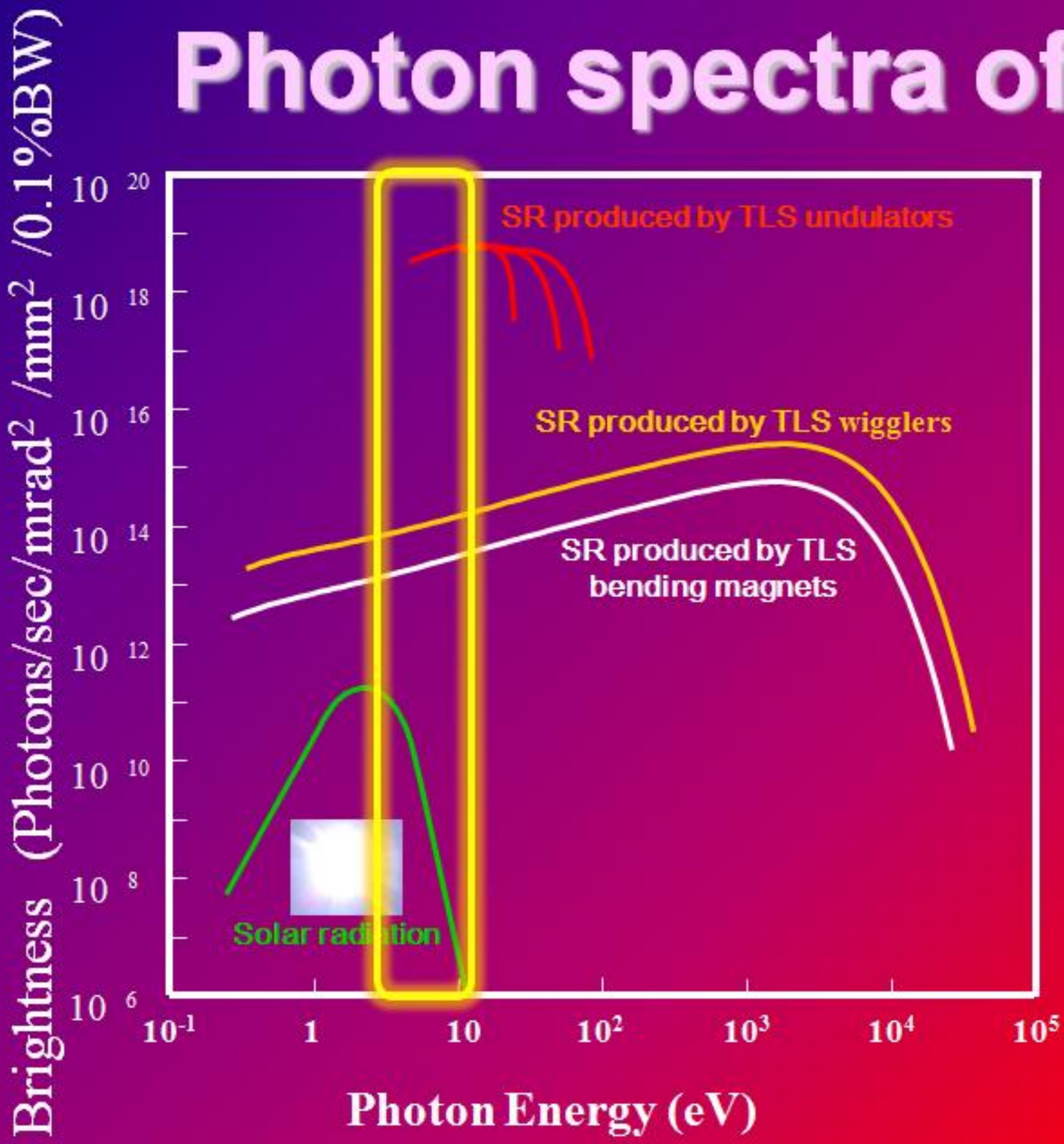
國家同步輻射研究中心
National Synchrotron Radiation Research Center

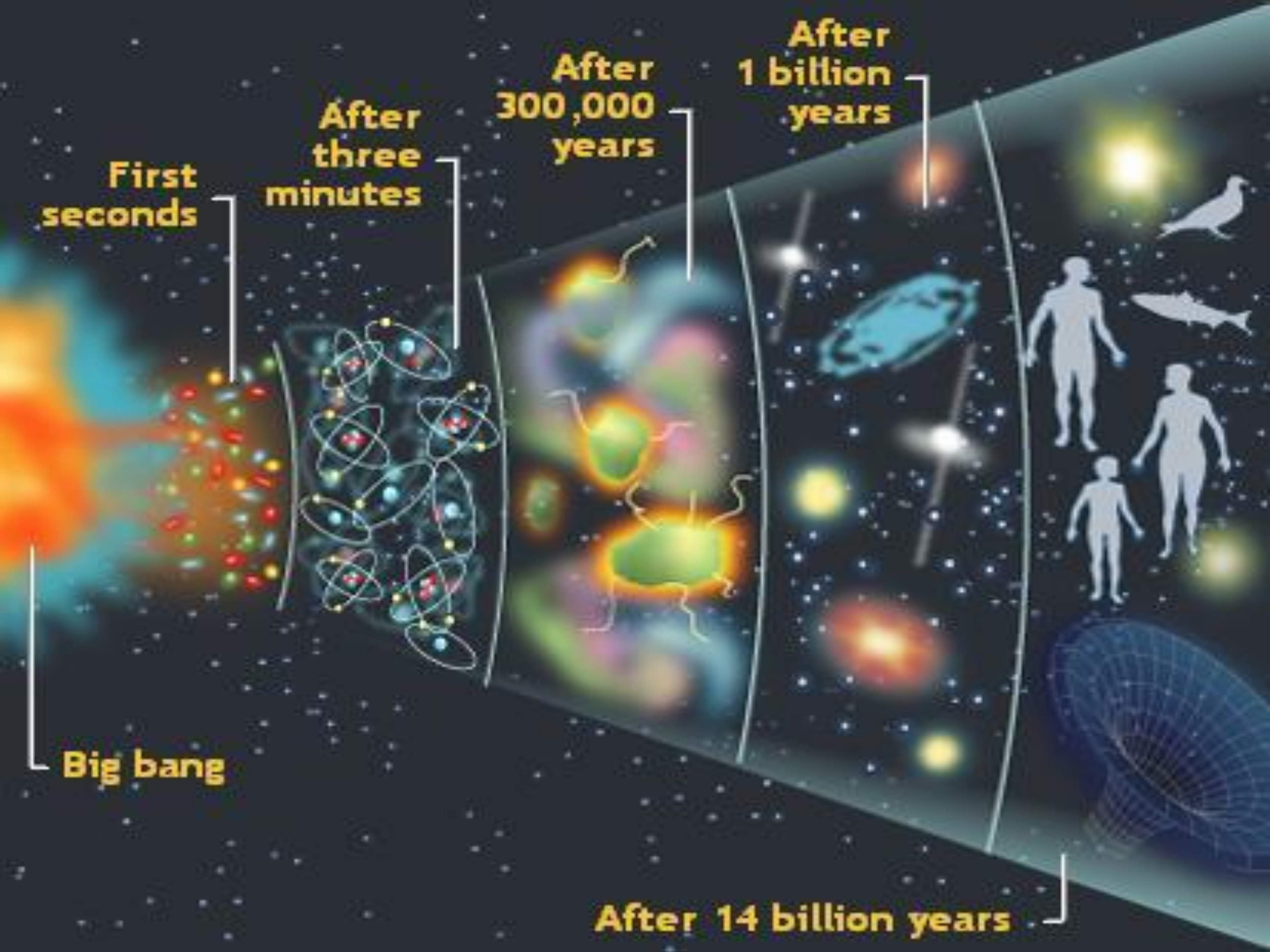
Looking into Space with Synchrotron Far-UV Spectroscopy

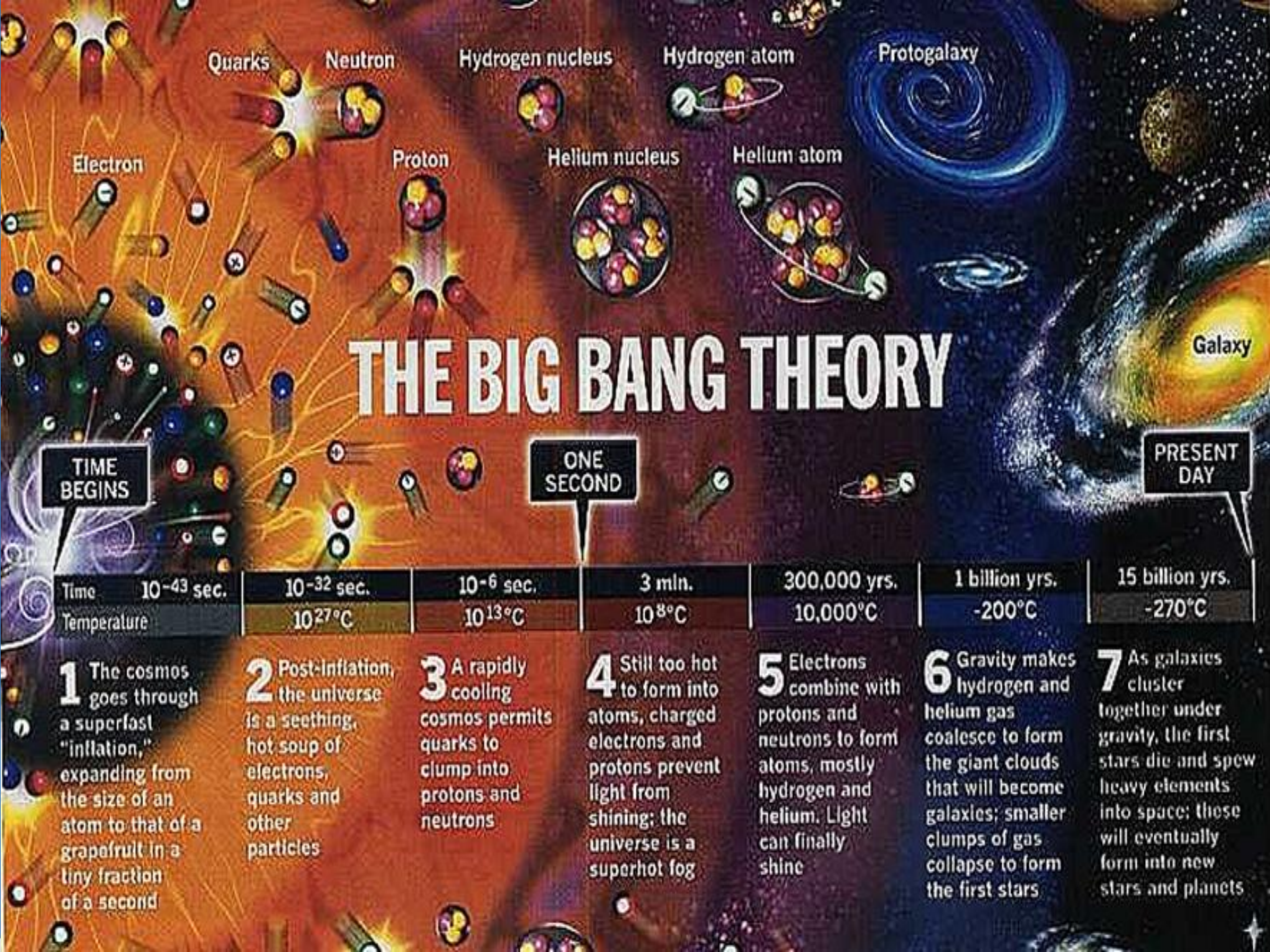
鄭 炳 銘

2015/10/21

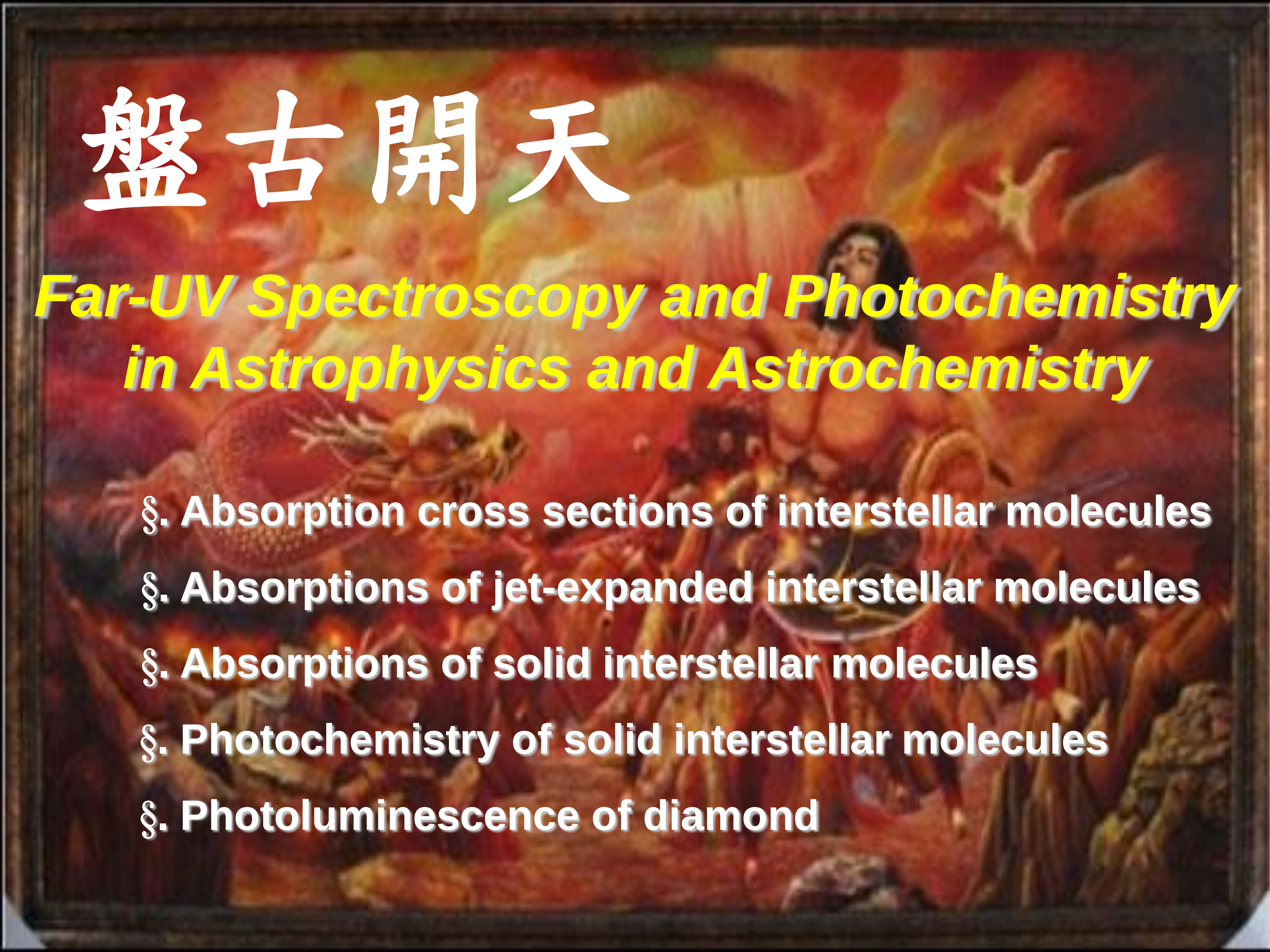
Photon spectra of TLS




First seconds
After three minutes
After 300,000 years
After 1 billion years
Big bang
After 14 billion years



Quarks
Neutron
Hydrogen nucleus
Hydrogen atom
Protogalaxy
Electron
Proton
Helium nucleus
Helium atom
THE BIG BANG THEORY
Galaxy
TIME BEGINS
ONE SECOND
PRESENT DAY
Time
Temperature
10^{-43} sec.
10^{-32} sec.
10^{27}°C
10^{-6} sec.
10^{13}°C
3 min.
10^{8}°C
300,000 yrs.
10,000°C
1 billion yrs.
-200°C
15 billion yrs.
-270°C
1 The cosmos goes through a superfast "inflation," expanding from the size of an atom to that of a grapefruit in a tiny fraction of a second
2 Post-inflation, the universe is a seething, hot soup of electrons, quarks and other particles
3 A rapidly cooling cosmos permits quarks to clump into protons and neutrons
4 Still too hot to form into atoms, charged electrons and protons prevent light from shining; the universe is a superhot fog
5 Electrons combine with protons and neutrons to form atoms, mostly hydrogen and helium. Light can finally shine
6 Gravity makes hydrogen and helium gas coalesce to form the giant clouds that will become galaxies; smaller clumps of gas collapse to form the first stars
7 As galaxies cluster together under gravity, the first stars die and spew heavy elements into space; these will eventually form into new stars and planets



盤古開天

Far-UV Spectroscopy and Photochemistry in Astrophysics and Astrochemistry

§. Absorption cross sections of interstellar molecules

§. Absorptions of jet-expanded interstellar molecules

§. Absorptions of solid interstellar molecules

§. Photochemistry of solid interstellar molecules

§. Photoluminescence of diamond


宇宙考古
Isotopic Fractionations

Isotopic Fractionations

A key issue in understanding the atmosphere and climate of an extraterrestrial body and their variation is the extent of isotopic fractionation, such as D/H, $^{18}O/^{16}O$, and $^{15}N/^{14}N$.

Isotopic Fractionations

^{1}H	^{2}H	^{3}H
1.00794	2.0141	
99.985%	0.015%	t½ =12.32yrs
Stable	Stable	Cosmogenic/ anthropogenic

^{3}He	^{4}He
3.016	4.0026
0.000137%	99.999863%
Stable	Stable/ Radiogenic

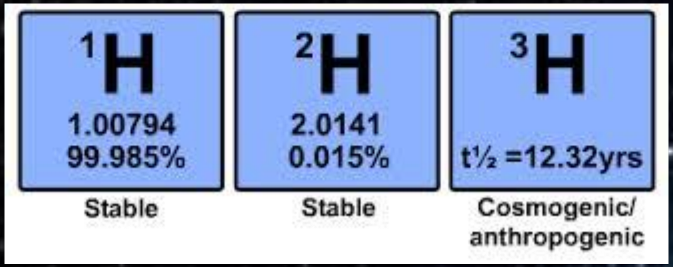
^{12}C	^{13}C	^{14}C
12.00000	13.00335	14.0
98.89%	1.11%	t½ = 5715yrs
Stable	Stable	Radioactive Cosmogenic/ anthropogenic

^{14}N	^{15}N
14.00307	15.0001
99.63%	0.37%
Stable	Stable

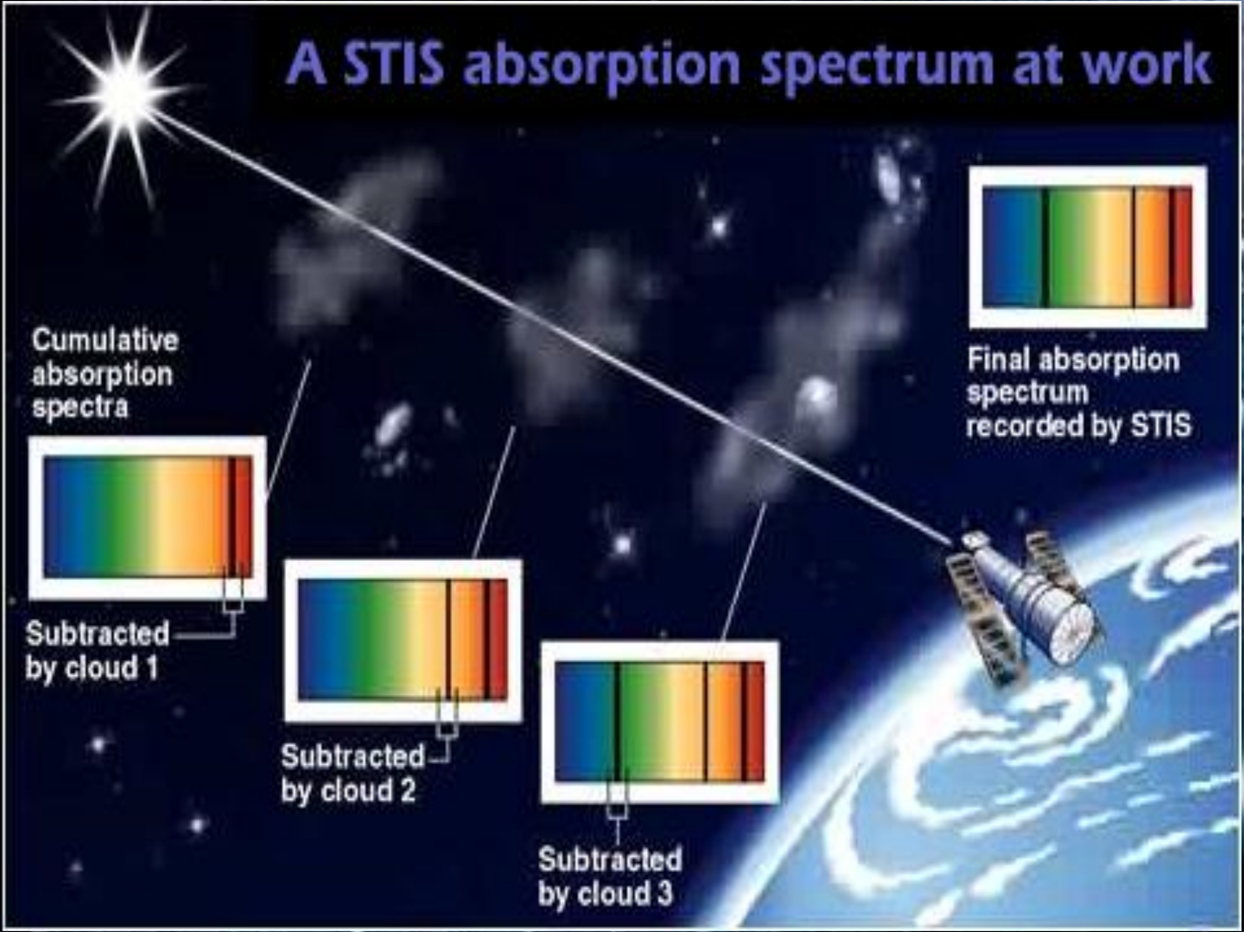
^{16}O	^{17}O	^{18}O
15.9949	16.9991	17.9991
99.76%	0.04%	0.20%
Stable	Stable	Stable

^{32}S	^{33}S	^{34}S	^{36}S
31.97207	32.97145	33.96786	35.96708
95.02%	0.75%	4.21%	0.02%
Stable	Stable	Stable	Stable

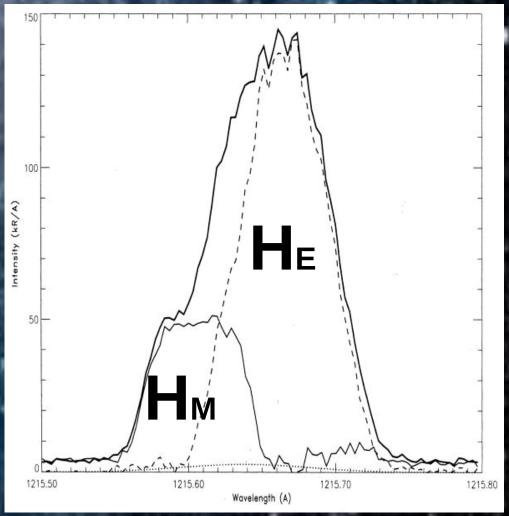
D/H ratios in the solar system and interstellar clouds

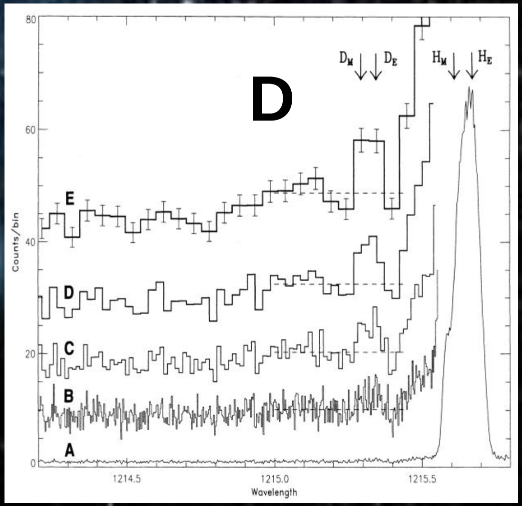


D/H ratios in the solar system and interstellar clouds

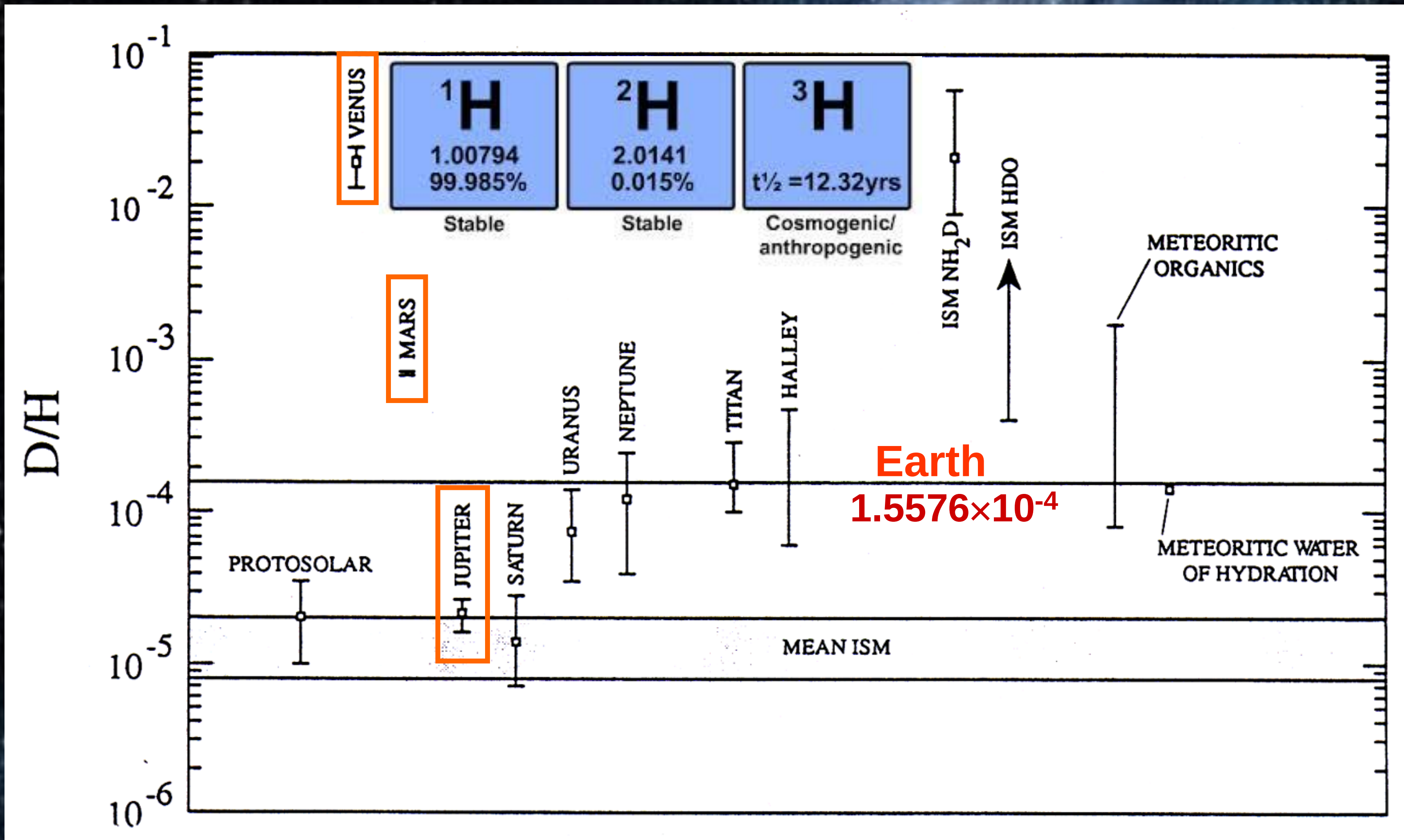


Hubble Space Telescope





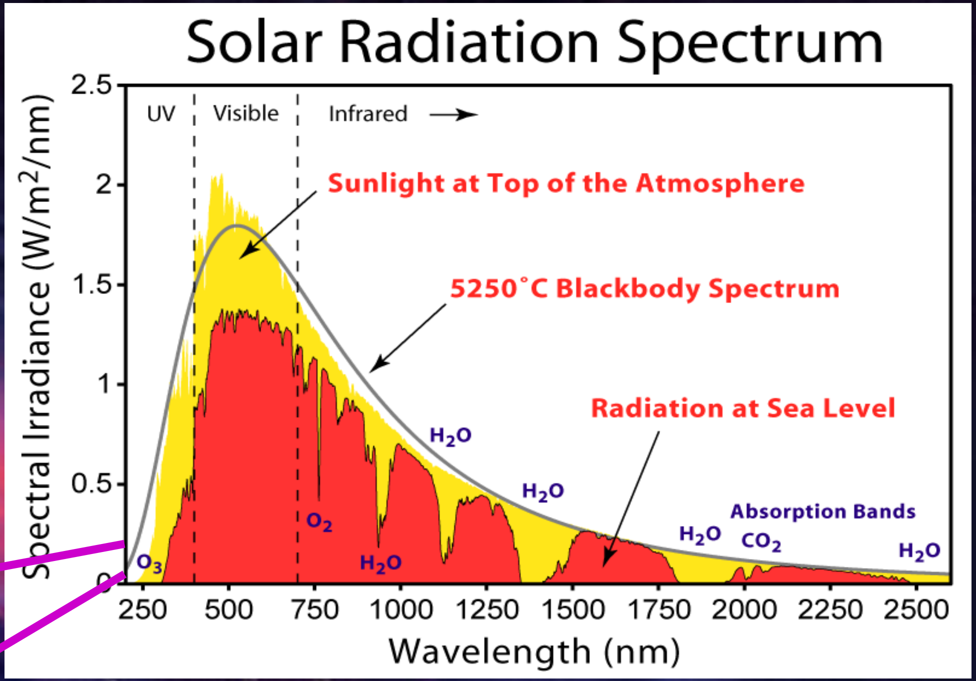
D/H ratios in the solar system and interstellar clouds

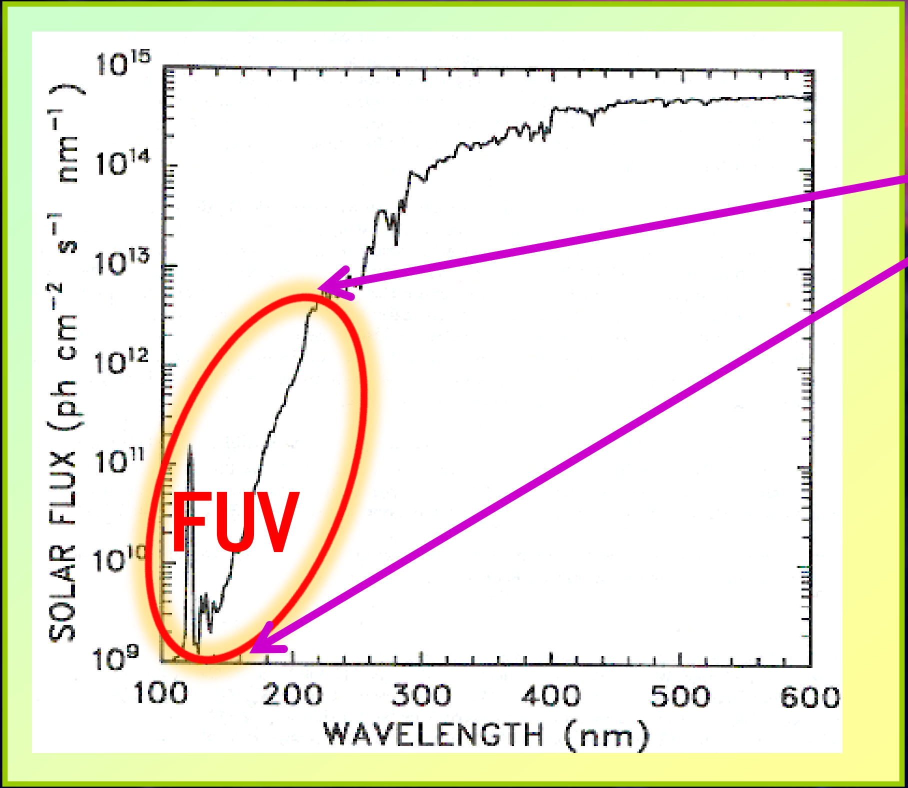


PHIFE

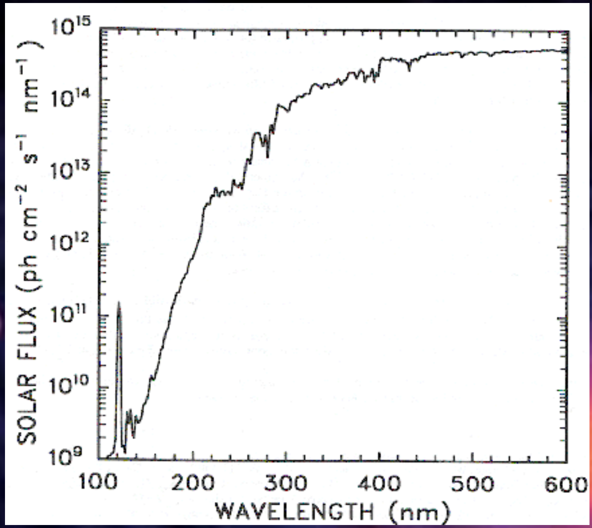
A mechanism proposed to explain variations in isotopic fractionation on astro-environments is called the photo-induced fractionation effect (PHIFE). The crucial idea is that the rate of photolysis differs greatly for various isotopes near the threshold region.

Spectrum of Solar flux

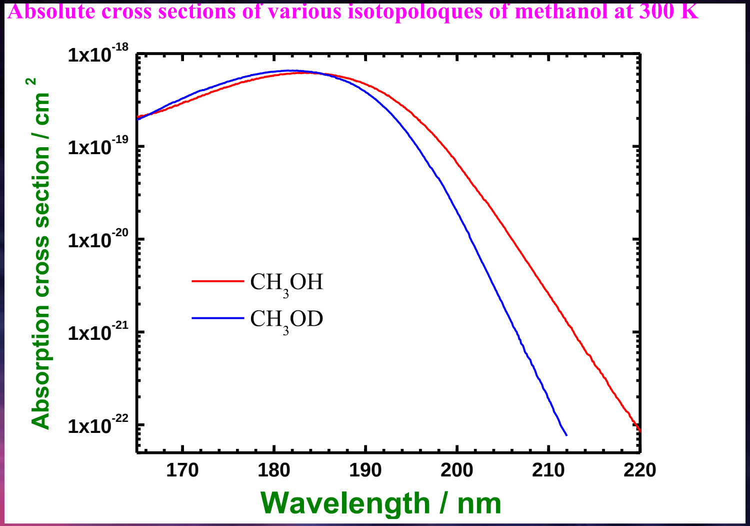


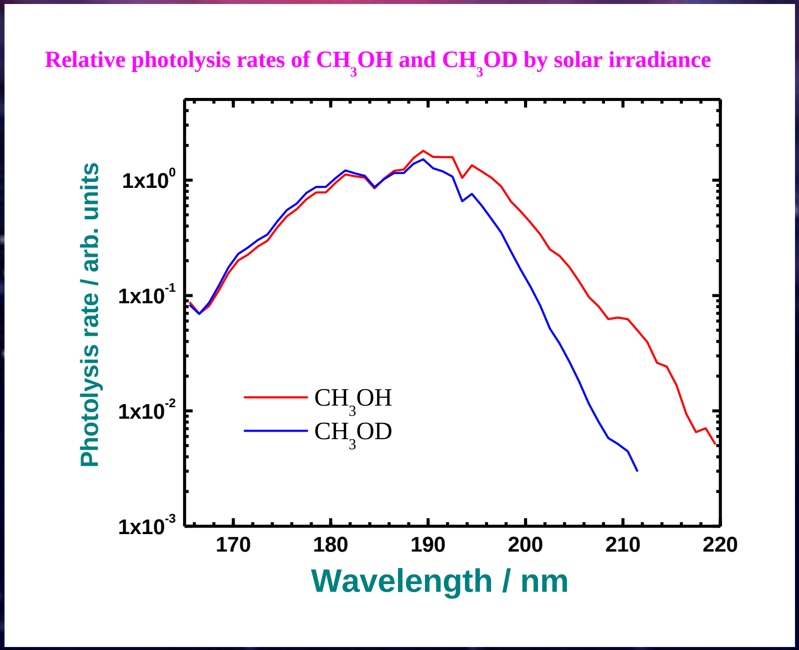


Dissociation Rates by Sun

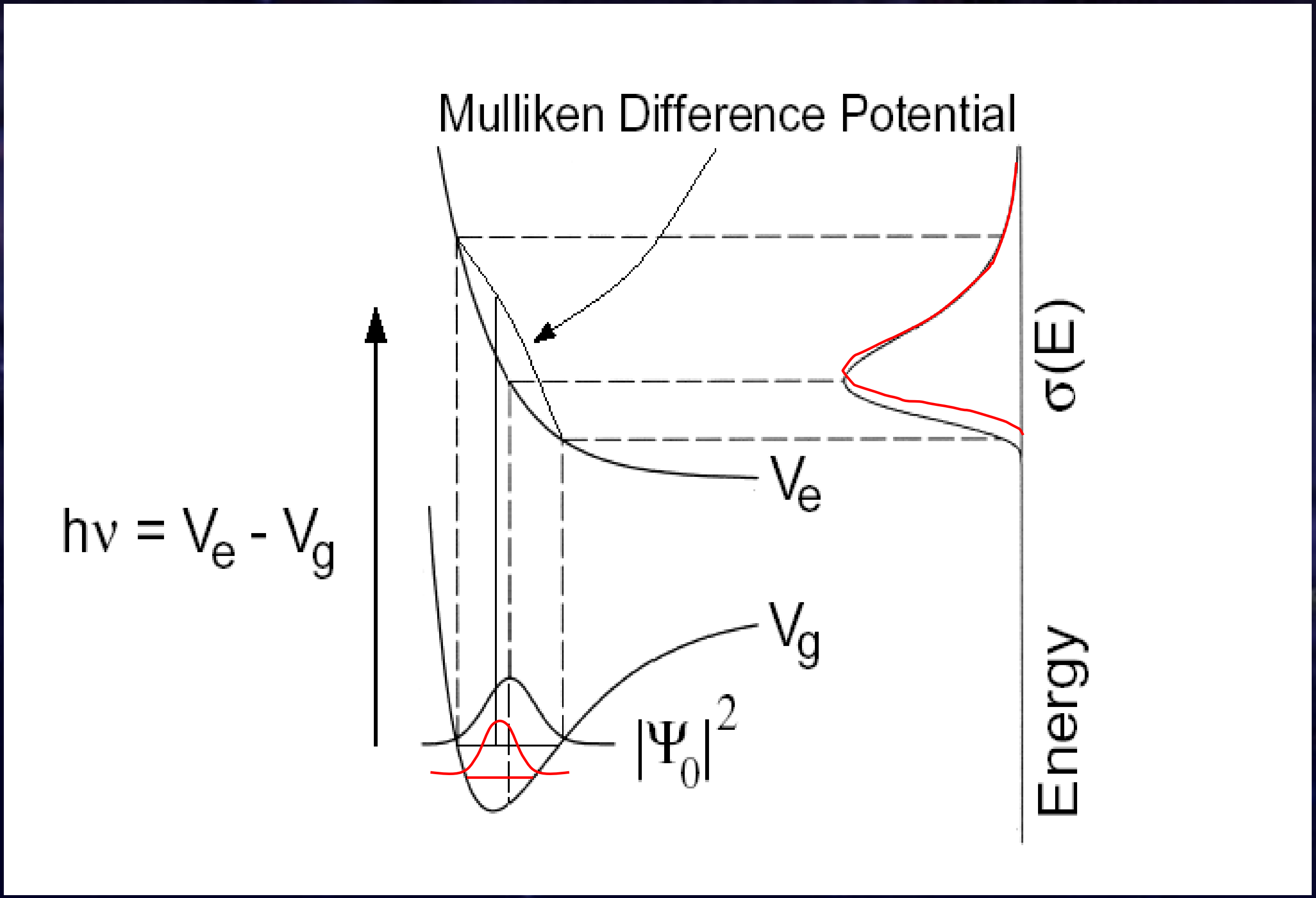


X





Electronic excitations vs absorption specta

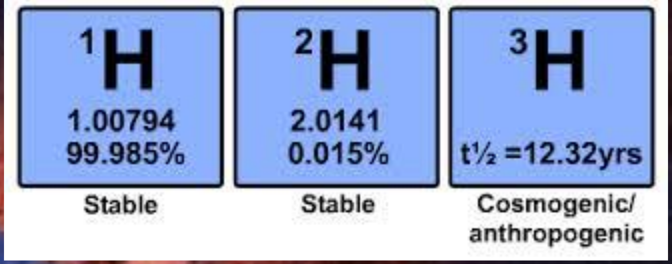


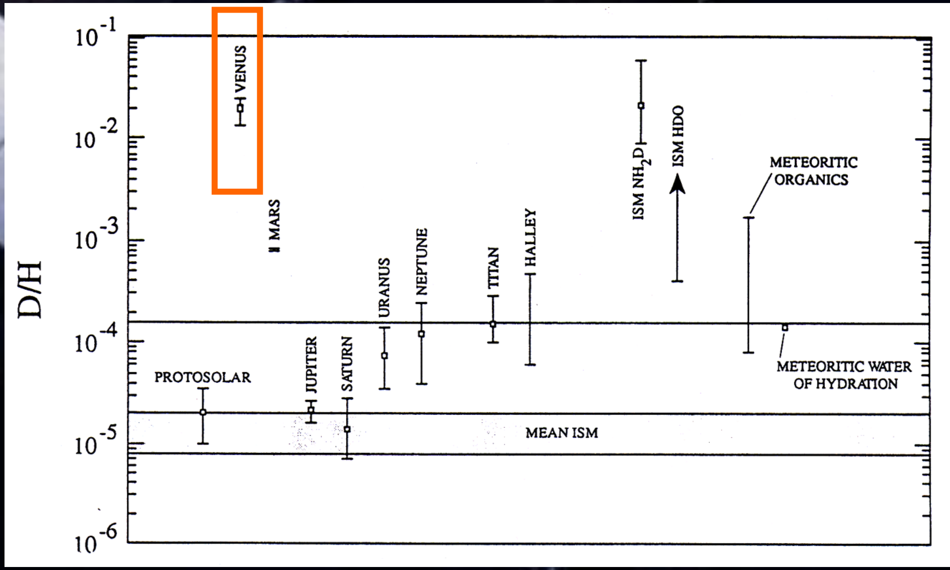
I. FUV absorption cross sections

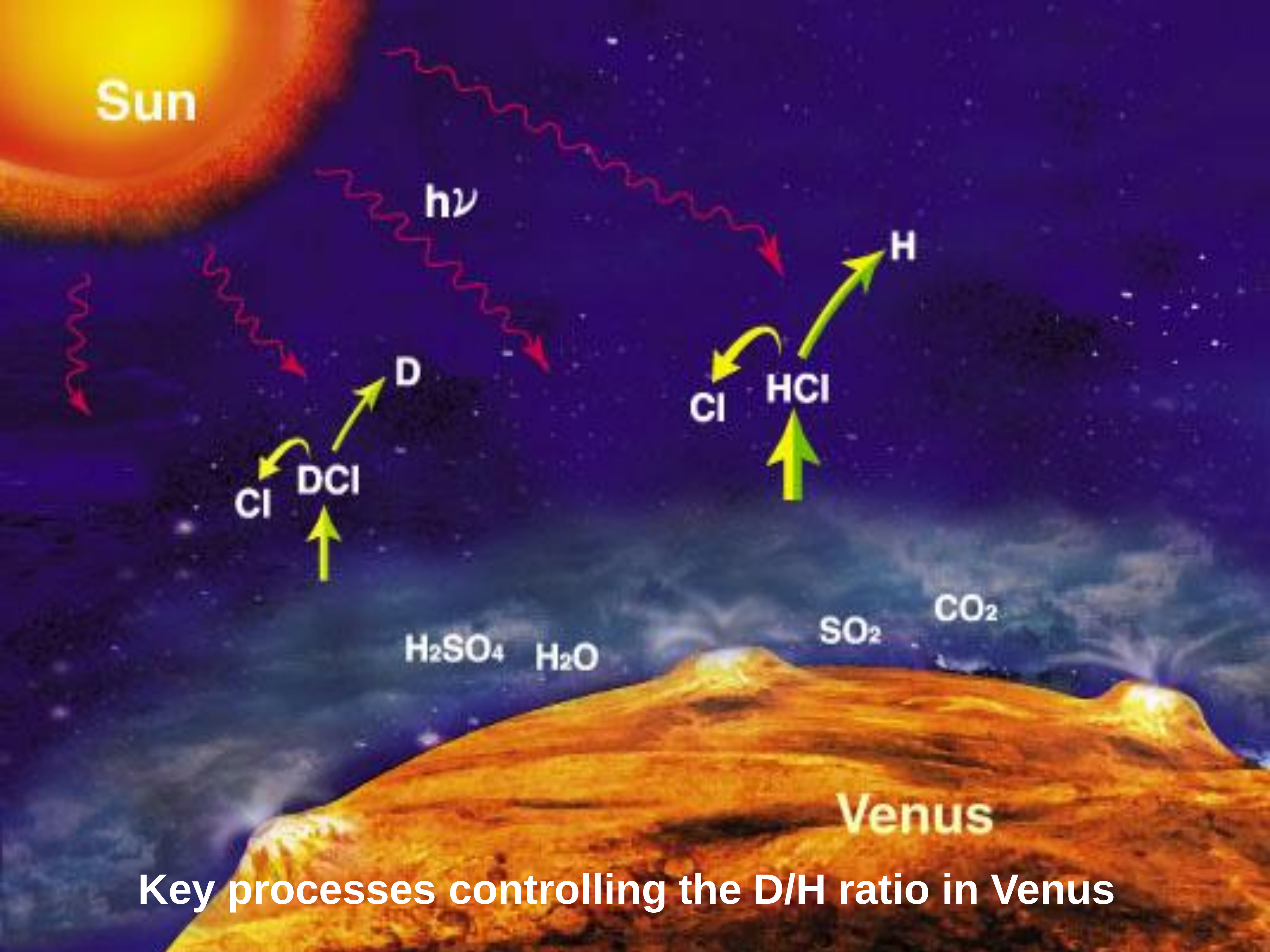
To assess the photochemically induced changes in planets, we must know absolute absorption cross sections of planetary molecules in the far-UV range.

