# ATTOSECOND PUMP – ATTOSECOND PROBE SPECTROSCOPY OF AUGER DECAY

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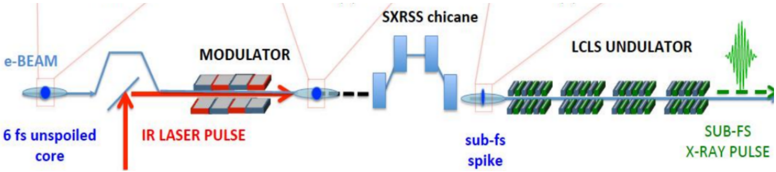
Frontiers of Physical Sciences with X-ray FELs

Imperial College London

November 13$^{th}$ 2019

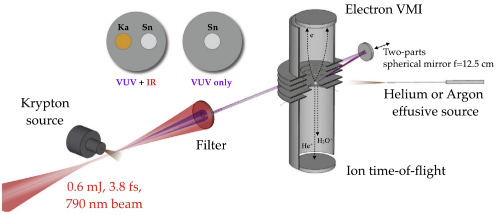
# Source development towards attosecond pump-probe

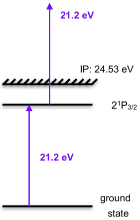
- X-LEAP: 0.5 fs duration pulses



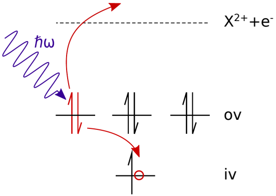

- HHG-based sources: Towards XUV pump-probe experiments

(Tisch and co., 2017)



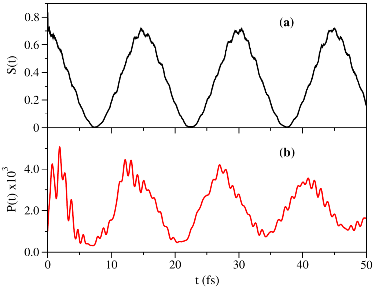
# *single-photon* laser-enabled Auger decay (*sp*LEAD)

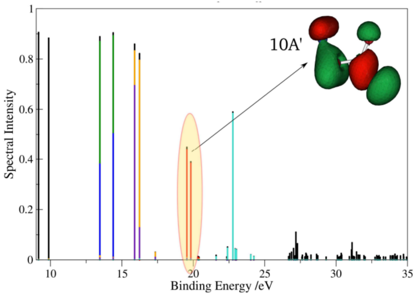



- *theory:* Cooper & Averbukh, PRL **111**, 083004 (2013)

  *recent experiment on spLEAD:*
  K. Ueda & co., PRL **119**, 073203 (2017)

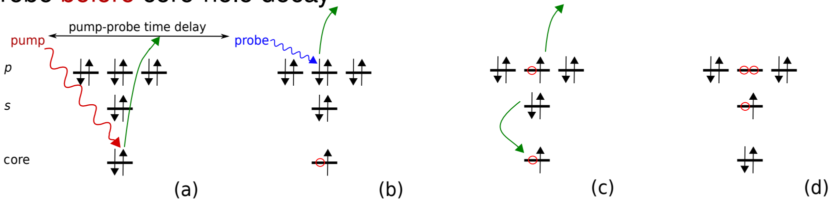
proposal: background-free observation of hole migration



# Pump-probe scheme

- probe before core-hole decay

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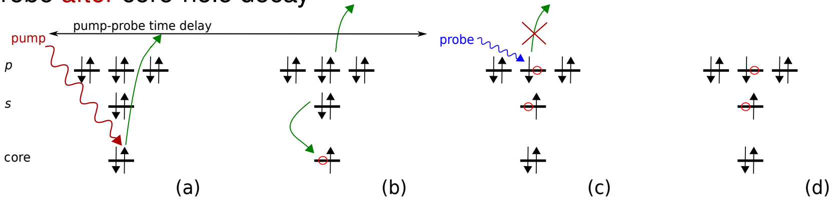

