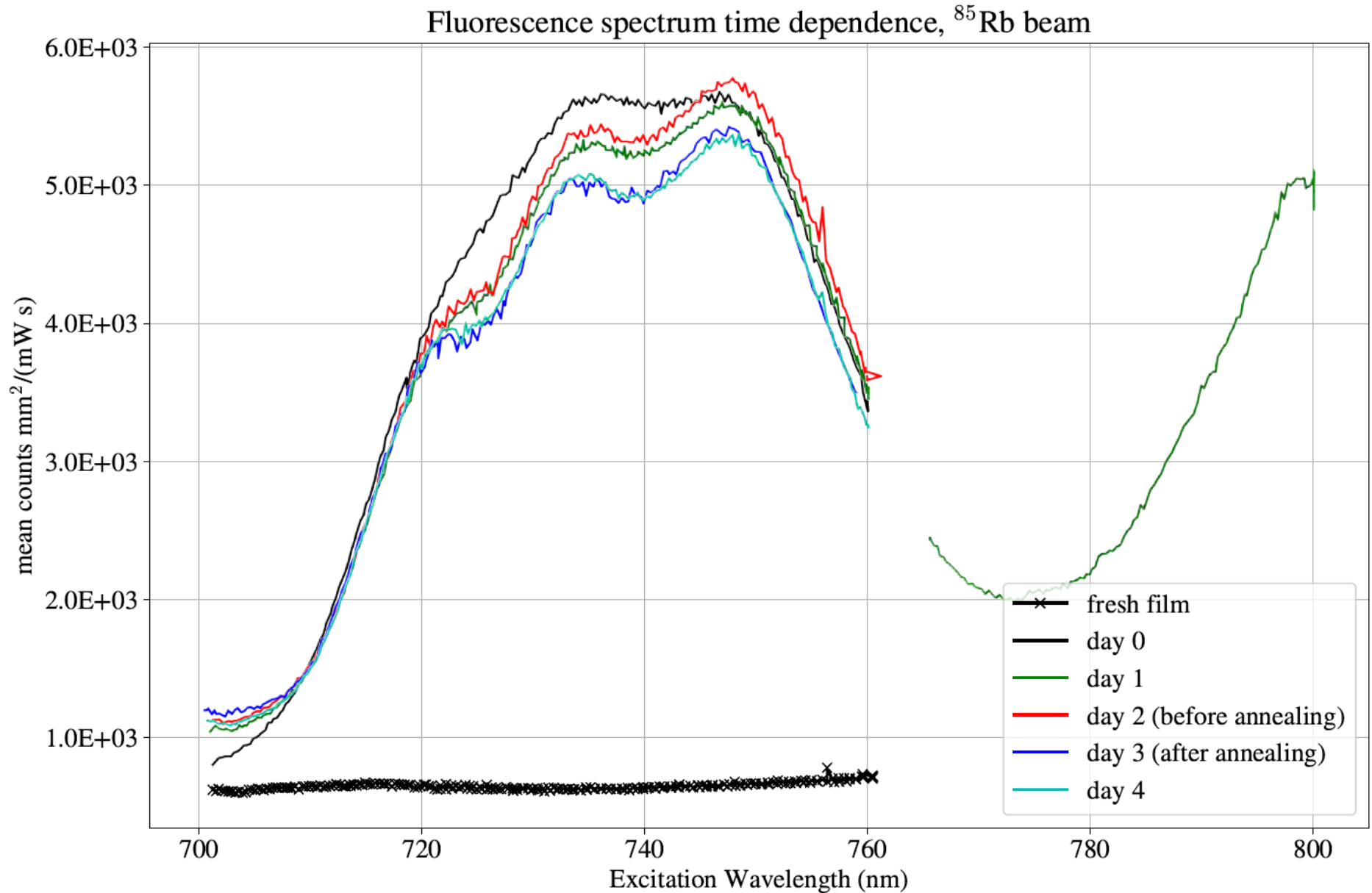
Rb Fluorescence in Solid Kr



Neutral Rb Fluorescence (>830 nm) in Solid Kr



Implanted Rb^{+31} spectrum resembles neutral Rb.