Absorption cross sections of interstellar molecules have been extensively measured in the far-UV range, but such data for their isotopes are lacking and need to be established.

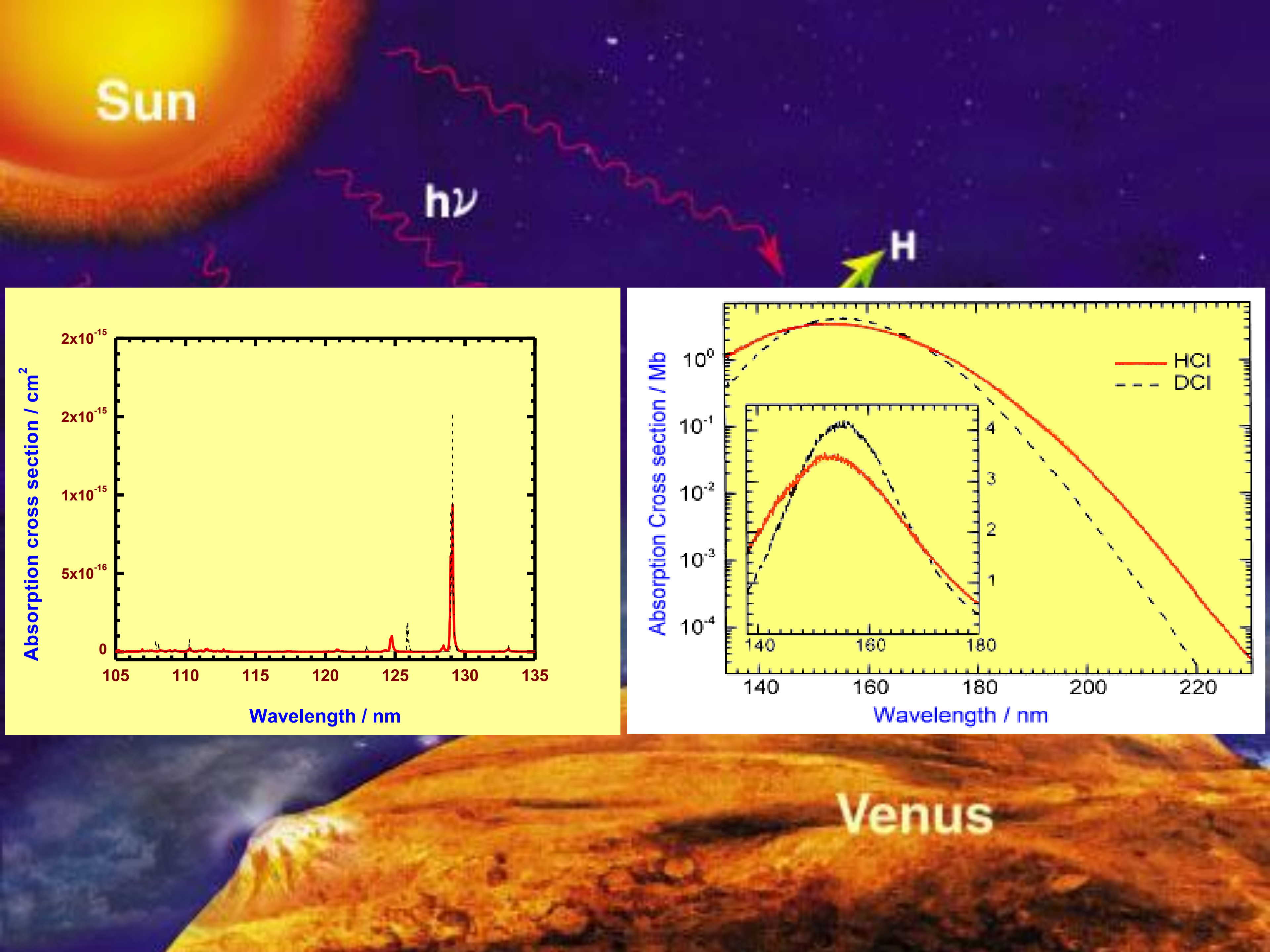
D/H ratios in the solar system and interstellar clouds



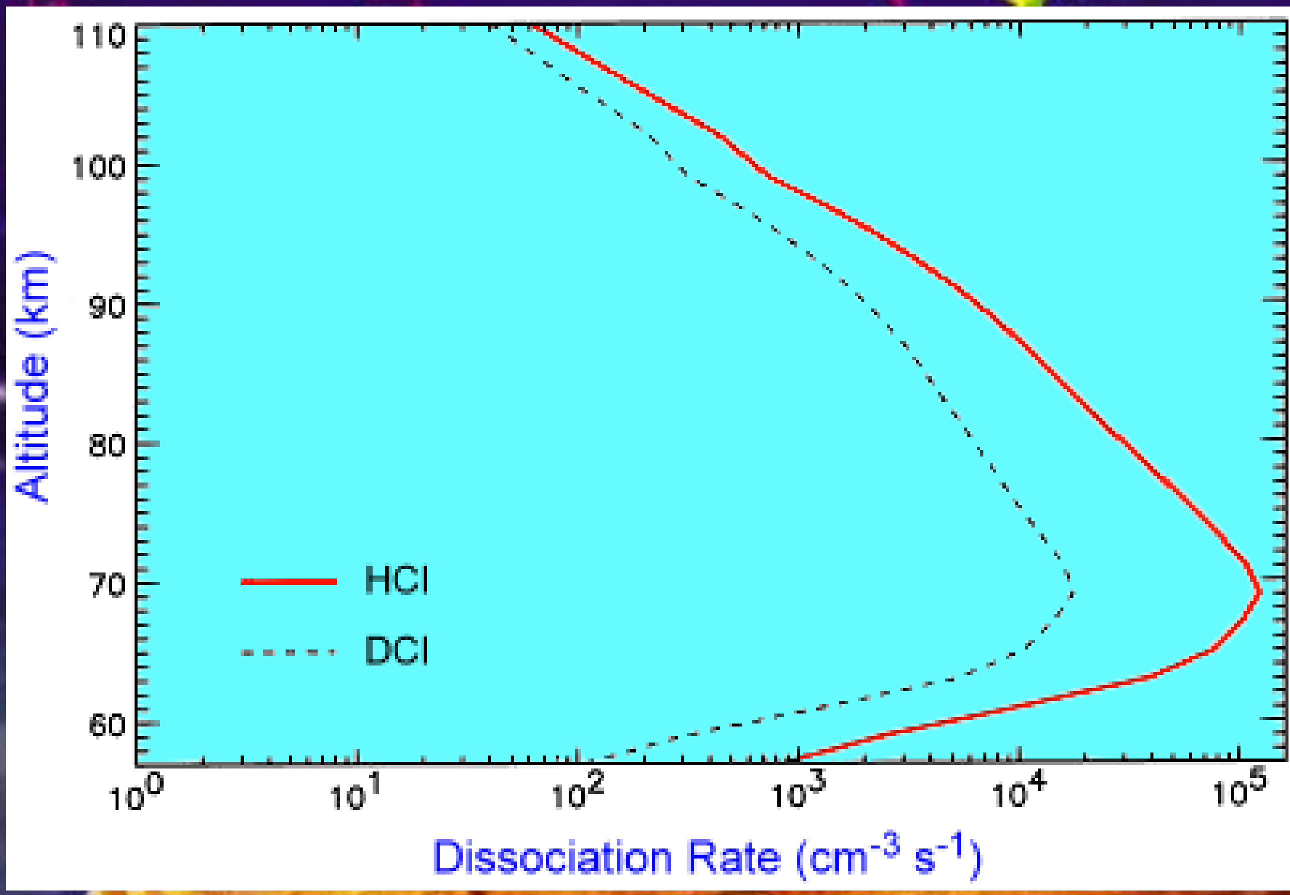




Key processes controlling the D/H ratio in Venus


Sun
hν
H
Venus
Absorption cross section / cm2
2x10-15
2x10-15
1x10-15
5x10-16
0
105
110
115
120
125
130
135
Wavelength / nm
Absorption Cross section / Mb
10^0
10^-1
10^-2
10^-3
10^-4
HCl
DCl
4
3
2
1
140
160
180
140
160
180
200
220
Wavelength / nm

Comparison of photolysis rates of HCl And DCl in the atmosphere of Venus

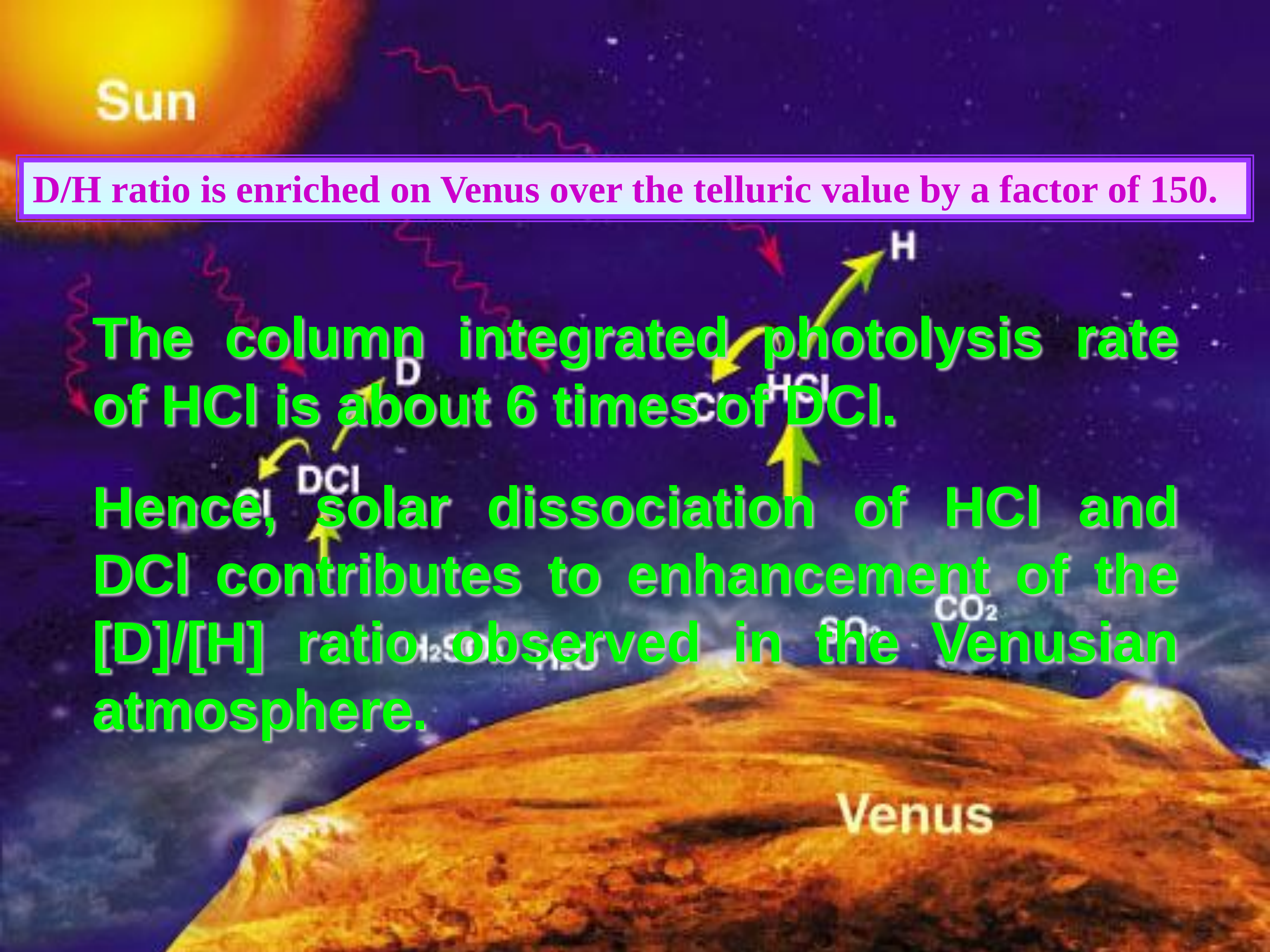


Key processes controlling the D/H ratio in Venus

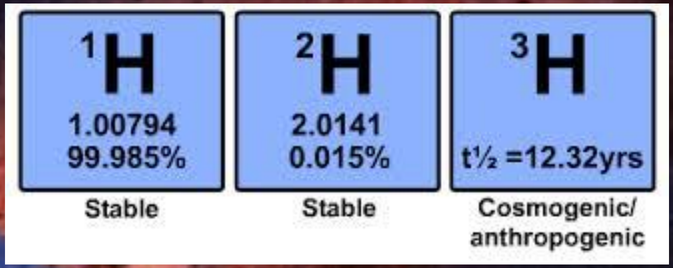
D/H ratio is enriched on Venus over the telluric value by a factor of 150.

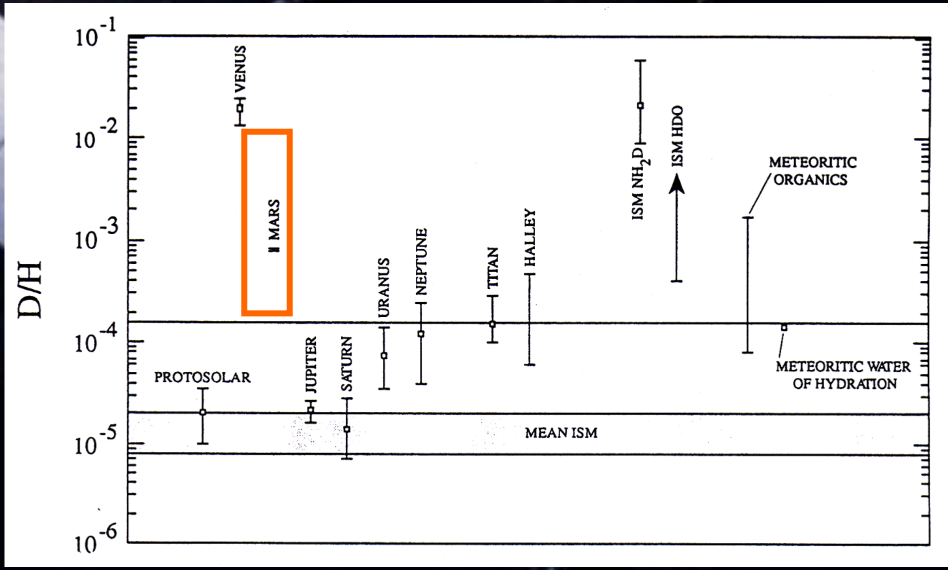
The column integrated photolysis rate of HCl is about 6 times of DCl.

Hence, solar dissociation of HCl and DCl contributes to enhancement of the [D]/[H] ratio observed in the Venusian atmosphere.



D/H ratios in the solar system and interstellar clouds

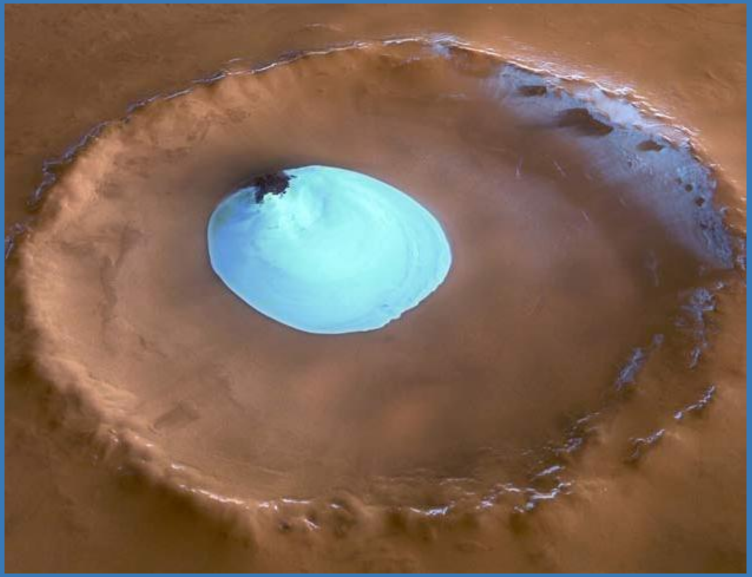




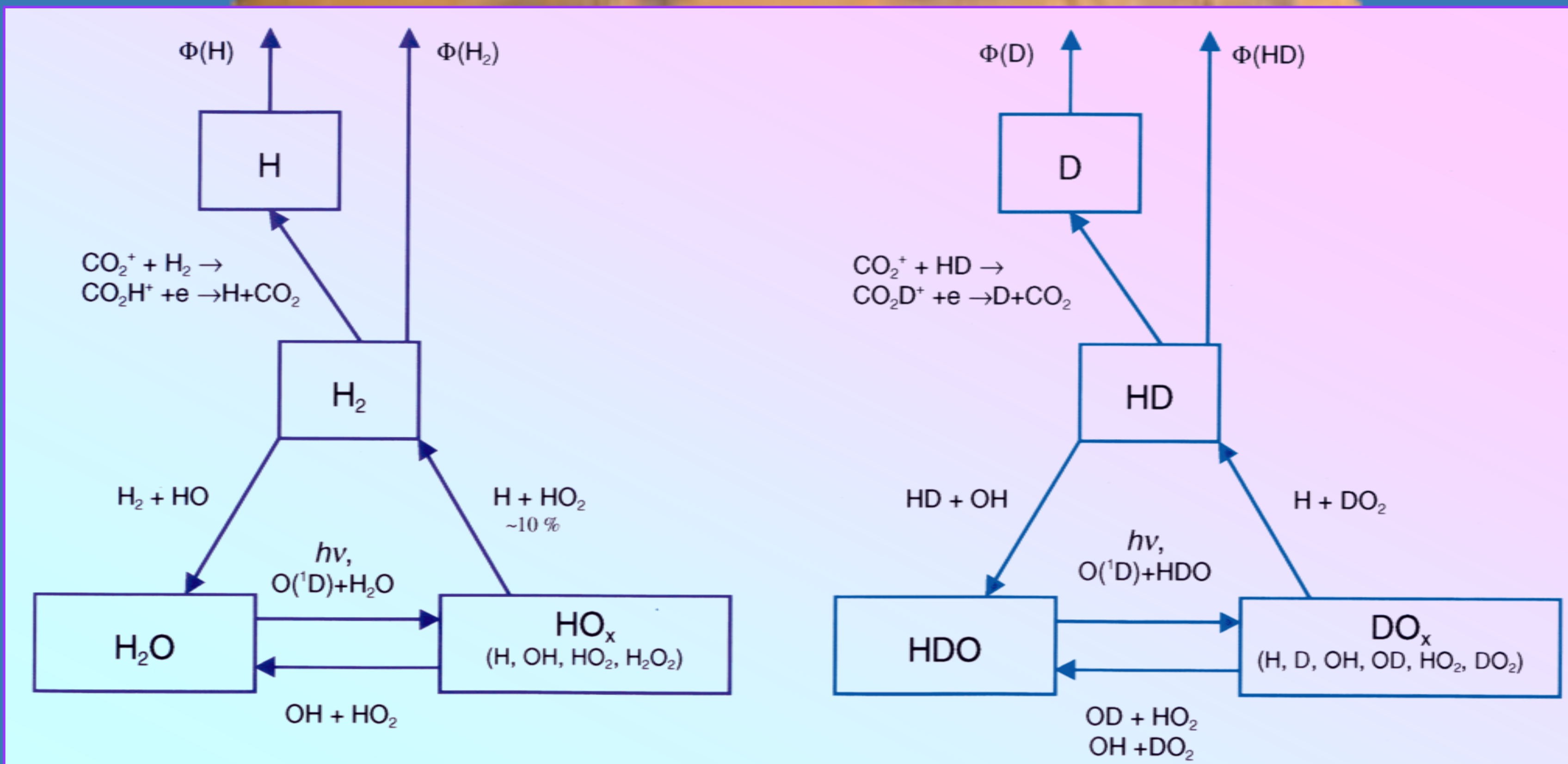
H_2O on Mars

Water on Mars

Mars had plenty of water once. Dissociation of water vapor with subsequent escape of H, H_2, and O is the primary mechanism of water loss from Mars. H_2O and HDO are the major sources of H and D in Mars.



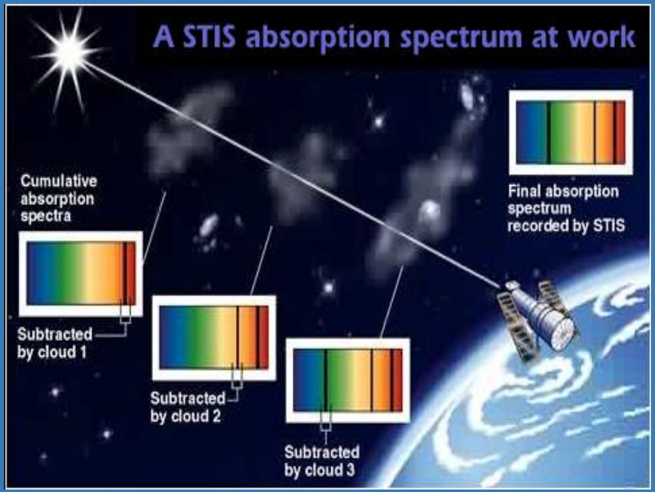
Photochemical model of Mars



Isotopic Fractionation of D/H on Mars

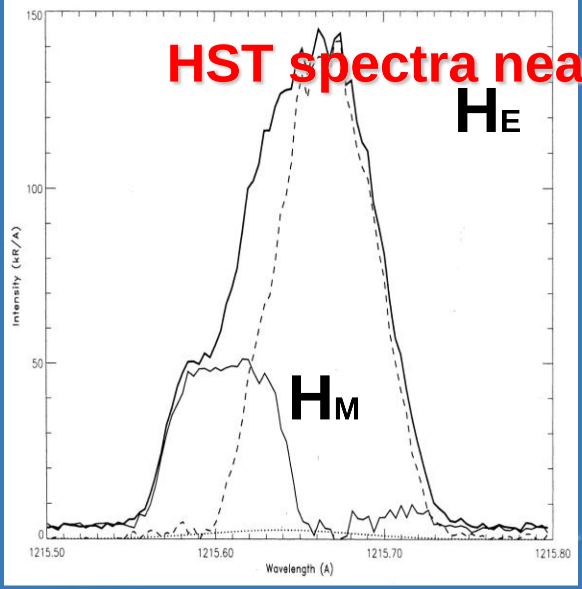
- Owen *et al.* reported that D/H = $(9 \pm 4) \times 10^{-4}$ on Mars in 1988.
  Compared: D/H = 1.56×10^{-4} on Earth.

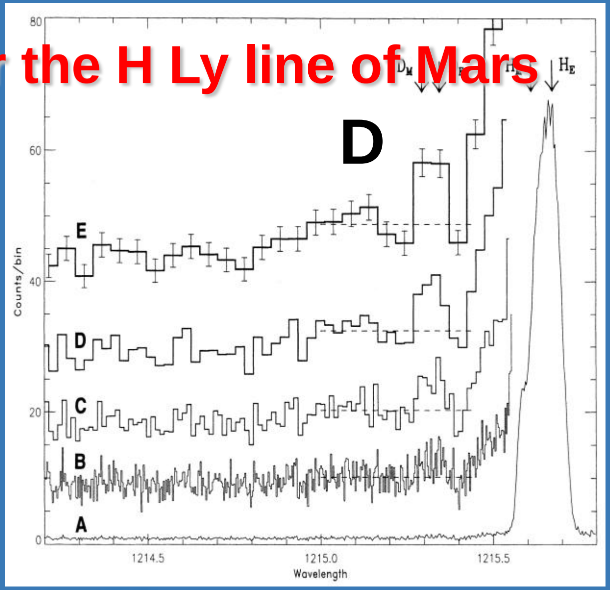
- In 1998, the measurement from HST confirmed the D/H ratio.



Hubble Space Telescope

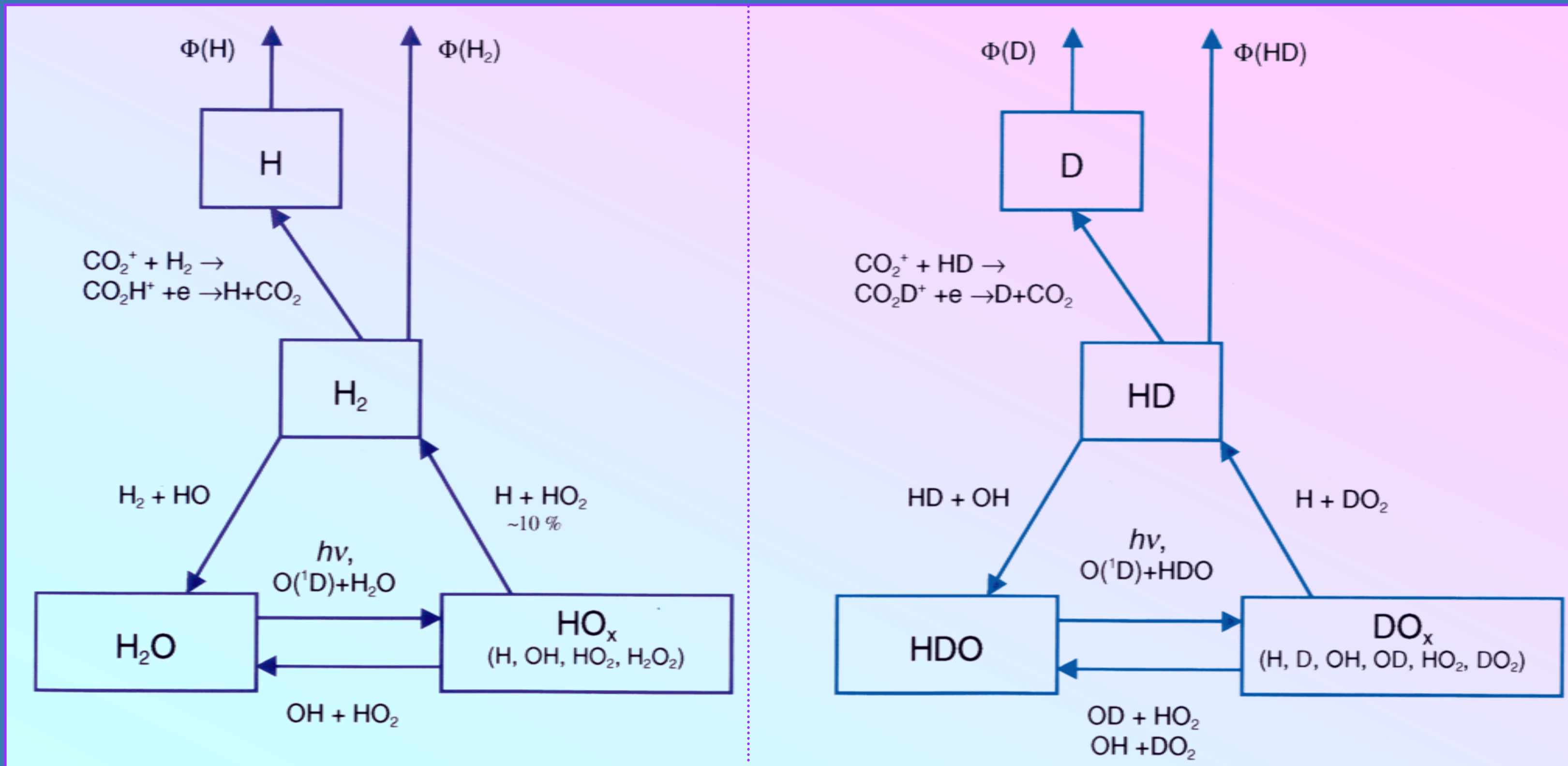
HST spectra near the H Ly line of Mars





Krasnopolky et al., Science, vol. 280, 1576 (1998).

Photochemical model in Mars



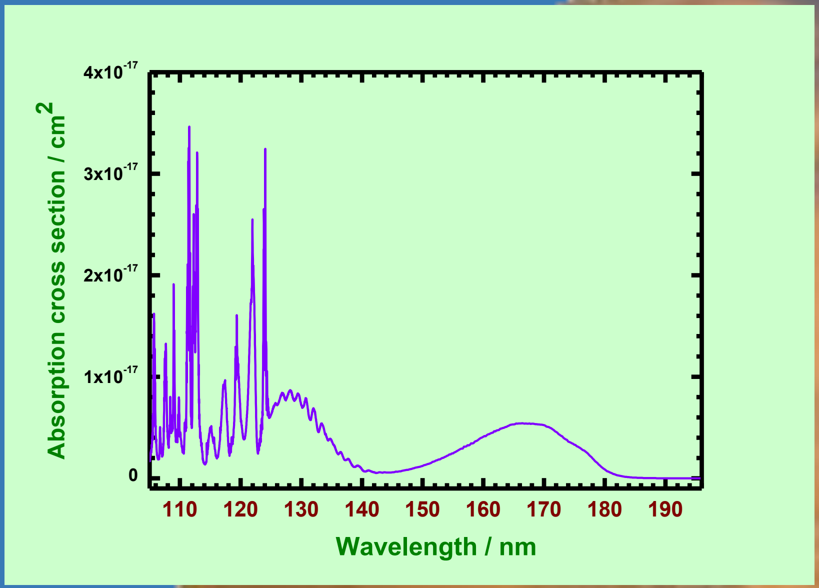
$\phi_H = \phi(H) + 2\,\phi(H_2) + \phi(HD)$ ~ 1.6×10^8 atoms cm^{-2} sec^{-1}
$\phi_D = \phi(D) + \phi(HD)$ ~ 8.0×10^3 atoms cm^{-2} sec^{-1}

Efficiency of escape of D relative to H:

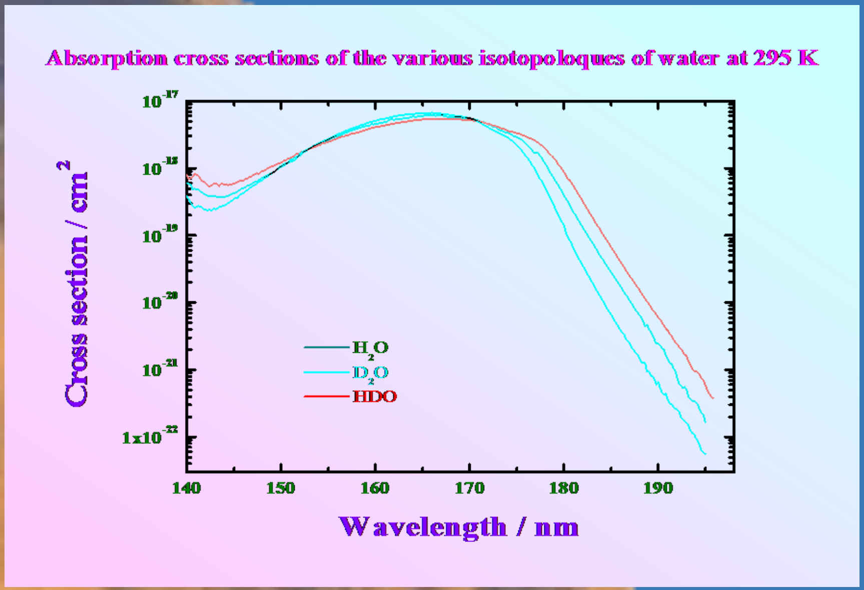
$$F = \frac{\phi_D / \phi_H}{[HDO]_0 / 2[H2O]_0}$$

~ **0.02** measured by Hubble Space Telescope

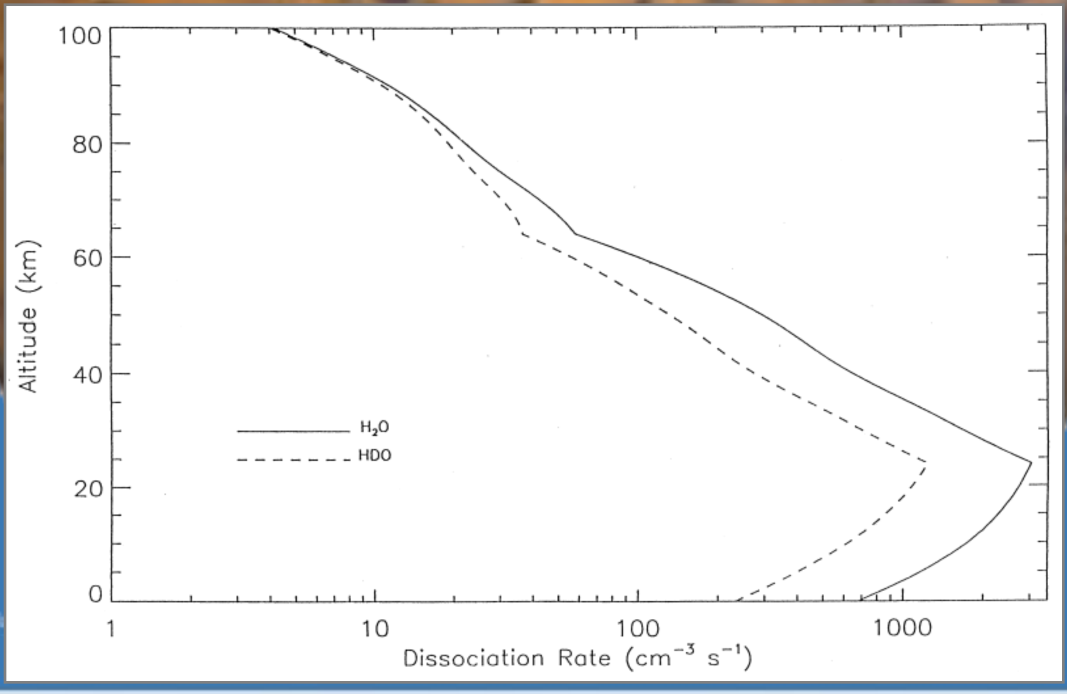
~ **0.32** calculated from the best model



Absorption cross section / cm2
4x10-17
3x10-17
2x10-17
1x10-17
0
110 120 130 140 150 160 170 180 190
Wavelength / nm

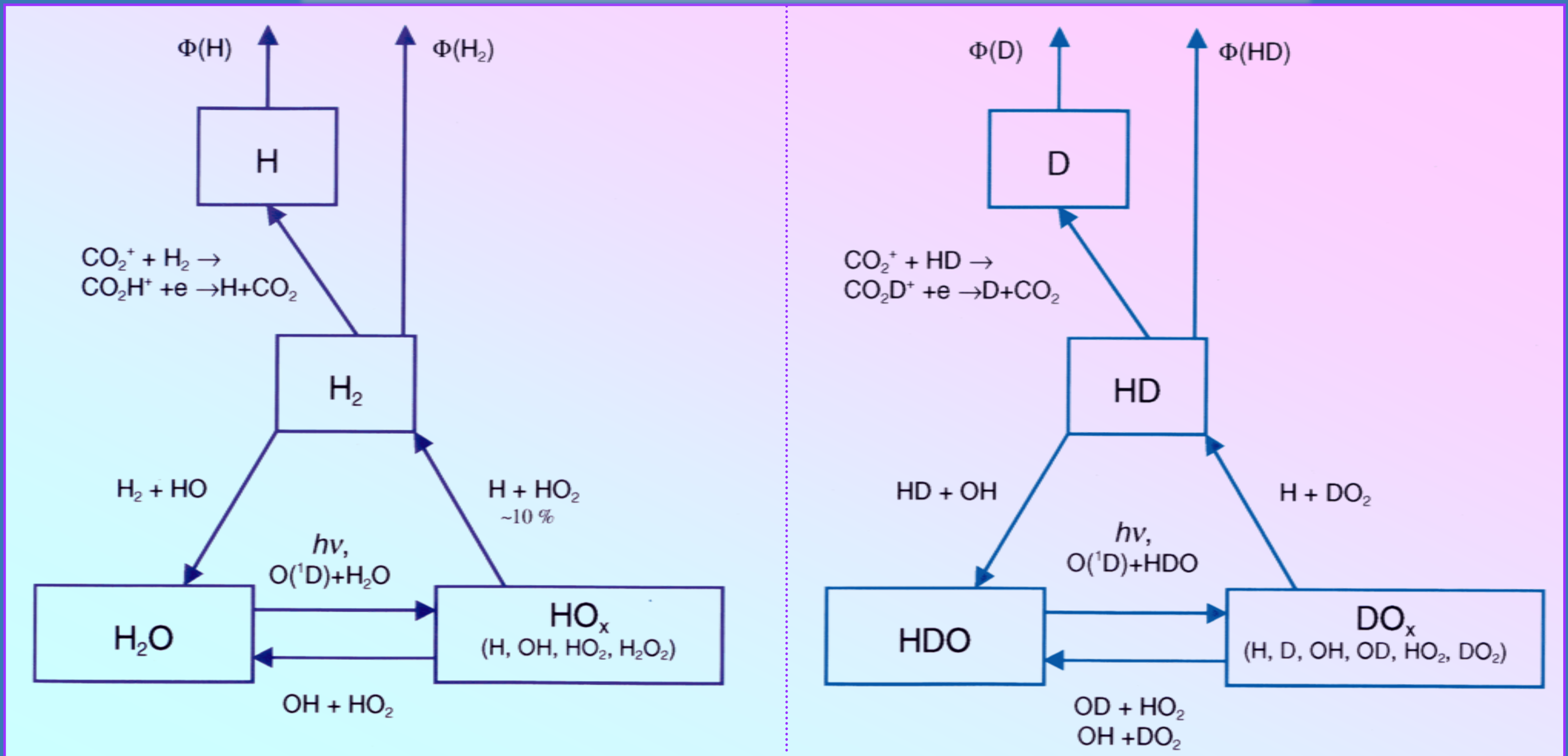


Absorption cross sections of the various isotopoloques of water at 295 K
Cross section / cm2
H2O
D2O
HDO
140 150 160 170 180 190
Wavelength / nm



