BEAMS OF REFRACTORY ELEMENTS

Laser break-up molecules and ionization

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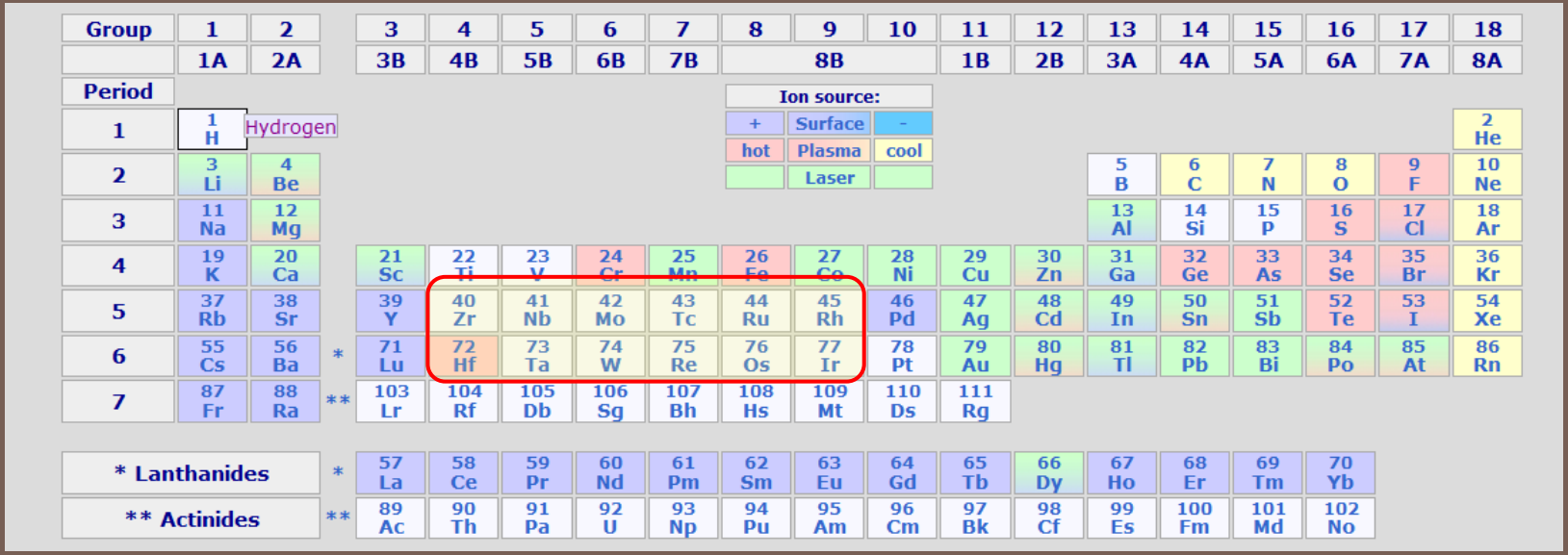
Refractory elements

- melting points ~ 2000 K



Group	1	2		3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
	1A	2A		3B	4B	5B	6B	7B	8B			1B	2B	3A	4A	5A	6A	7A	8A
Period									Ion source:										
1	1 H Hydrogen								+	Surface	-								2 He
2	3 Li	4 Be							hot	Plasma	cool			5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg								Laser				13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca		21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr		39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	*	71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	**	103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg							

* Lanthanides	*	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb
** Actinides	**	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No

Molecular beams

- ZrF_3^+ @ ORSAY

  by isocele2 Z. Phys. A 309, 185 (1982)