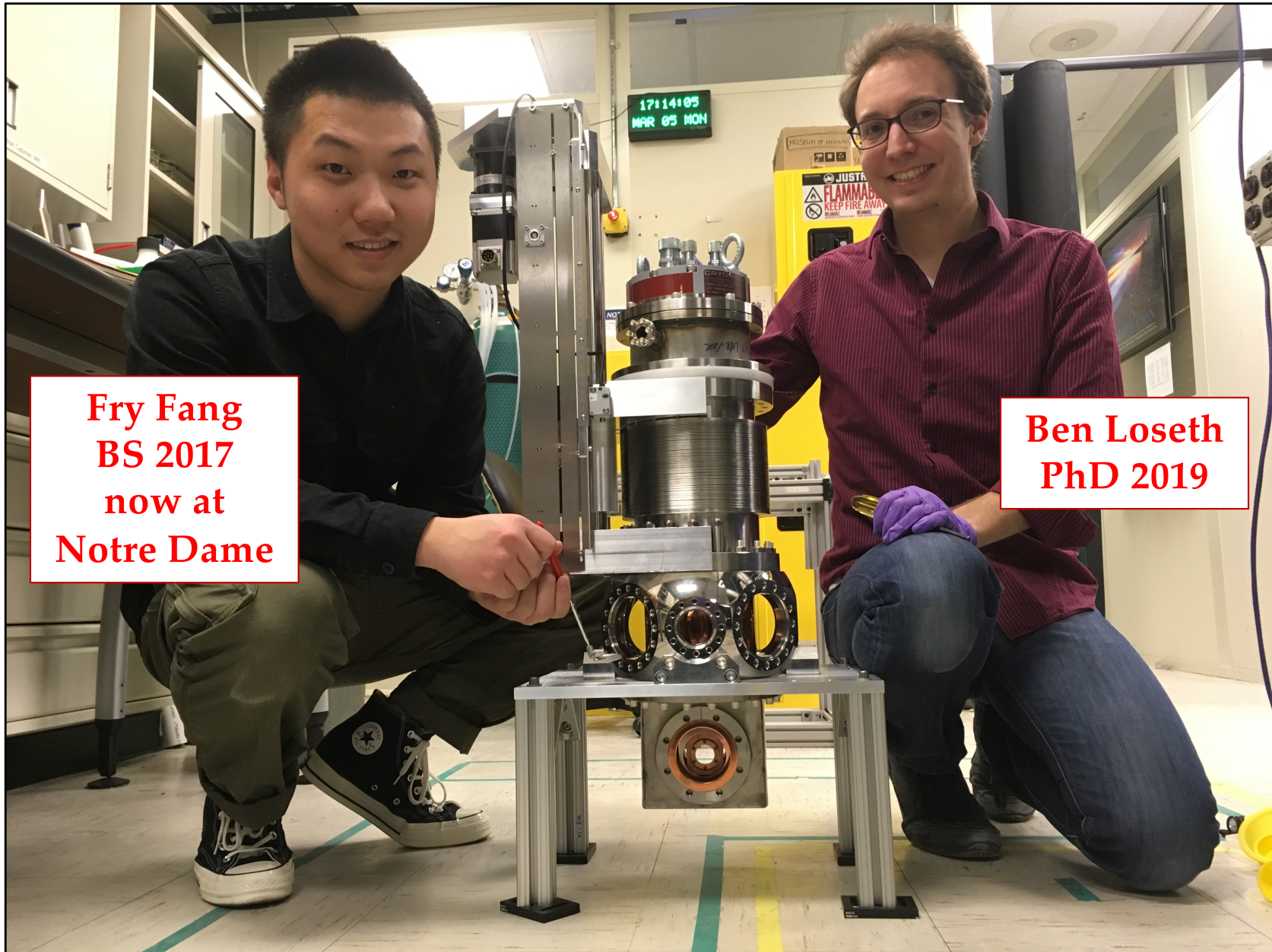


Prospects of Single Atom Sensitivity

parameter	Neutral Rb from effusive oven	Implanted Rb ⁺³¹ ions from ReA3	Barium in solid Xenon for nEXO	“Typical” Single Molecule Experiment
measured background rate	100000x10³ cps/mW		1x10 ³ cps / mW	0.4x10 ³ cps / mW
fluorescence cross section	(2.3 +/- 1.7) x10⁻¹⁶ cm²	>(9.2 +/- 2.8) x10⁻¹⁶ cm²	(0.01 to 0.1) x10 ⁻¹⁶ cm ²	~0.3 x10 ⁻¹⁶ cm ²
quantum efficiency	(3.1 +/- 0.8)%	>(12 +/- 8)%	>(0.1 to 1)%	>10%
Refs	this work		PRA 91, 022505 (2015) Nature 569 203 (2019)	ARPC 1997. 48:181 RSI 74 3597 (2003)

- 1. Challenge: Background counts is 10⁵ times more than successful single emitter experiments. Remedy: illuminate a smaller area of substrate + spatial filtering**
- 2. Good: Most ions appear to be completely neutralized in medium.**
- 3. Good: Rb fluorescence cross section appears to be x10 larger than for other successful single emitter imaging experiments.**
- 4. Next up: study the effect of photobleaching.**

Thanks for your attention!



Fry Fang
BS 2017
now at
Notre Dame

Ben Loseth
PhD 2019

“...stars somehow produce Tc as they go along...”

Technetium in the Stars

Paul W. Merrill

Mount Wilson and Palomar Observatories

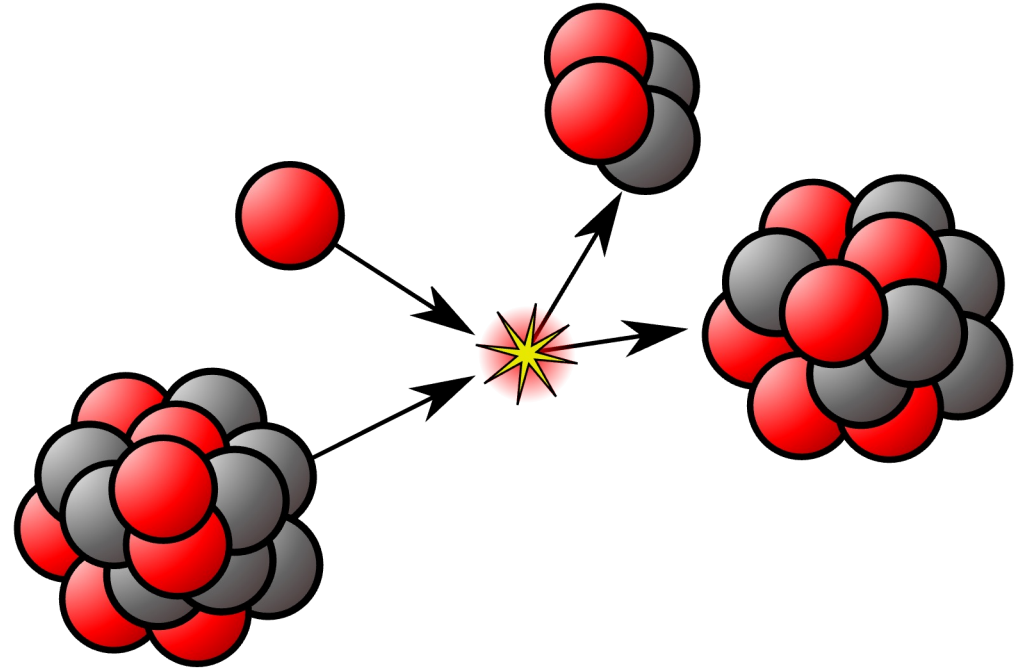
Technetium, the first “artificial” element, was identified in 1937 by Perrier and Segrè in a piece of molybdenum that had been bombarded with neutrons in the cyclotron at Berkeley. Technetium has also been detected among the products of fission of heavy atoms. No completely stable isotope is known; the most nearly stable has a half-life less than a million years.

The spectrum of technetium was thoroughly investigated in 1950 by Meggers and Scribner at the National Bureau of Standards. Their work has made astronomical investigations possible. In 1951 Charlotte E. Moore announced the possible presence of weak lines of ionized technetium in the solar spectrum.

It is surprising to find an unstable element in the stars. Either (1) a stable isotope actually exists although not yet found on earth; or (2) S-type stars somehow produce technetium as they go along; or (3) S-type stars represent a comparatively transient phase of stellar existence.

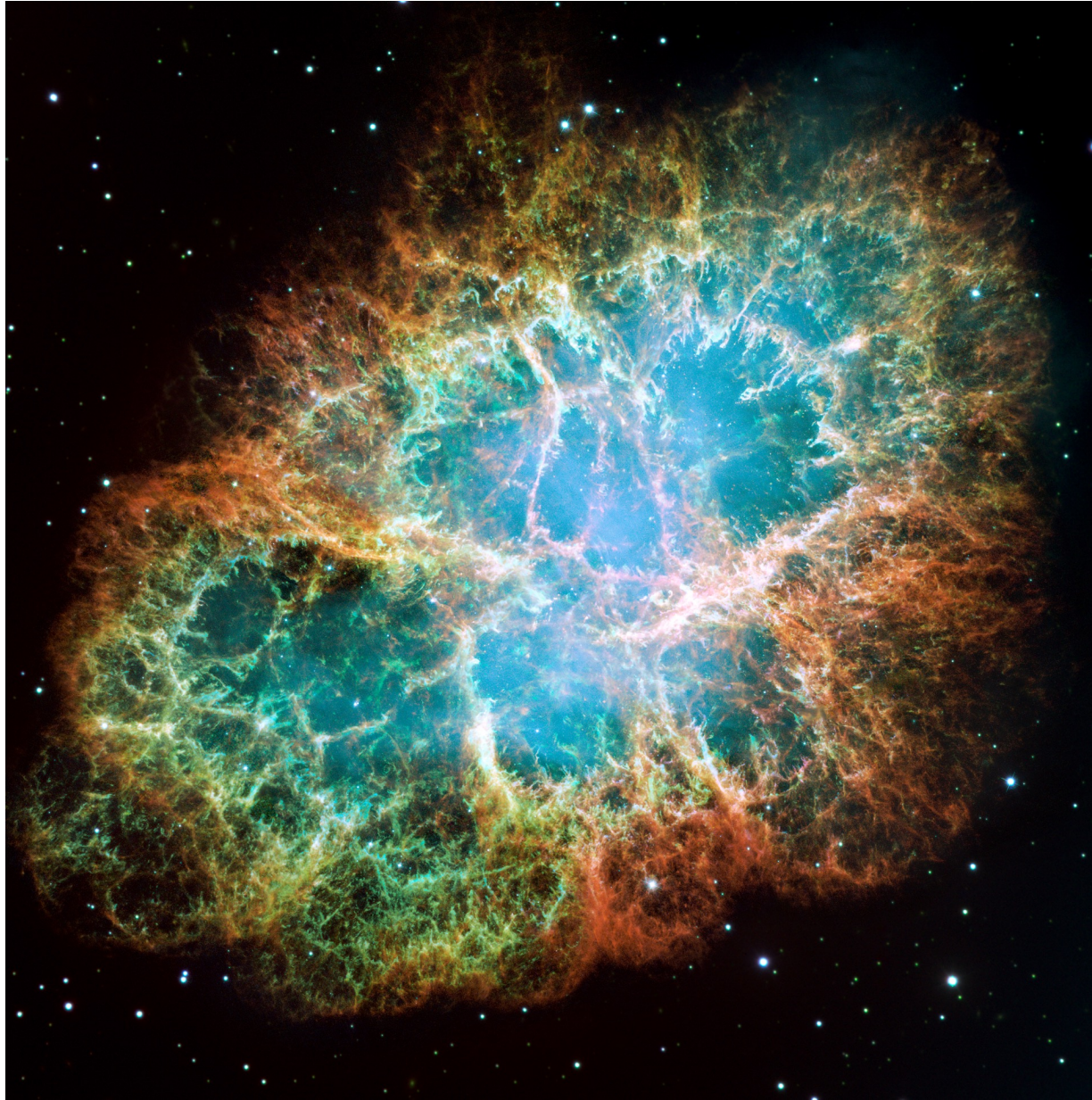
Science 115, 484 (1952)