Altitude (km)
100
80
60
40
20
0
H2O
HDO
1 10 100 1000
Dissociation Rate (cm-3 s-1)

Photochemical model in Mars



$\phi_H = \phi(H) + 2\,\phi(H_2) + \phi(HD) \sim 1.6\times10^8$ atoms cm^{-2} sec^{-1}

$\phi_D = \phi(D) + \phi(HD) \sim 1.5\times10^3$ atoms cm^{-2} sec^{-1}

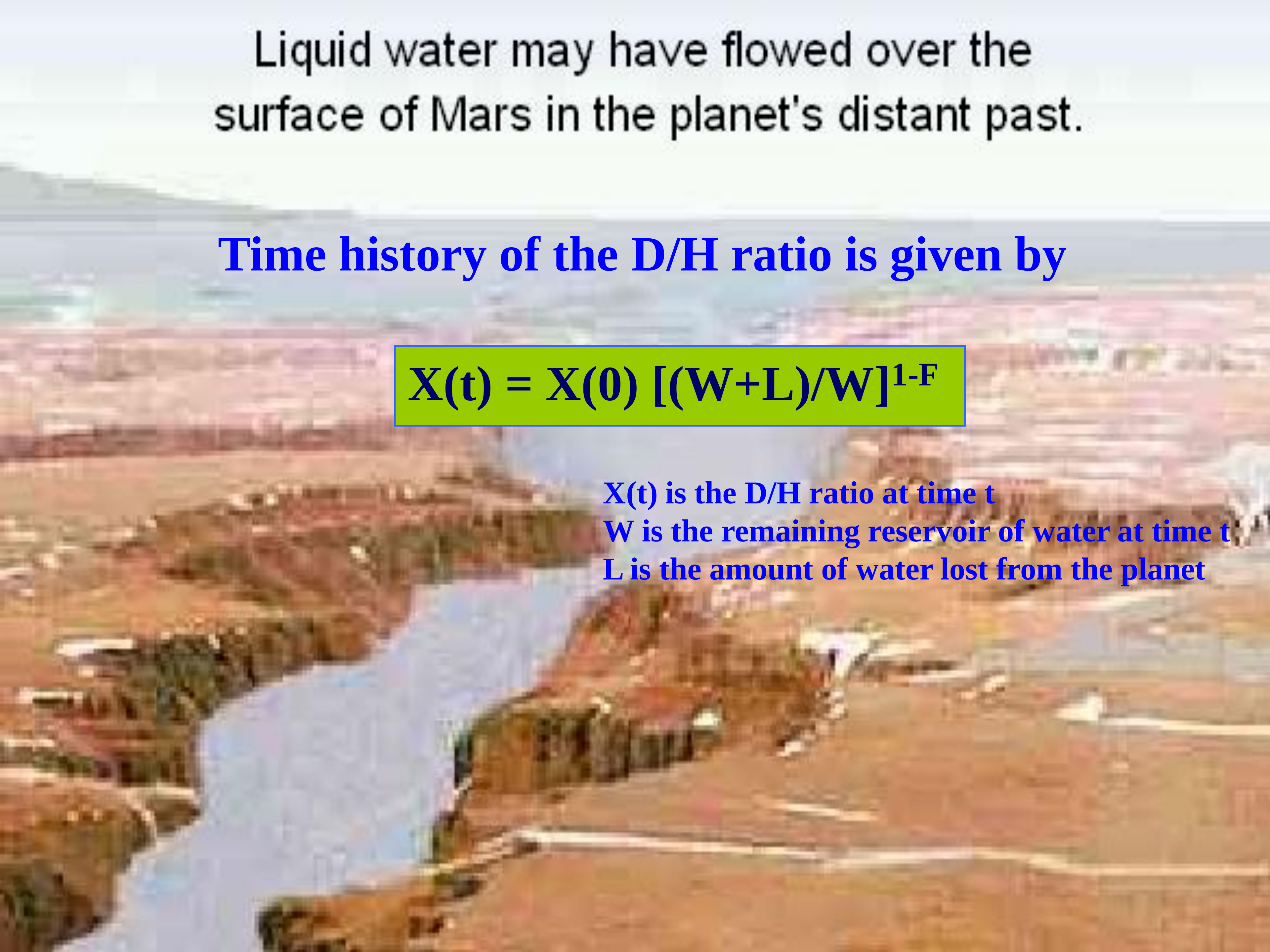
Efficiency of escape of D relative to H:

$$F = \frac{\phi_D / \phi_H}{[HDO]_0 / 2[H2O]_0}$$

~ **0.02** measured by Hubble Space Telescope

~ **0.32** calculated from the best model

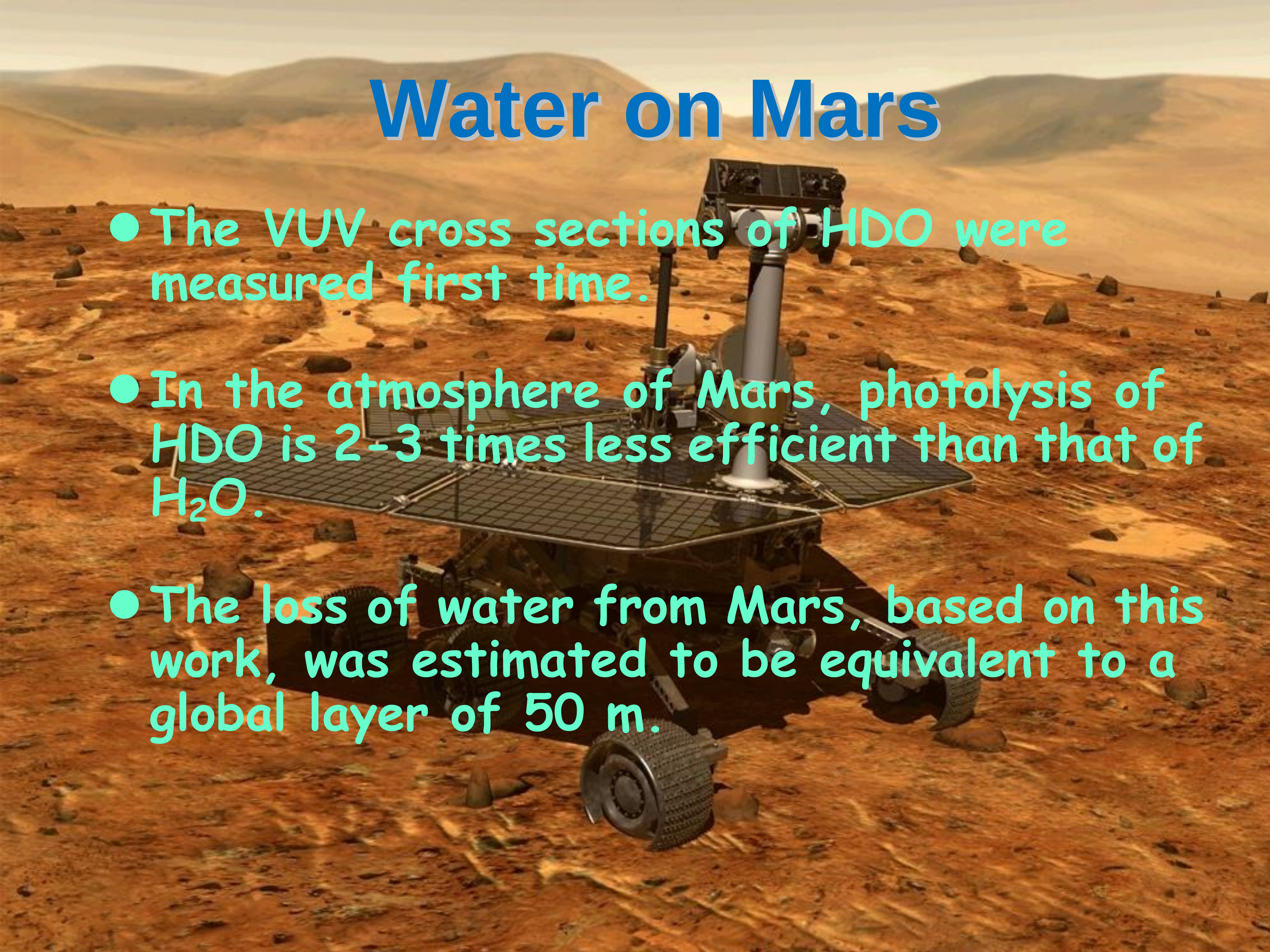
~ **0.06** calculated from our data



Time history of the D/H ratio is given by

$$\boxed{X(t) = X(0)\,[(W+L)/W]^{1-F}}$$

X(t) is the D/H ratio at time t
W is the remaining reservoir of water at time t
L is the amount of water lost from the planet

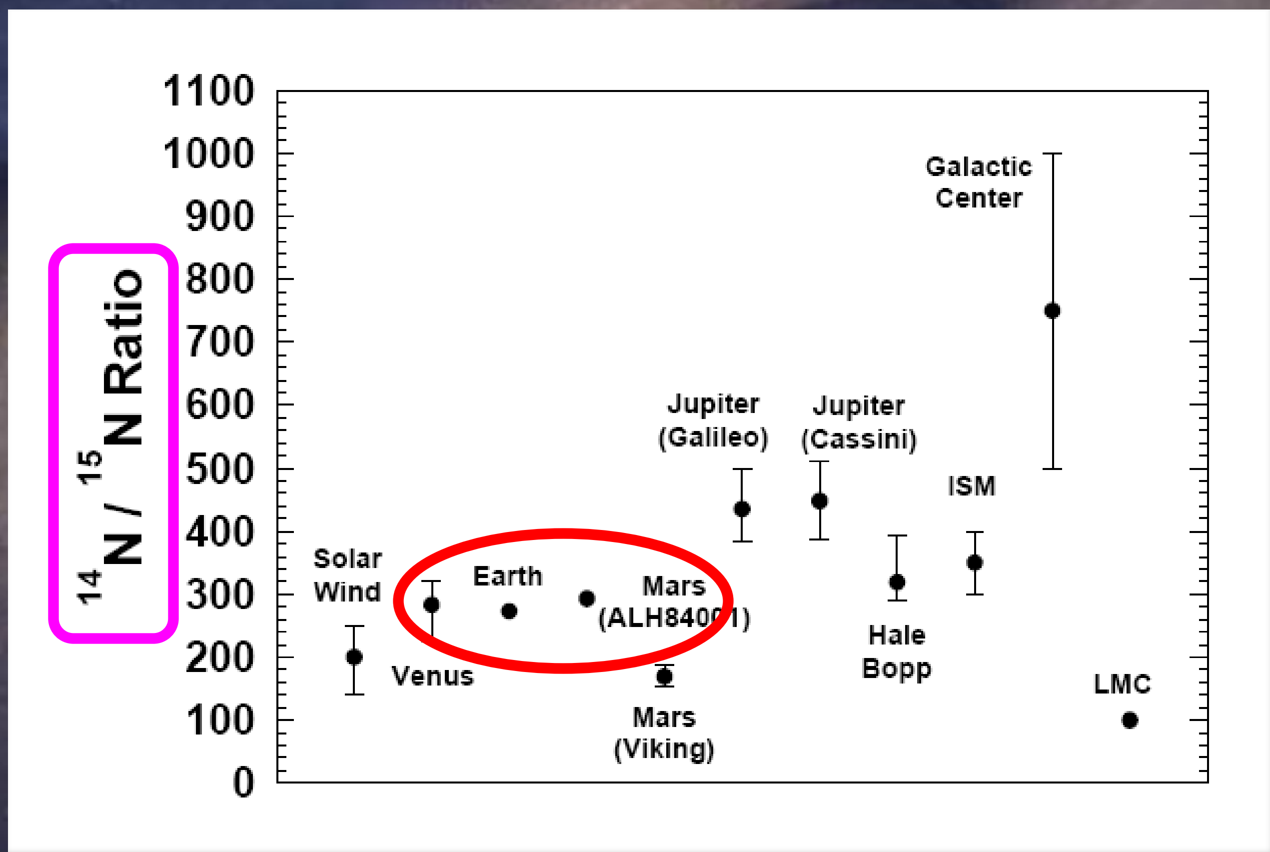


Water on Mars

- The VUV cross sections of HDO were measured first time.
- In the atmosphere of Mars, photolysis of HDO is 2-3 times less efficient than that of H_2O.
- The loss of water from Mars, based on this work, was estimated to be equivalent to a global layer of 50 m.

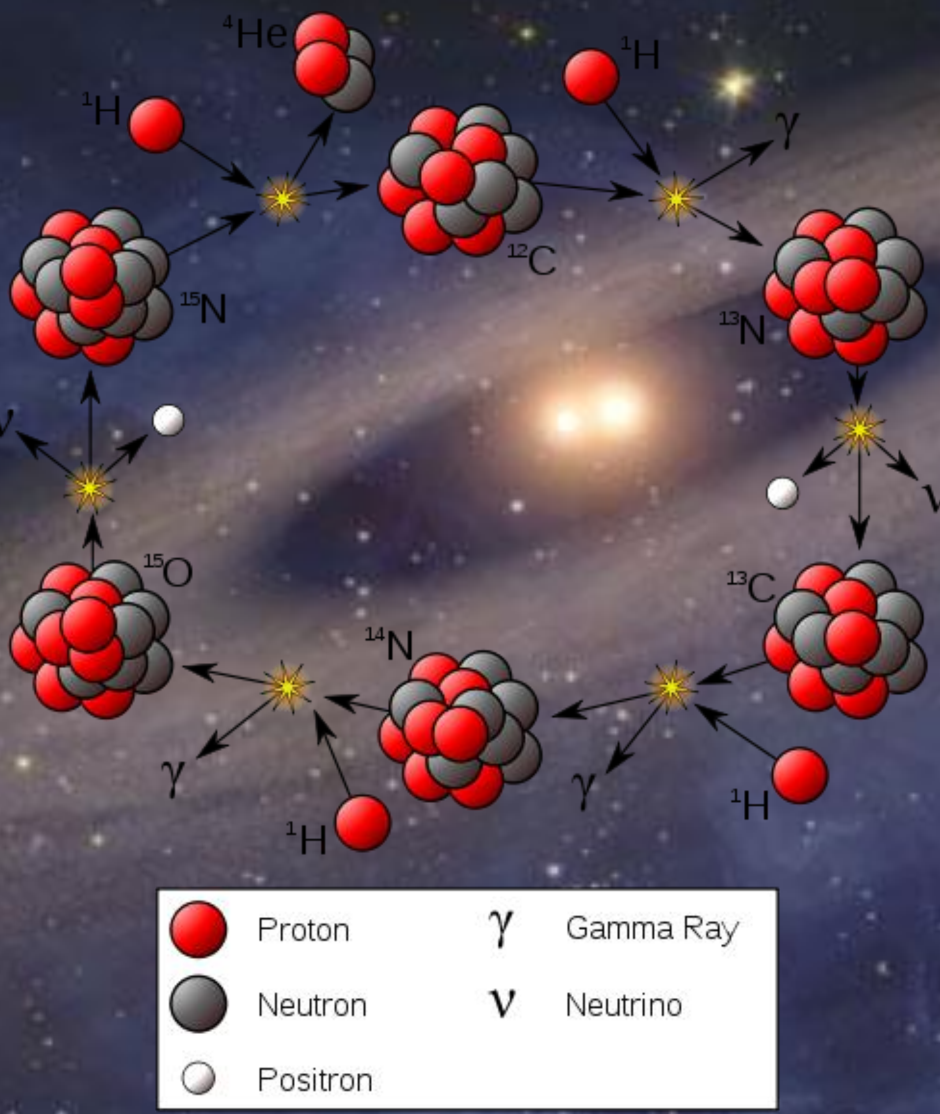
Nitrogen Isotopic Fractionation

Ratio on Earth



Nitrogen Isotopic Fractionation

CNO cycle

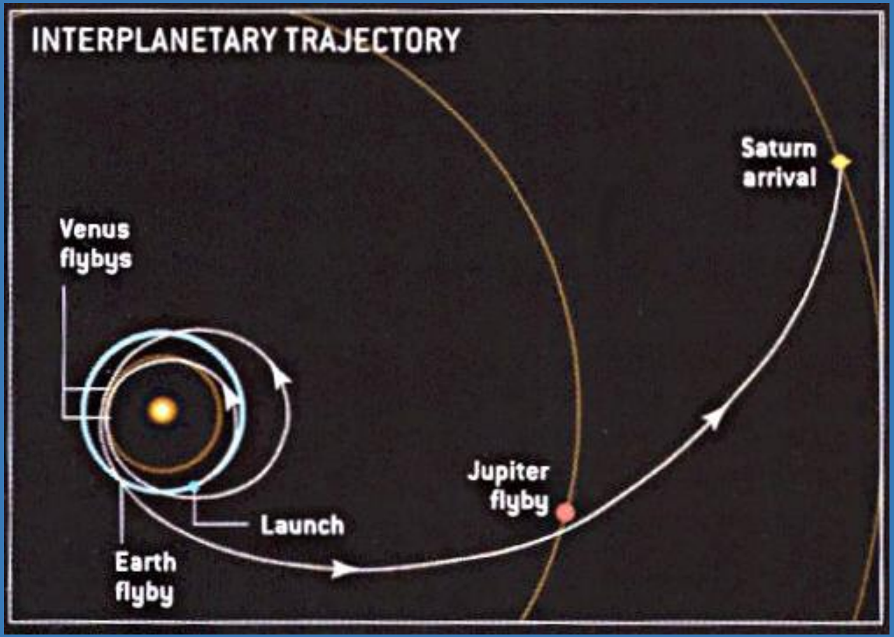


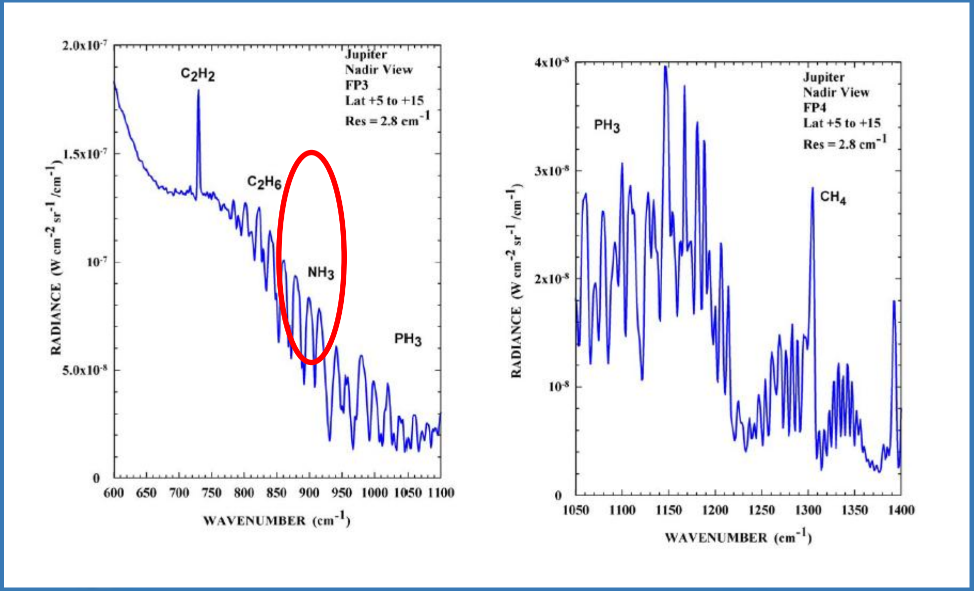
Cassini-Huygens Mission to Saturn

The Cassini spacecraft was launched on 10/7/1997 for a four year orbital tour of Saturn and its satellites scheduled beginning in July 2004.

NH_3 observed in planets

Cassini-Huygens Mission

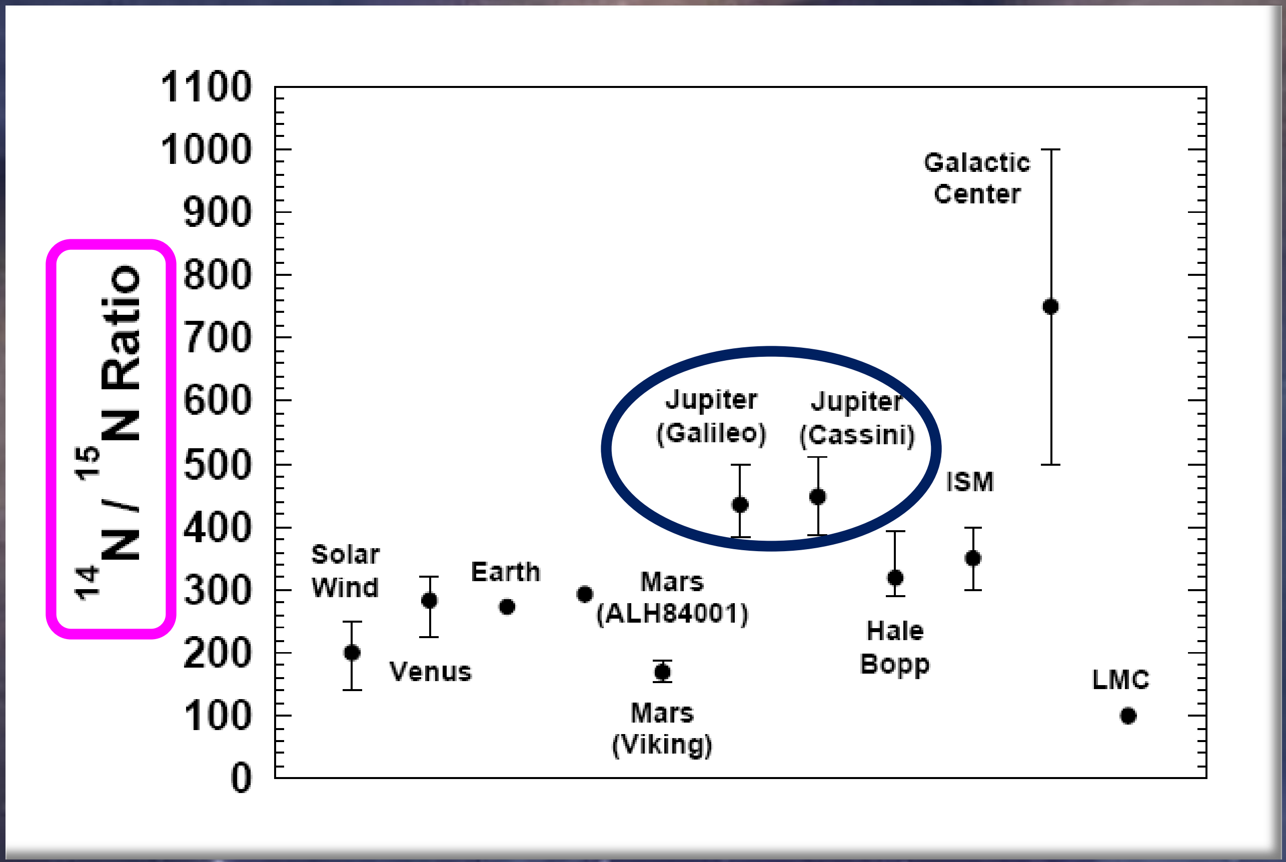




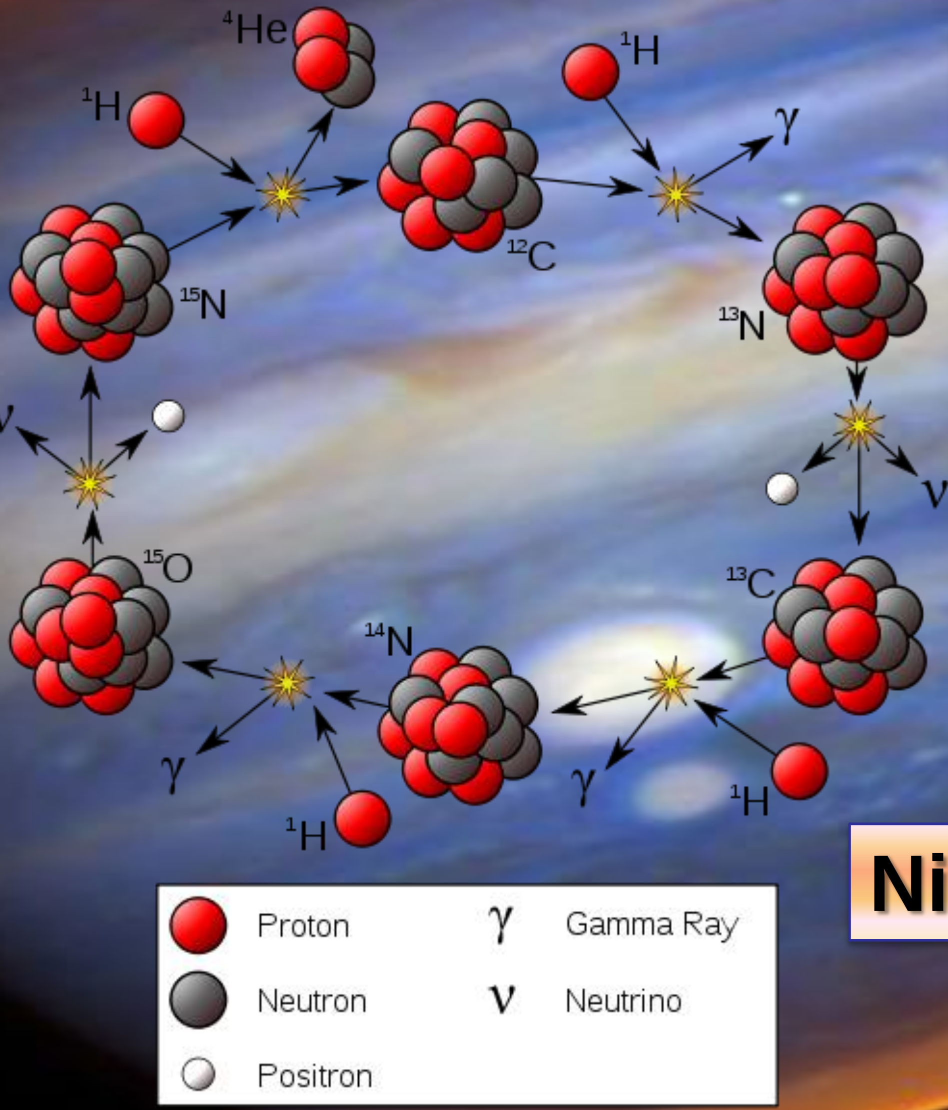
The spacecraft flew by Jupiter in Dec. 2000 ~Jan. 2001: Unique global infrared spectral maps of Jupiter obtained.

Nitrogen Isotopic Fractionation

Nitrogen puzzle



$^{14}N/^{15}N$ ratio in Jupiter

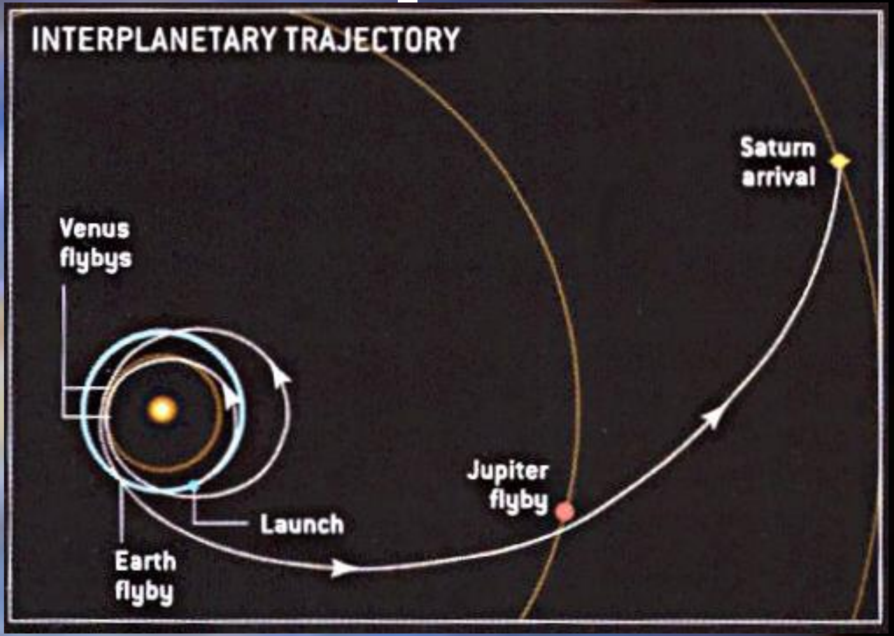


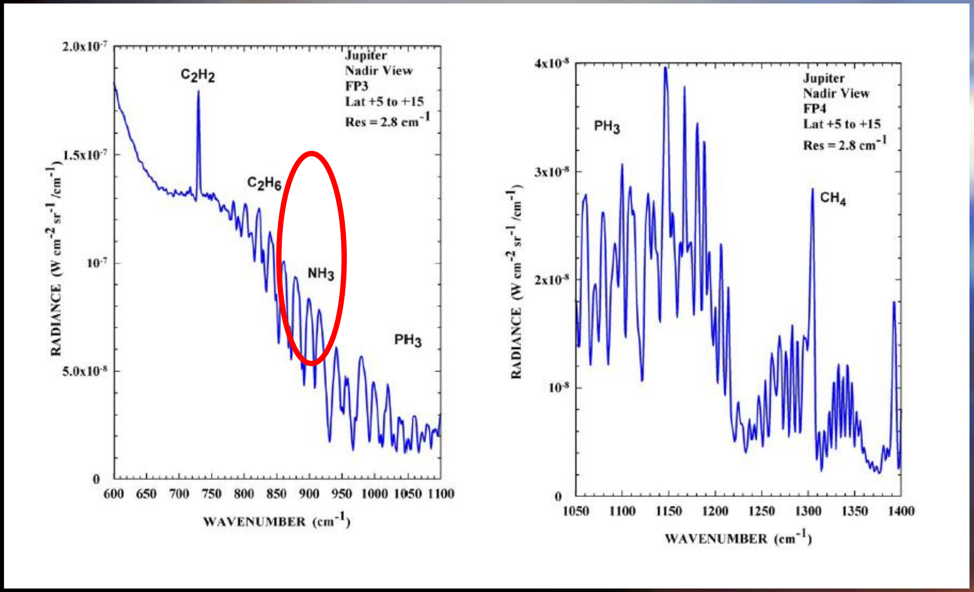
CNO cycle

Nitrogen puzzle

Jupiter

$^{14}N/^{15}N$ ratio in Jupiter

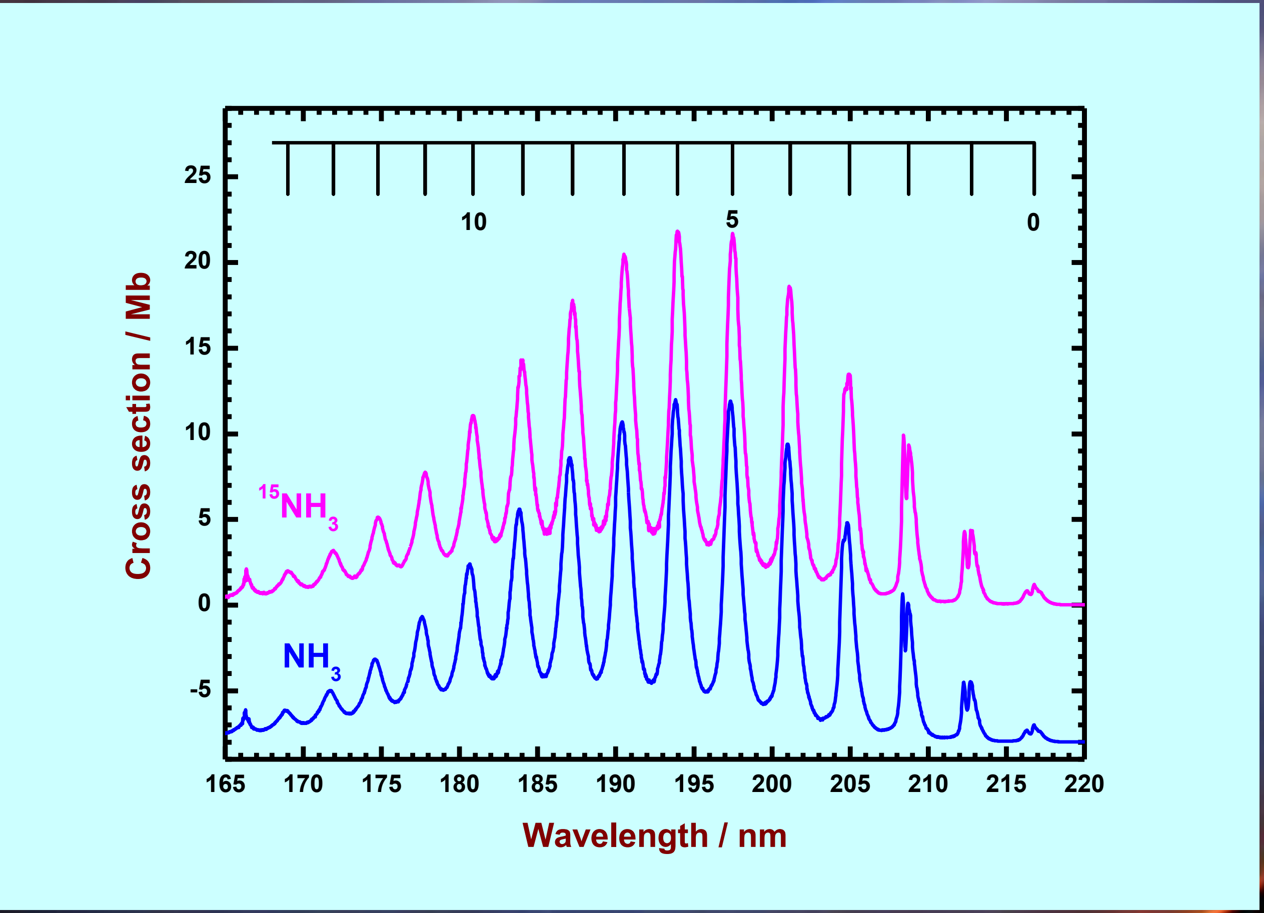




Nitrogen puzzle

Jupiter

$^{14}N/^{15}N$ ratio on Jupiter



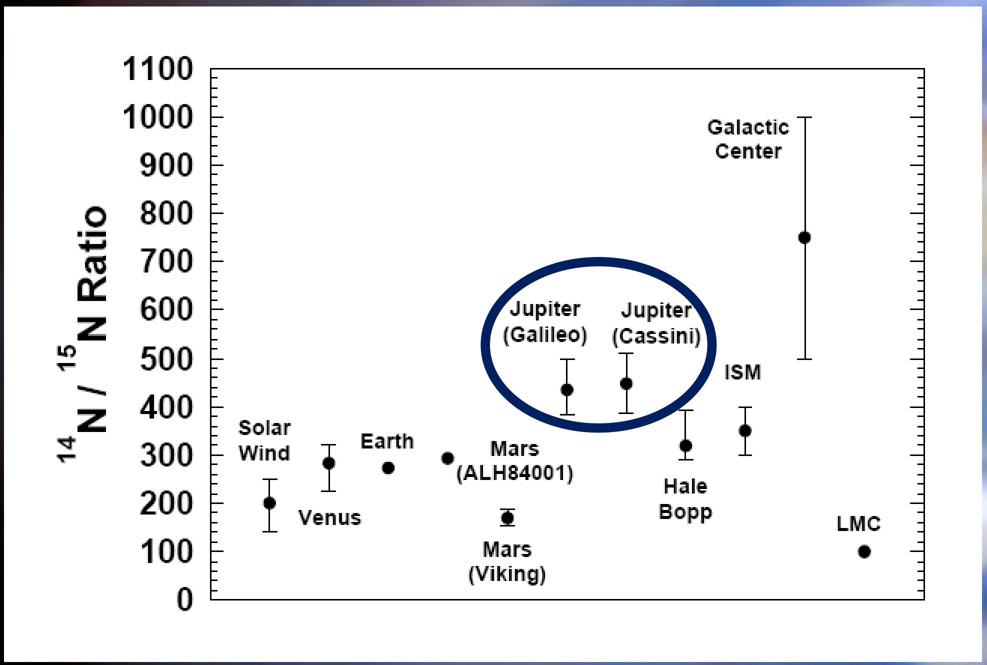
VUV absorption cross sections of $^{14}NH_3$ and $^{15}NH_3$

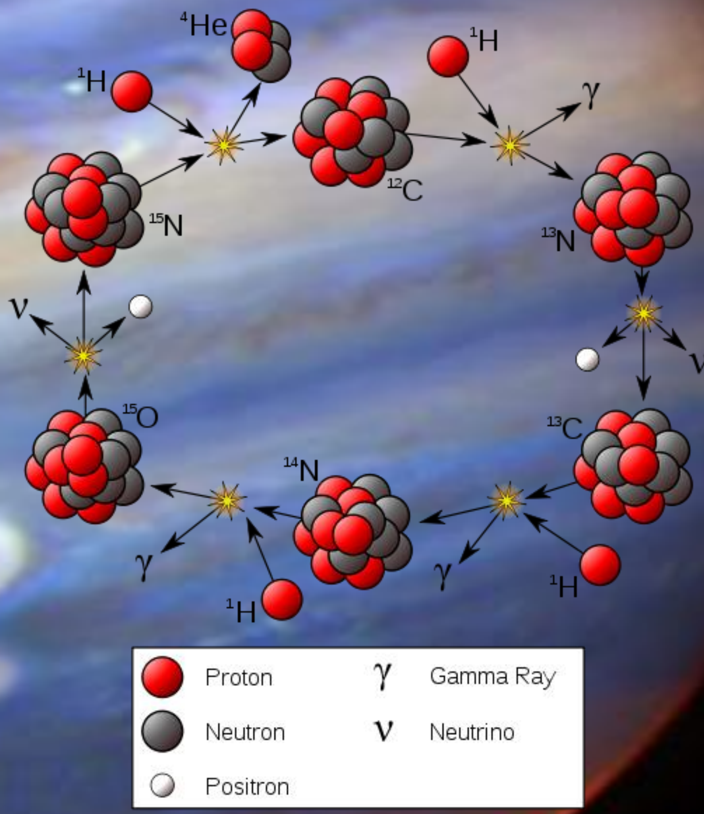
Jupiter

$^{14}NH_3$ & $^{15}NH_3$ on Jupiter

Nitrogen puzzle

CNO cycle

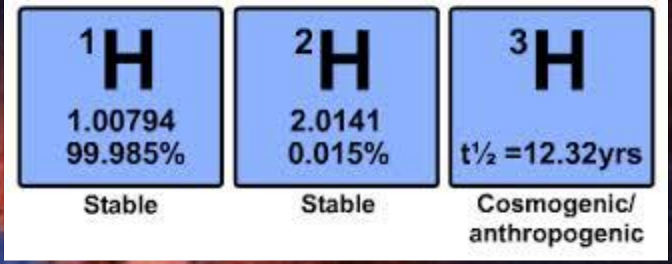


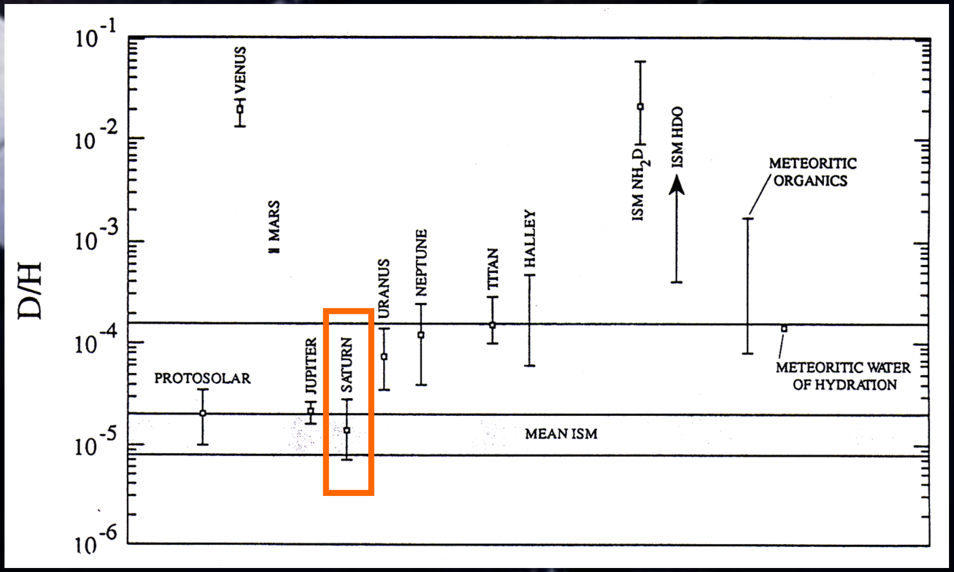


$^{14}N/^{15}N$ ratio in Jupiter

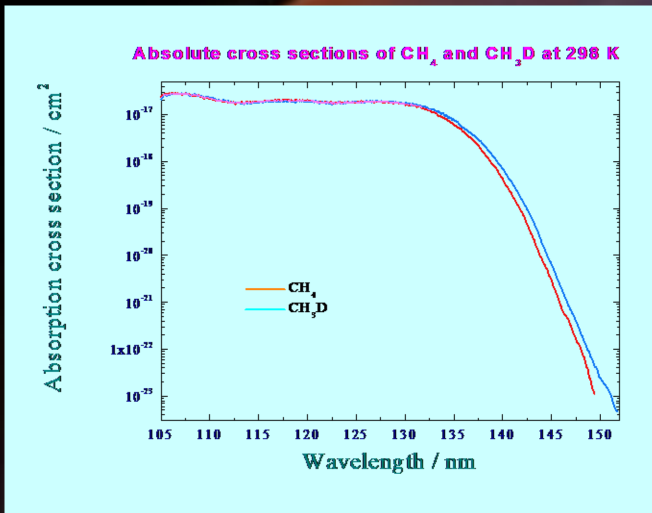
Jupiter

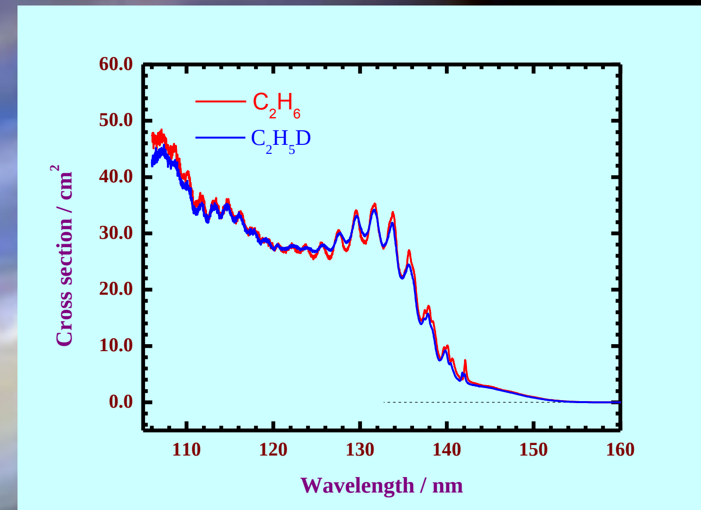
D/H ratios in the solar system and interstellar clouds