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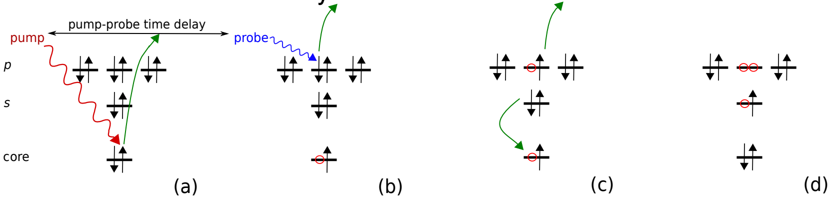
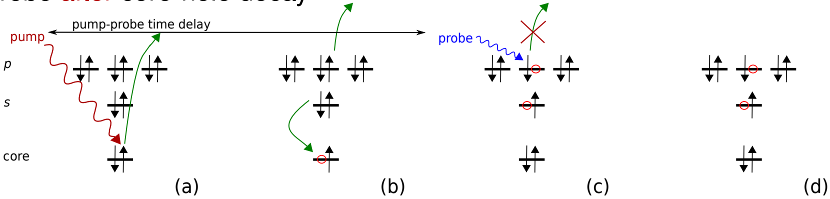



- probe **after** core-hole decay




$X^{3+}$ yield proportional to core-hole survival probability

# Probe pulse requirements

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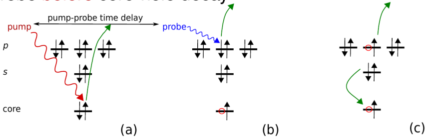


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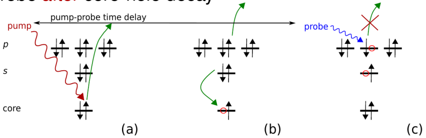

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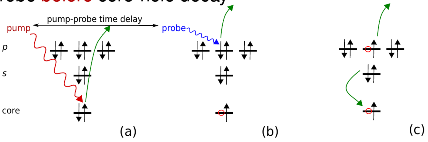


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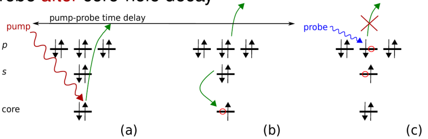


1. $\mathrm{SIP}(\mathrm{core}^{-1}) + \hbar\omega \geq \mathrm{DIP}(\mathrm{core}^{-1}\mathrm{val}^{-1})$
2. $\mathrm{SIP}(\mathrm{core}^{-1}) + \hbar\omega \geq \mathrm{TIP}(\mathrm{val}^{-3})$

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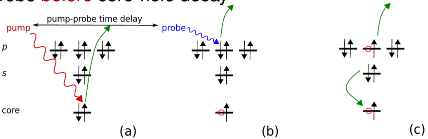


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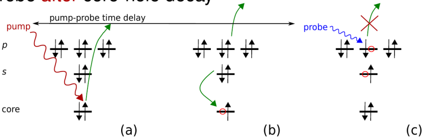


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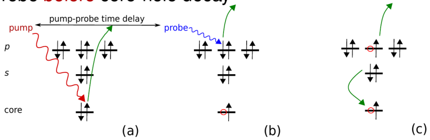


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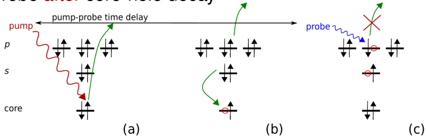


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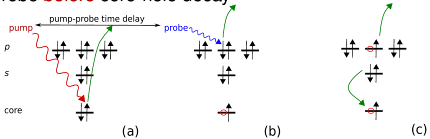


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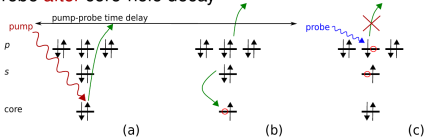