Nuclear reactions inside of stars...



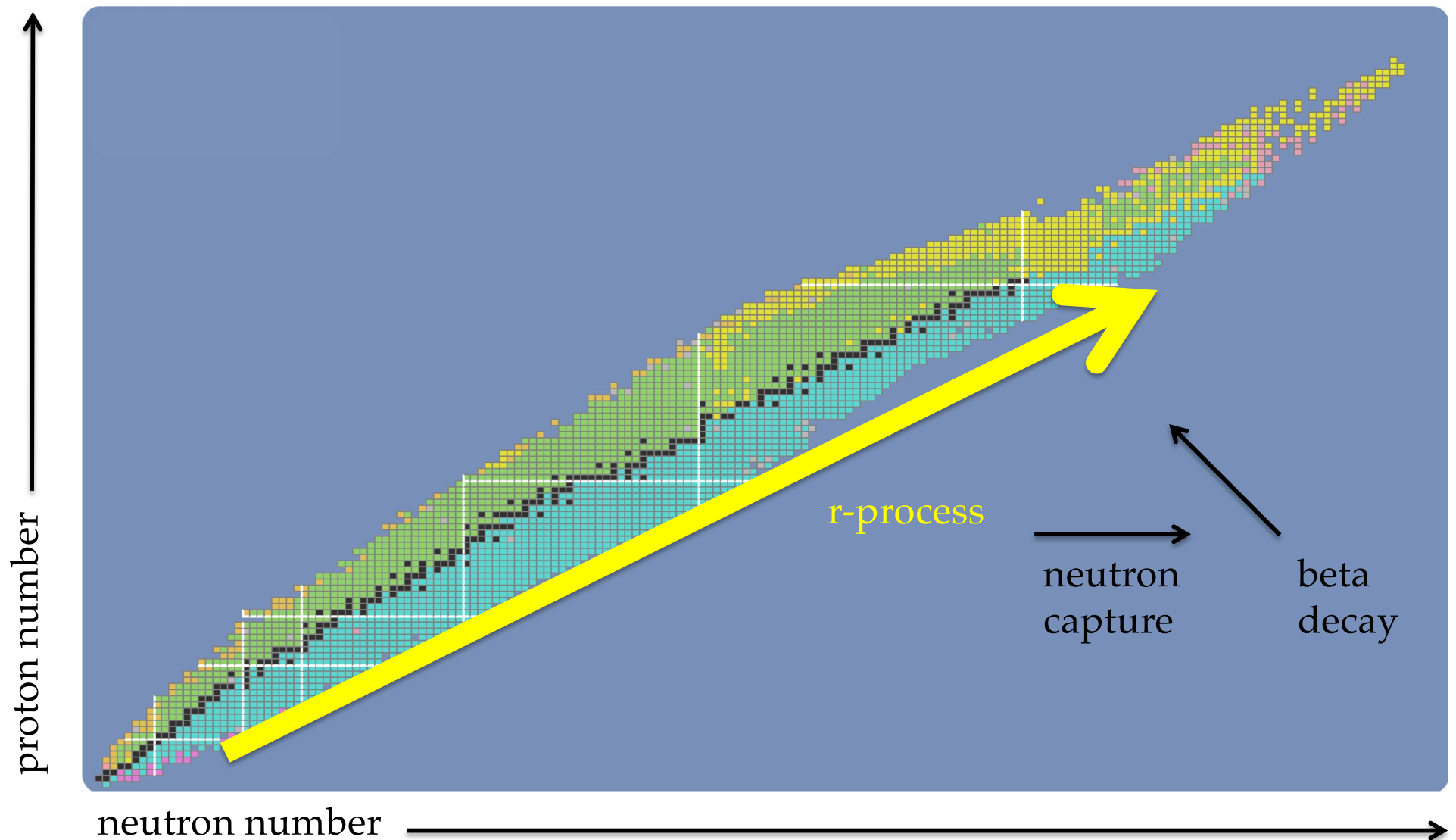
By Kjerish - Own work, CC BY-SA 4.0,
<https://commons.wikimedia.org/w/index.php?curid=54378478>

We are all made of dead stars!



By NASA, ESA, J. Hester and A. Loll (Arizona State University) -
HubbleSite: gallery, release, Public Domain,
<https://commons.wikimedia.org/w/index.php?curid=516106>

“Rapid” Neutron Capture Process



<https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html>

Neutron Star Mergers: Site of the r-process?