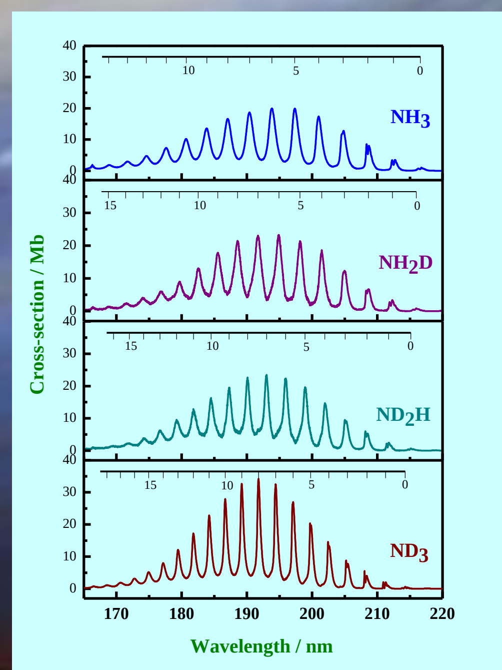




CH_4, C_2H_6, and NH_3 in Jupiter





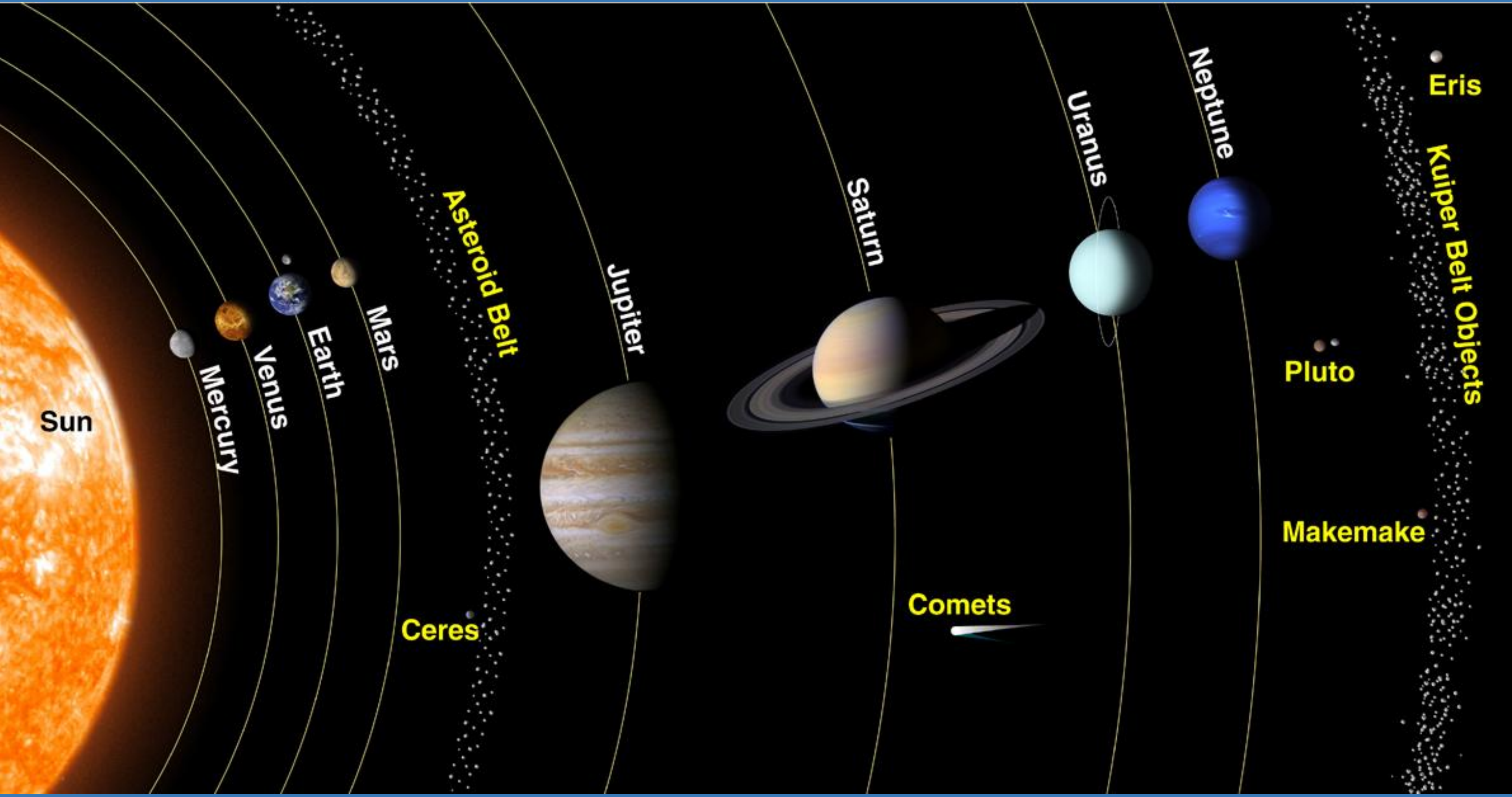


Works have been done

- HCl / DCl → Venus
- H_2O / HDO / D_2O → Mars
- CH_4 / CH_3D → Jupiter
- C_2H_6 / C_2H_5D → Jupiter
- NH_3 / NH_2D / NHD_2 / ND_3 / $^{15}NH_3$ → Jupiter
- CH_3OH / CD_3OH / CH_3OH / CD_3OD → Comets

Temperatures in the Solar System

167 43



543 287 139 109 76 55 44

735 227

National Synchrotron Radiation Research Center

Ave. temperature (K)

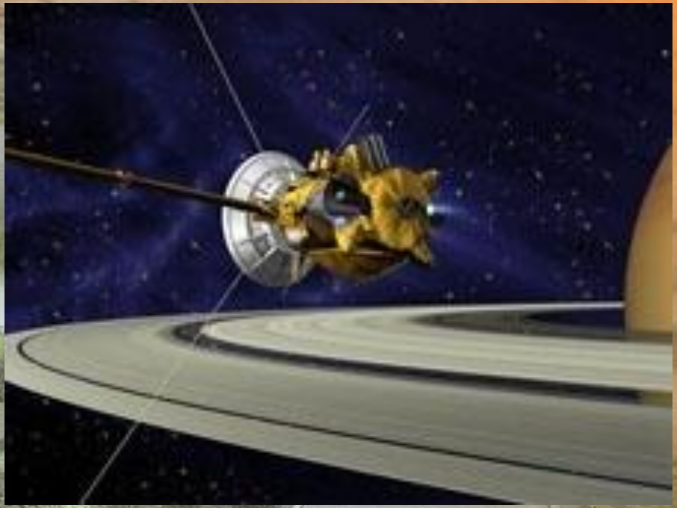
II. FUV absorption below 100 K

To assess the photochemically induced changes in space, we must know absorption cross sections of pertinent planetary species in the far-UV range.

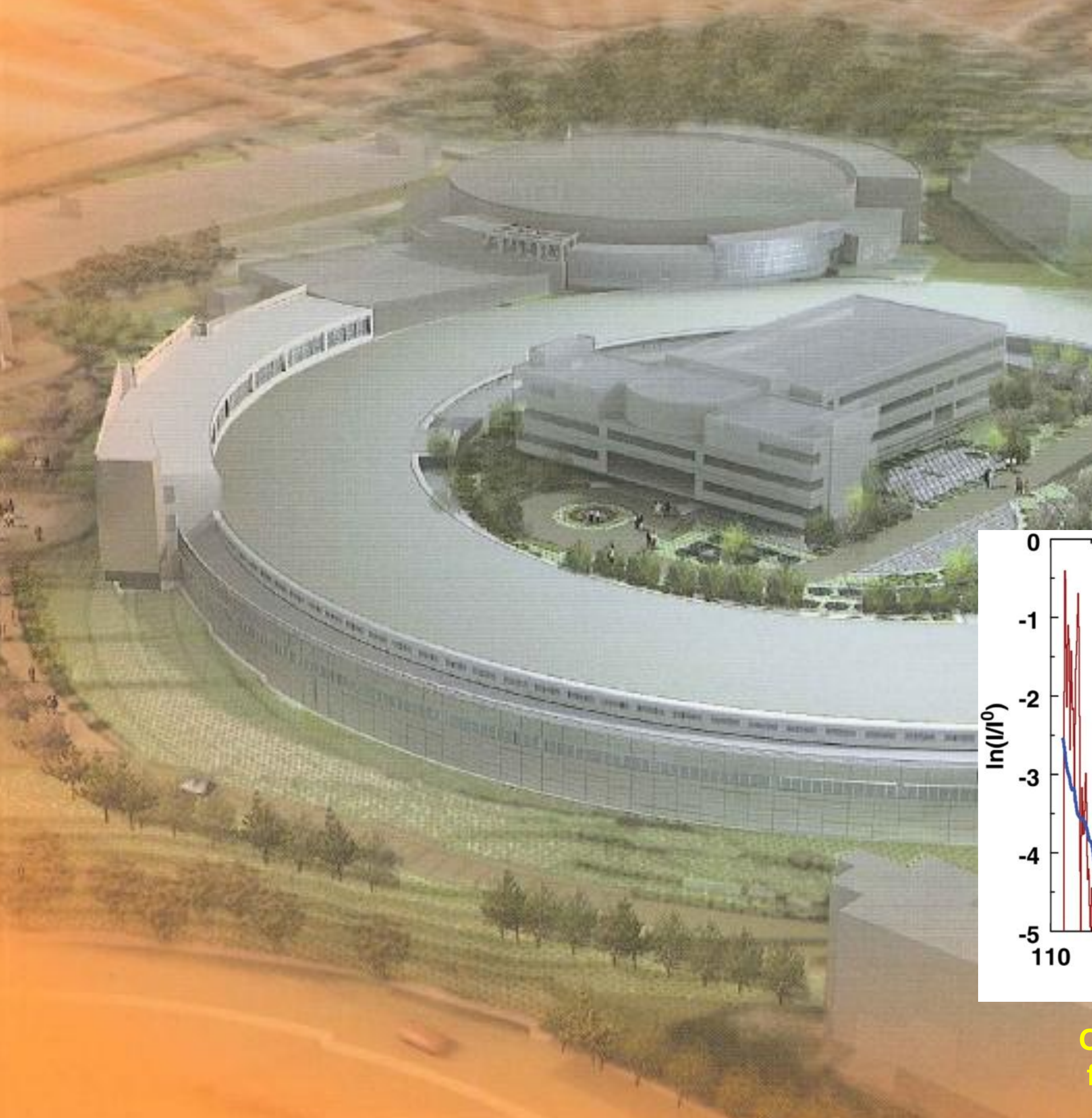
Absorption cross sections of interstellar molecules have been extensively measured in the far-UV range near room temperature, but such data for low temperature are lacking and need to be established.

FUV Astro-science

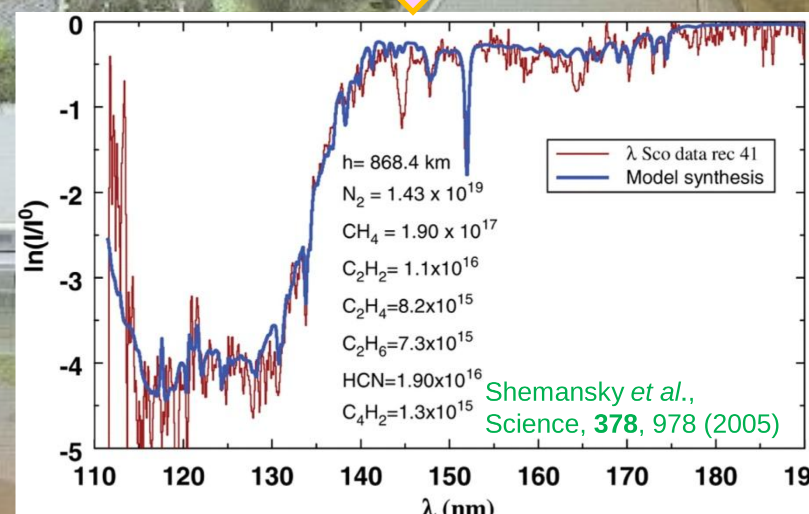
Cassini-Huygens Mission to Saturn



FUV Astro-science

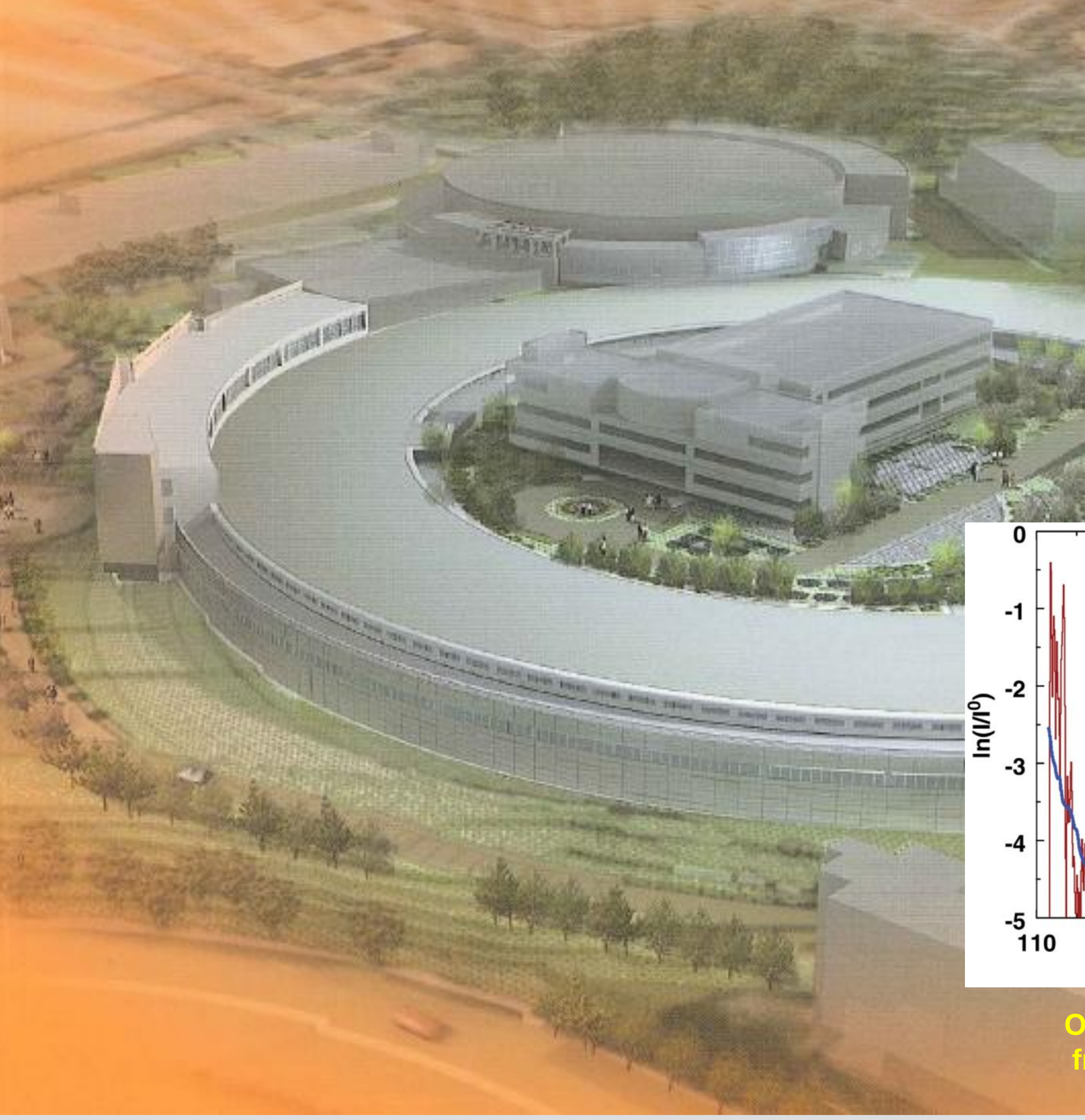


Cassini-Huygens Mission to Saturn



Observed Titan's transmission spectrum from the Cassini UVIS FUV spectrograph.

FUV Astro-science



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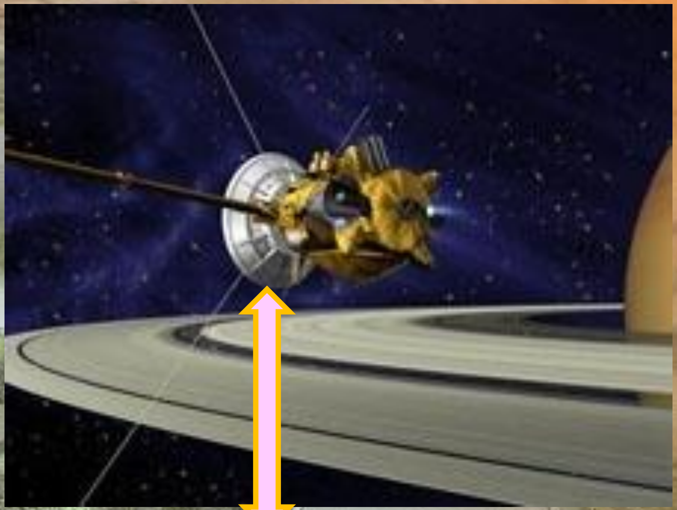

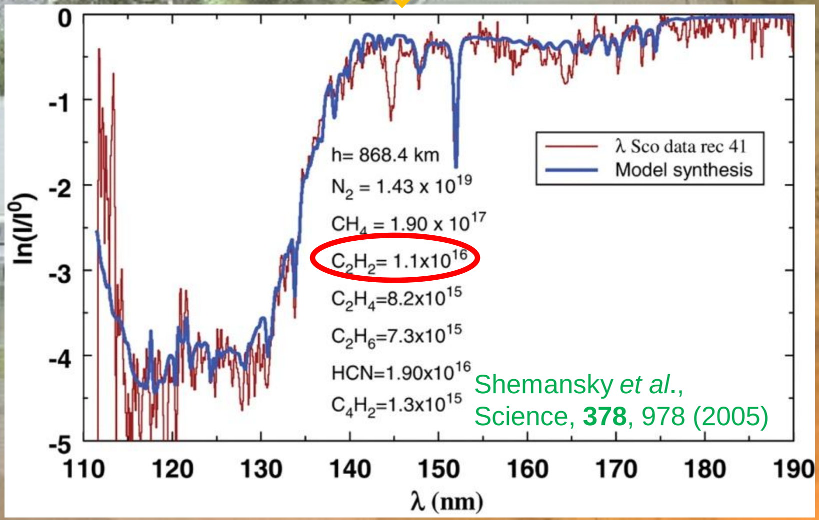

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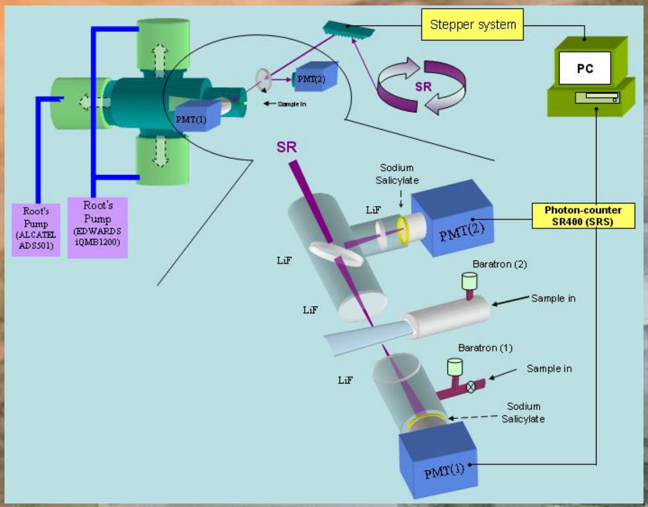
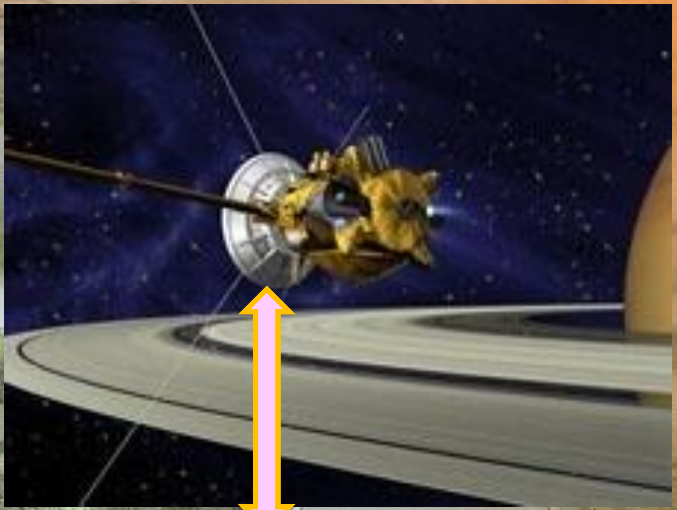
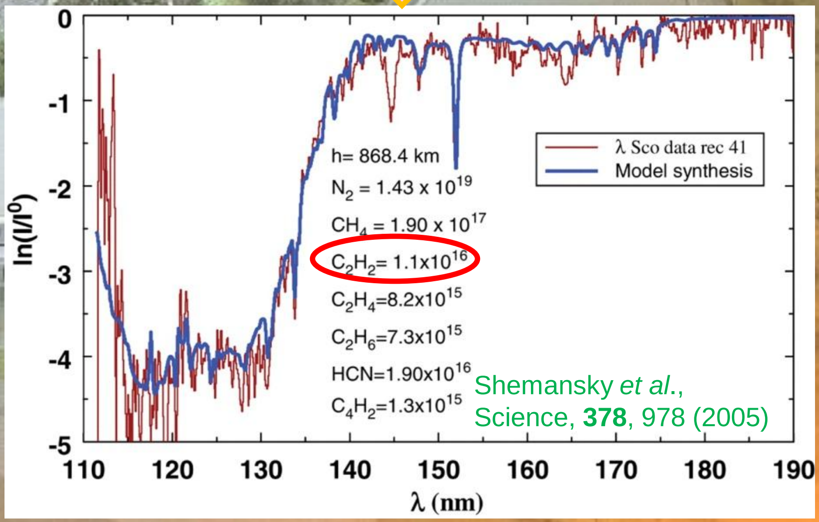
FUV Astro-science

VUV Slit-jet Absorption System



Cassini-Huygens Mission to Saturn





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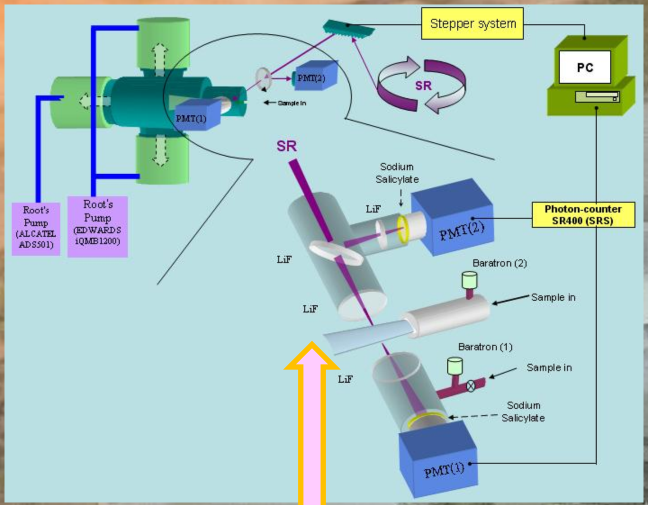
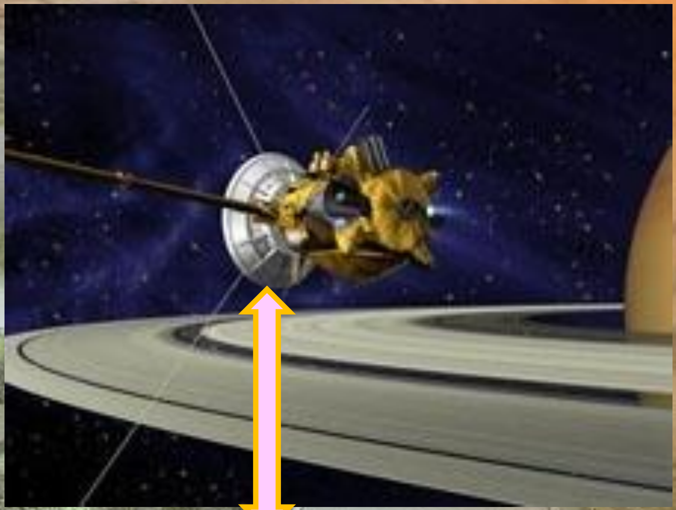
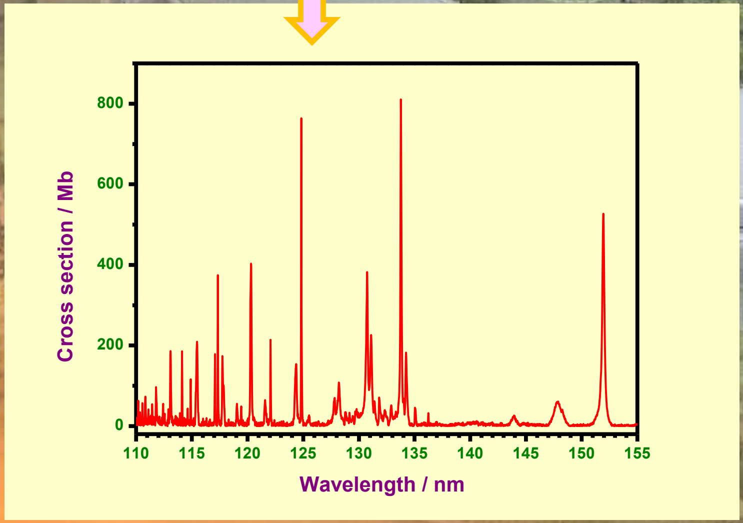
Absorption Cross Section of Gaseous Acetylene at 85 K in Wavelength Range 110-155 nm

VUV Slit-jet Absorption System

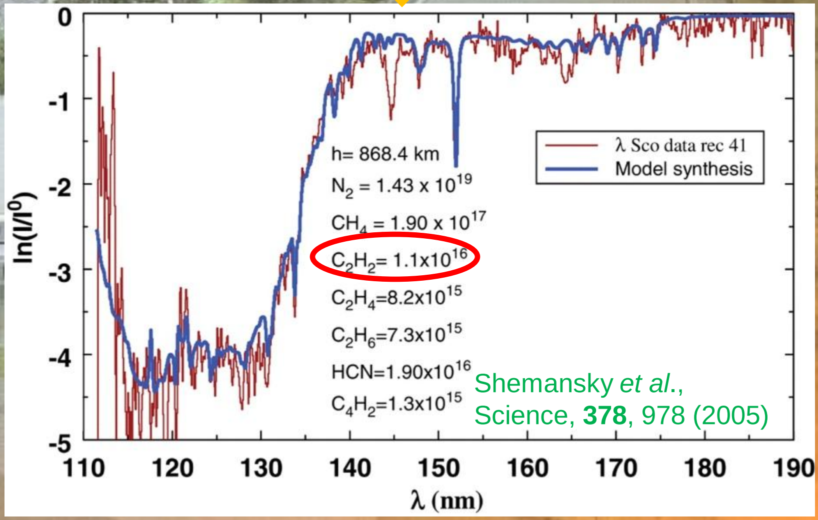


Cassini-Huygens Mission to Saturn





Measured absorption cross sections of C_2H_2 at 85 K with VUV slit-jet absorption system



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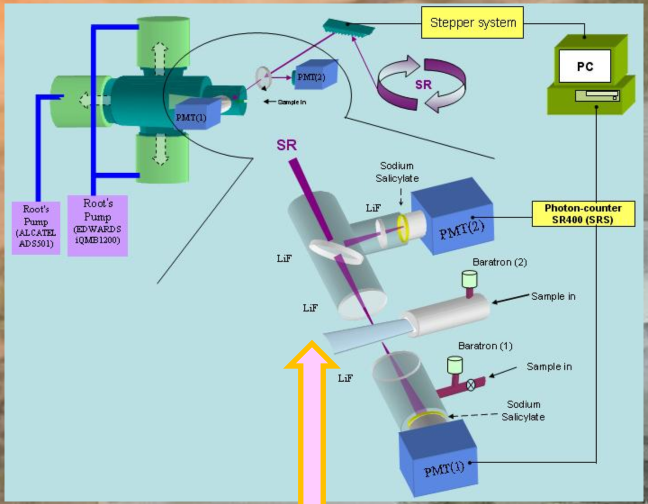
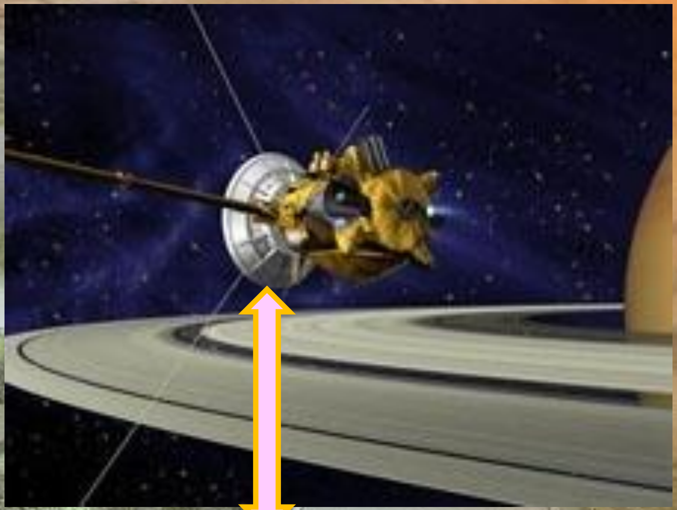
Absorption Cross Section of Gaseous Acetylene at 85 K in Wavelength Range 110-155 nm

Cheng *et al*., ApJS, **196**, 3 (2011)

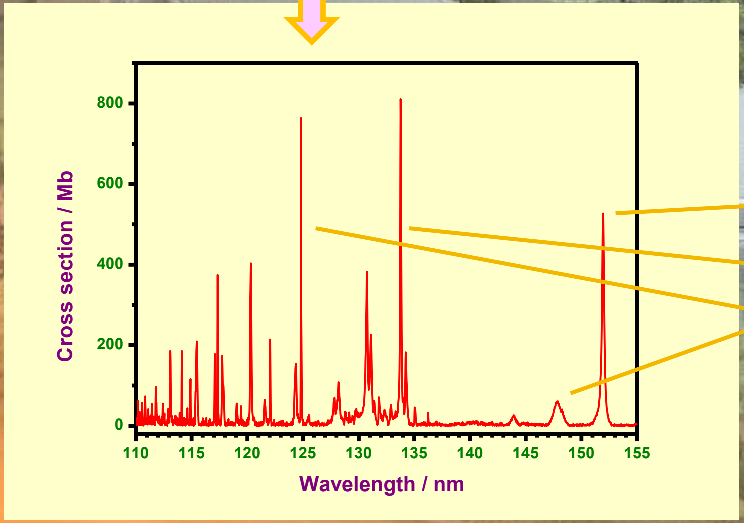
VUV Slit-jet Absorption System



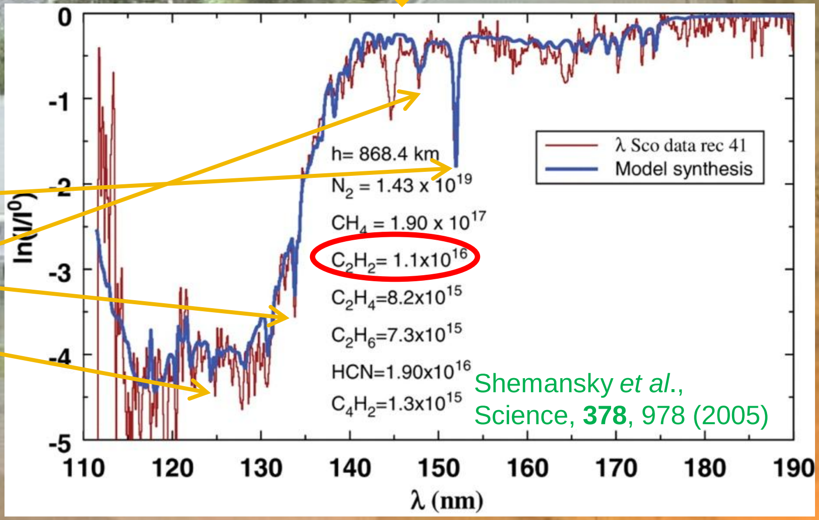
Cassini-Huygens Mission to Saturn



Absorption cross sections of gaseous C_2H_2 at 298 and 85 K were measured in wavelength range 110-155 nm with a VUV slit-jet absorption system. The cross sections of C_2H_2 are applicable to determine properties sensitive to temperature for diagnostic work on Saturn and Titan.

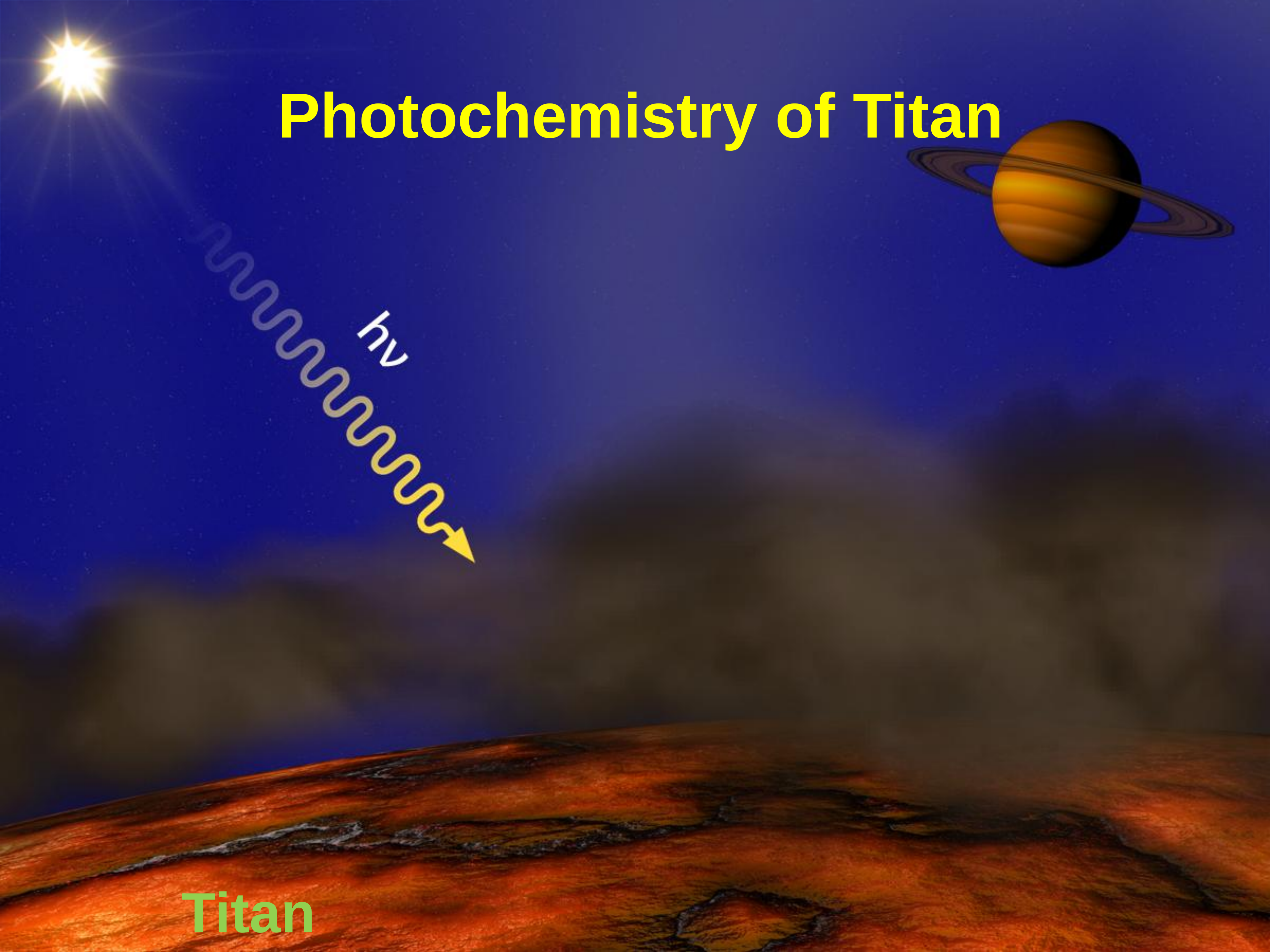


Measured absorption cross sections of C_2H_2 at 85 K with VUV slit-jet absorption system

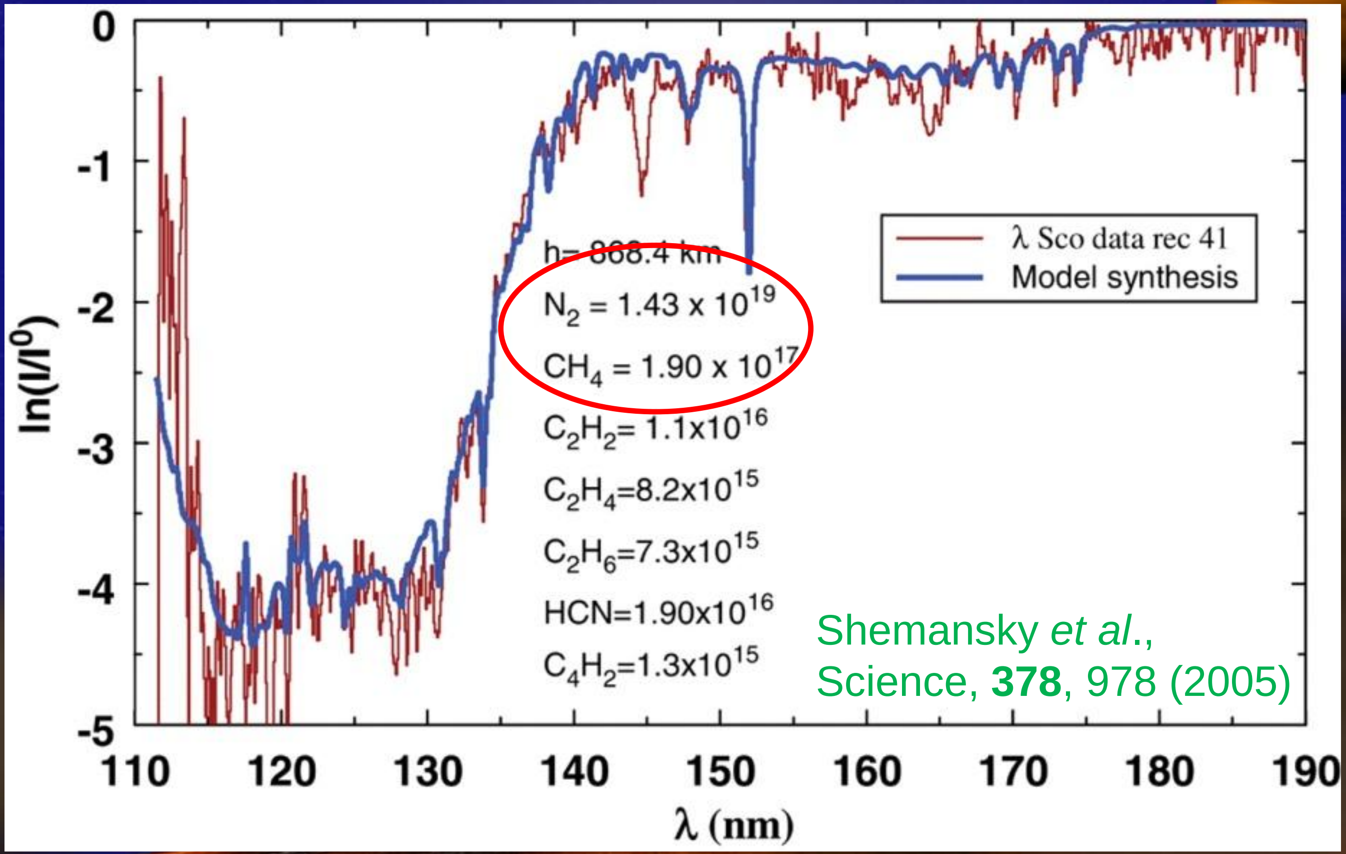


Observed Titan's transmission spectrum from the Cassini UVIS FUV spectrograph.