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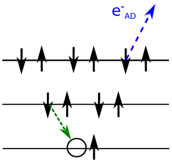
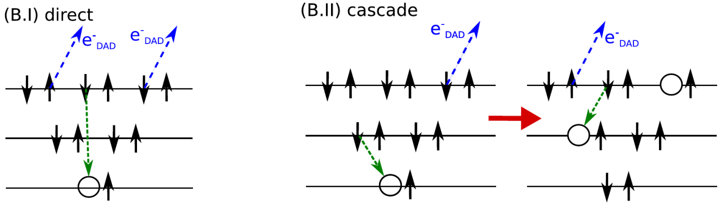
$$\mathrm{TIP}(\mathrm{val}^{-3}) - \mathrm{SIP}(\mathrm{core}^{-1}) \leq \hbar\omega \pm \hbar\,\delta\omega < \mathrm{TIP}(\mathrm{val}^{-3}) - \mathrm{DIP}(\mathrm{val}^{-2})$$

# Double Auger decay

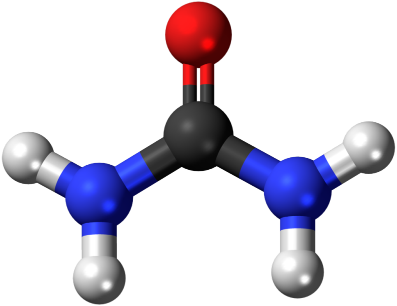
- **A** Auger decay




- **B** double Auger decay – uncontrollable production of $X^{3+}$