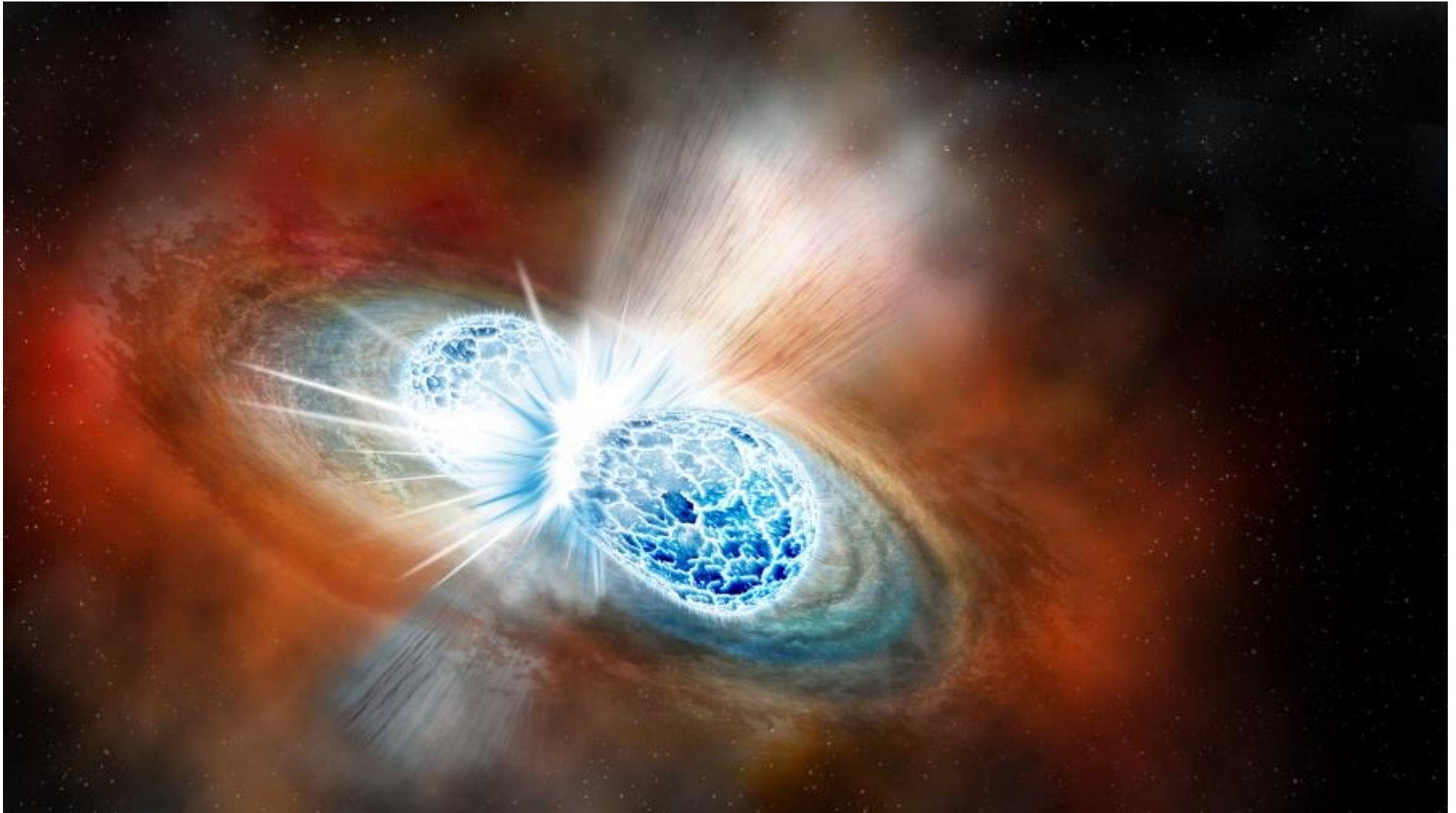
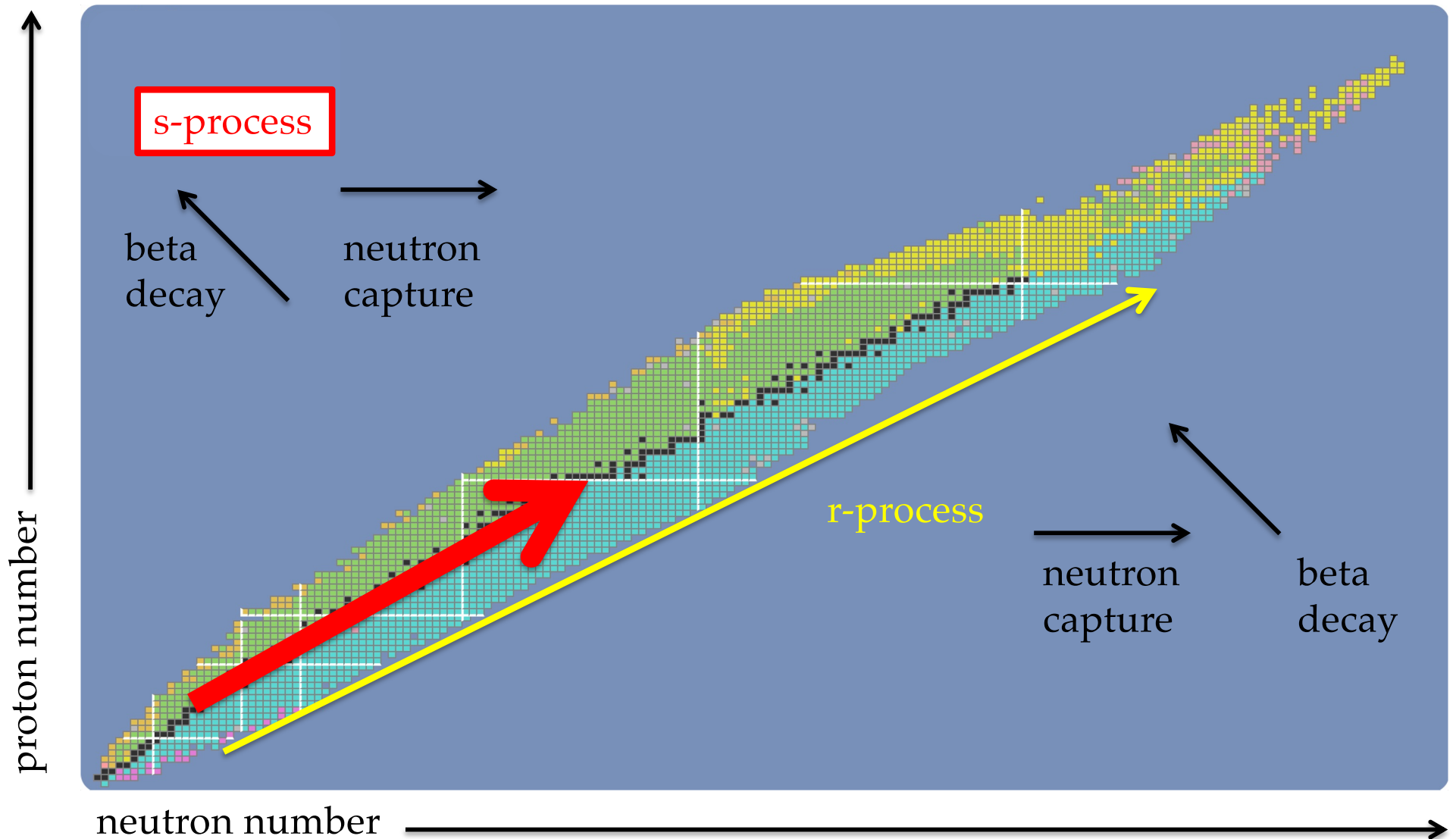


Illustration by Robin Dienel courtesy of the Carnegie Institution for Science.