Photochemistry of Titan



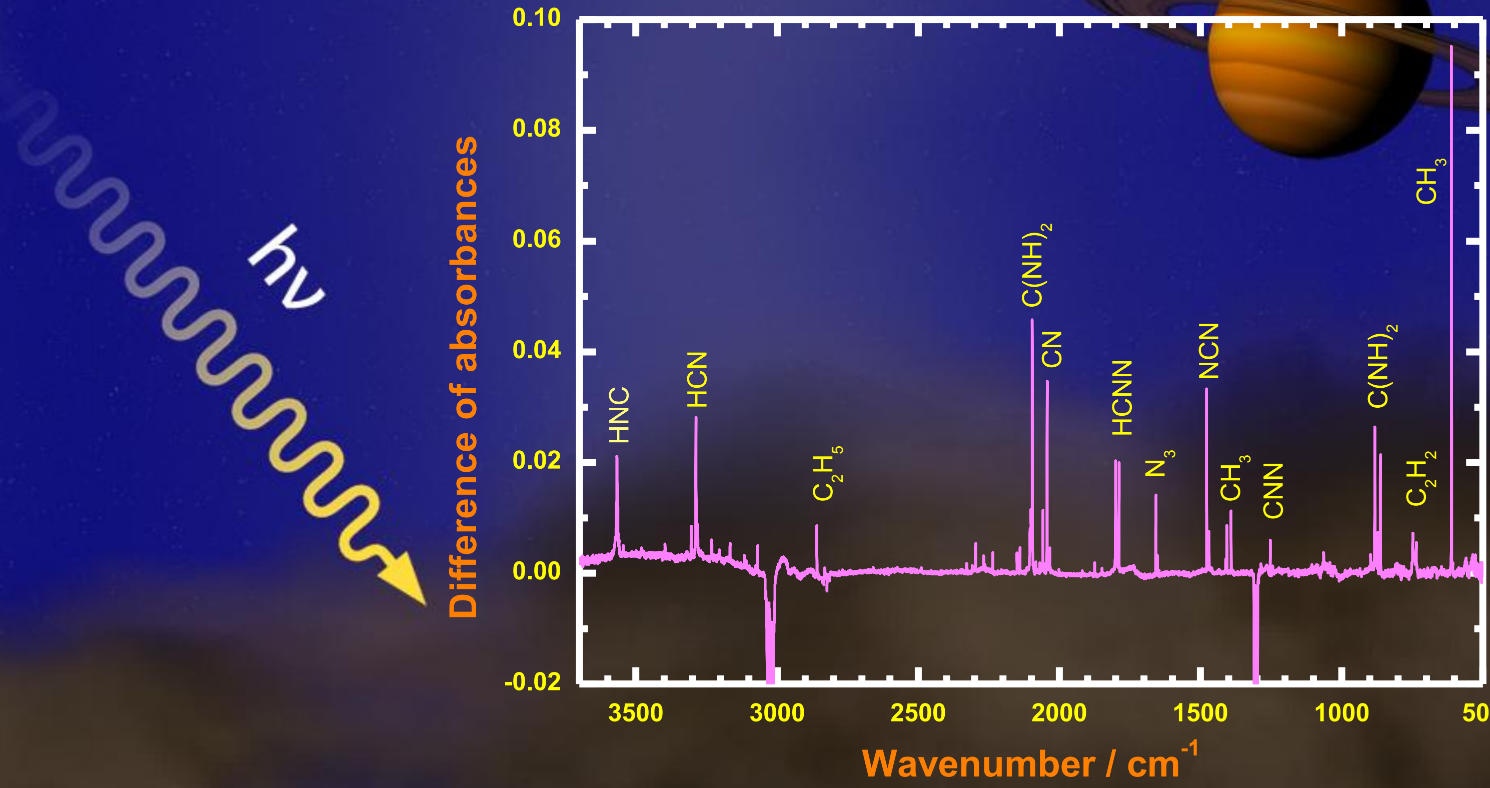
Photochemistry of Titan



Observed Titan's transmission spectrum from the Cassini UVIS FUV spectrograph.

Titan

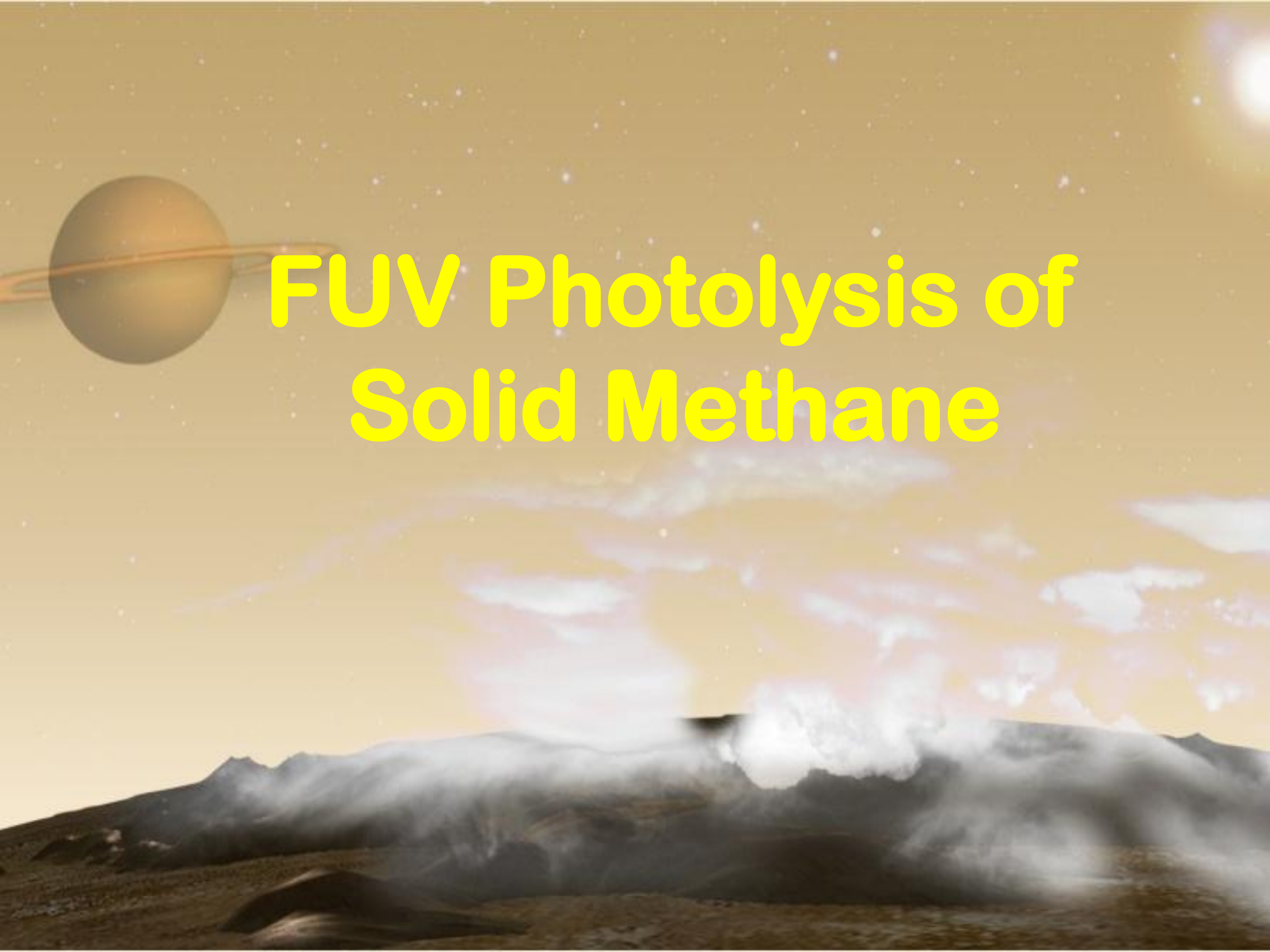
FUV Photolysis of CH_4 Dispersed in Solid N_2



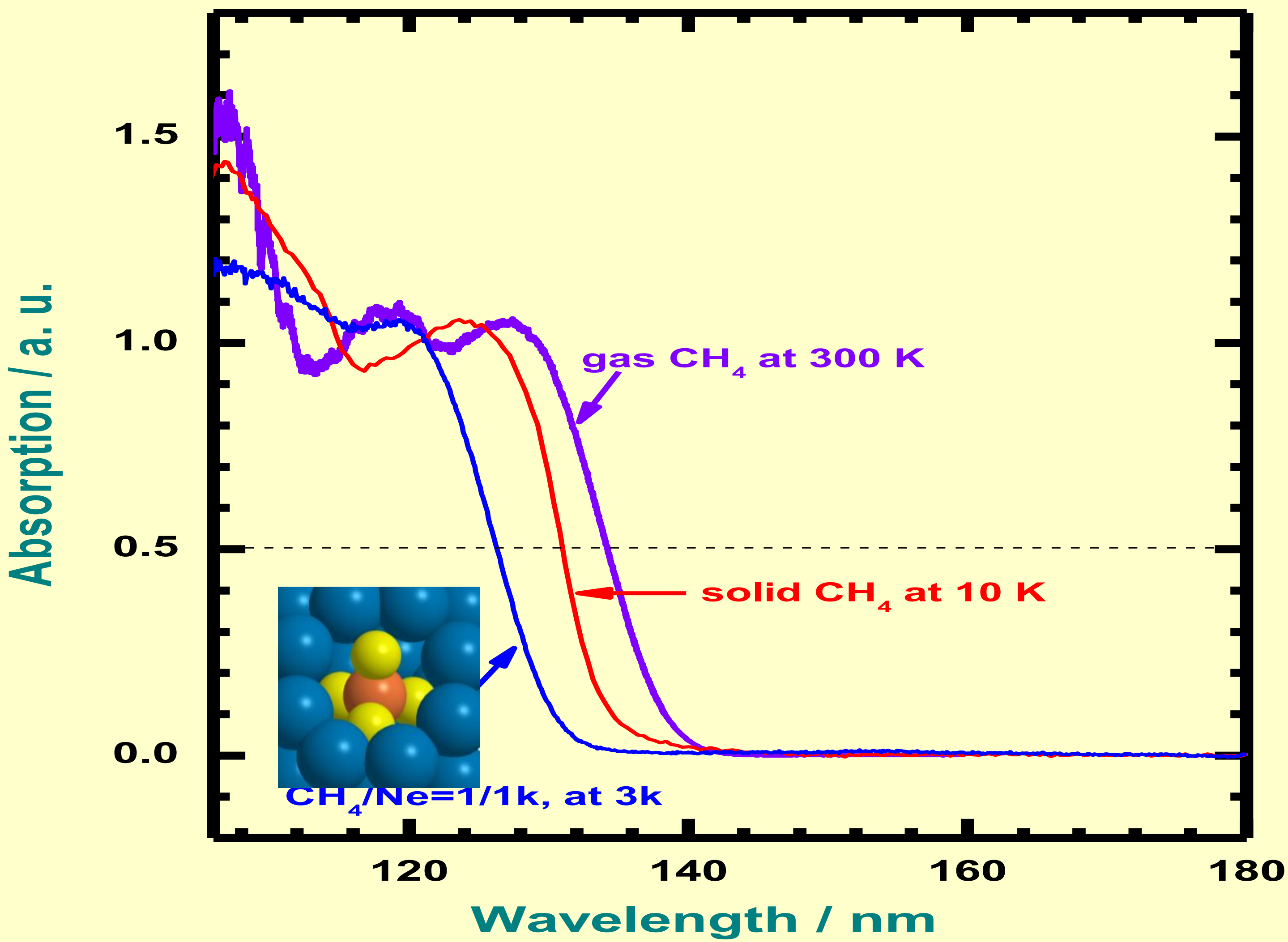
CH_4-N_2 clouds → N_3, C_nN (n = 1-3), CN_2, $(CN)_2$, HCN_2, HC_2N, $C(NH)_2$, HN_3, HNC, HCN, $HCCNH^+$ and $NCCN^+$

Titan

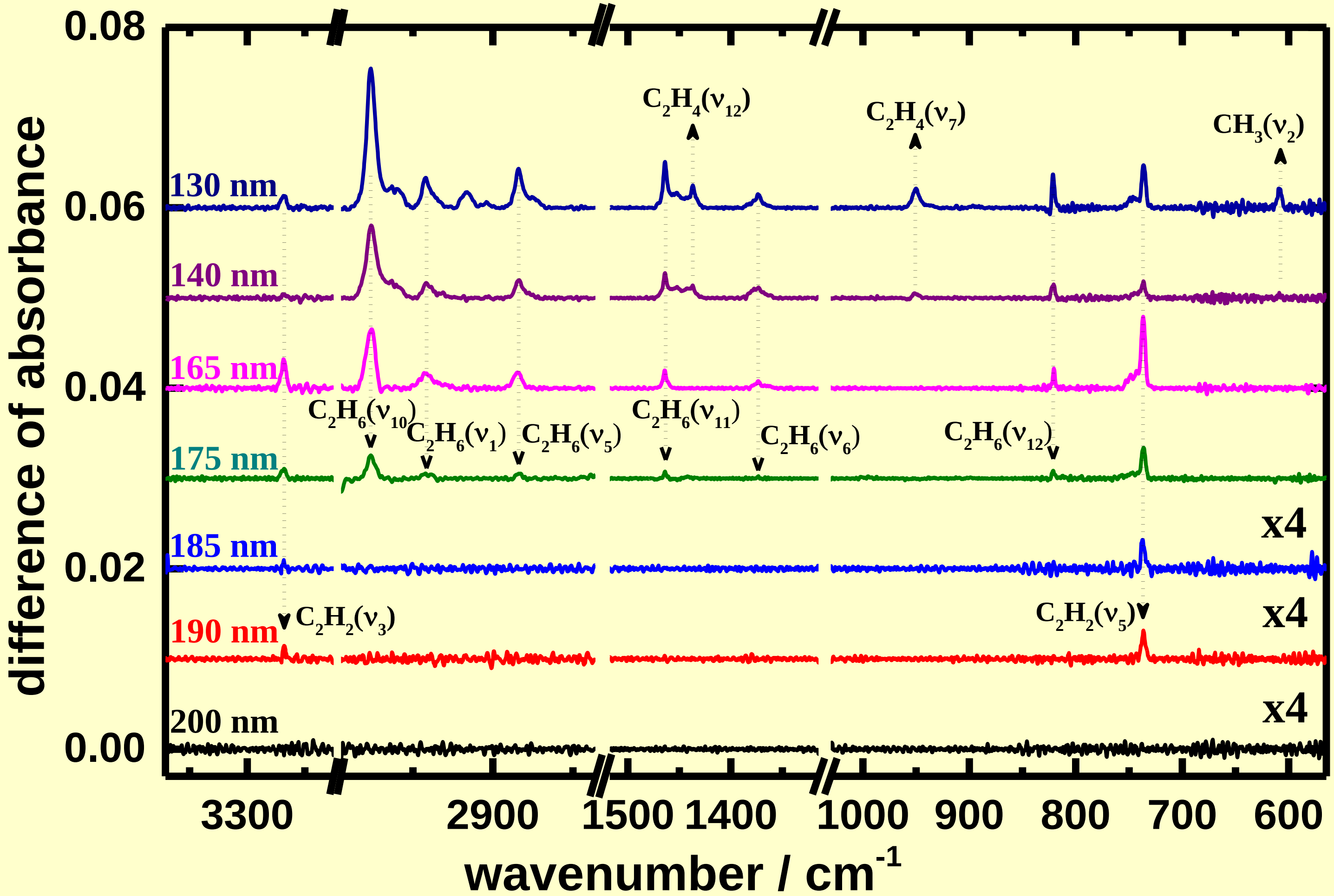

100K
80K
Titan


FUV Photolysis of
Solid Methane

FUV absorption spectra of methane

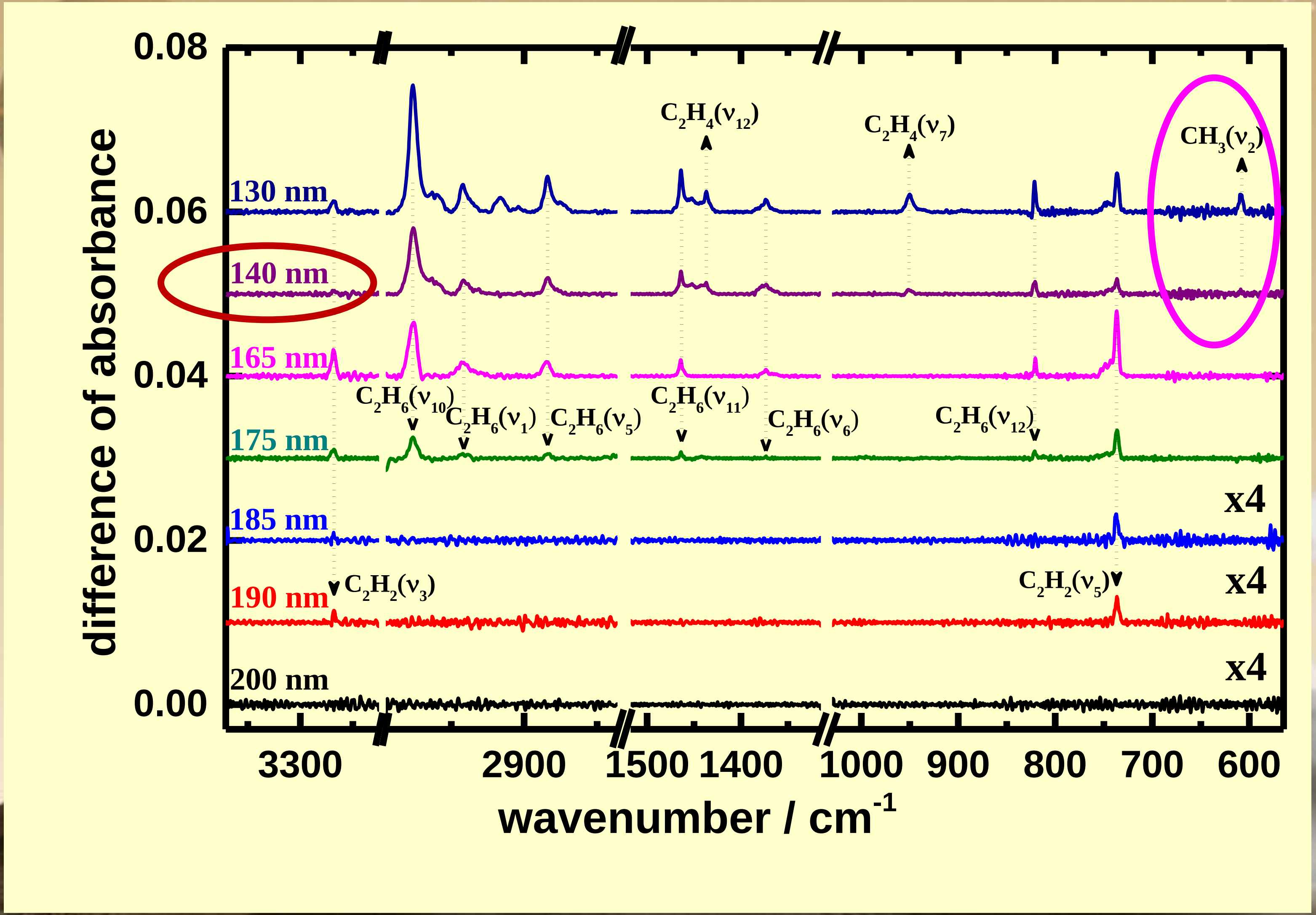


FUV photolysis on pure solid CH_4



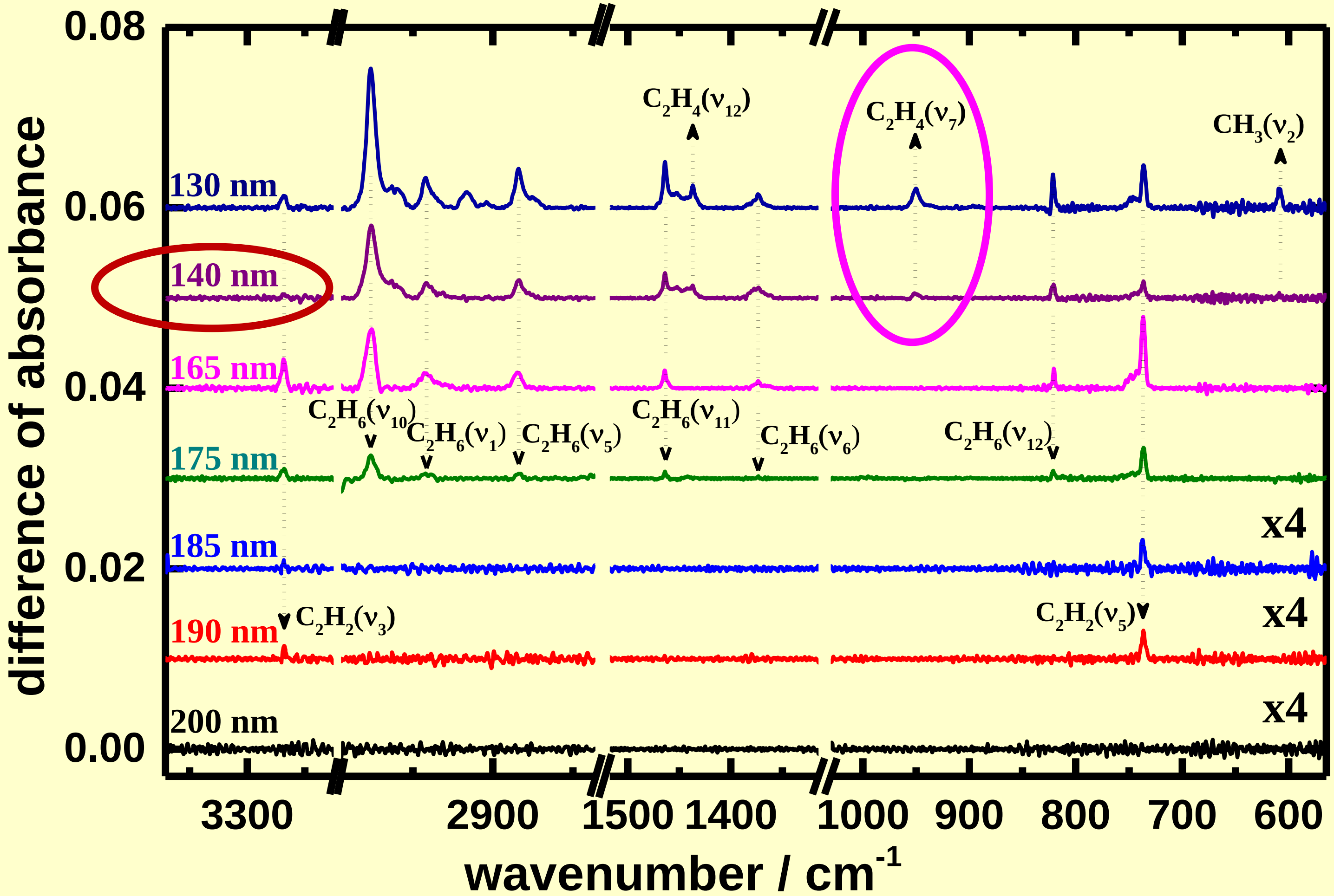
FUV photolysis on pure solid CH_4

CH_3



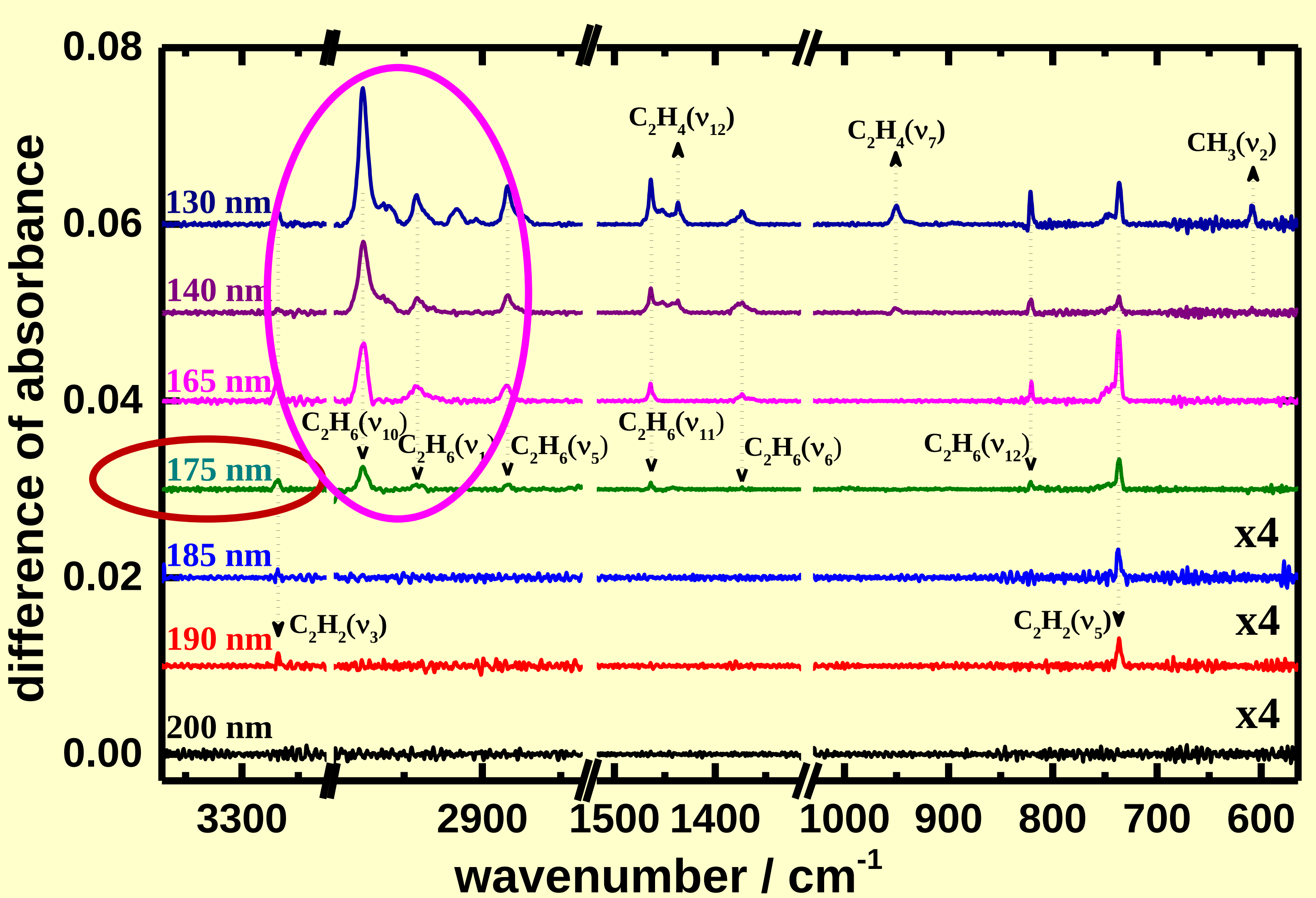
FUV photolysis on pure solid CH_4

C_2H_4



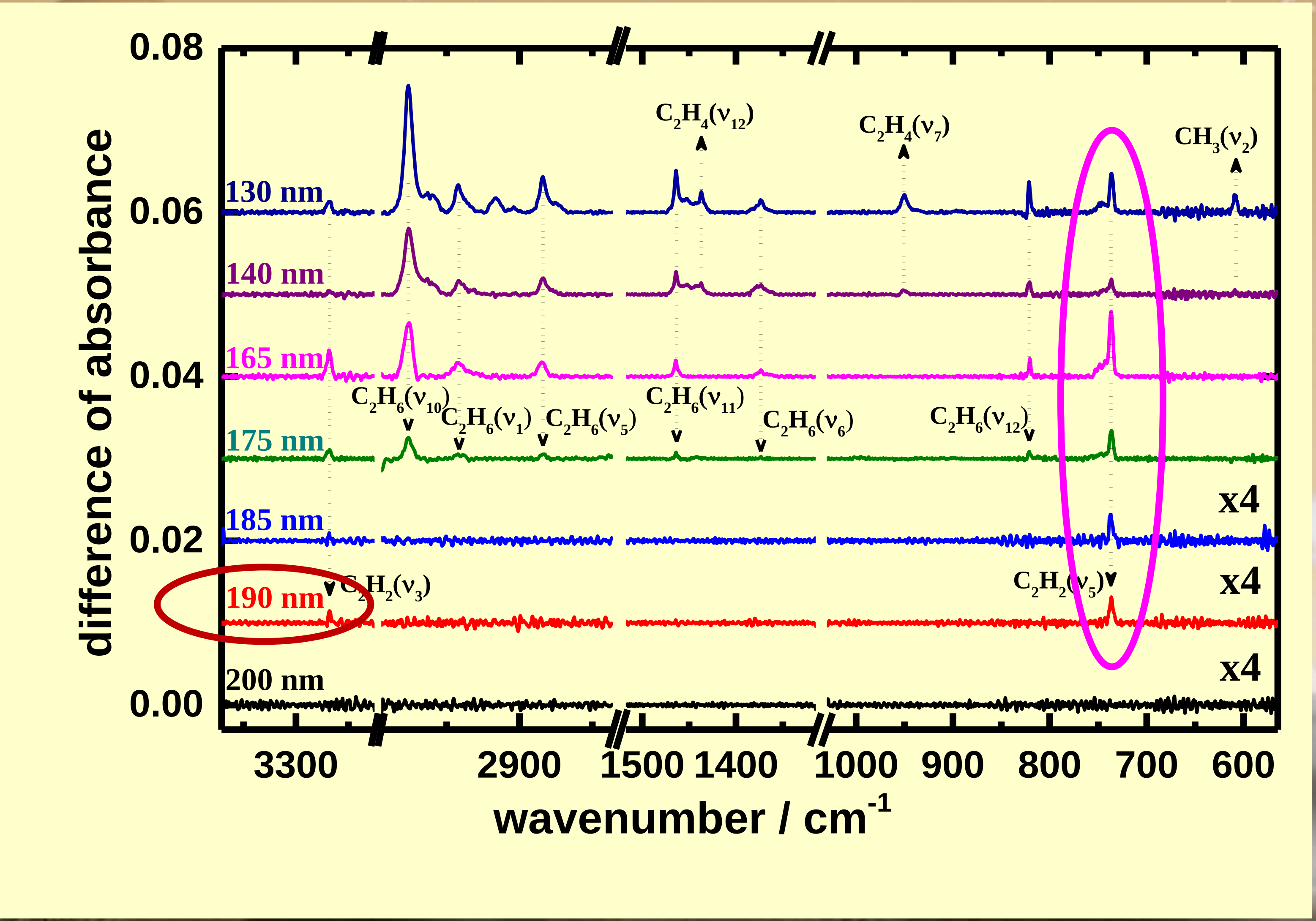
FUV photolysis on pure solid CH_4

C_2H_6

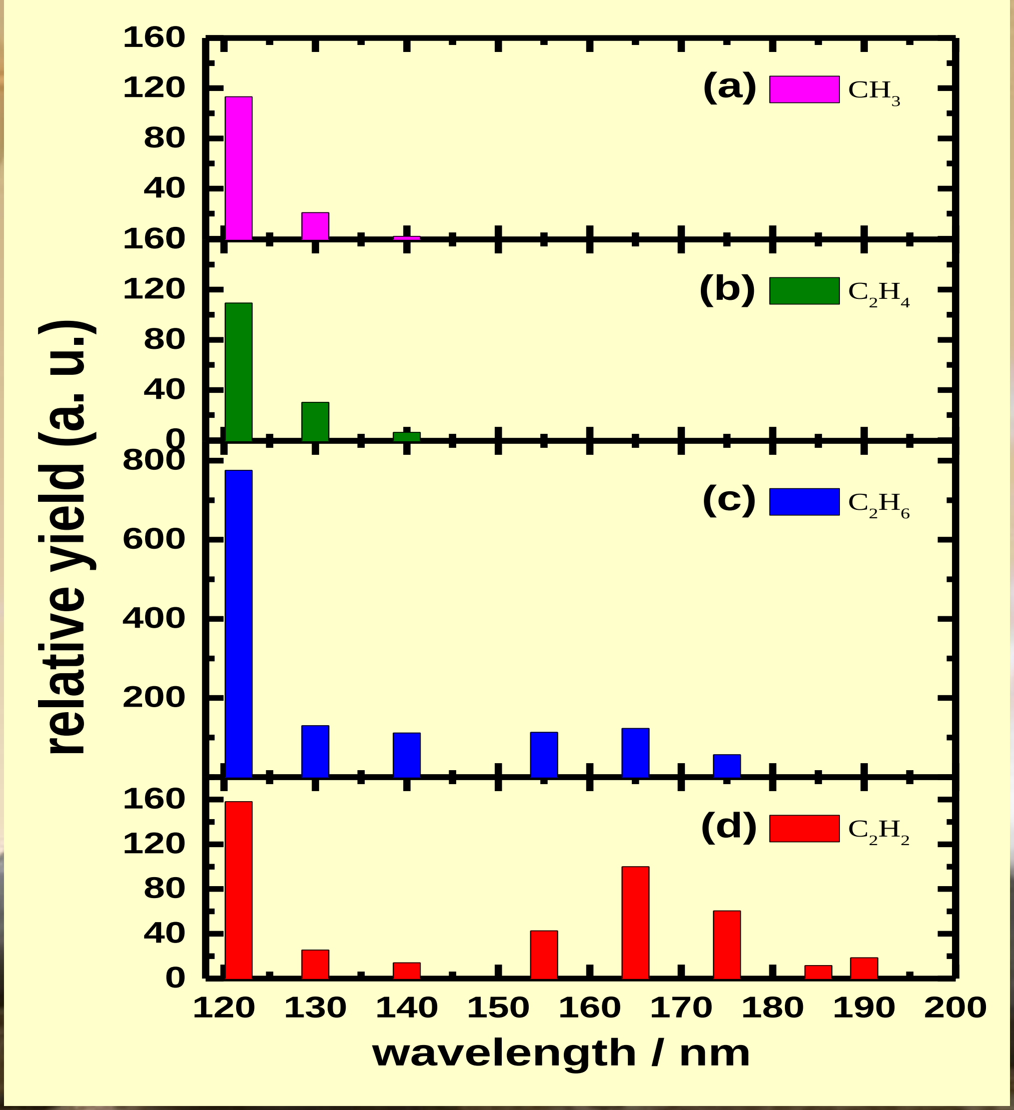


FUV photolysis on pure solid CH_4

C_2H_2

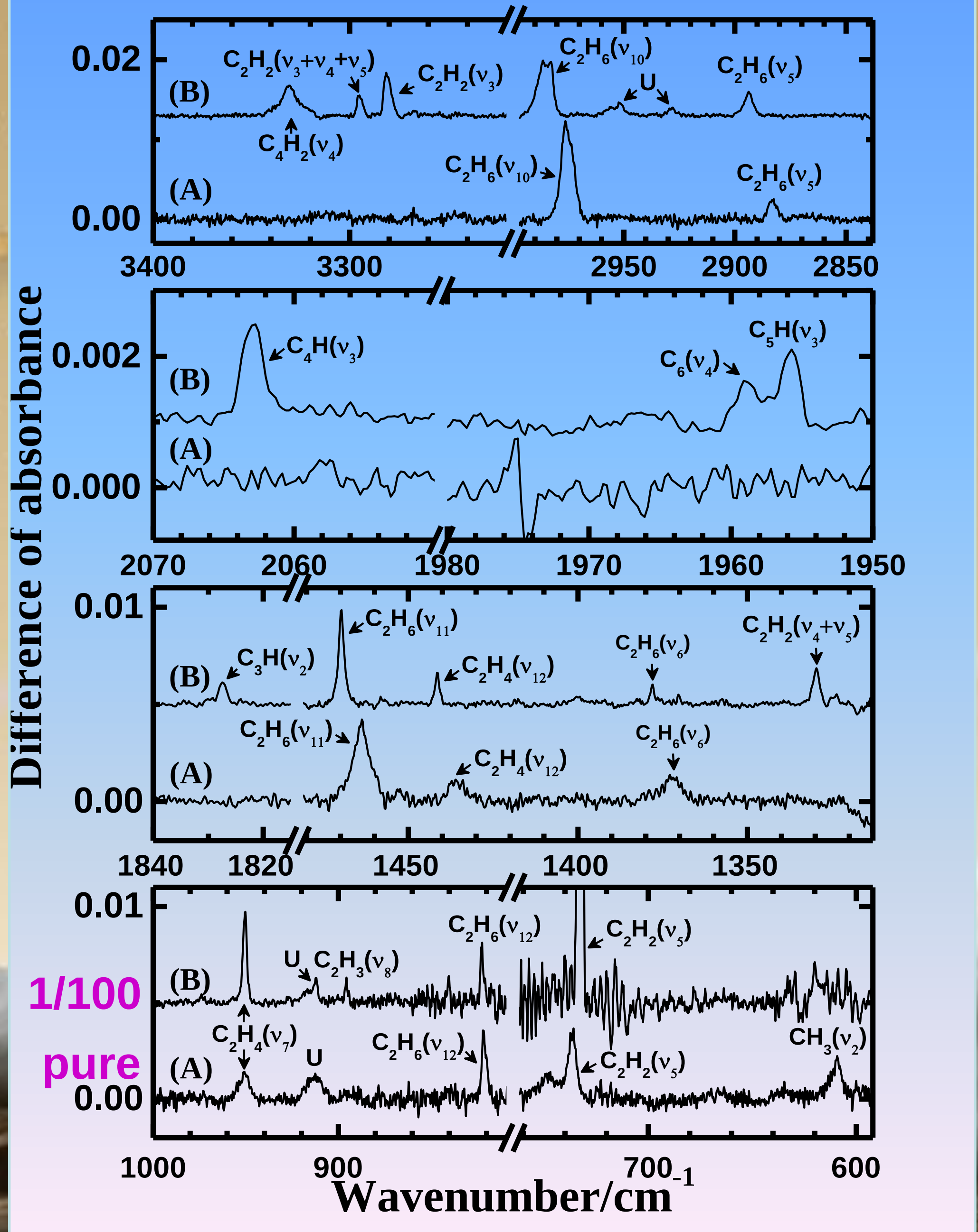


Relative yields vs photolysis wavelength for solid methane at 3 K



Accepted by MNRAS

Photolysis of solid CH_4 121.6 nm at 3 K



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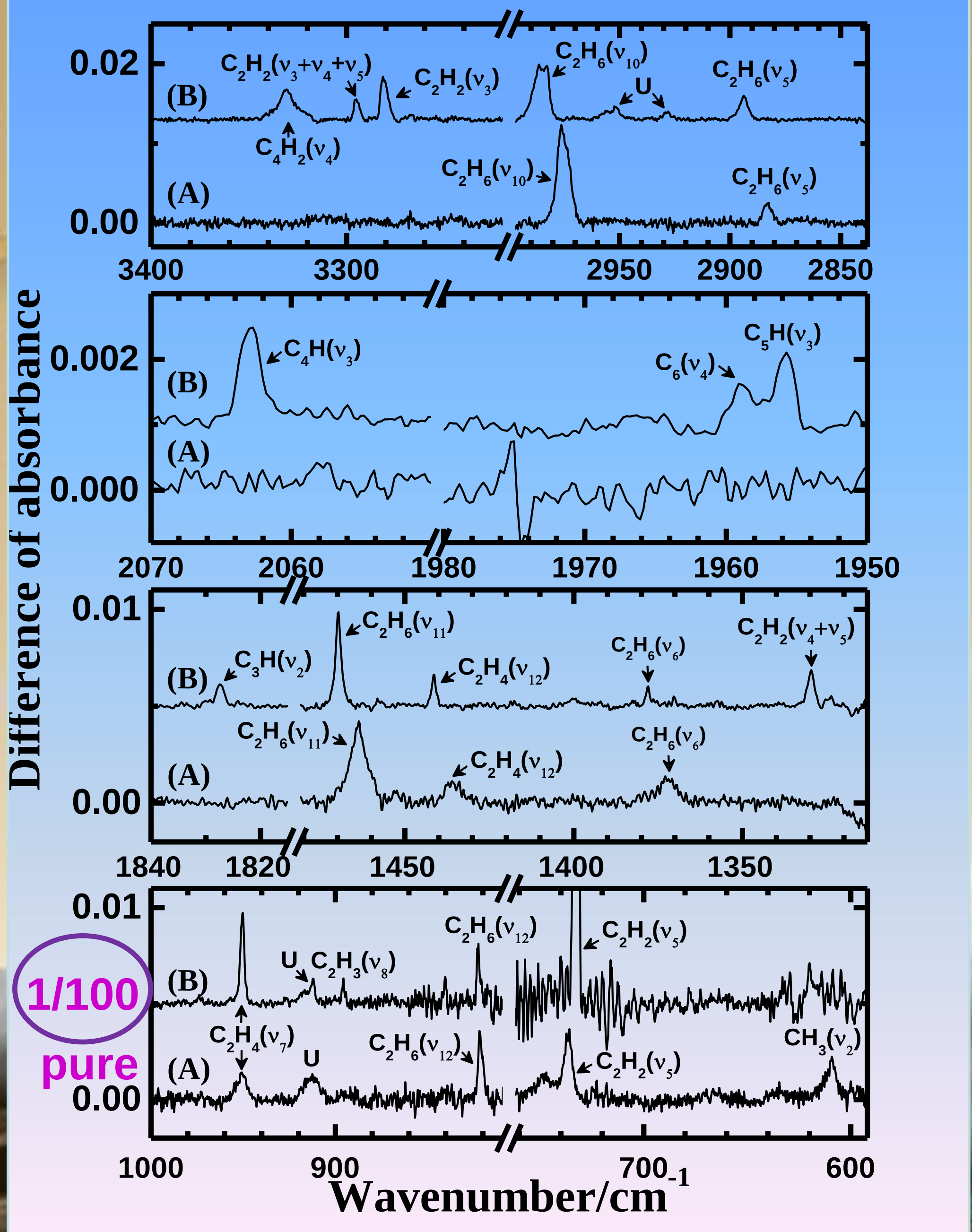