  SrF_2 powder + CF_4 gas
  ^{3}He beam

  → ZrF_3^+ was extracted by Plasma ion source

- HfF_3^+ @ isolde

  NIMB 62(1992)535

  Ta foil + CF_4 gas
  p beam

  → HfF_3^+ was extracted by Plasma ion source

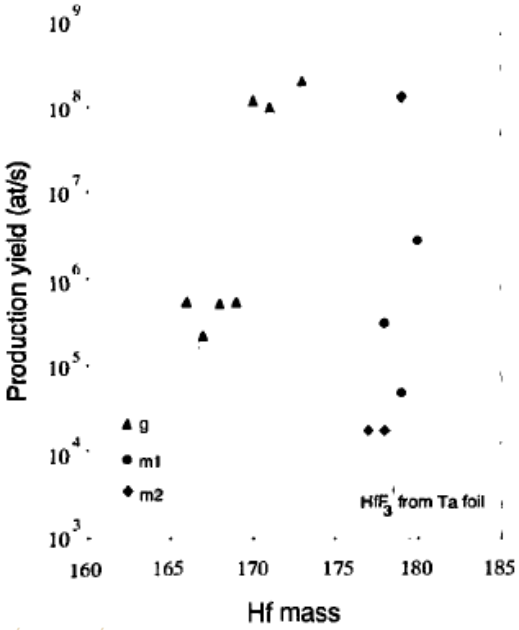
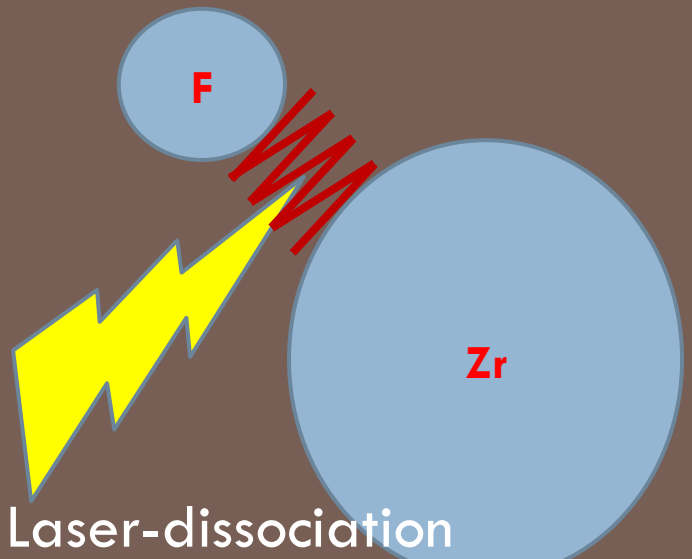
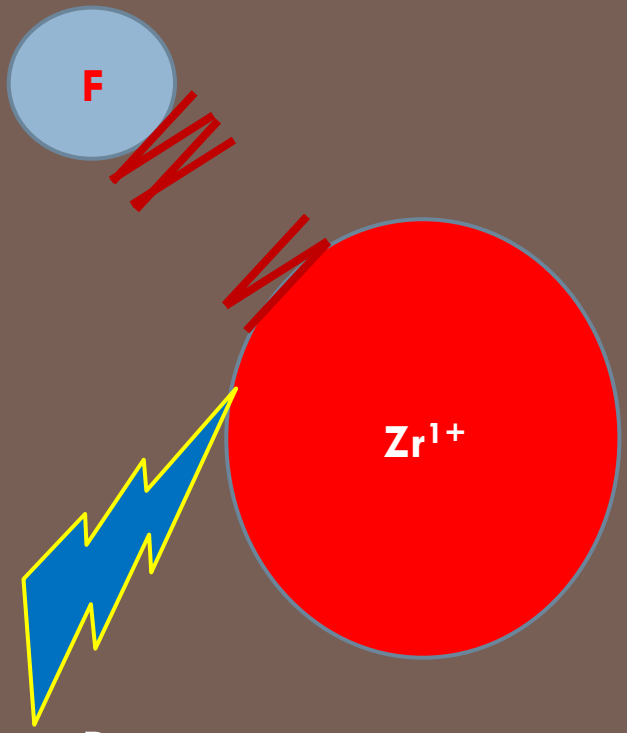


Fig. 5. Production yields of HfF_3^+ ions from a 122 g/cm^2 Ta foil target equipped with a hot plasma-discharge ion-source and a CF_4 leak (g = ground state, m1 = first isomer, m2 =

Laser-induced breakup + ionization



Laser-dissociation



Resonant-ionization

Feasibility study

1. Producing ZrF^+

S. Soorkia et al.,
J. Phys. Chem. A, **2011**, *115* (34), pp 9620–9632,

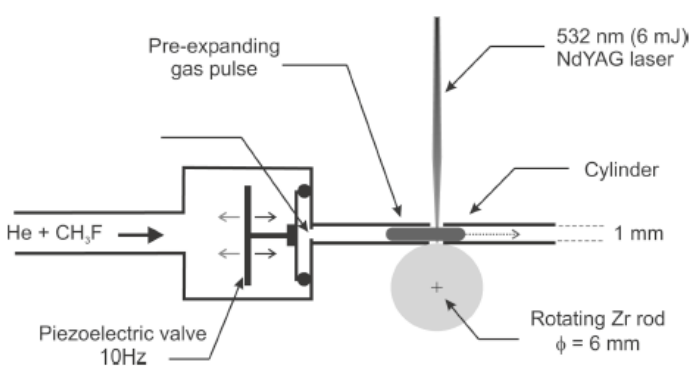


Figure 1. Outline of the laser ablation - molecular beam experimental setup used to obtain cold ZrF radicals.

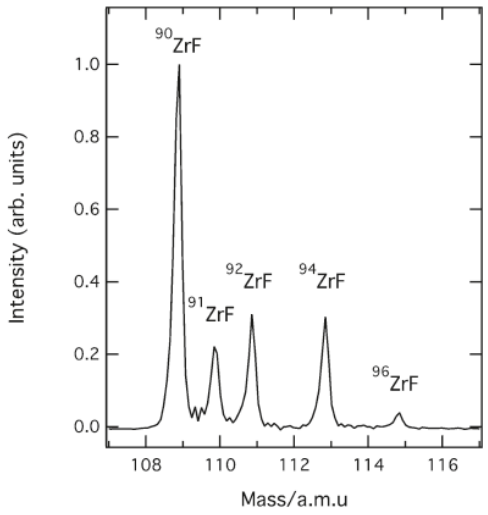


Figure 2. Mass spectrum of the zirconium fluoride radical. The relative intensities of the five naturally occurring isotopologues, i.e., ^{90}ZrF, ^{91}ZrF, ^{92}ZrF, ^{94}ZrF, and ^{96}ZrF, are in the ratio of the isotopic abundances of Zr.