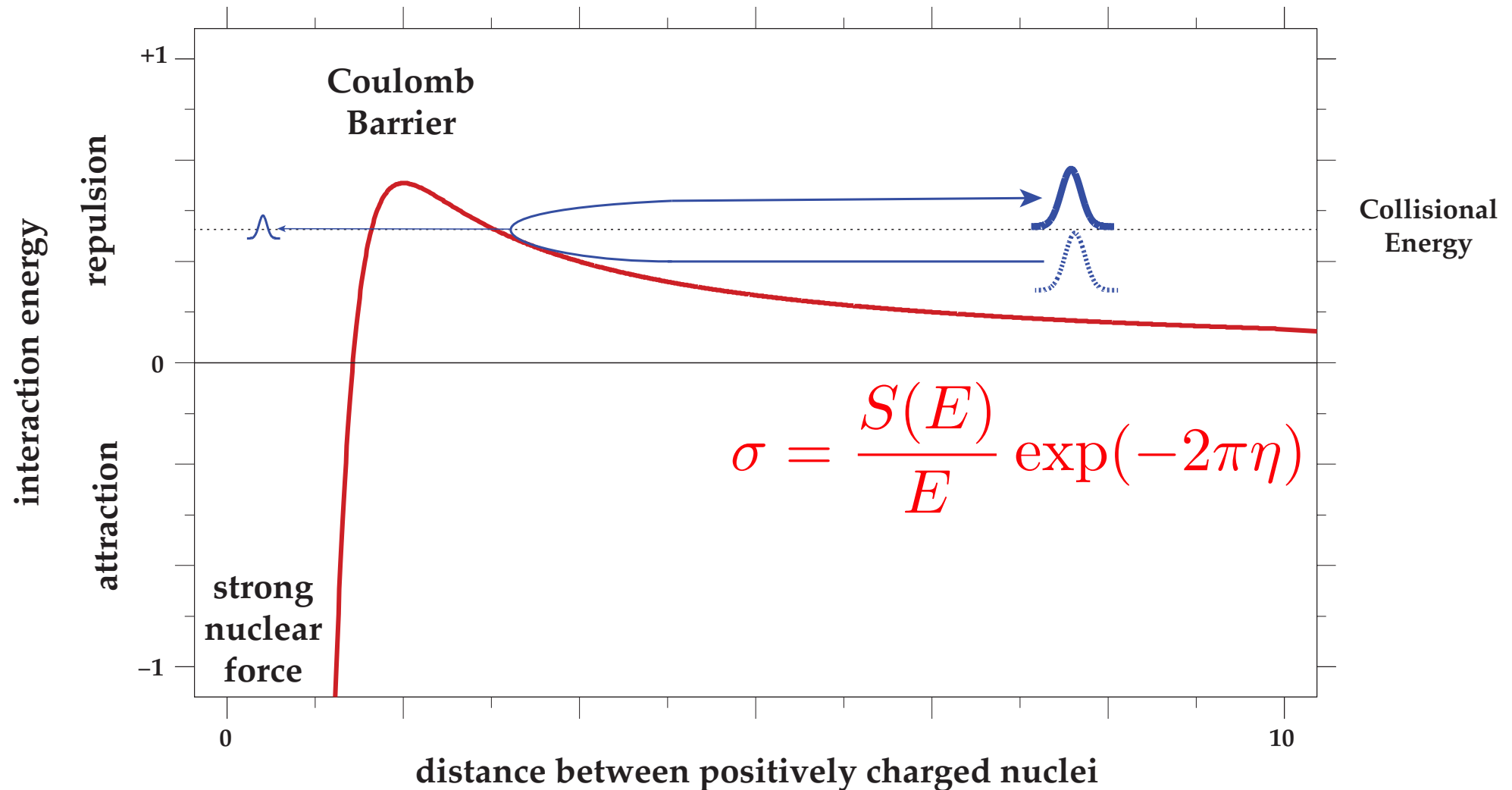
<https://carnegiescience.edu/news/new-era-astronomy-begins-first-ever-observation-two-neutron-stars-colliding>

“Slow” Neutron Capture Process



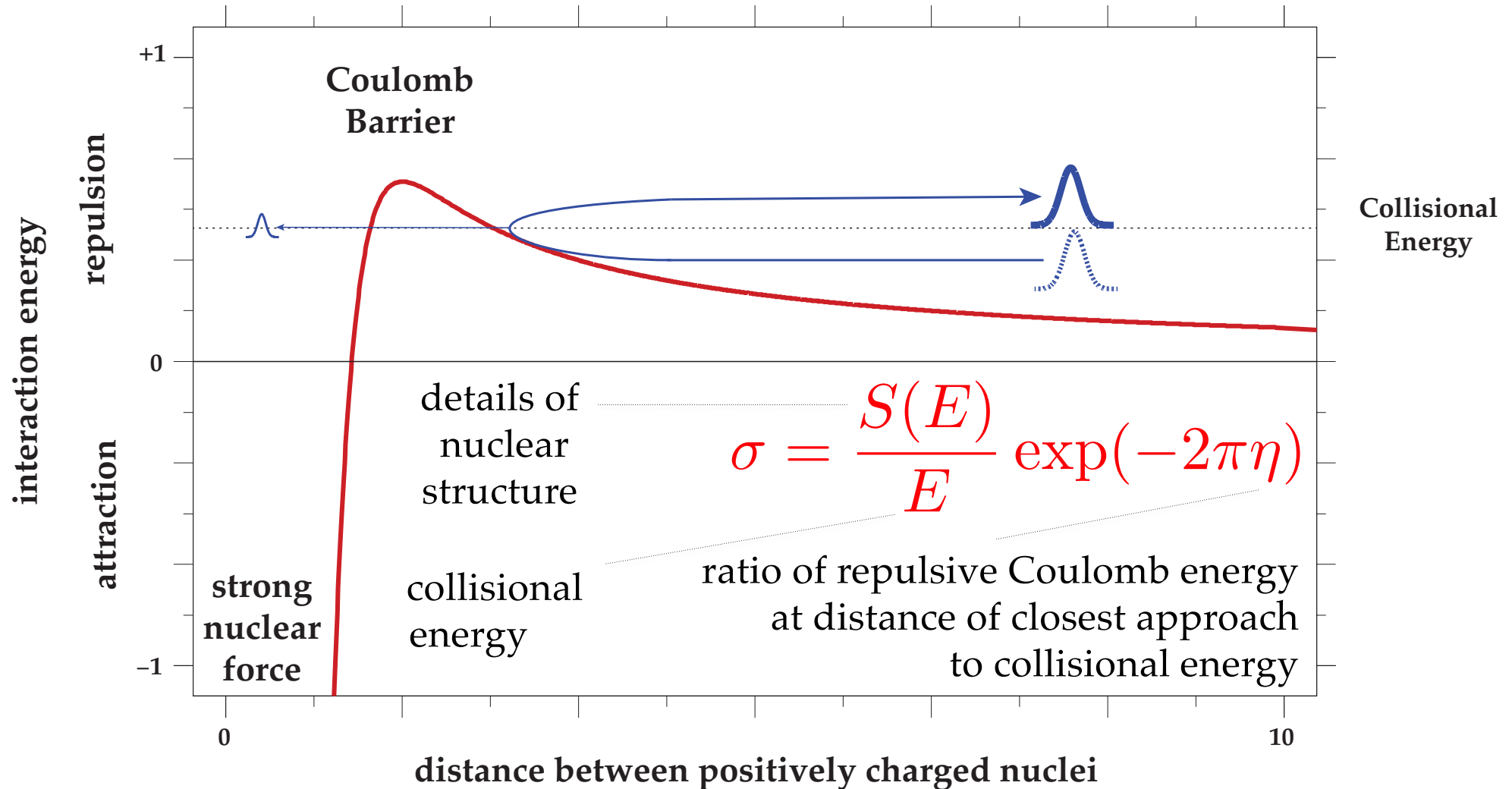
<https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html>