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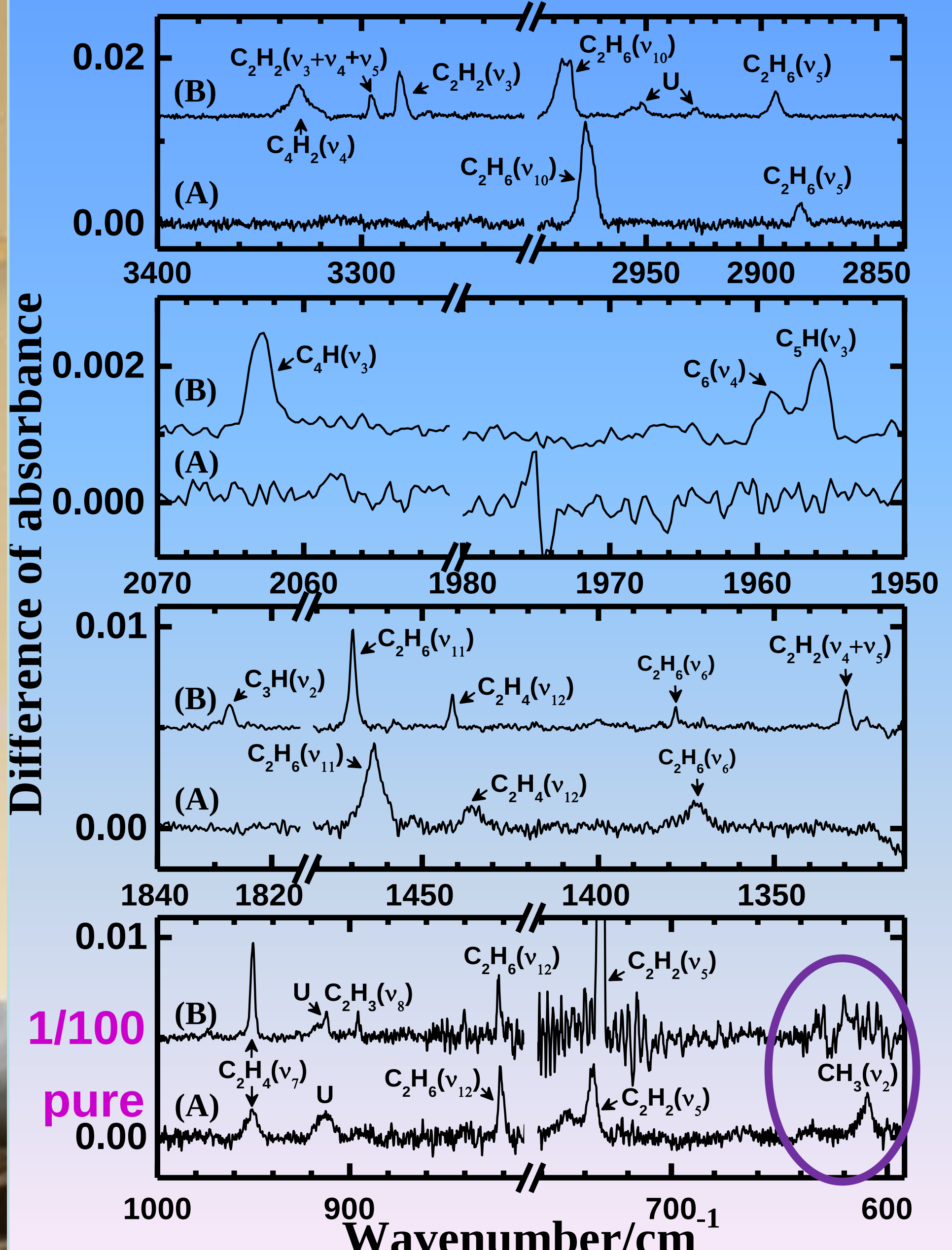

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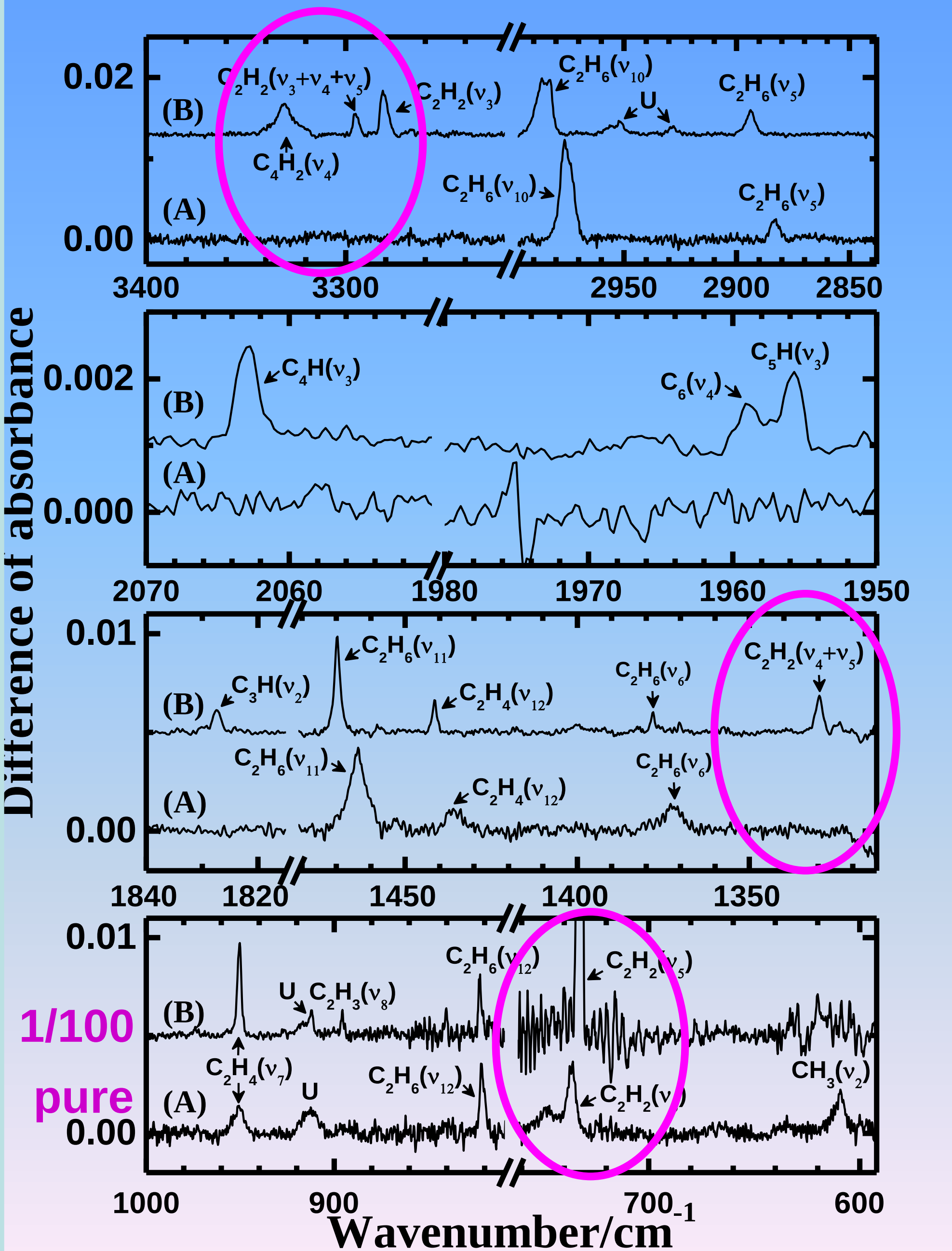

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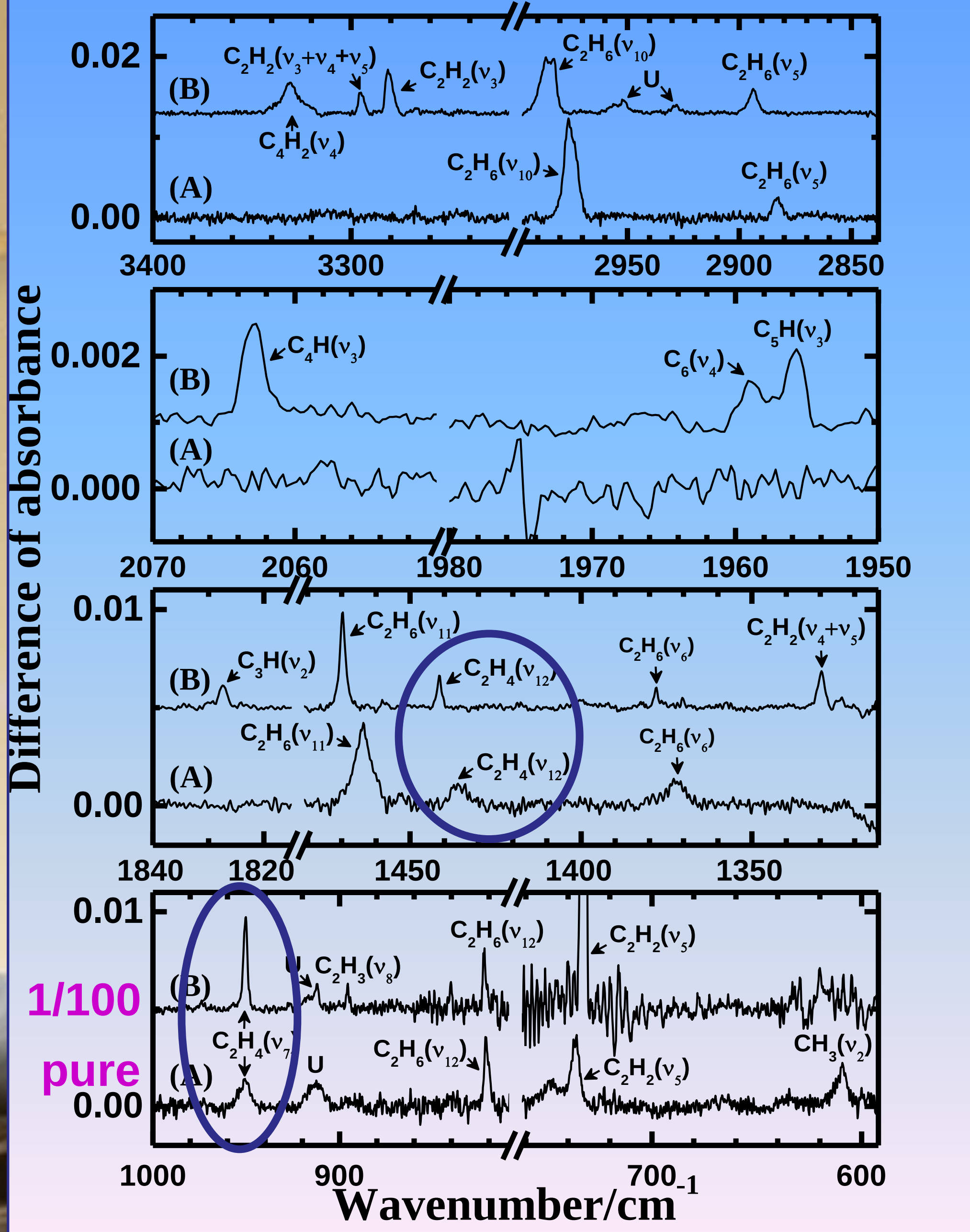

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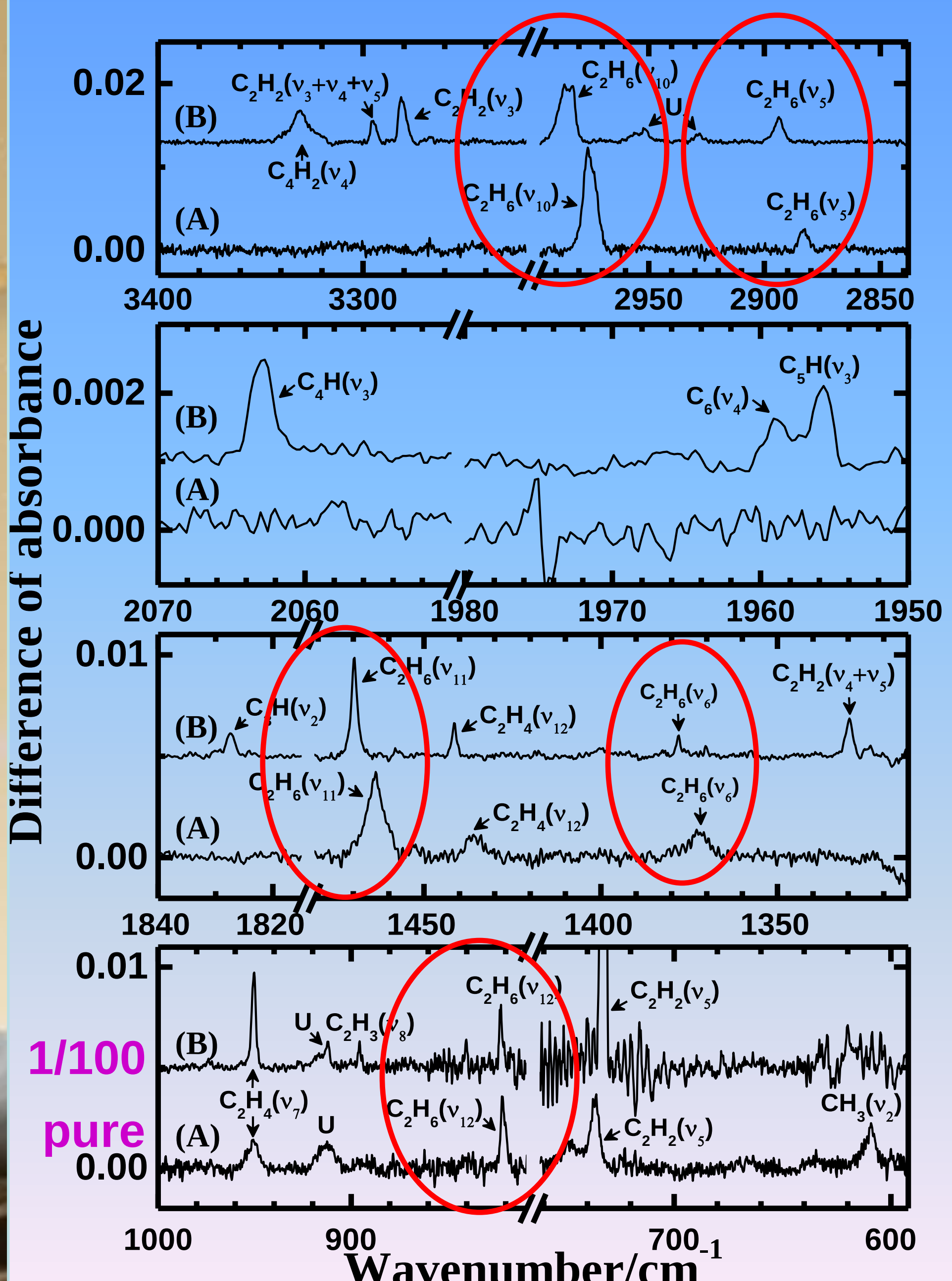

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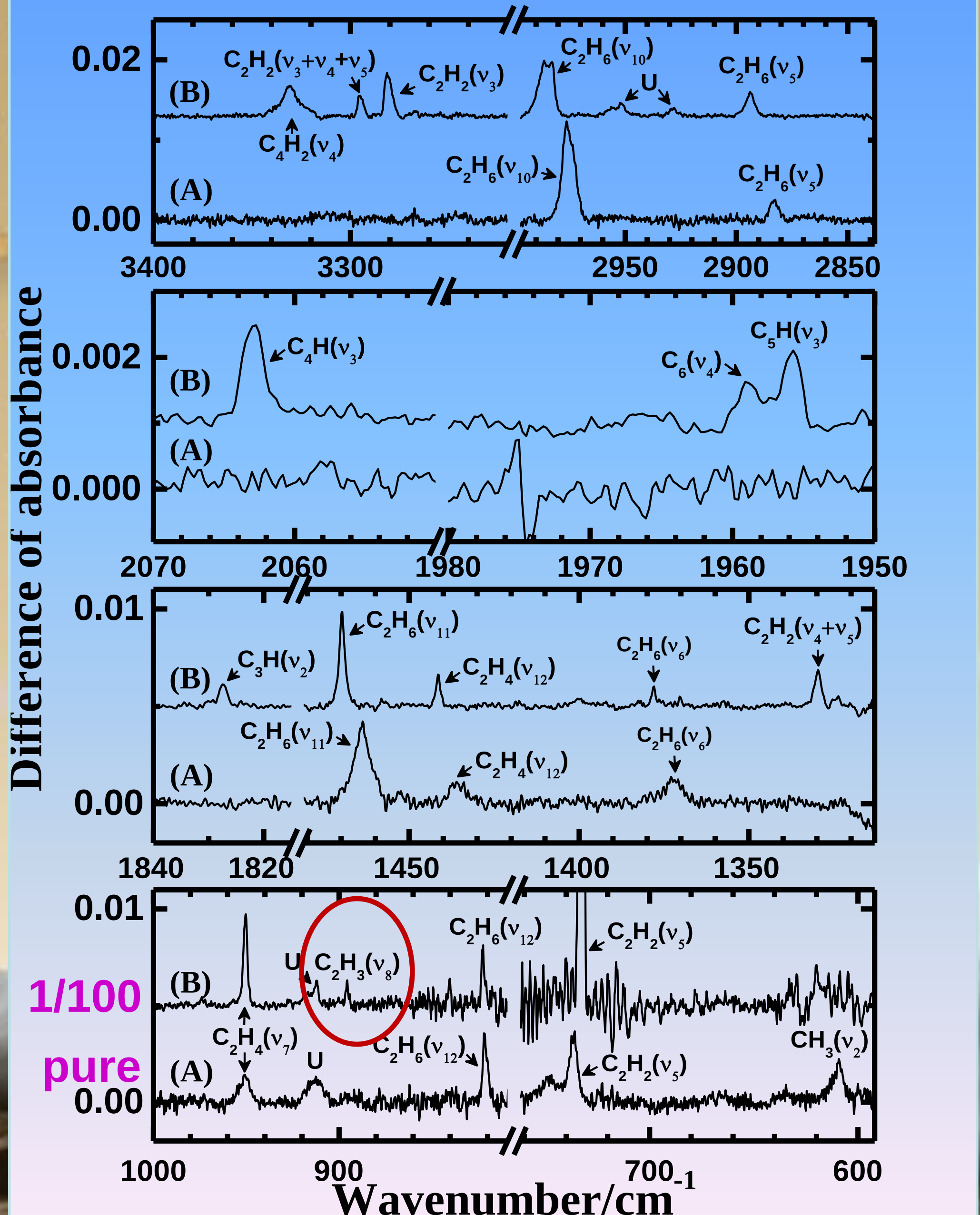

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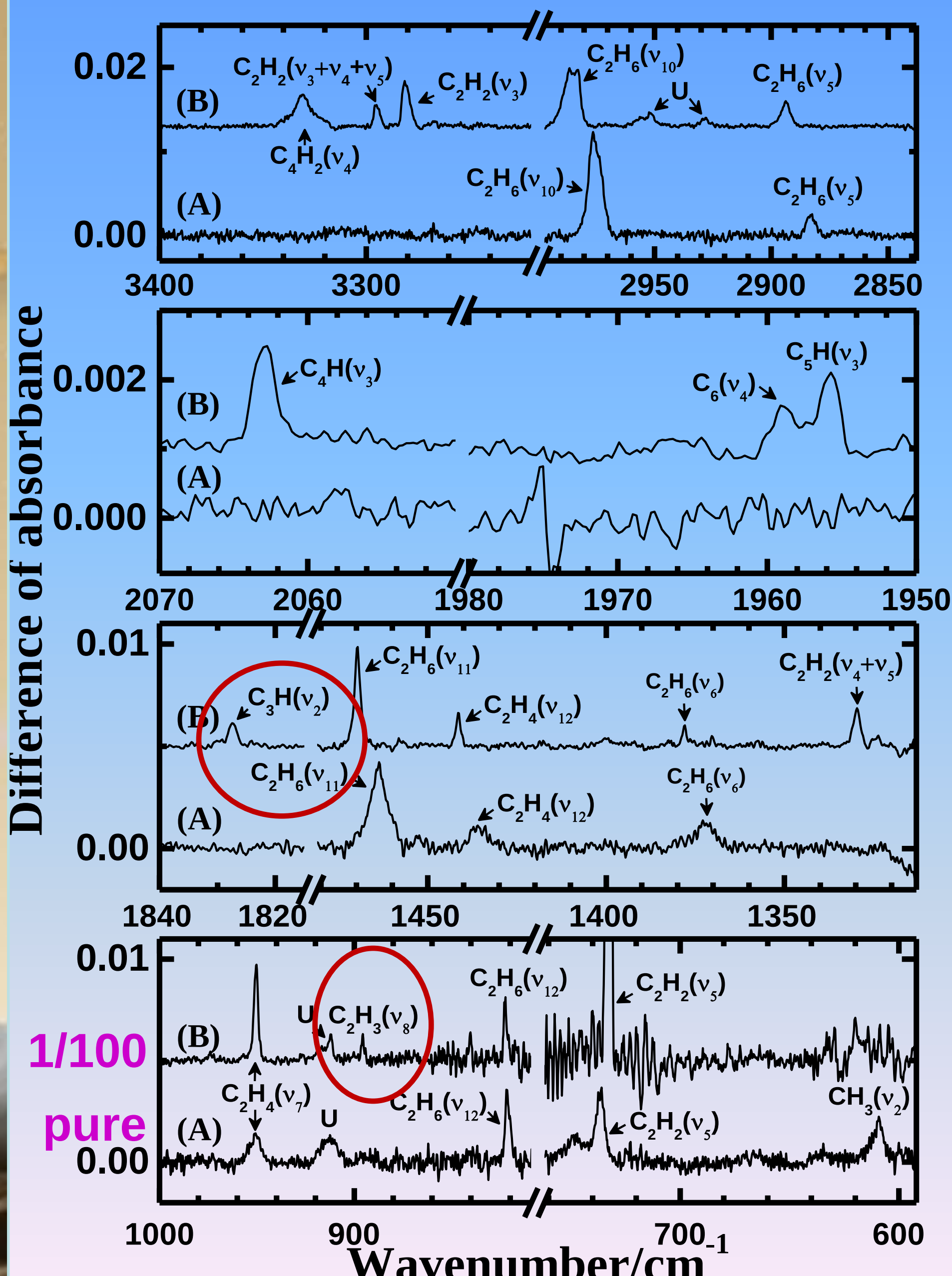

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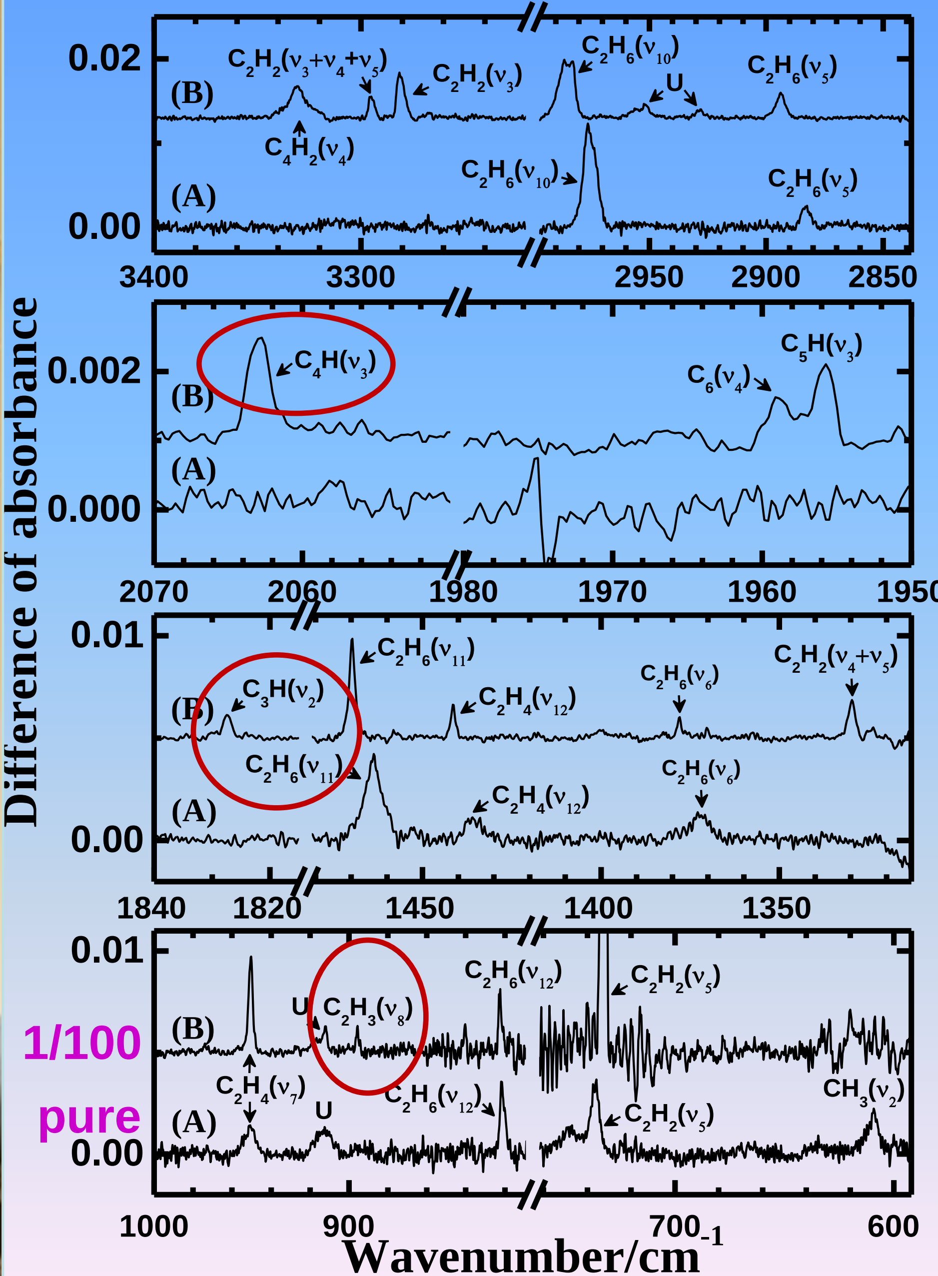

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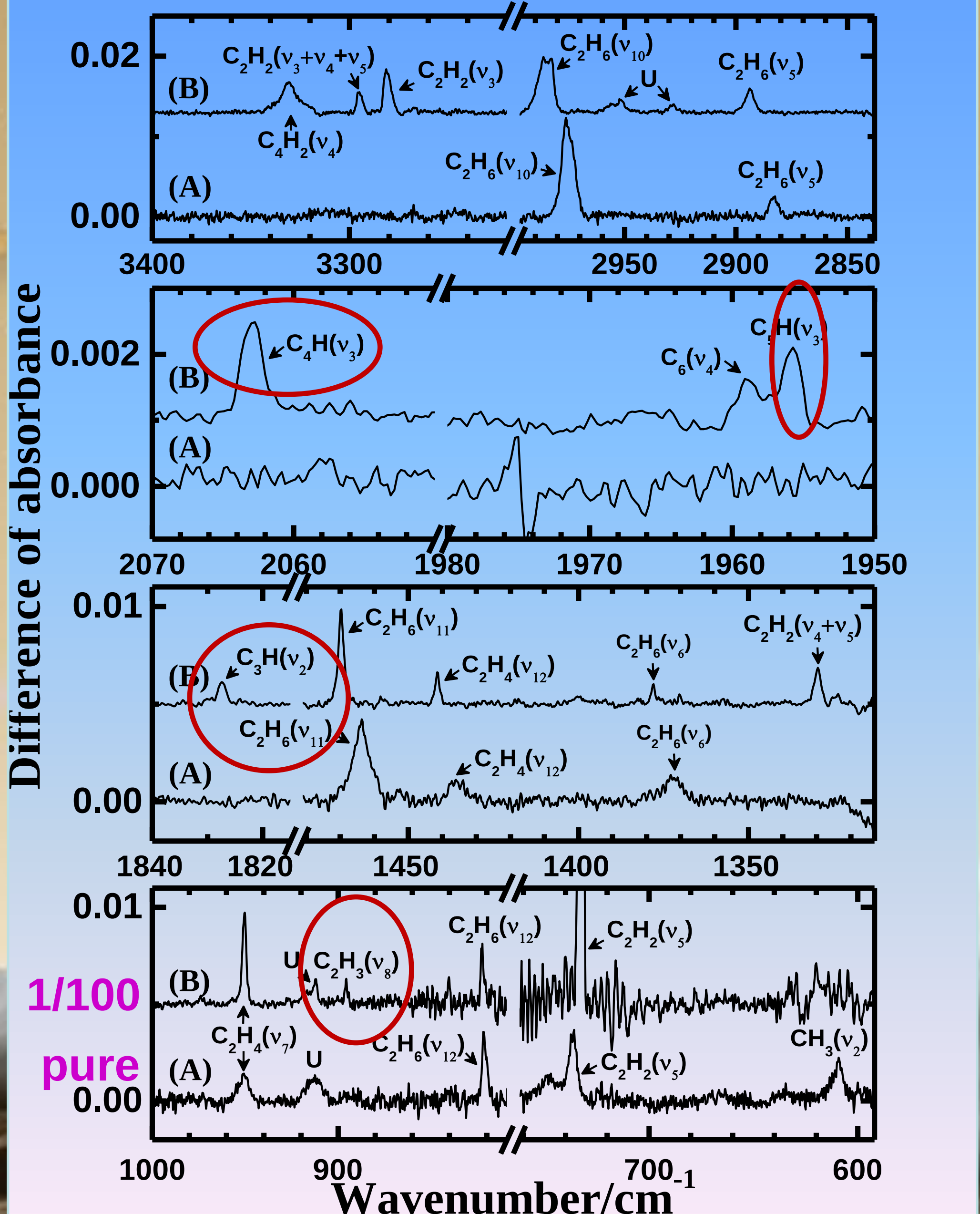

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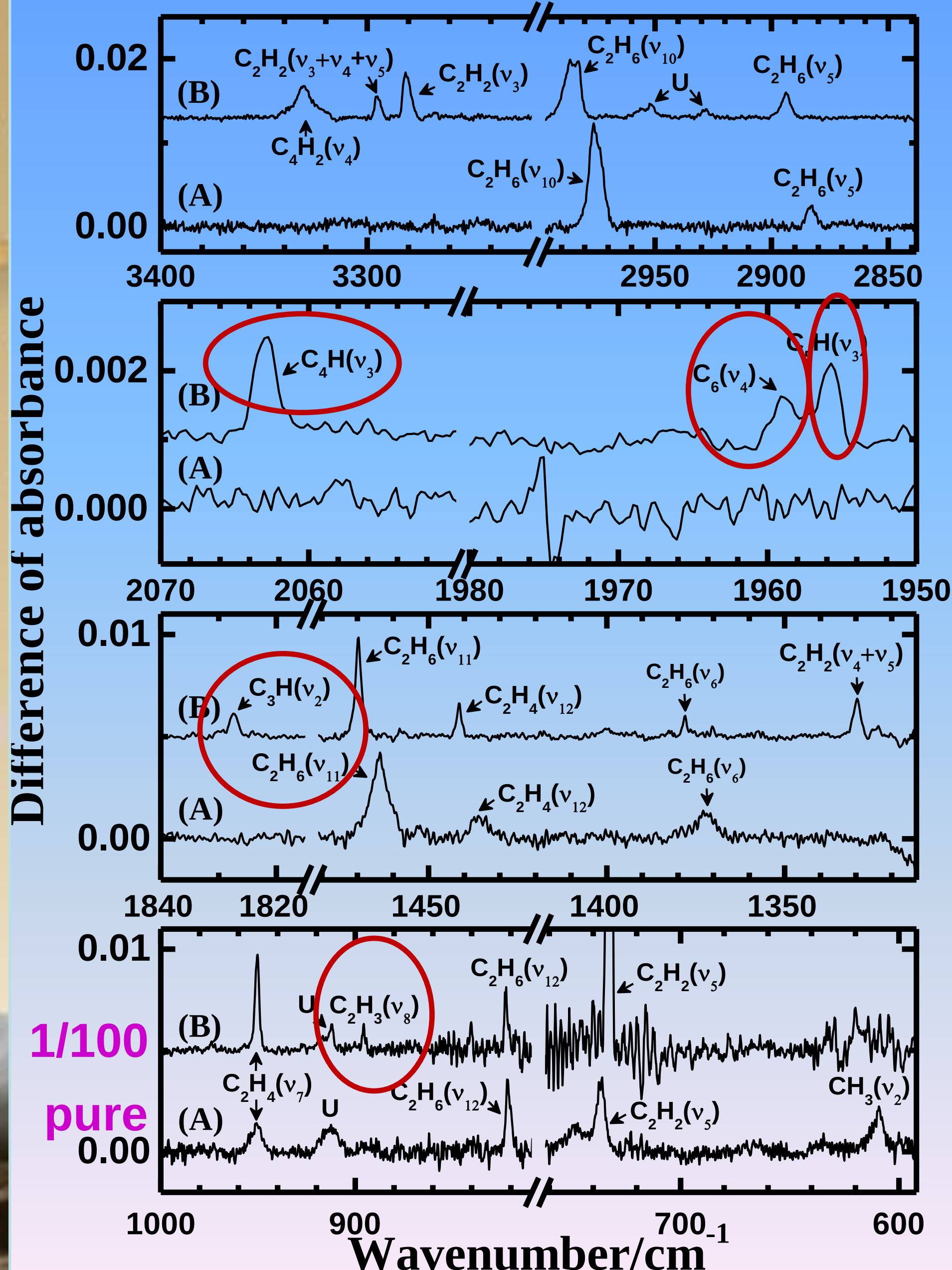

FUV Photolysis of CH_4/Ne=1/1000 at 3 K

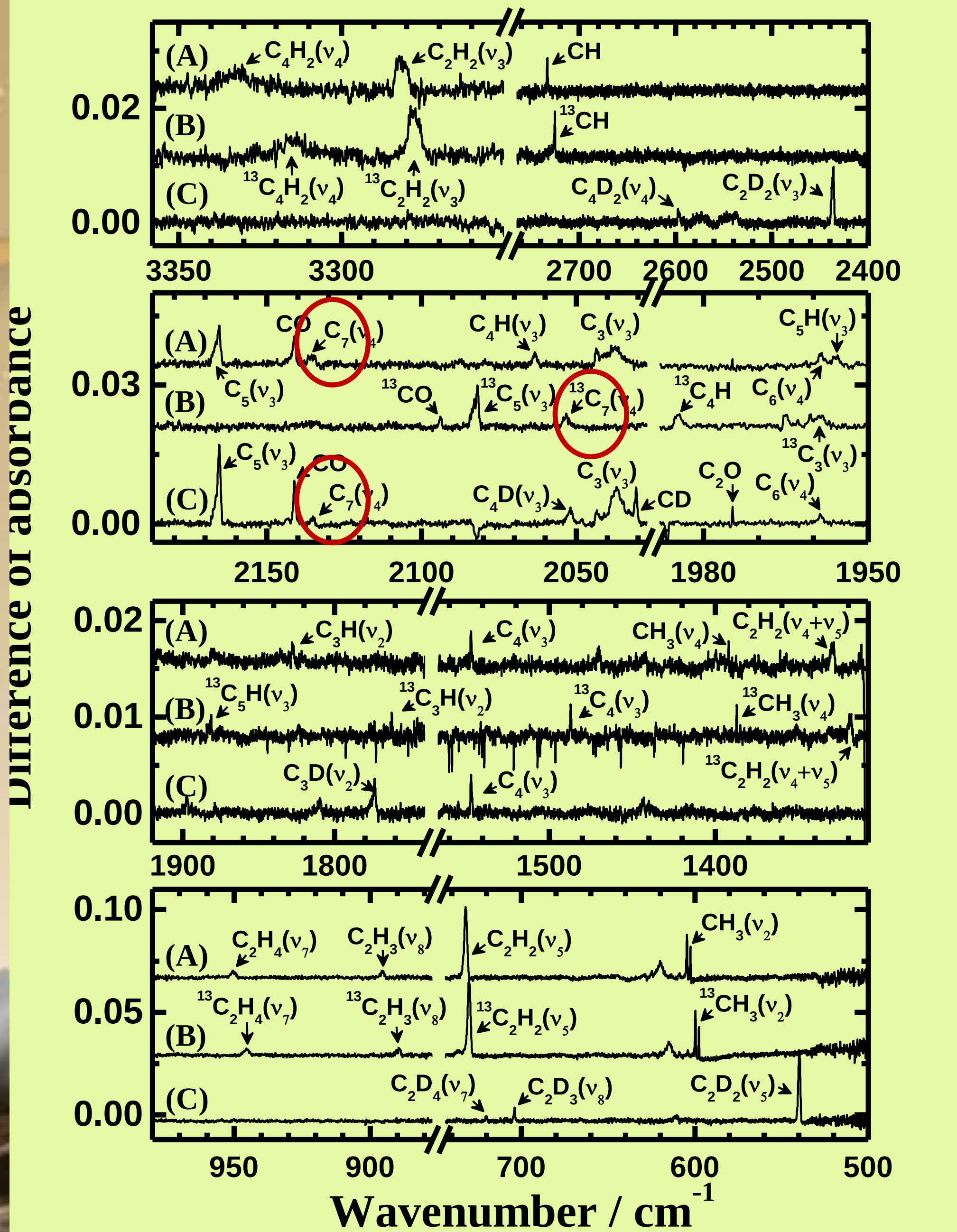

Table 2. Products of Photolysis at 120 nm of Isotopic Methanes Dispersed (1:1000) in Neon at 3 K and Wavenumbers of Their Absorption Lines Assigned from the Indicated Sources

CH_4/Ne		$^{13}CH_4$/Ne		CD_4/Ne	
species	wavenumber (cm^{-1})	species	wavenumber (cm^{-1})	species	wavenumber (cm^{-1})
CH	2732.9	^{13}CH	2725.3	CD	2030.6
CO	2141.1	^{13}CO	2094.0	CO	2141.2
CH_3	602.6/604.5/620.2 (ν_2), 1392.0 (ν_4)	$^{13}CH_3$	597.7/599.6/614.9 (ν_2), 1387.1 (ν_4)	CD_3	1027.8 (ν_4), 2378.9 (ν_3)
C_2H	1835.9 (ν_3)	$^{13}C_2H$	1776.0 (ν_3)	C_2D	1737.6 (ν_3)
C_2O	1974.8 (ν_1)	$^{13}C_2O$	1964.9 (ν_1)	C_2O	1974.8 (ν_1)
C_2H_2	732.2 (ν_5), 1329.6 ($\nu_4+\nu_5$), 3282.2 (ν_3), 3296.3 ($\nu_3+\nu_4+\nu_5$)	$^{13}C_2H_2$	730.1 (ν_5), 1319.1 ($\nu_4+\nu_5$), 3278.7 (ν_3)	C_2D_2	539.7 (ν_5), 2436.3 (ν_3)
C_2H_3	895.3 (ν_8)	$^{13}C_2H_3$	889.5 (ν_8)	C_2D_3	703.9 (ν_8)
C_2H_4	950.4 (ν_7)	$^{13}C_2H_4$	945.5 (ν_7)	C_2D_4	720.1 (ν_7)
C_3	2036.4/2038.7/2041.1/2043.5 (ν_3)	$^{13}C_3$	1958.4/1960.5/1962.9/1964.9 (ν_3)	C_3	2037.0/2038.5/2041.1/2043.5 (ν_3)
C_3H	1827.5 (ν_2)	$^{13}C_3H$	1762.2 (ν_2)	C_3D	1773.6 (ν_2)
C_4	1547.2 (ν_3)	$^{13}C_4$	1487.1 (ν_3)	C_4	1547.0 (ν_3)
C_4H	2063.5 (ν_3)	$^{13}C_4H$	1984.6 (ν_3)	C_4D	2051.9 (ν_3)
C_4H_2	3330.5 (ν_4)	$^{13}C_4H_2$	3313.7 (ν_4)	C_4D_2	2597.4 (ν_4)
C_5	2165.4 (ν_3)	$^{13}C_5$	2082.0 (ν_3)	C_5	2165.4 (ν_3)
C_5H	1955.6 (ν_3)	$^{13}C_5H$	1881.2 (ν_3)	C_5D	1917.5 (ν_3)
C_6	1958.4 (ν_4)	$^{13}C_6$	1890.2 (ν_4)	C_6	1958.7 (ν_4)
C_7	2135.2 (ν_4)	$^{13}C_7$	2053.4 (ν_4)	C_7	2135.1 (ν_4)
				U[a]	1937.0

[a]Unidentified line.