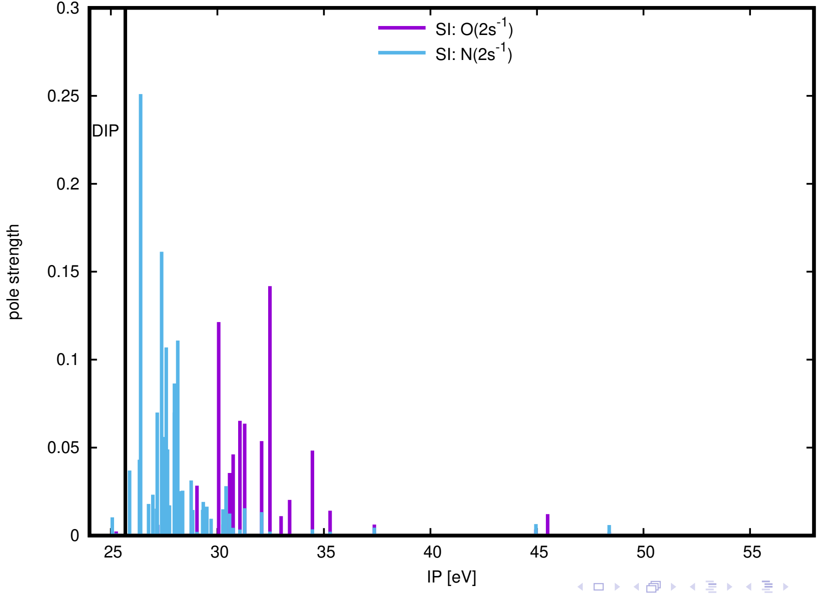


# Example – dynamics of O($2s^{-1}$) hole in urea



$CO(NH_2)_2$

# $O(2s^{-1})$ hole in urea – ADC(2)x calculations



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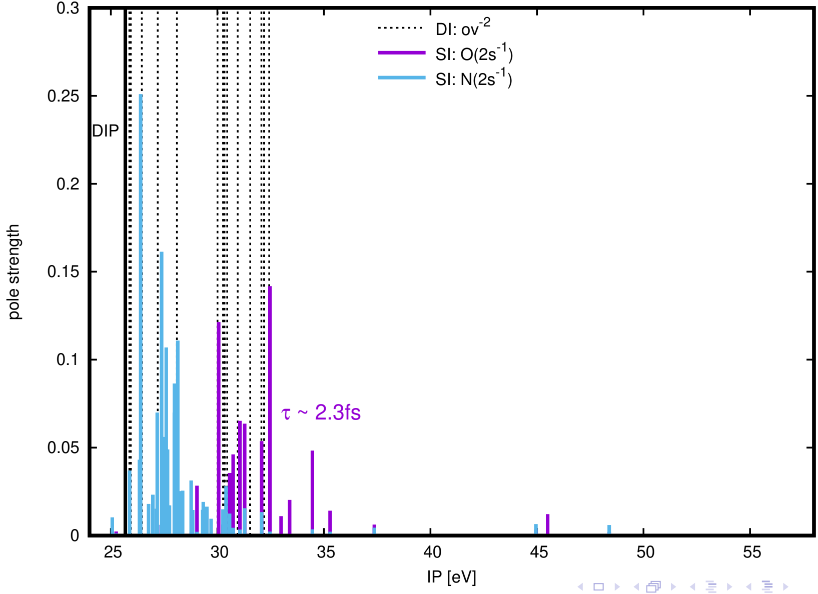

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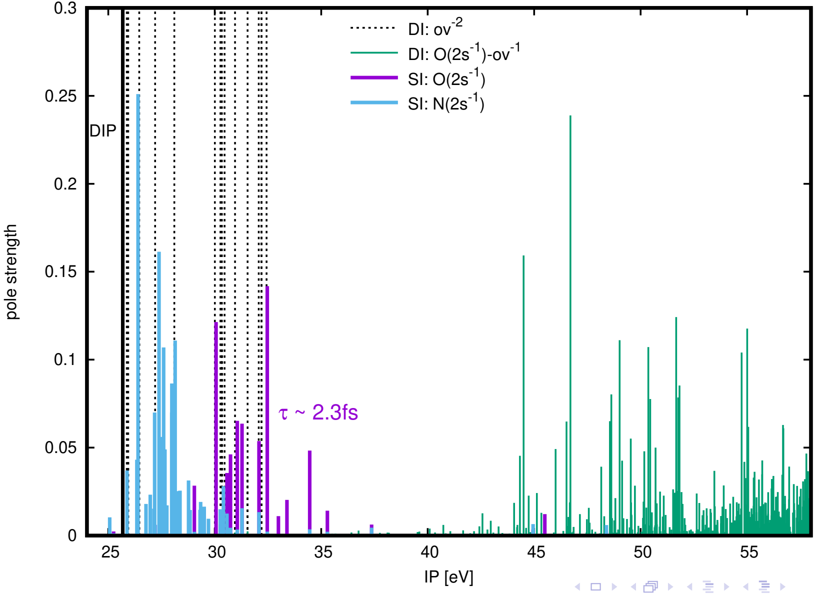

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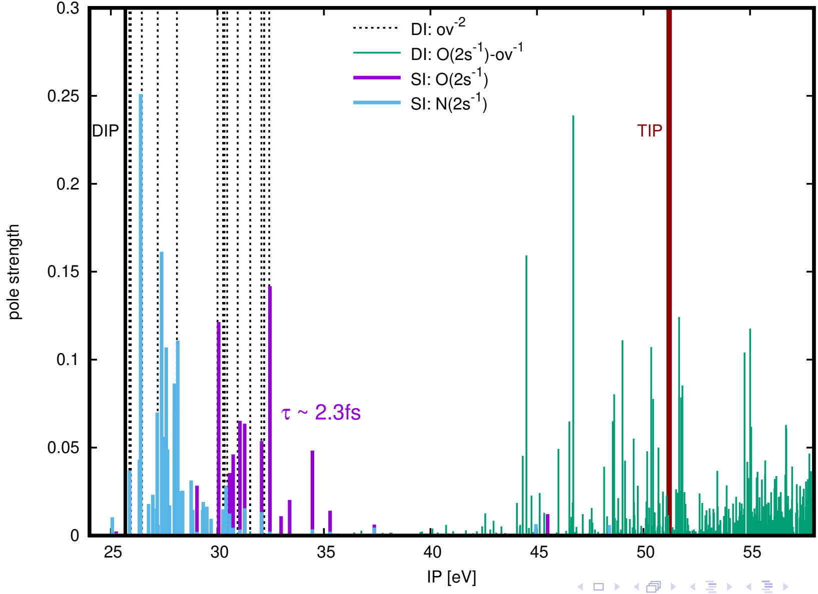