Fusion Reaction is Unlikely Due To “Coulomb Barrier”



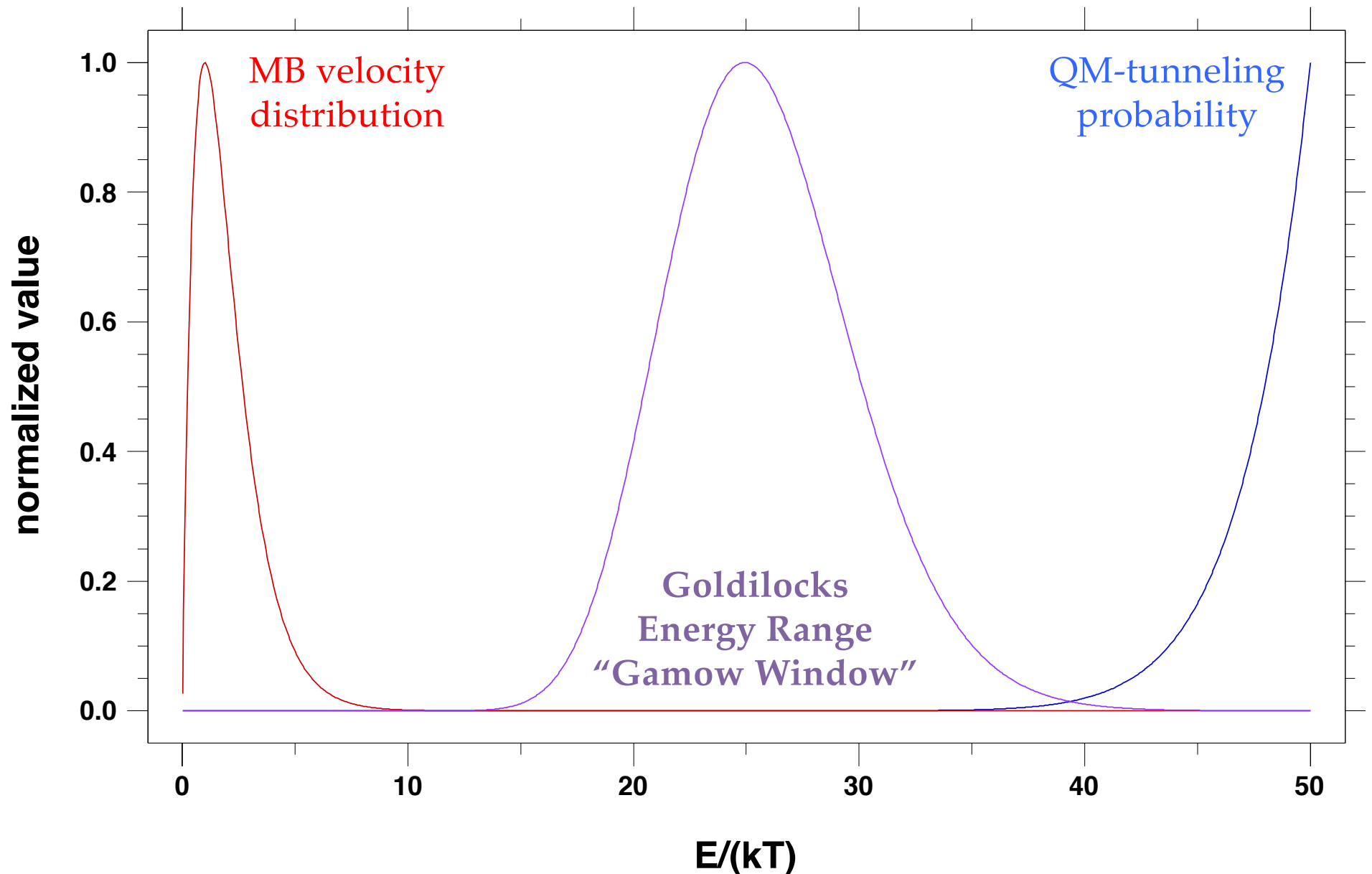
In the quantum mechanical picture, the positively charged nuclei repel each other but are able to interact via “Tunneling.”

Reaction is Unlikely Due To “Coulomb Barrier”

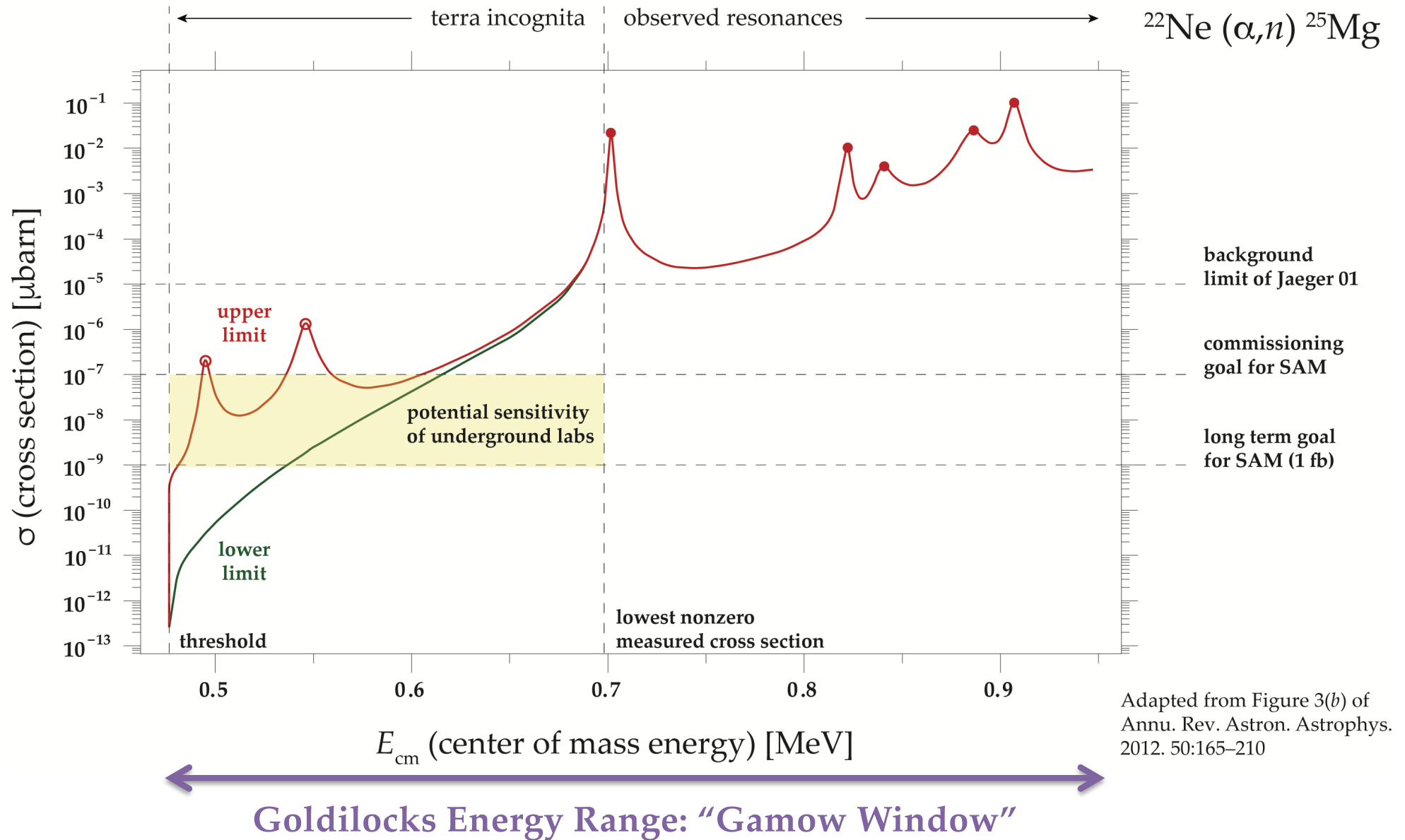


In the quantum mechanical picture, the positively charged nuclei repel each other but are able to interact via “Tunneling.”