2. Laser radiation of 2 steps:

breaking up molecules,

ionizing Zr

TOF spectrum
w/He+ CH_3F gas (2%)

Basic parameters

- Ionization of Zr : $Zr \rightarrow Zr^{+} + 1e^{-}$

  Ionization energy Zr(I)

  = 6.63390 eV ~2 x 355nm (3.5 eV)

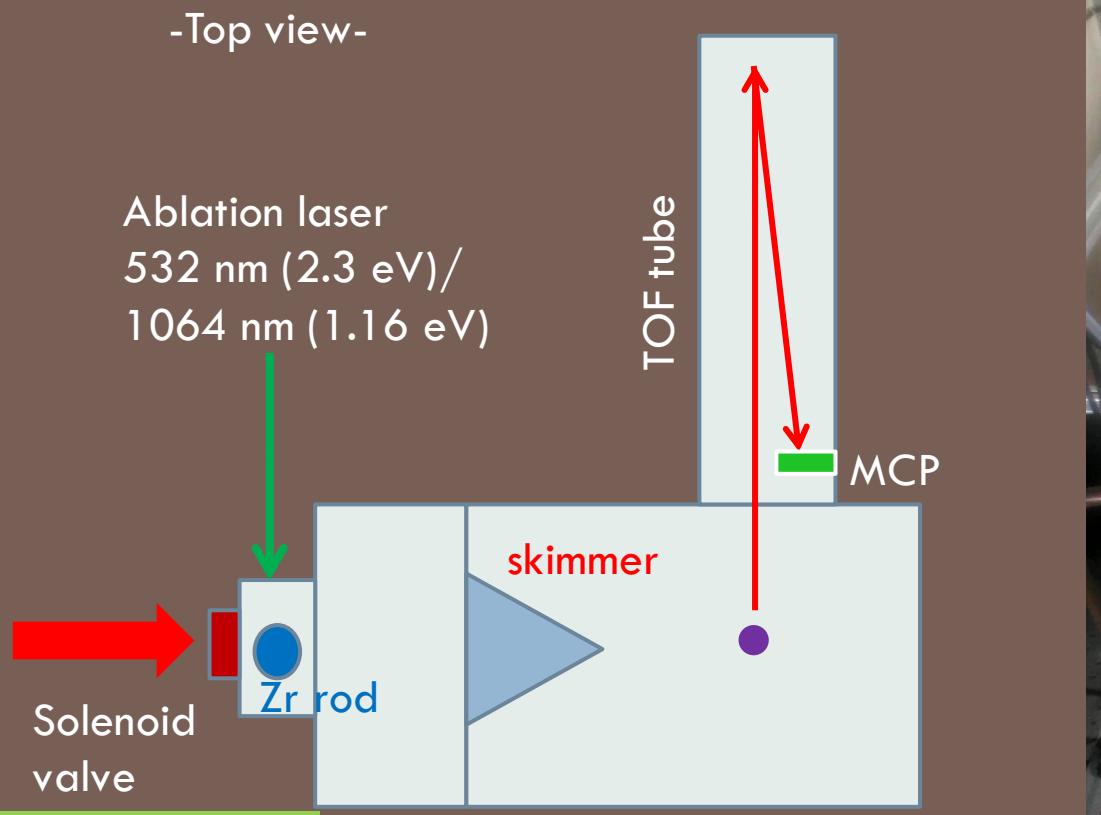
  cross section ~ 4×10^{-17} cm^2 (Fe)

- Bonding energy of ZrF : $ZrF \rightarrow Zr + F$

  = 6.3 eV (!?) ~2 x 355nm (3.5 eV)

  cross section ~ $5 \times 10^{-19} cm^2 (SF_6)$!?

Test with 355nm@ LARIS lab.



Pulsed gas flow
Ar
+
CF_4 gas

UV laser: 355 nm (3.5 eV)
of Nd/Yag

Valve and lasers are operated with 10 Hz.