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C_3	2036.4/2038.7/2041.1/2043.5 (ν_3)	$^{13}C_3$	1958.4/1960.5/1962.9/1964.9 (ν_3)	C_3	2037.0/2038.5/2041.1/2043.5 (ν_3)
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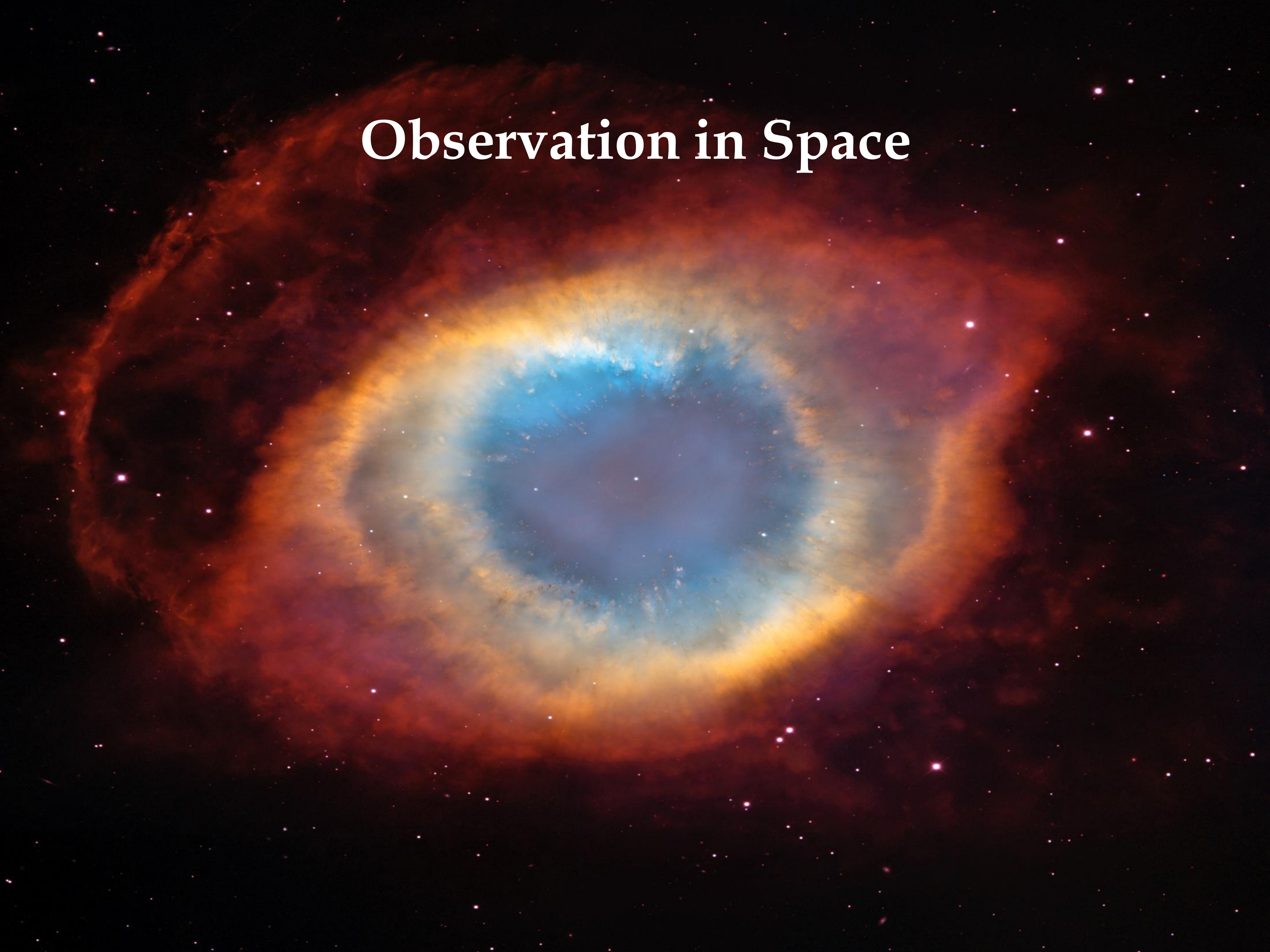
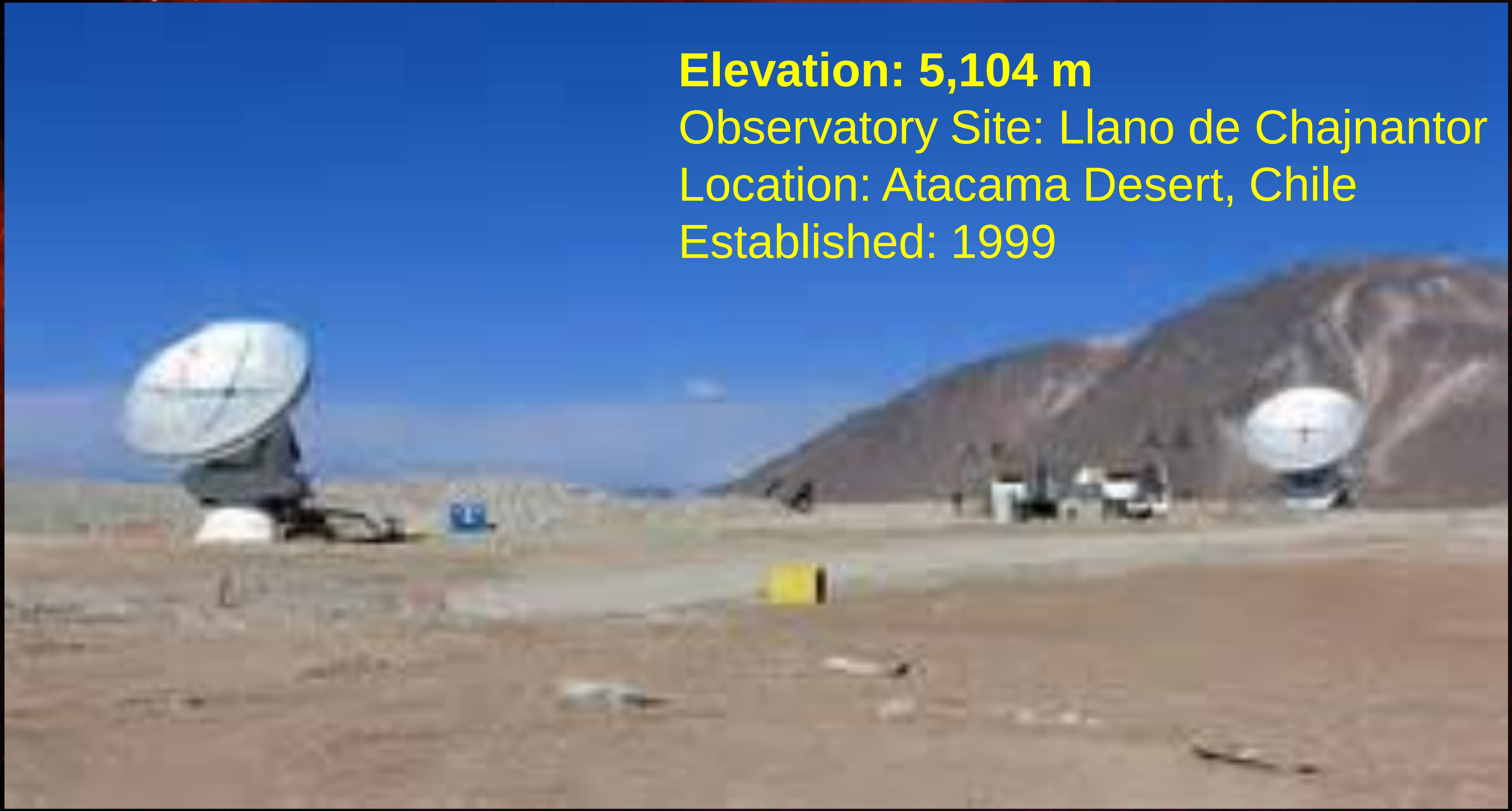
C_4H	2063.5 (ν_3)	$^{13}C_4H$	1984.6 (ν_3)	C_4D	2051.9 (ν_3)
C_4H_2	3330.5 (ν_4)	$^{13}C_4H_2$	3313.7 (ν_4)	C_4D_2	2597.4 (ν_4)
C_5	2165.4 (ν_3)	$^{13}C_5$	2082.0 (ν_3)	C_5	2165.4 (ν_3)
C_5H	1955.6 (ν_3)	$^{13}C_5H$	1881.2 (ν_3)	C_5D	1917.5 (ν_3)
C_6	1958.4 (ν_4)	$^{13}C_6$	1890.2 (ν_4)	C_6	1958.7 (ν_4)
C_7	2135.2 (ν_4)	$^{13}C_7$	2053.4 (ν_4)	C_7	2135.1 (ν_4)
				U[a]	1937.0

[a]Unidentified line.

Observation in Space



Observation in Space



Observatory: millimeter wave, submillimeter, microwave

Transient species observed in IRC + 10216

Carbon star
IRC +10216

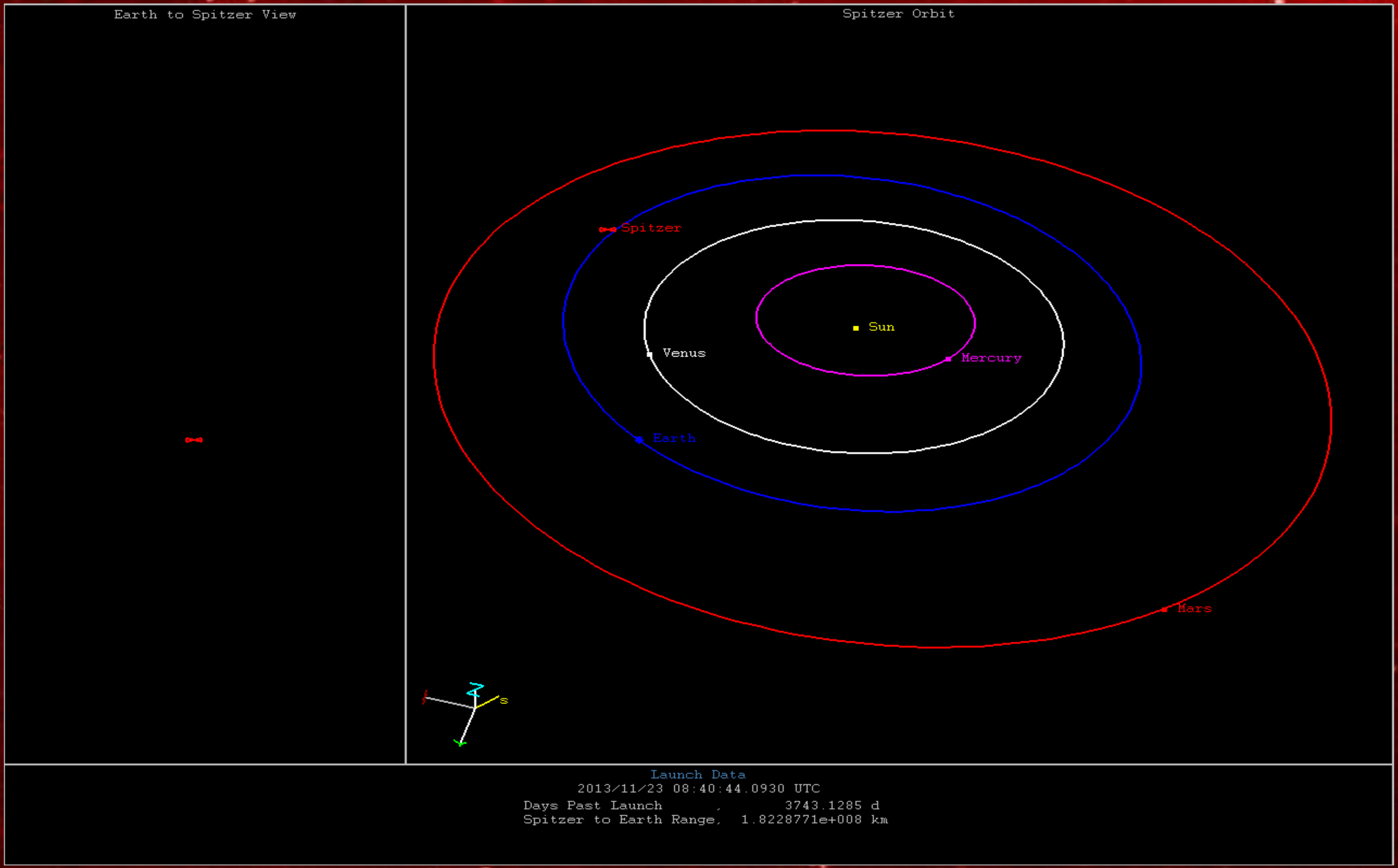
Telescope

C_2, C_3, C_4…C_n
C_2H, C_3H, C_4H,
C_5H, C_6H,…C_nH

NASA's Infrared Spitzer Space Telescope

http://www.spitzer.caltech.edu/mission/38-Science

Where Is Spitzer Now?

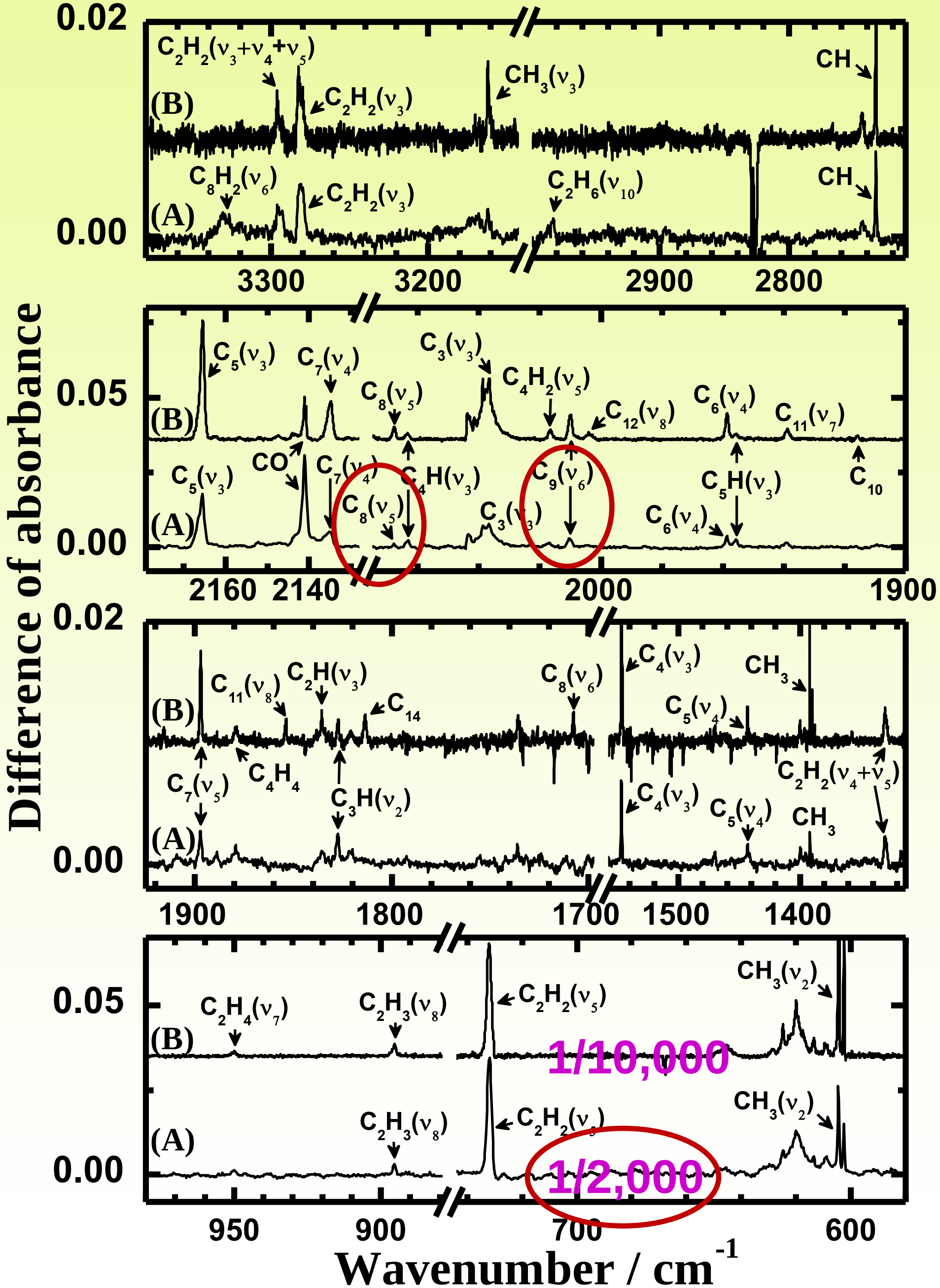


http://www.spitzer.caltech.edu/mission/where_is_spitzer

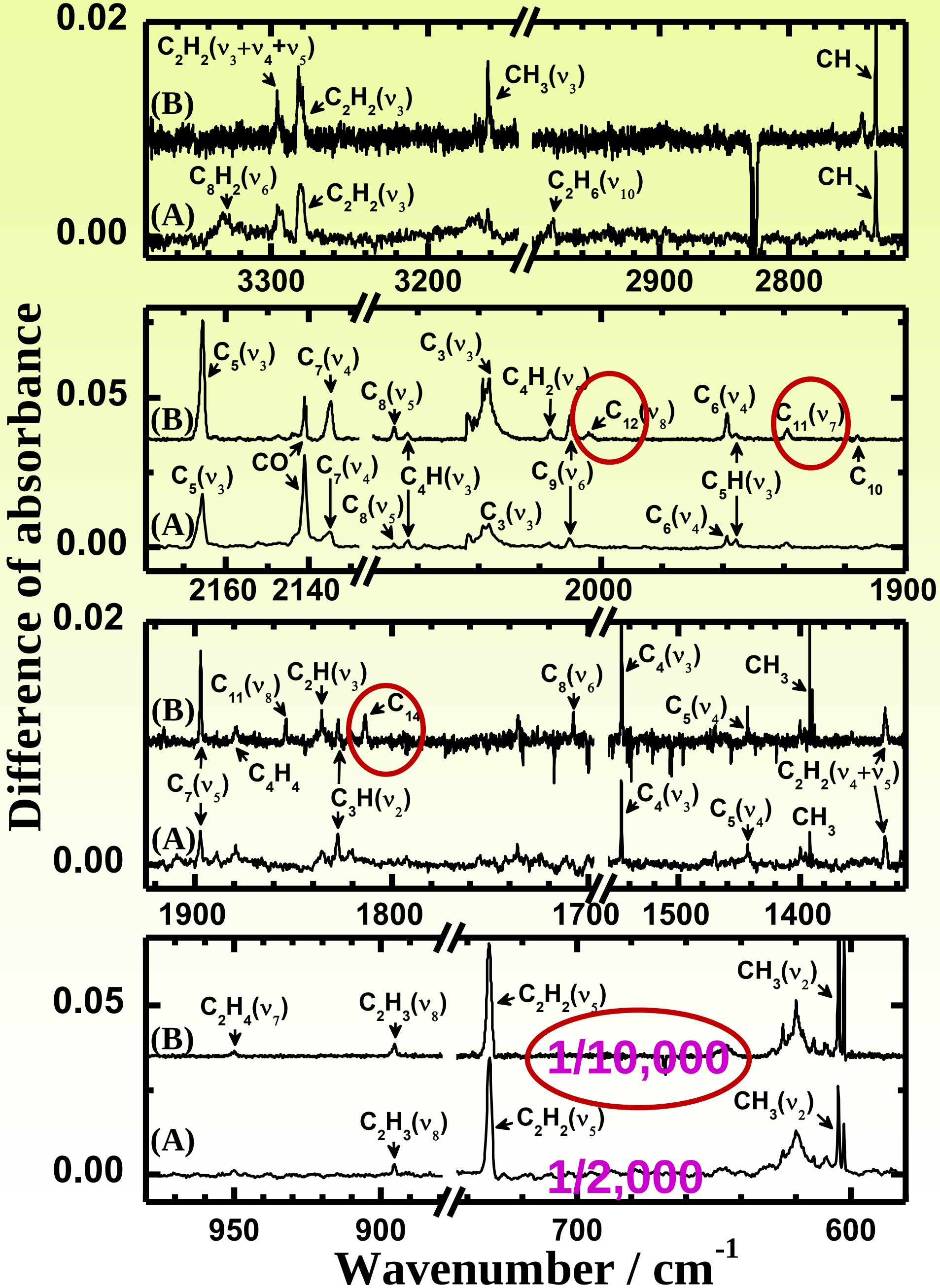
Molecules identified in IRC+ 10216

CO	C_2H	HC_3N	C_2S	SiO	NaCl
CS	C_3H	HC_5N	C_3S	SiS	AlCl
CN	C_4H	HC_7N	C_3N	SiC	KCl
HCN	C_5H	HC_9N	C_5N	SiN	AlF
HNC	C_6H	C_4H_2	HC_4N	SiC_2	MgNC
CH_4	C_7H	C_6H_2	C_3H_2	SiC_3	MgCN
NH_3	C_8H	HC_2N	CH_3CN	SiCN	AlNC
H_2S		C_3	CP	SiC_4	KCN
C_2H_2		C_5	PN	SiH_4	NaCN
C_2H_4					

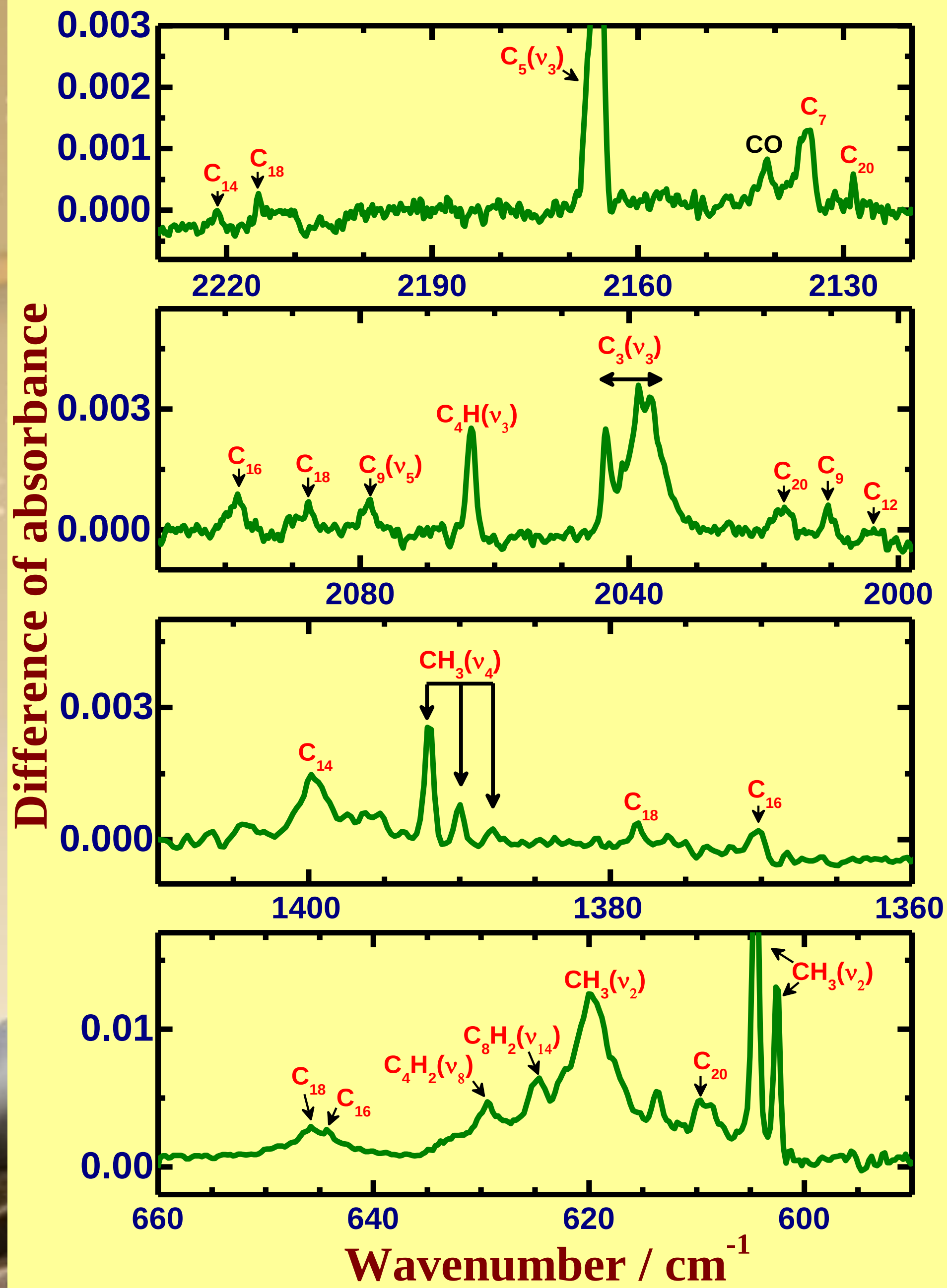
FUV Photolysis of methane/Ne at 3 K



FUV Photolysis of methane/Ne at 3 K

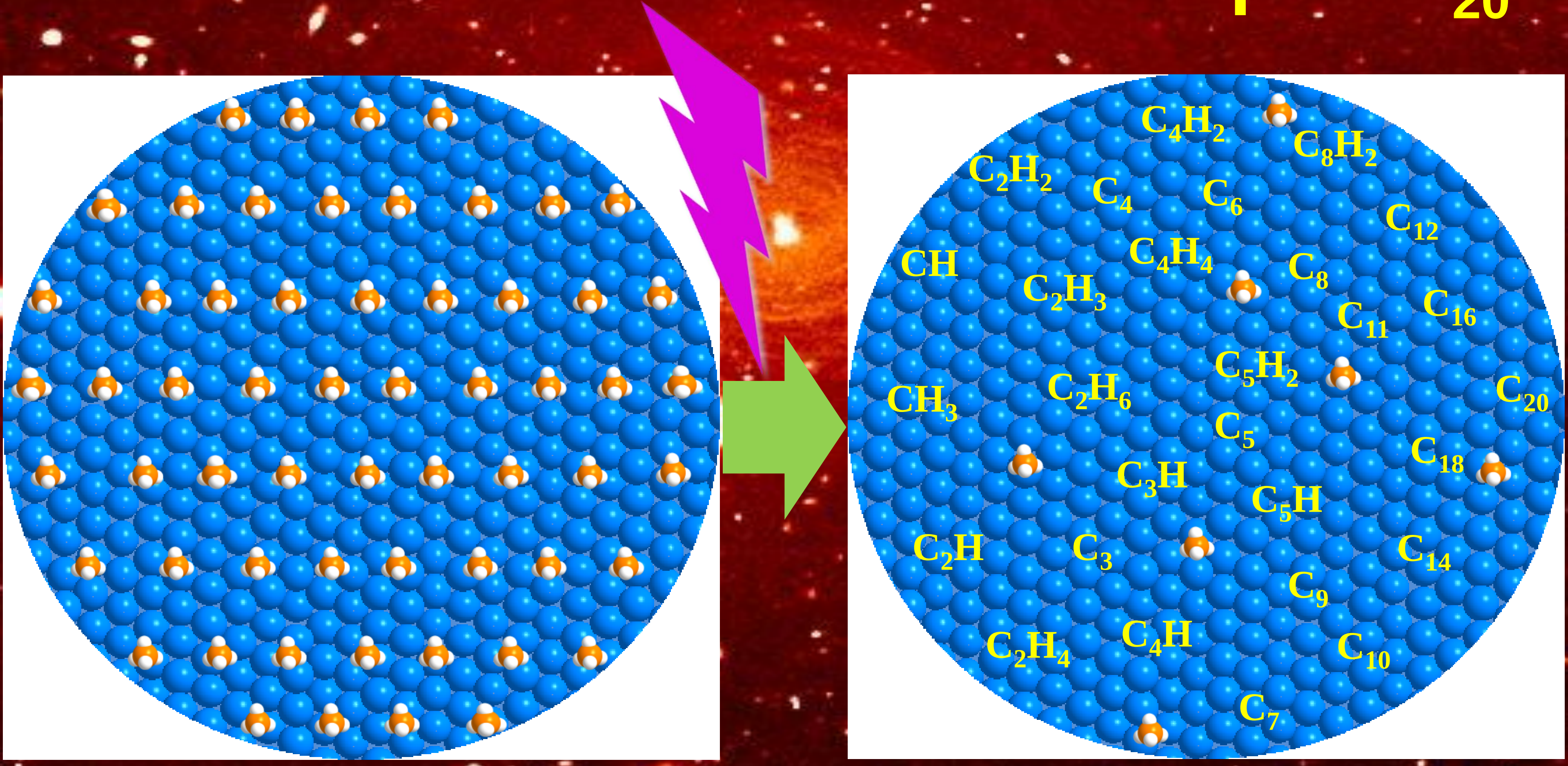


FUV Photolysis of $CH_4/H_2/Ne=1/6/1,000$ at 3 K



FUV Photolysis on CH_4

Production of Carbon Chains up to C_{20}

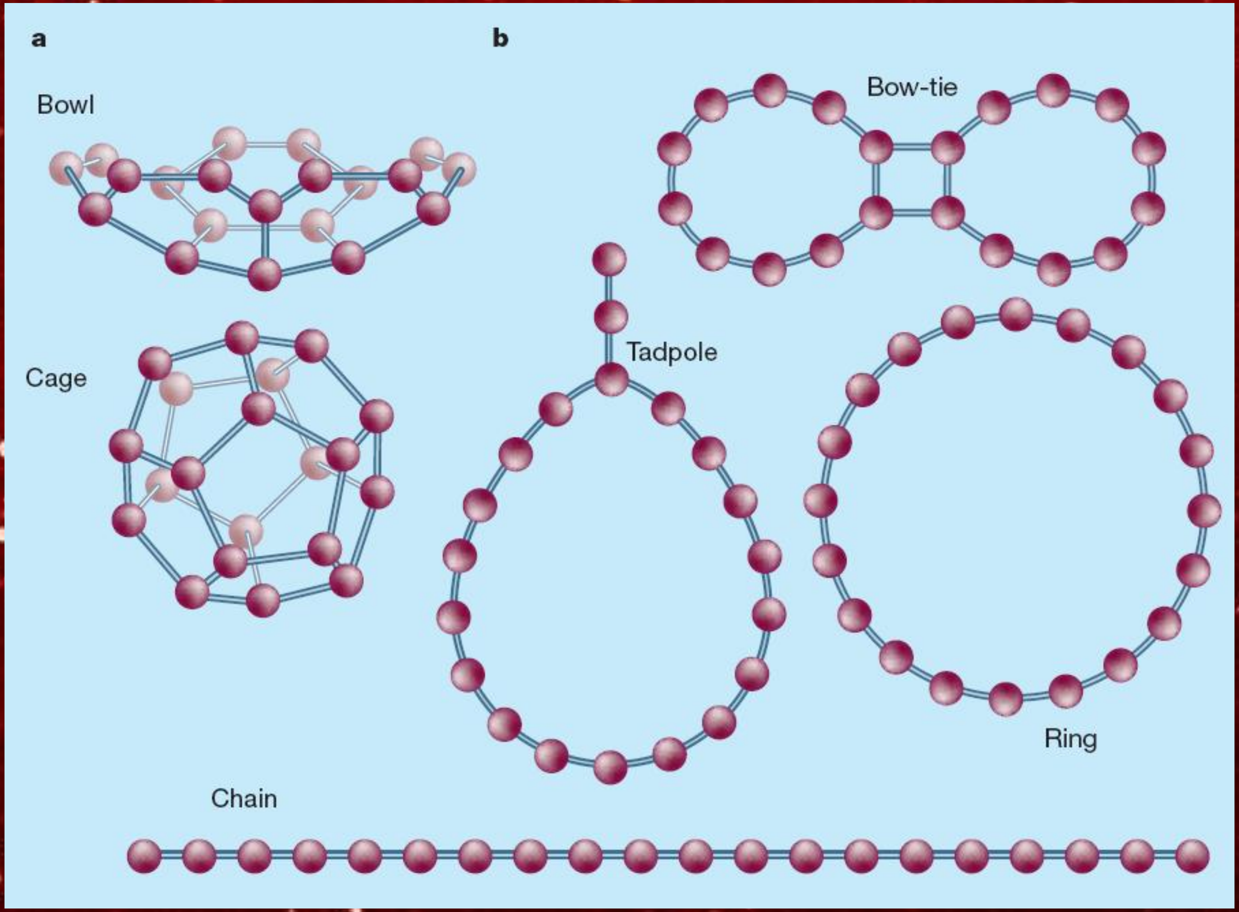


Production of Carbon Chains up to C_{20}

CH_4 : Ne	1:0	1:100	1:1000	1:2000	1:10,000	1:1000 + 6 H_2
C_n	C_2	C_6	C_7	C_9	C_{18} or C_{20}	C_{20}

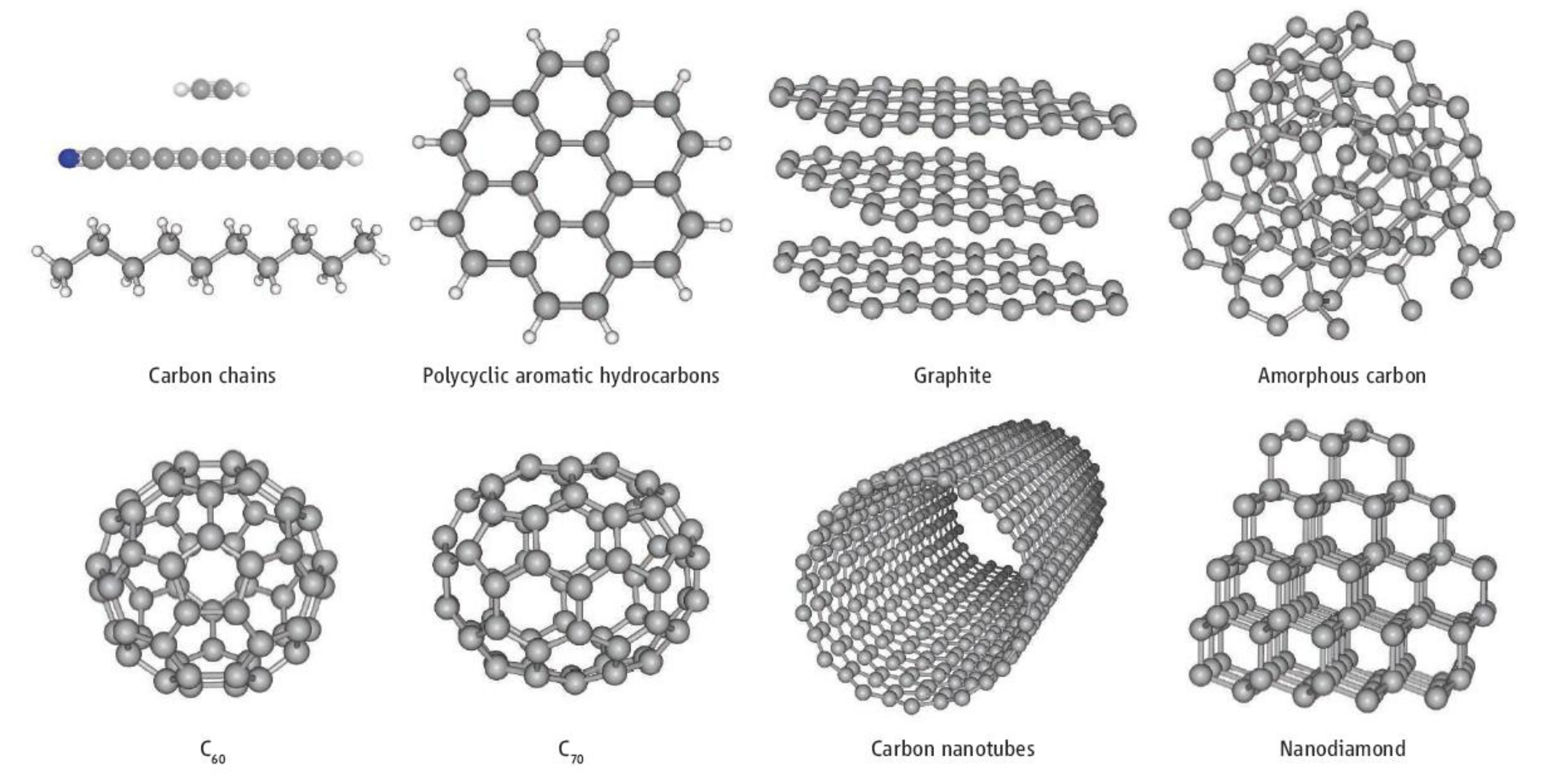
Lin et al. J. Phys. Chem. A, 2014, vol. 118, 3438

The smallest fullerene: isomers of C_{20}



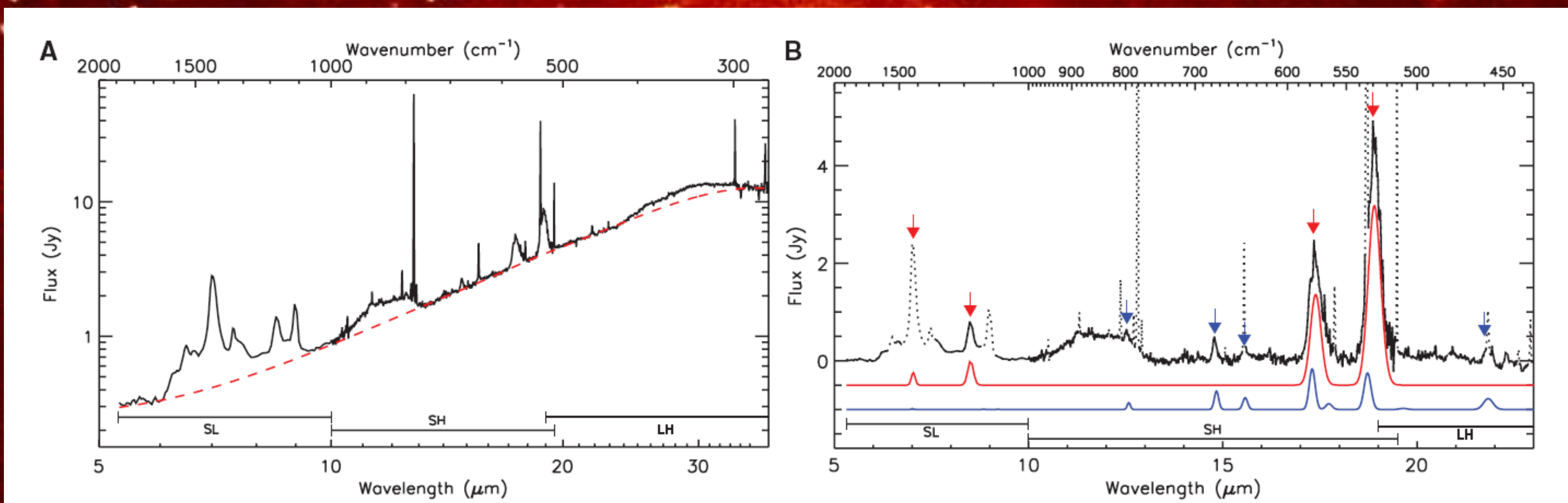
M. F. Jarrold, Nature, 2000, **407**, 26

Carbons in Space



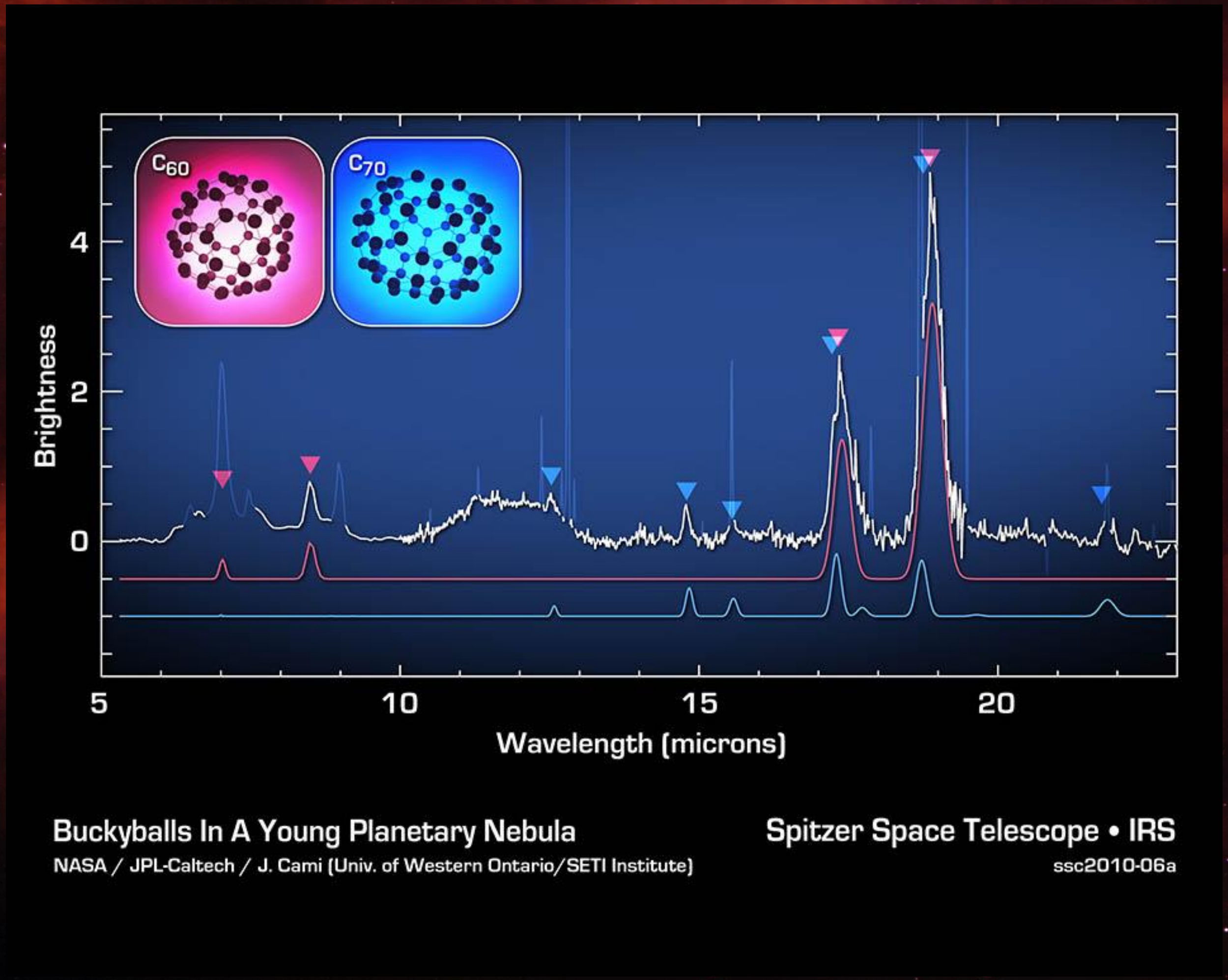
Ehrenfreund & Foing, Science, 2010, **329**, 1159