Collisional Energy \rightarrow Temperature of Star

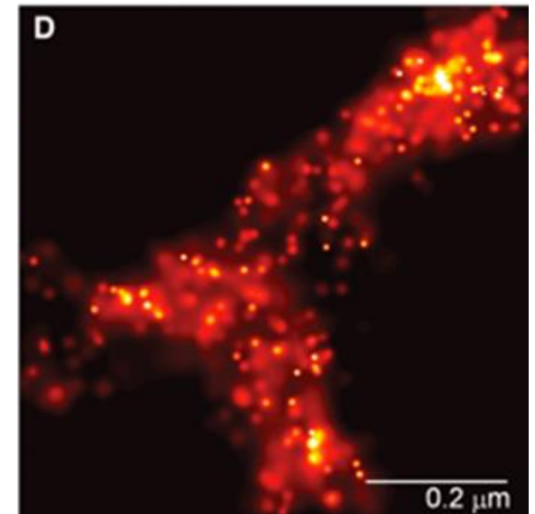
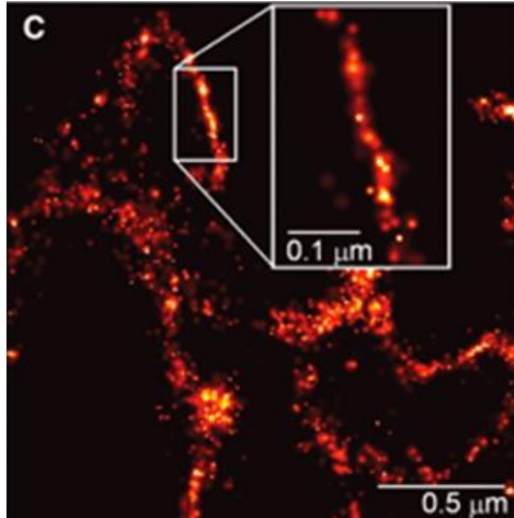
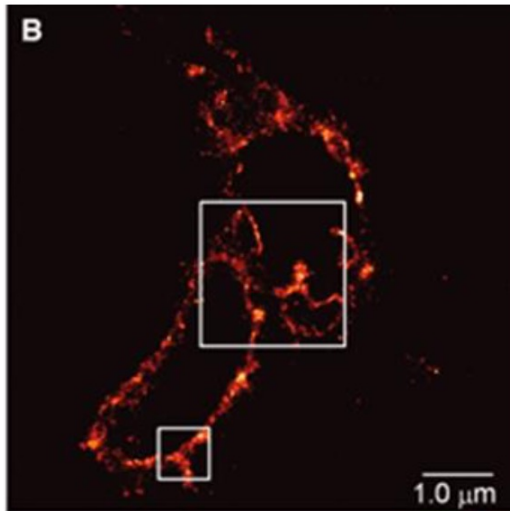


$^{22}\text{Ne} + ^4\text{He}$: Key Source of Neutrons for s-Process



Adapted from Figure 3(b) of
Annu. Rev. Astron. Astrophys.
2012. 50:165–210

Single Emitter Imaging in Condensed Media



Betzig et al., *Science*, 2006, 313, 1642-1645

Nobel Prize Chemistry 2014

Mature tools & techniques can be borrowed from Single Molecule Biophysics!

Ba tagging for EXO

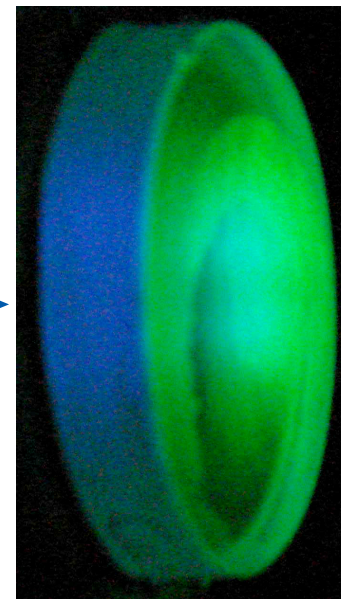
M. K. Moe PRC 44 R931 (1991)

W. Fairbank et al. @ Colorado State

Single Ba detection in s-Xe and laser scanning have been demonstrated!

Nature 569, 203 (2019)

Blue
Laser
Light
(388 nm) →



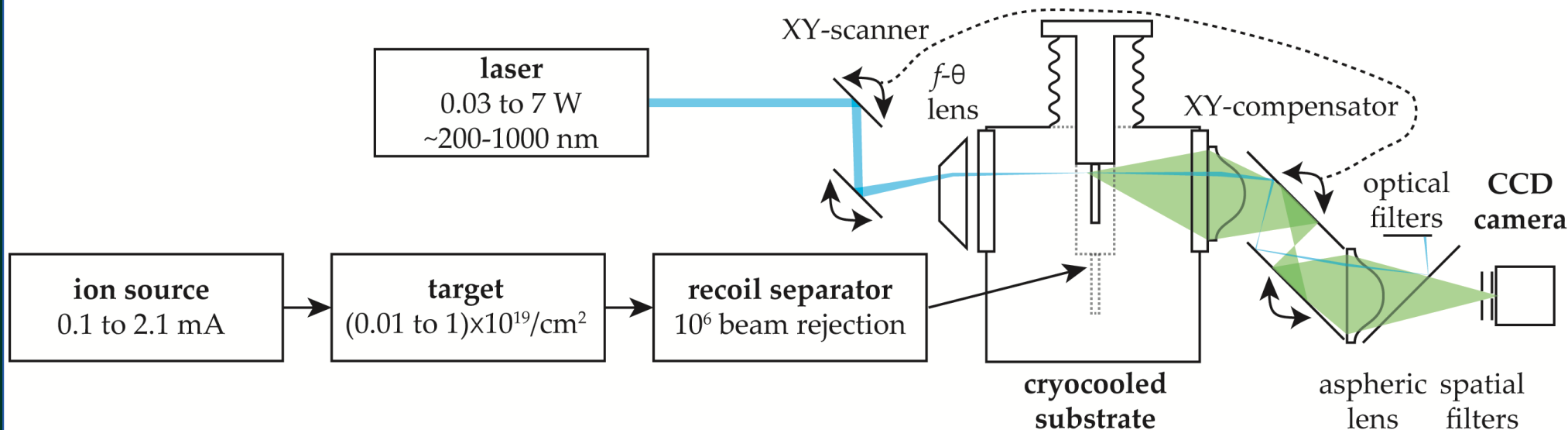
1 Yb : 10^6 Ne

1" diameter
CaF₂ substrate

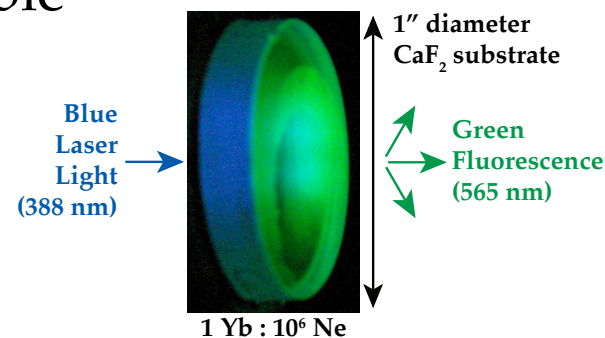
Green
Fluorescence
(565 nm)