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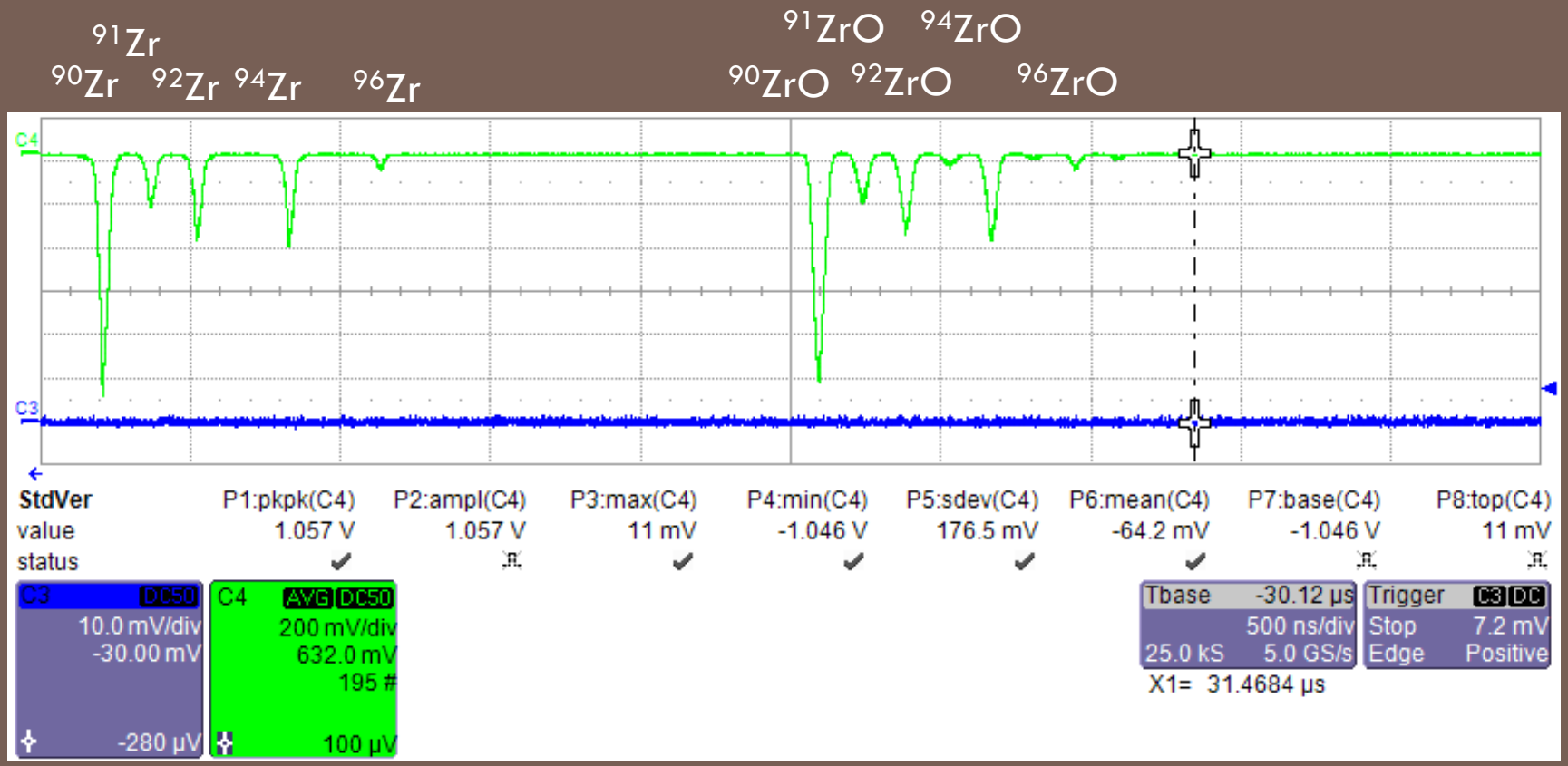
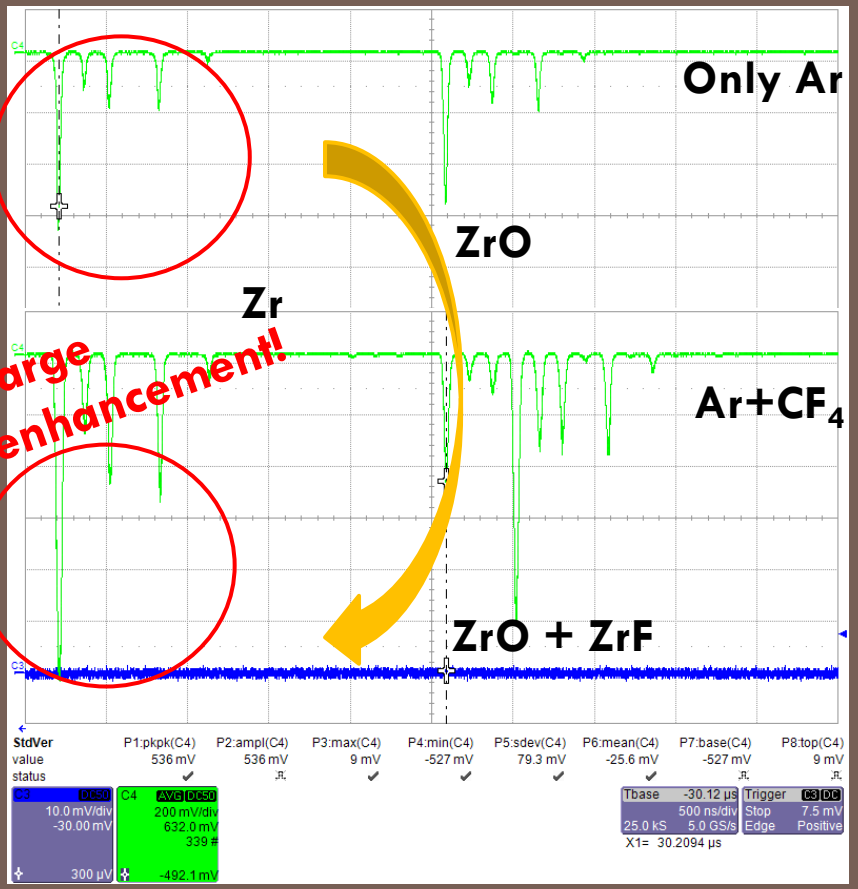
TOF spectrum w/only Ar



Ablation laser: 355 nm 8 mJ

UV(non-resonant ionization): 355 nm 2.3 mJ

TOF spectrum w/Ar + CF_4(6%)



- ZrF^+ peaks were seen but disappeared in several minuets.
- Zr^+ peaks were enhanced and seemed stable.
- We couldn't distinguish the breakup and ionization effect.