Detection of C_{60} and C_{70} in a Young Planetary Nebula



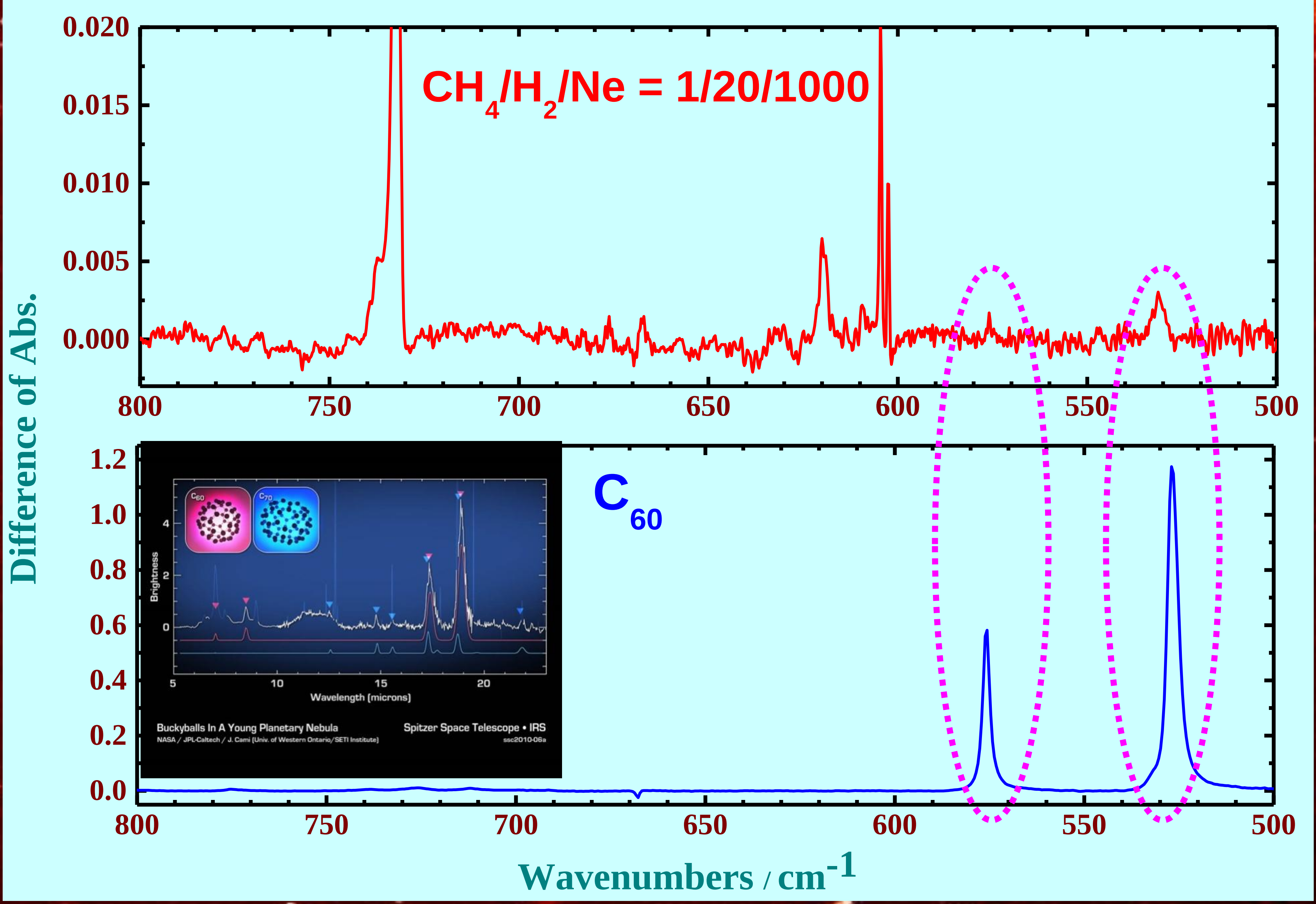
Cami et al., Science, 2010, **329**, 1180

Buckyballs in a Young Planetary Nebula

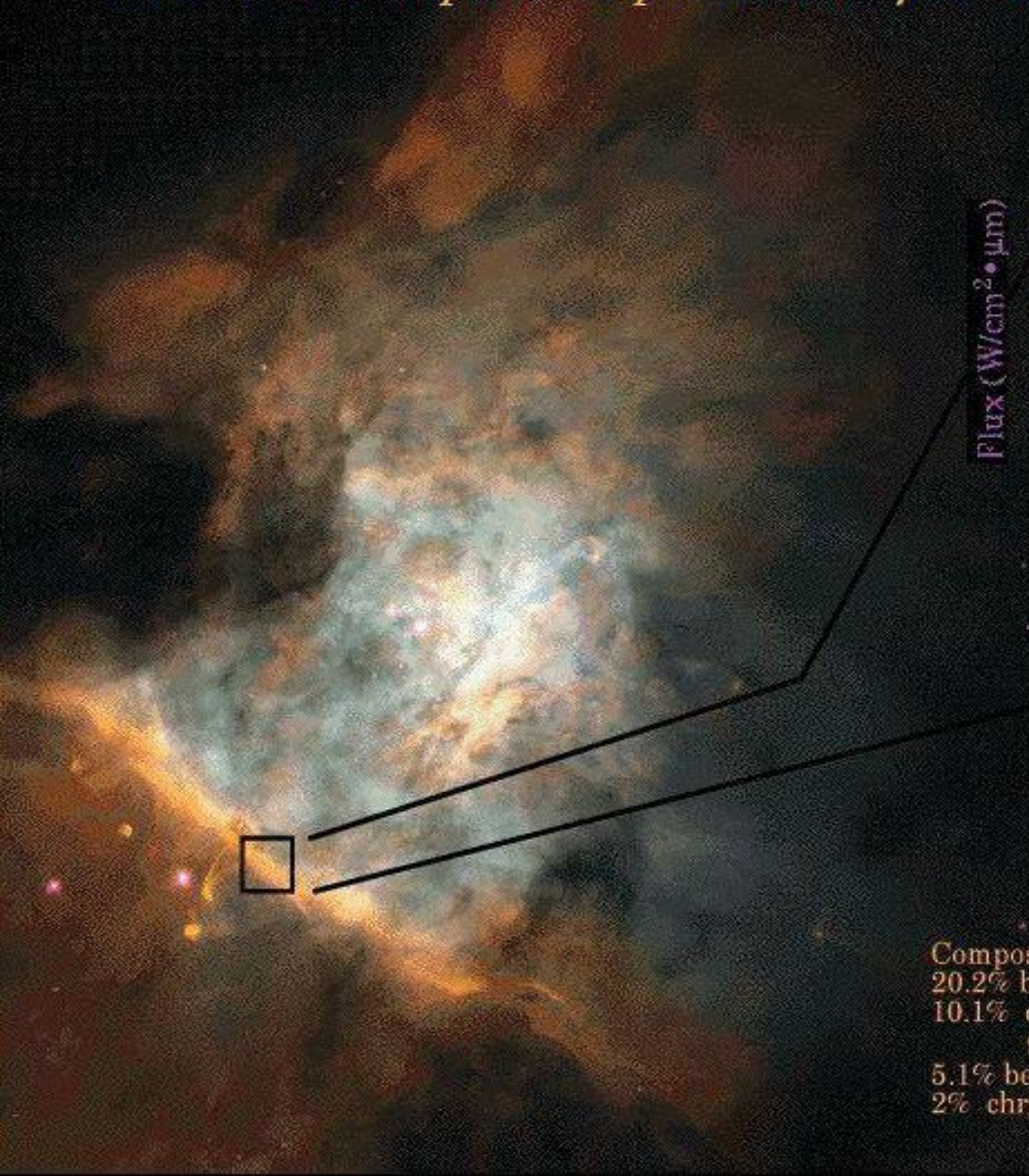


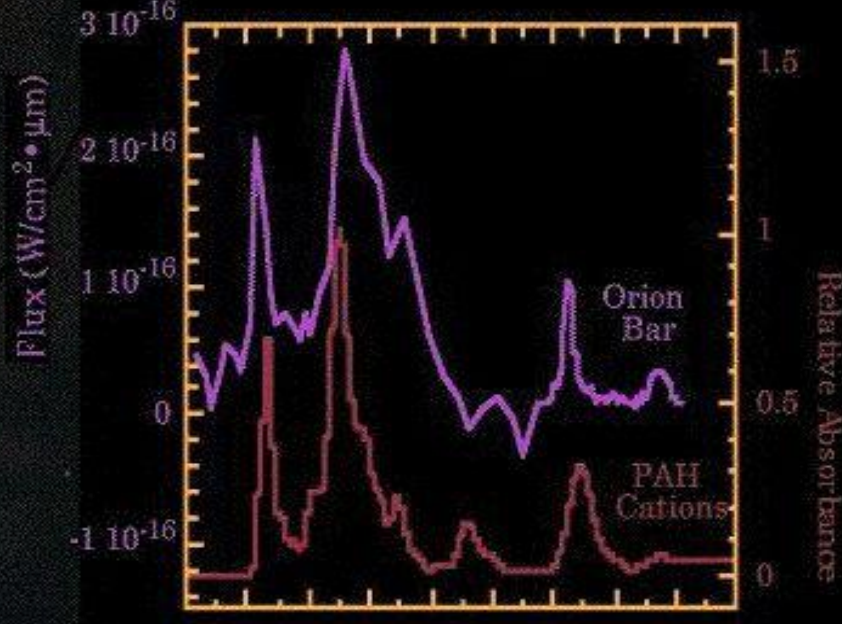
http://www.spitzer.caltech.edu/images/3213-ssc2010-06a-Jiggling-Soccer-Ball-Molecules-in-Space

FUV Photolysis of Solid Methane



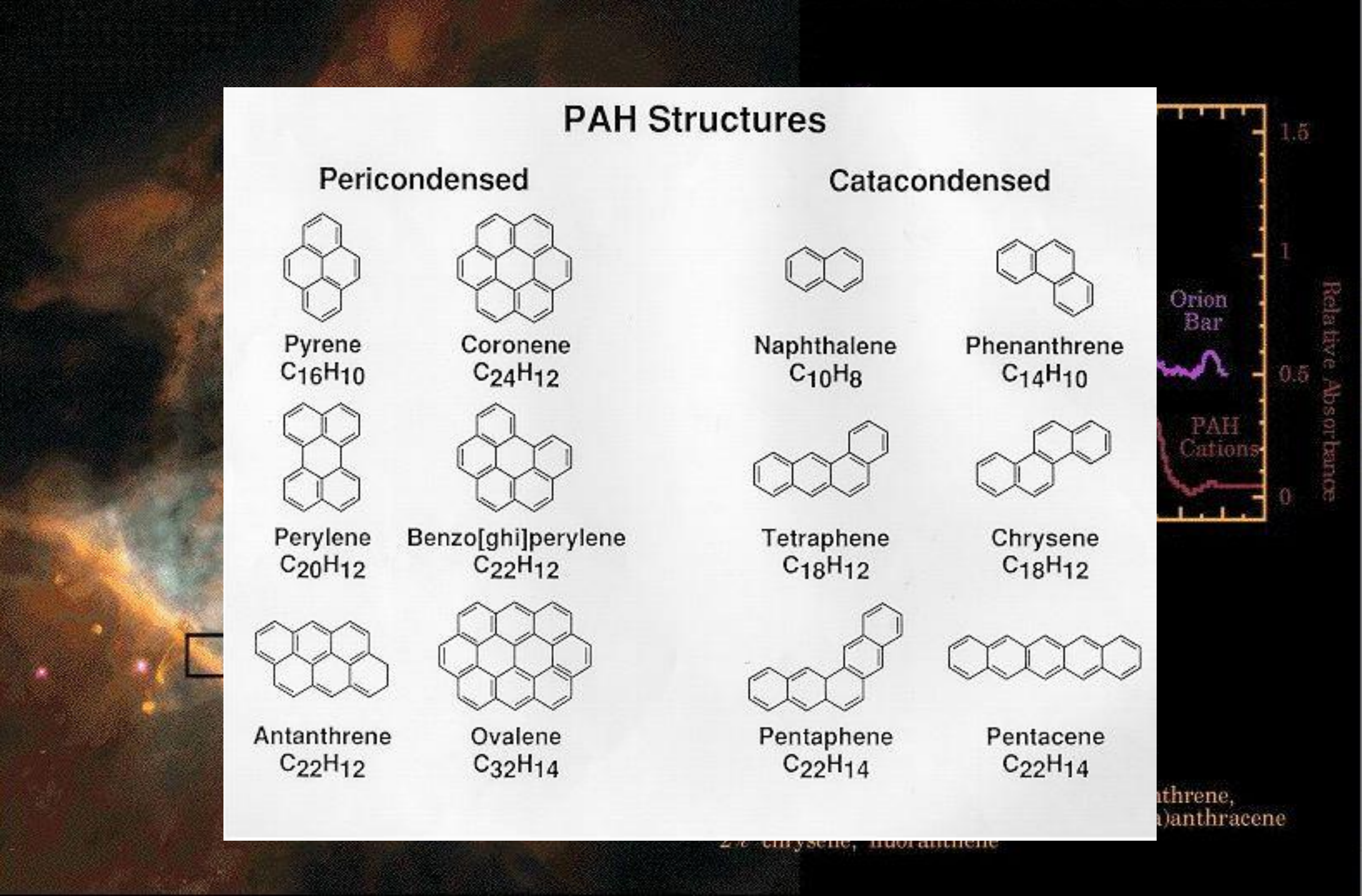
Comparison of the Infrared Emission Spectrum of the Orion Bar to the Absorption Spectrum of a Mixture of PAH Cations.



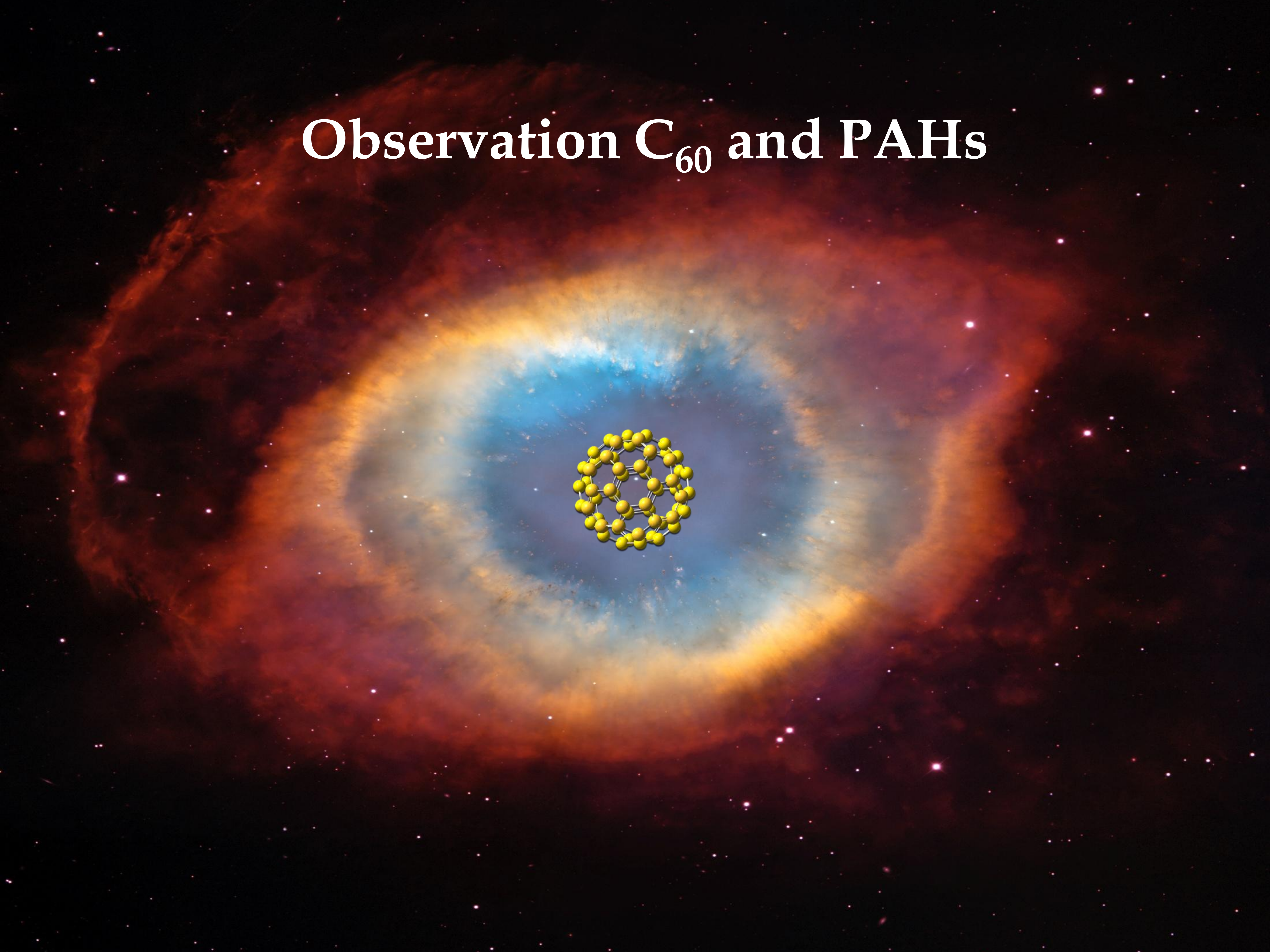


Composition:
20.2% benzo[k]fluoranthene, dicoronylene
10.1% coronene, benzo[b]fluoranthene,
9,10-dihydrobenzo(e)pyrene, phenanthrene,
5.1% benzo[ghi]perylene, tetracene, benz(a)anthracene
2% chrysene, fluoranthene

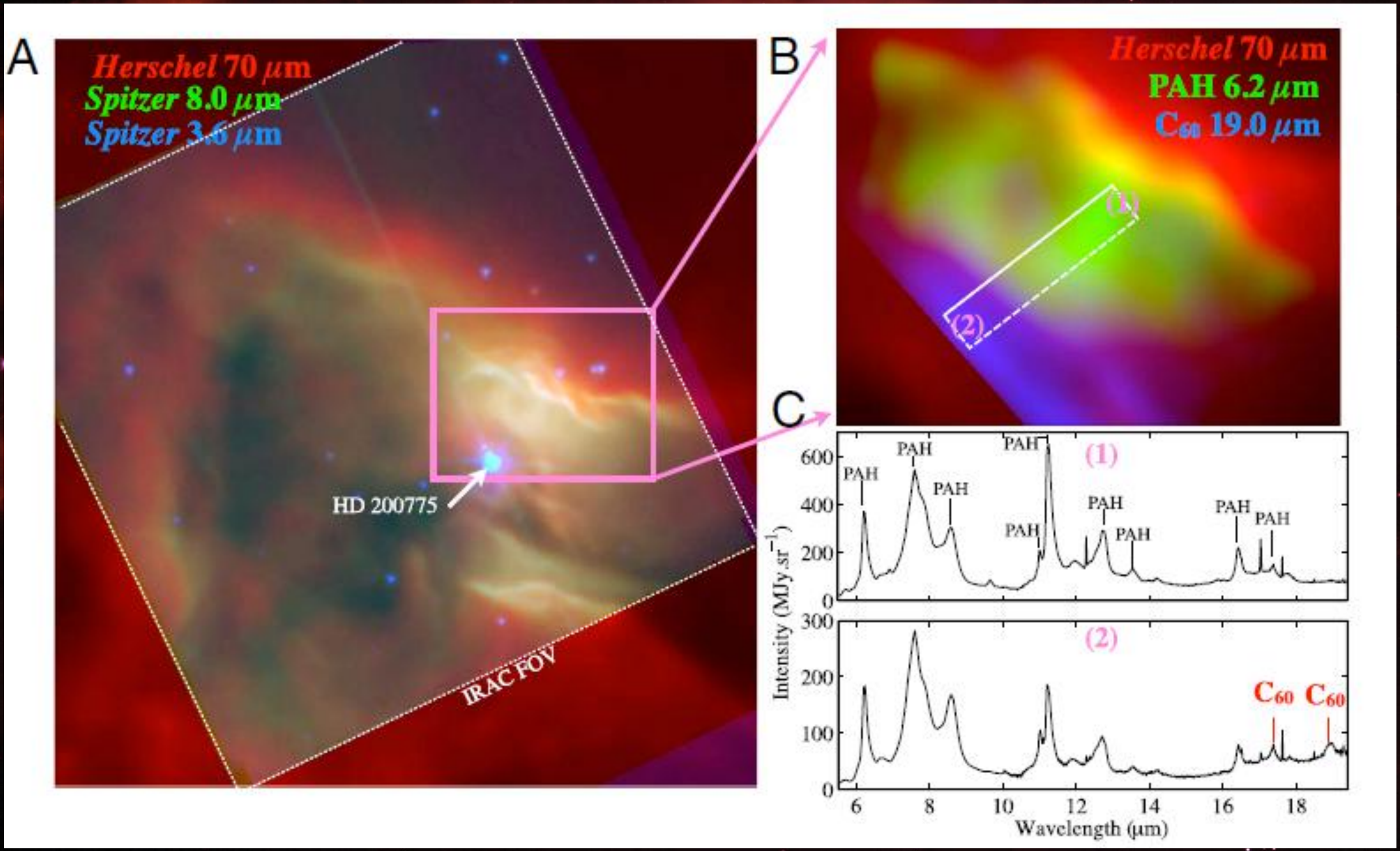
Comparison of the Infrared Emission Spectrum of the Orion Bar to the Absorption Spectrum of a Mixture of PAH Cations.



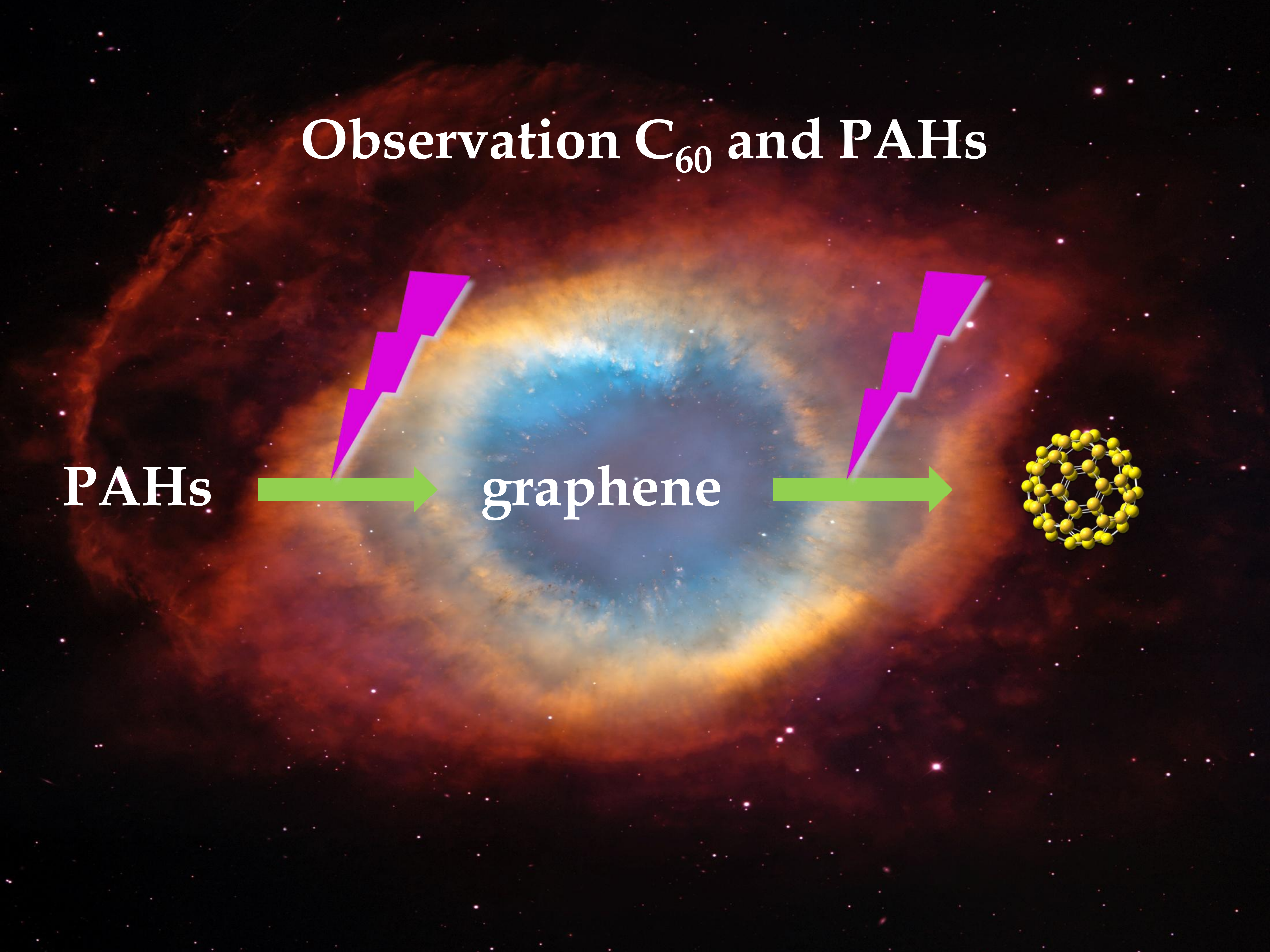
Observation C_{60} and PAHs



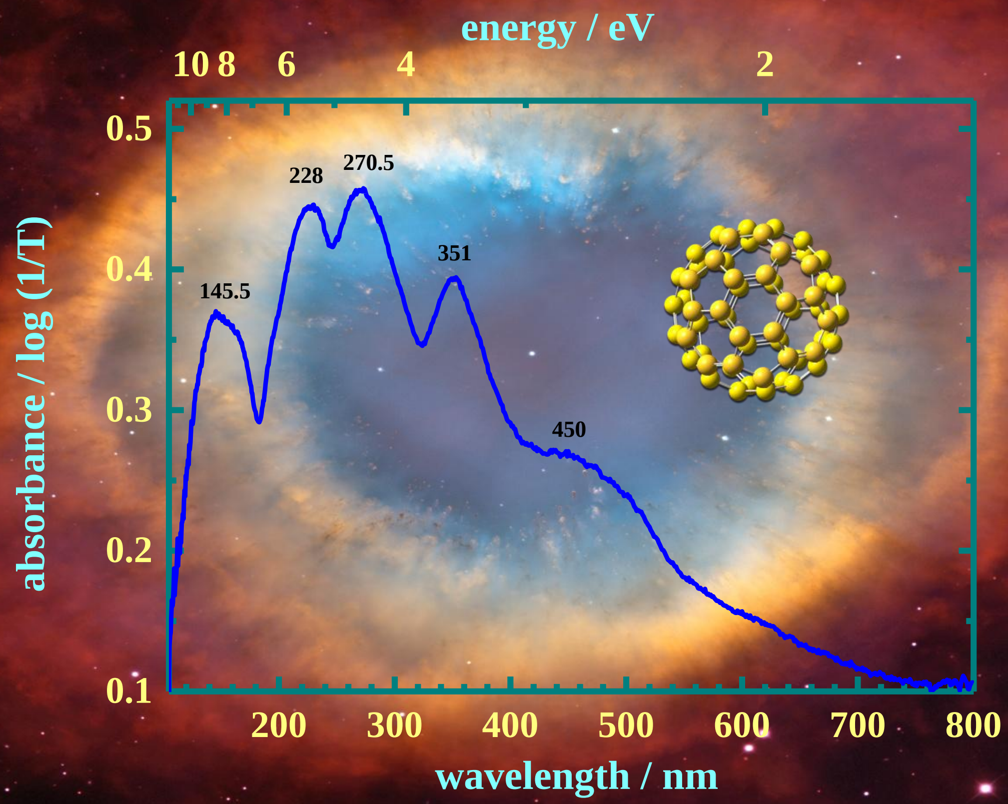
Observation in Space



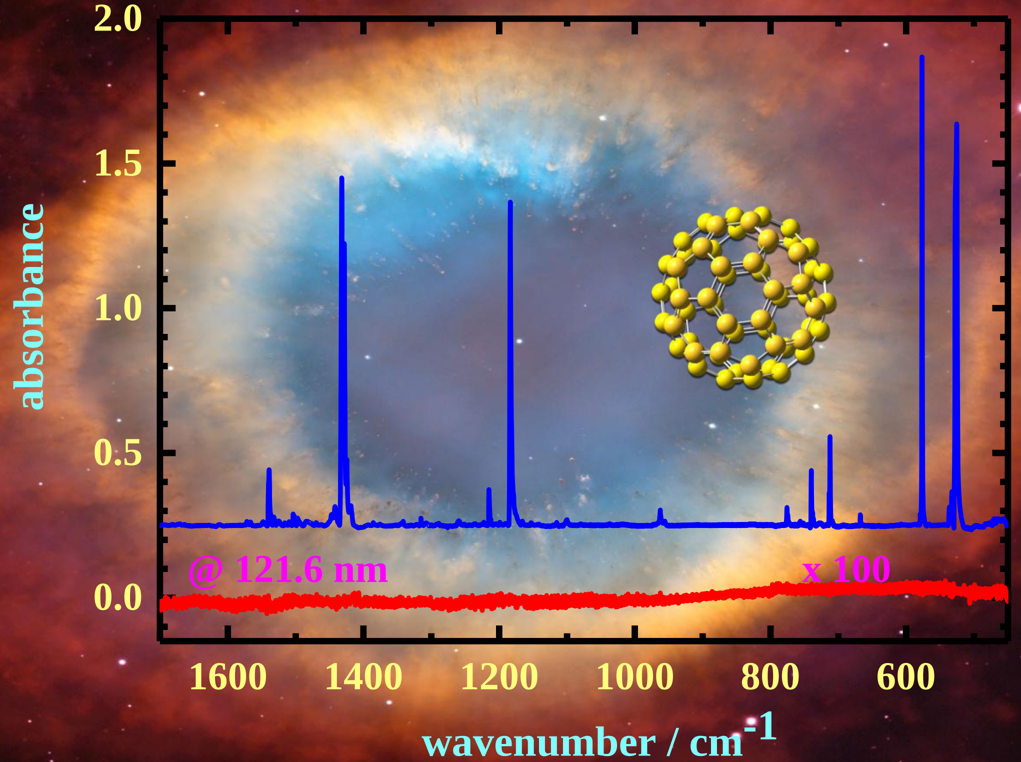
Berné and Tielens, PNAS, vol. 109, 401 (2012)


Observation C_{60} and PAHs
PAHs
graphene

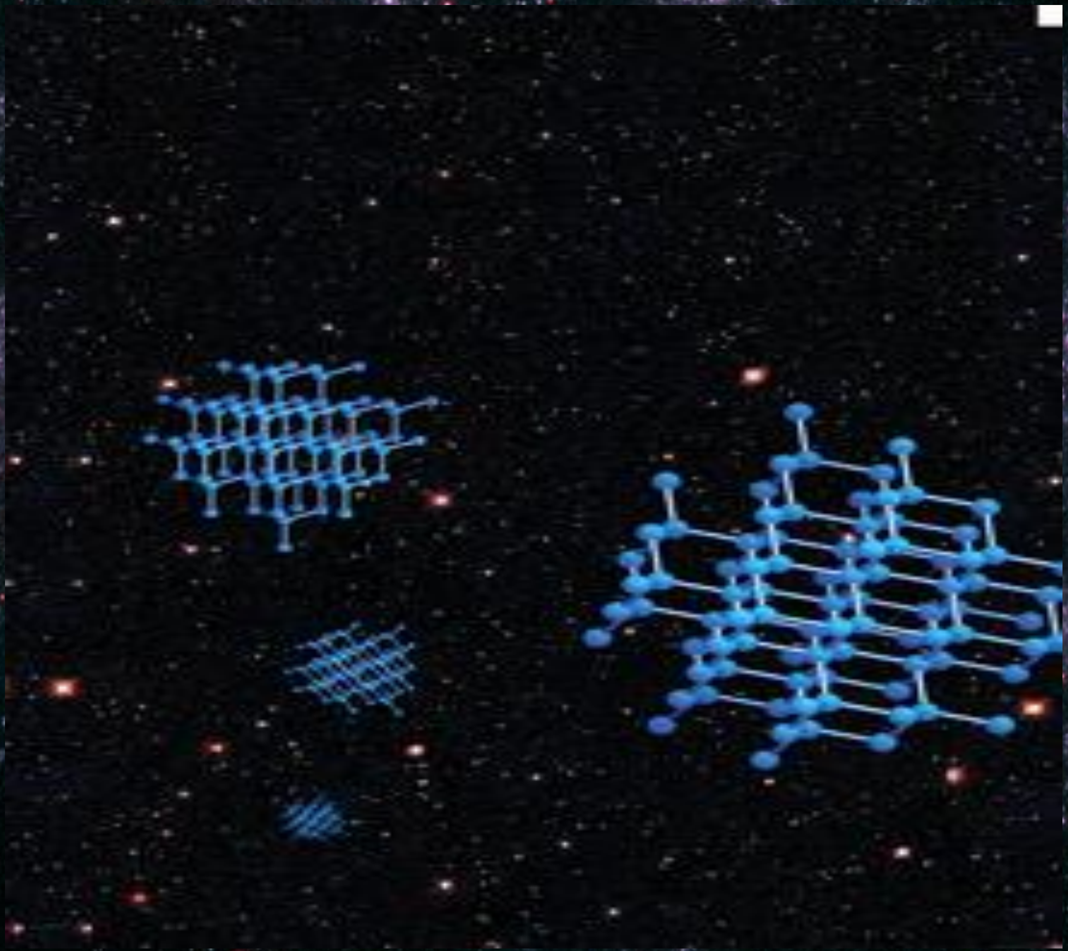
UV-visible spectrum of C_{60}



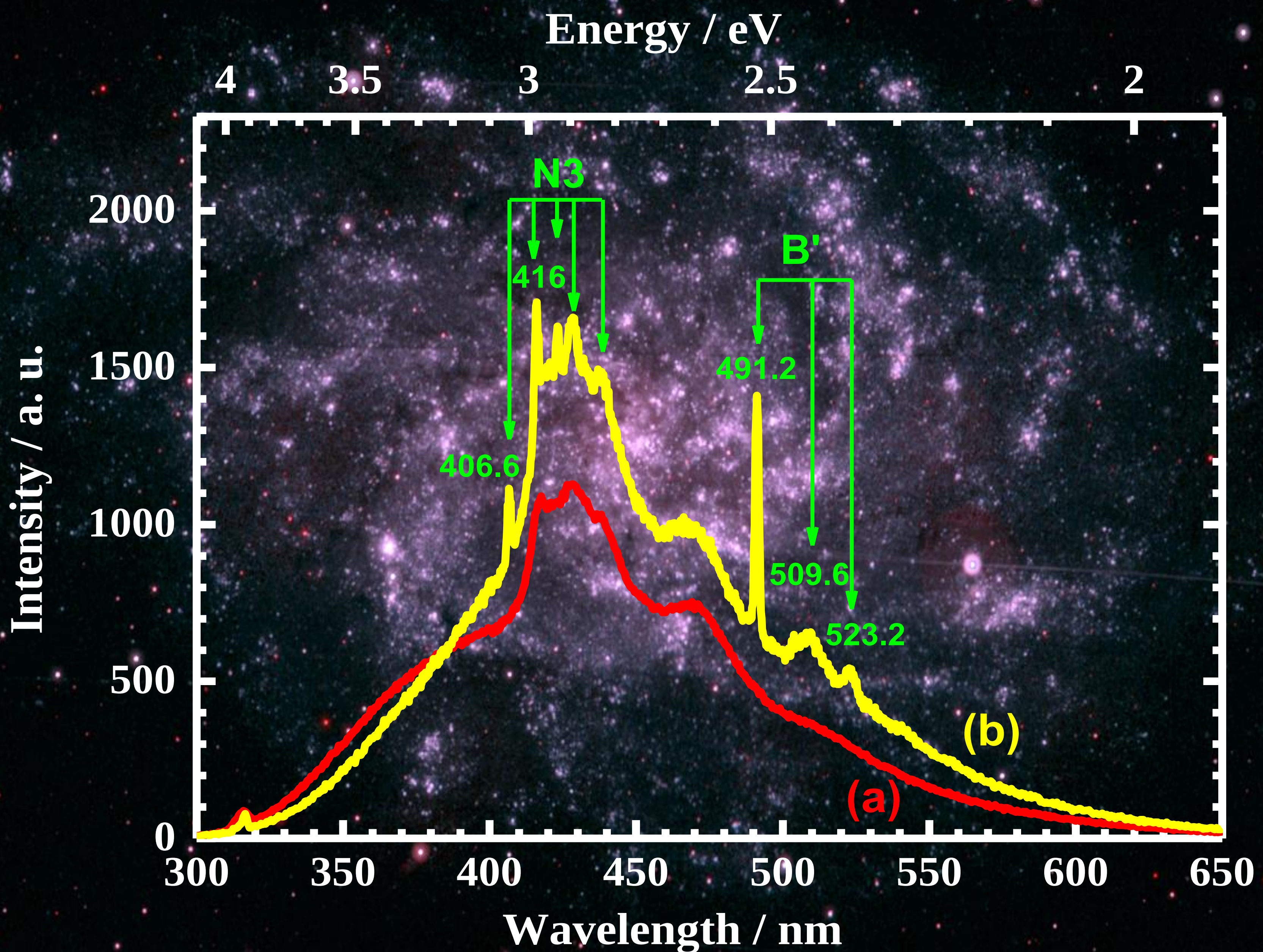
Far-UV Stability of C_{60}



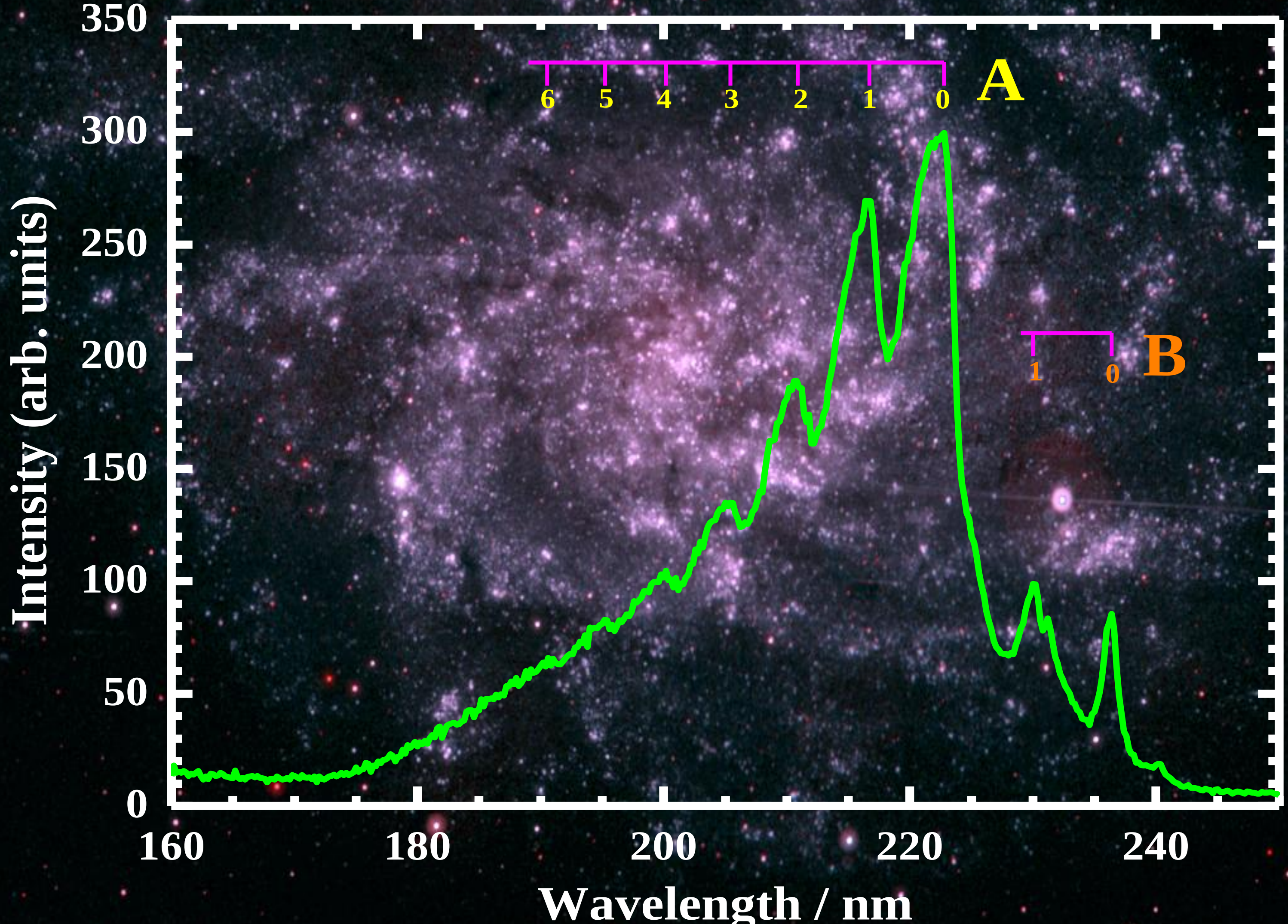
Sky with nano-diamonds



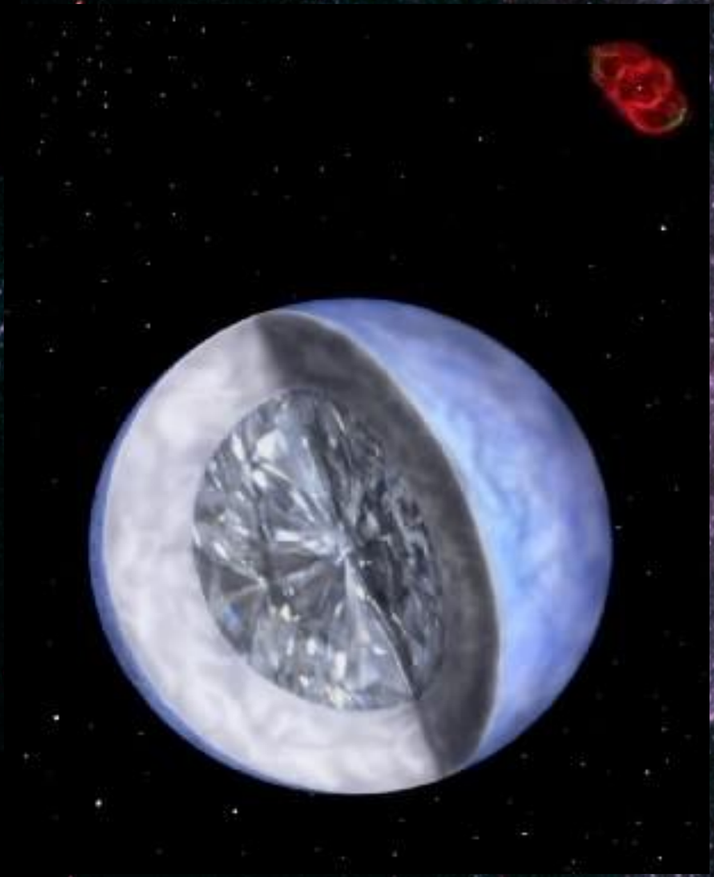
PL of Diamond



PLE of Diamond



Lucy: a diamond star



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Thank you for your attention!