Yb in s-Ne:
PRL 107, 093001
PRL 113, 033003

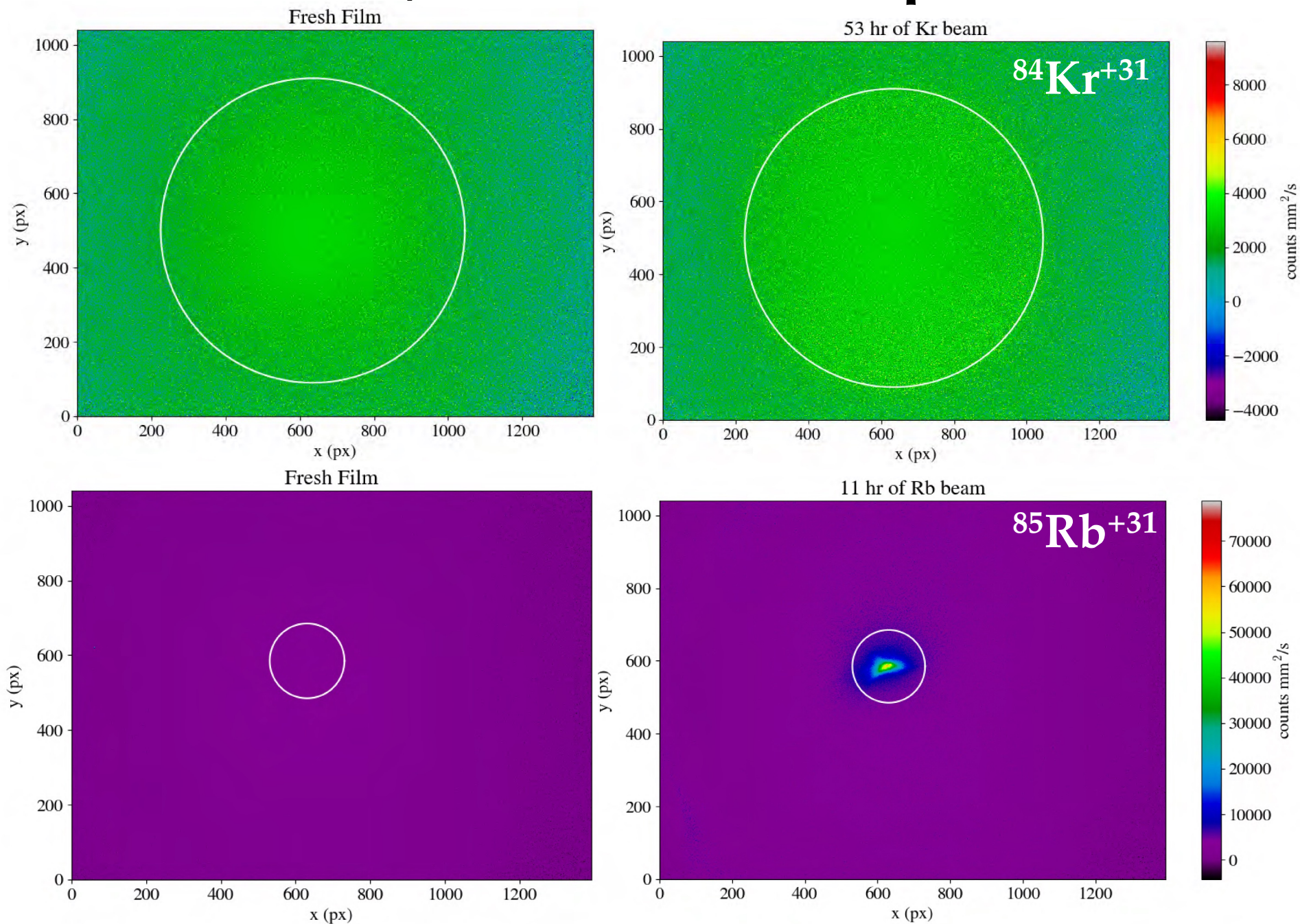
Basic Concept of Measurement Using SAM



- Efficient: cryogenic NG film captures everything (both products and beam)
- Selective: product atoms identified by localized resonant laser excitation
- Sensitive: large shift (few nm to 100's of nm) between **excitation spectrum** and **emission spectrum** coupled with spatial & optical filtering makes optical single atom detection feasible
- **Recoil separator is needed to:**
 - minimize heat load on NG film from beam
 - discriminate between isotopes



Fluorescence yields from ion-implanted beams



Main Technical Challenge: Suppressing Sources of Optical Background

Impurity	Source	Wavelength	Notes
all surfaces	excitation light	blue	optical filter
Nitrogen	vacuum residual gas	< 200 nm	too far off resonance
Oxygen	vacuum residual gas	< 245 nm	too far off resonance
Ozone	vacuum residual gas	< 350 nm	too far off resonance
Water	vacuum residual gas	< 210 nm	too far off resonance
“Stuff”	UVFS viewports	~green	needs more study
Cr³⁺	sapphire substrate	690 nm + broadband tail	needs more study
Apiezon N	inside cryostat	broadband green	don't use this
“Stuff”	surface of substrate	broadband green	needs more study

Plans to mitigate this:

- pre-photobleaching of impurities before measurement
- confocal optics
- aggressive surface treatments
- low impurity substrate materials

Some SAM-Friendly Atoms

Species	Excitation (nm)	Emission (nm)	Brightness (Hz)	Notes
Be	(225)	(455 & 332)	4.2E-1	(estimated)
Be	(225)	(245)	5.5E8	(estimated)
Mg	275	472 & 518	2.5E1	JCP 101, 10354-65, 1994
Mg	275	296	5.0E8	LTP 38, 679–87, 2012
Ca	(423)	(657 & 1953)	2.6E3	(estimated)
Sr	(461)	(689 & 2739)	4.7E4	(estimated)
Cd	(229)	(326 & 480)	4.1E5	(estimated)
Yb	388	(408)	1.9E8	(estimated)
Yb	388	546 & (1540)	1.1E6	PRL 107:093001, 2011
Li	670	890	~5E7	JCP 73, 3103-6, 1980
Na	587	720	4.6E7	JCP 69, 1670-5, 1978
K	762	900	~5E7	JCP 70, 2404-8, 1979
Rb	776	830	~5E7	JCP 78, 592-3, 1983
Cs	834	970	~5E7	JCP 78, 592-3, 1983