Ablation laser: 532 nm 8 mJ
UV(non-resonant ionization): 355 nm 2.3 mJ

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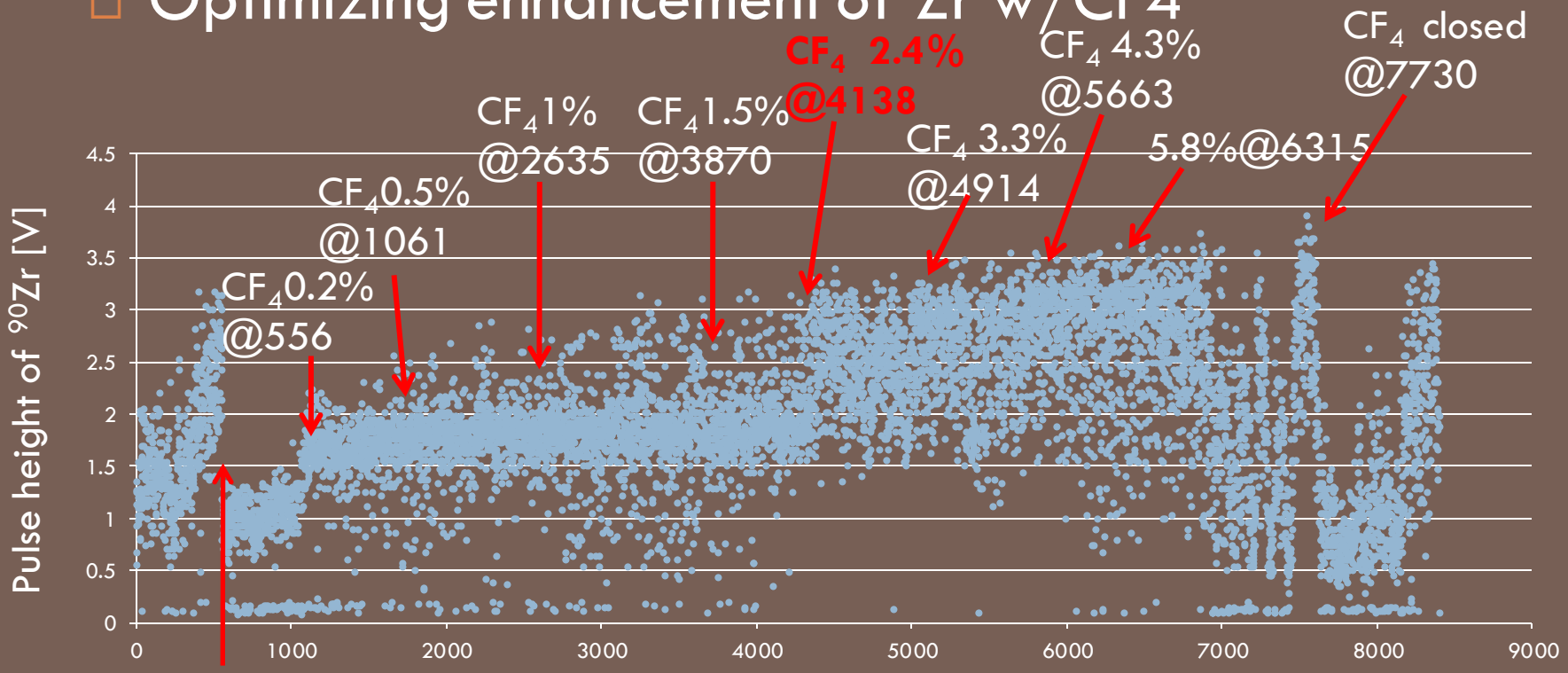
UV as dissociator and excitator

- ~~Confirmation of ZrF~~
- Optimizing enhancement of Zr w/CF4



HV of MCP
-3000 → -2800V

CF_4 > 6% may be too much.
A few % is enough to enhance the ^{90}Zr.

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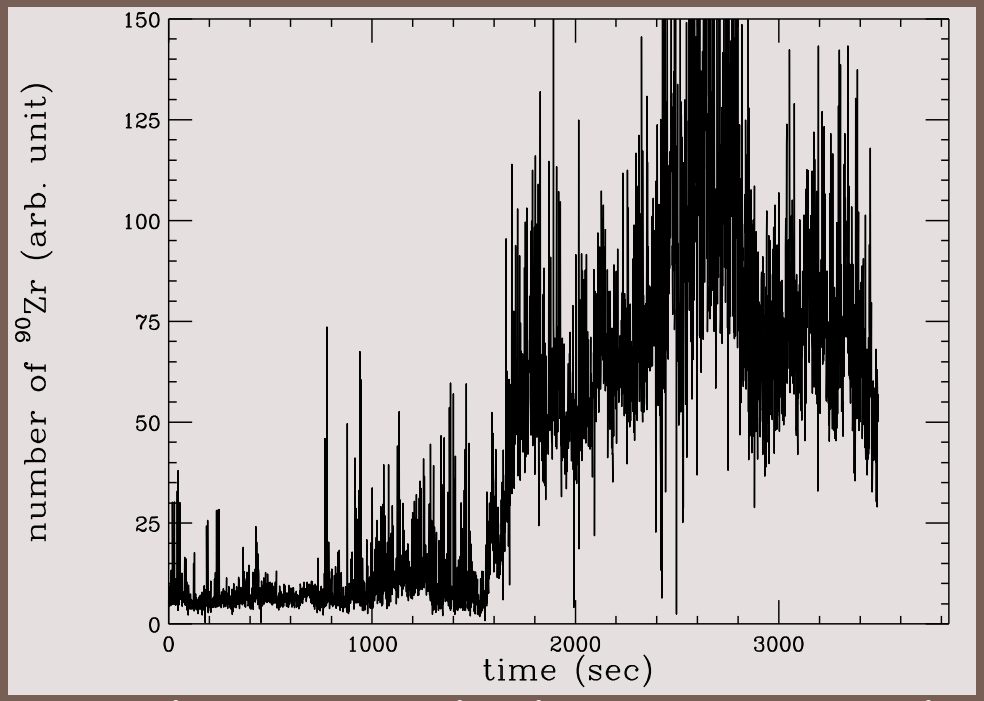
Non resonant ionization of Zr



Only Ar

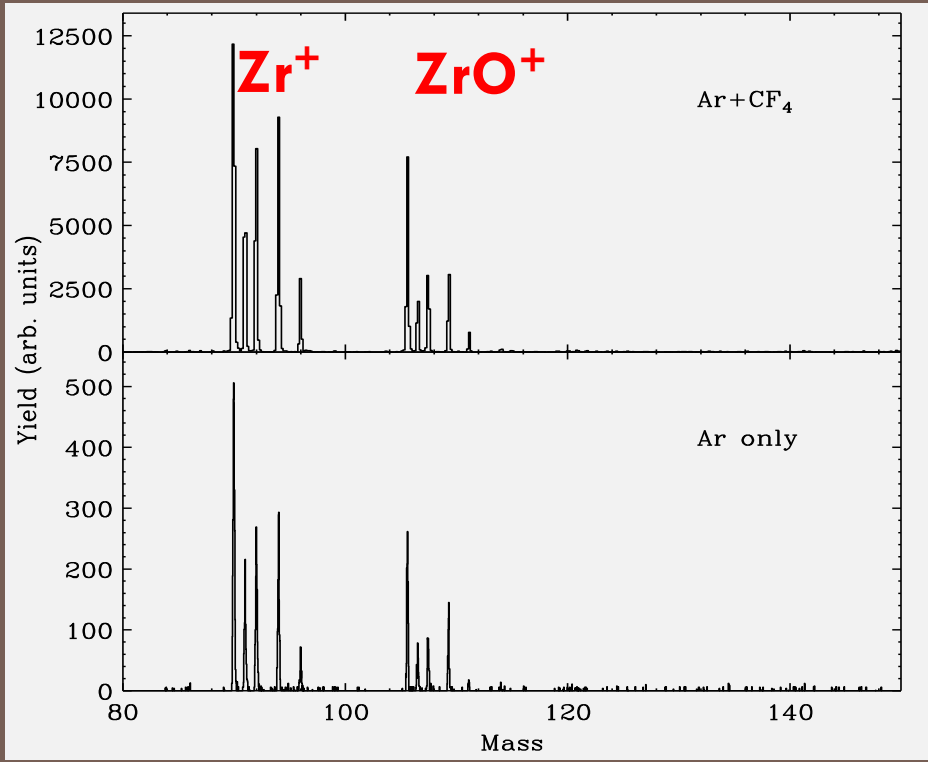
Ar+ CF_4(2.4%)

Ablation laser:
1064 nm 3mJ

Dissociation + Ionization:
355 nm 2mJ

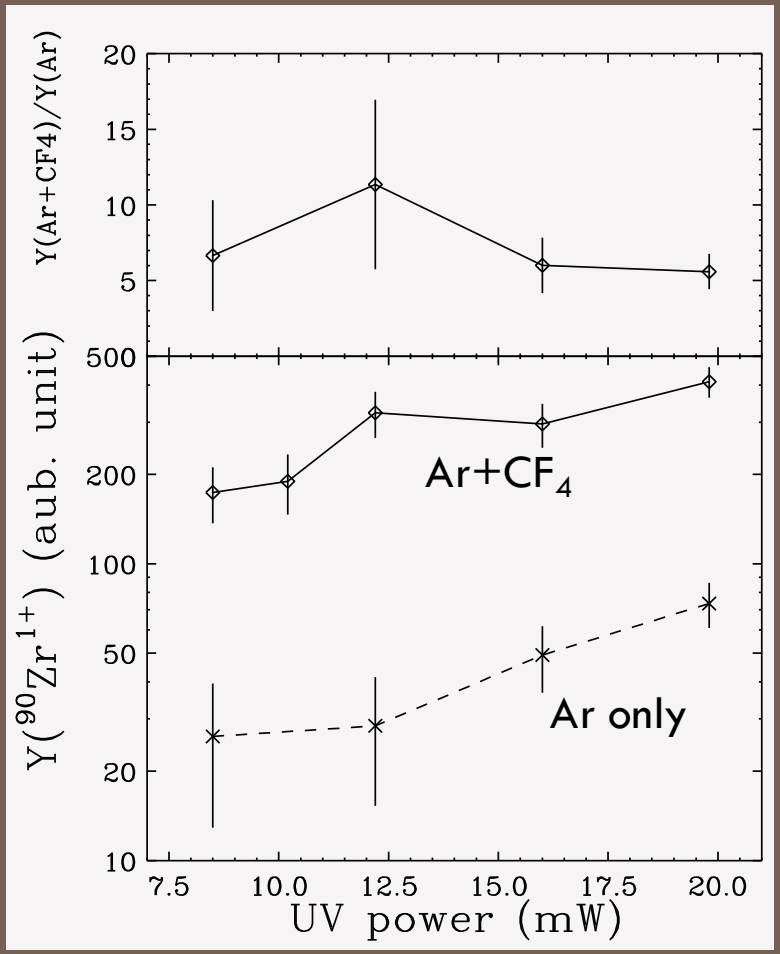
Yield of $^{90}Zr^+$ was enhanced **by 1 order of magnitude**!

Mass spectrum



- No ZrF^+ was seen w/Ar +CF_4
- Ratio of Zr/ZrO doesn't change so much

UV (355nm) power dependence



1. Only Ar gas flow

   Zr → Zr^+ : 6.6 eV

2. Ar + CF_4 (2.4%) gas flow

   Zr → Zr^+

   and

   ZrF → Zr + F

   Zr → Zr^+

No difference between Ar and Ar+CF_4

Breaking-up molecules seems saturated.

Possible mechanism

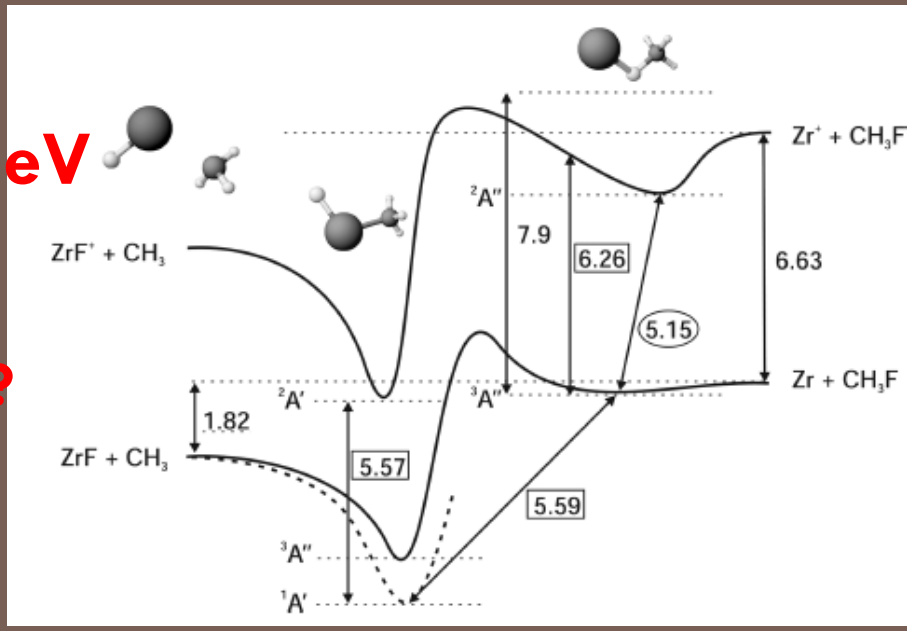
- $ZrF = Zr + F$ **– 6.3 eV**
- $ZrF + CH_3 = Zr + CH_3F$ **-1.82 eV**

  S. Soorkia et al.,
  J. Phys. Chem. A, **2010**, *114* (18), pp *5655*

- **$ZrF+CF_3 = Zr+CF_4$ -0.83 eV**

0.83 eV = 1520 nm

1064 nm Nd:Yag is enough ?

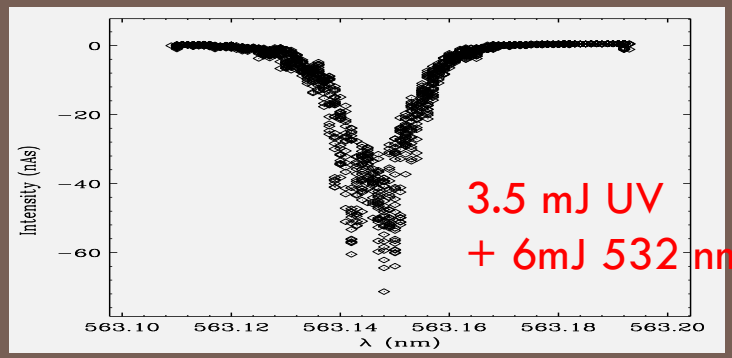


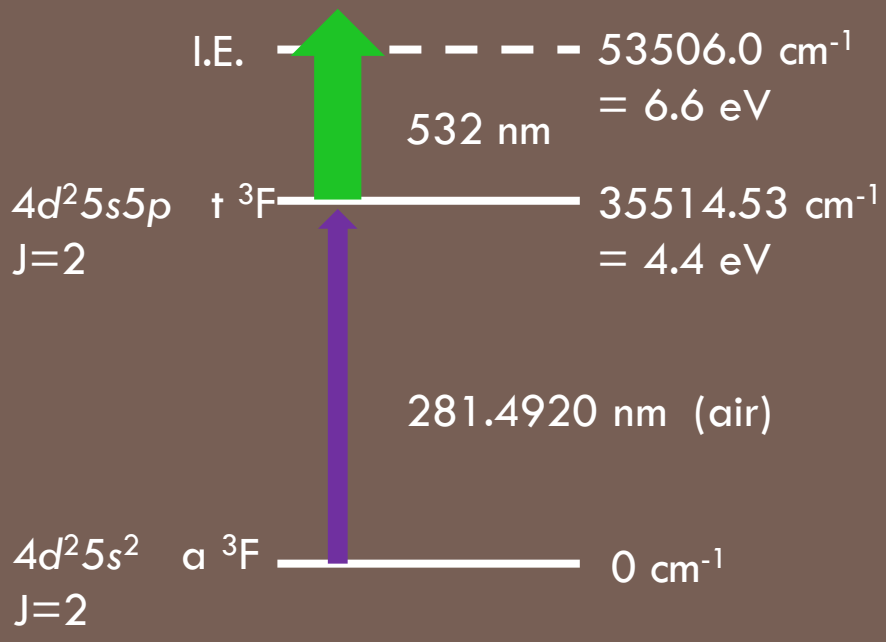
On-going works…

- ✓ Resonant ionization:

  1st step 281.4920 nm + 532 nm

  = 543218 cm^{-1}





- Molecular breakup:

  3rd hw (355nm), 2nd hw (532nm), 1064 nm of Nd:Yag

  power dependence/time difference will be checked.

- Removing of ZrO from Zr rod

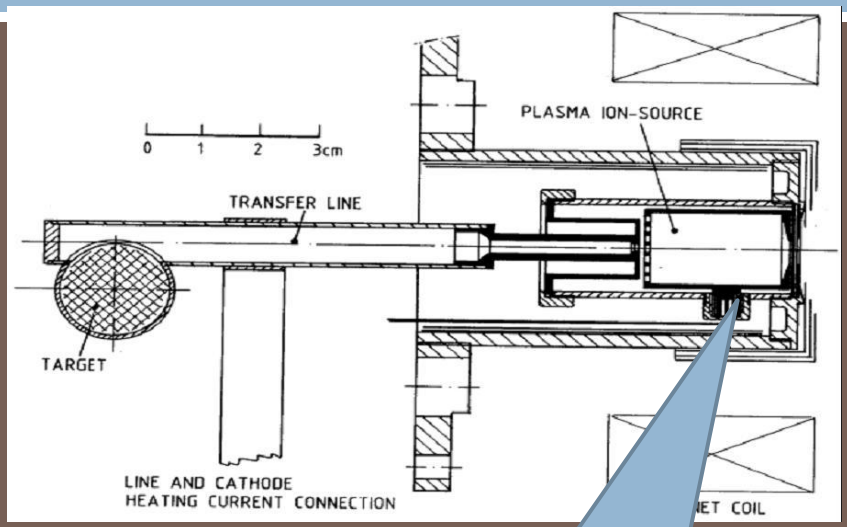
  Etching the Zr rod with acid prior to ablation

How to implement at ISOLDE?

- VADIS + RILIS
  - **Breaking-up molecule:**

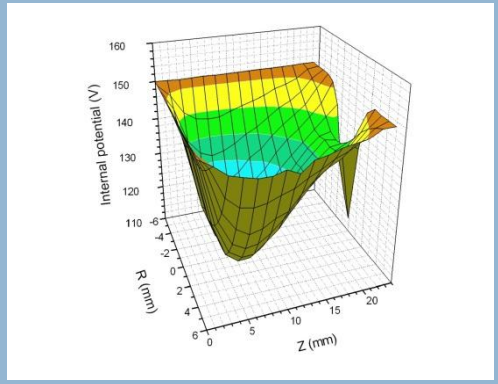
    VADIS cavity w/several eV ?

    or breakup laser ?



  - **Resonant ionization: RILIS**

    The resonant ionization scheme hasn't been established yet.



Summary

- Feasibility study of breaking-up refractory element fluoride is on-going with ablation + TOF spectrometer of Zr.
- In the case of non-resonant ioniztion, with small injection of CF_4 in Ar gas, large enhancement of Zr^+ peaks were observed.
- Both laser for resonant ionization of Zr and break-up molecules are ready.
- Further investigation of the phenomenon provides promising experimental opportunity.

Outlooks

- Implementation with VADIS + RILIS

- Establishment of resonant-ionization:
  hopefully 3 steps-resonant ionization

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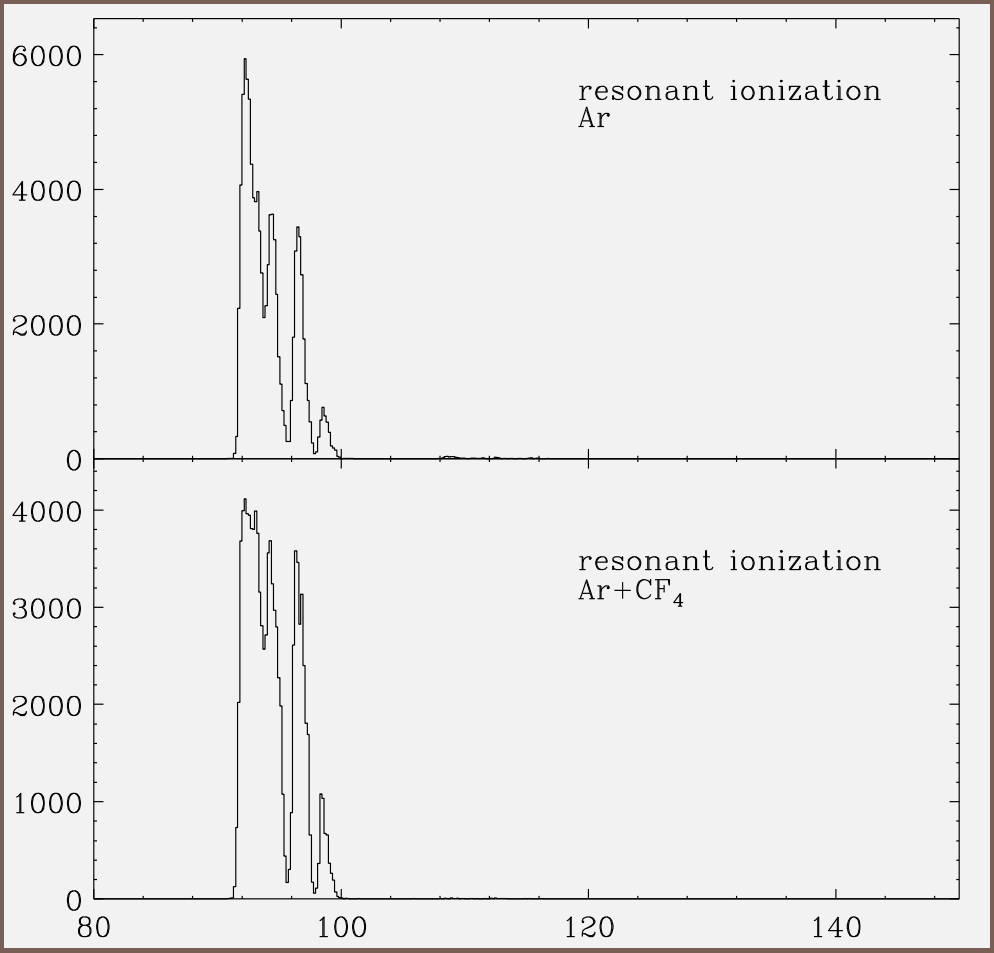
Mass spectrum of resonant ionization



In both cases, only Zr peaks were observed.