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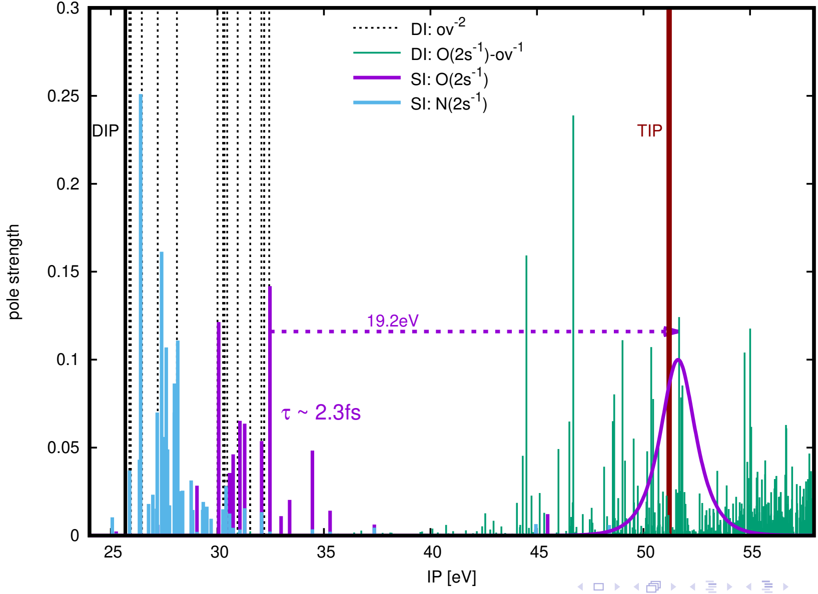

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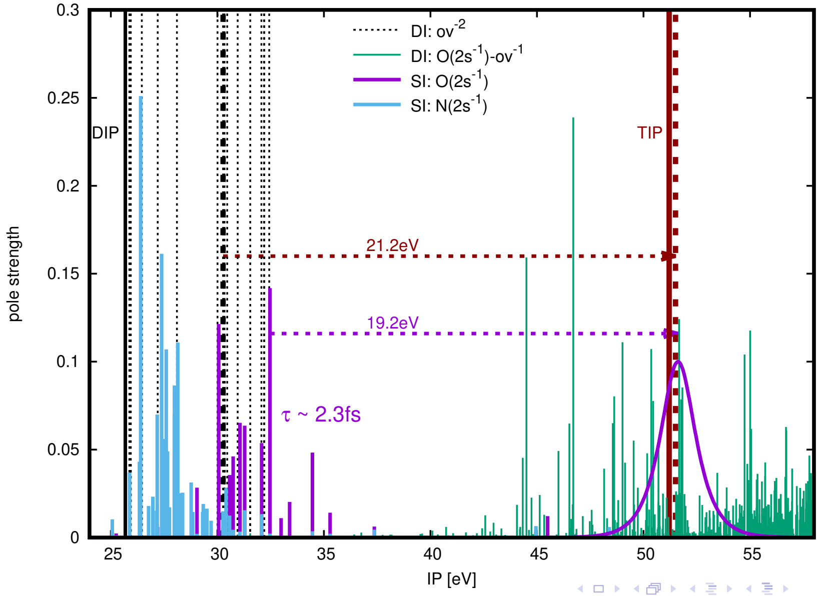

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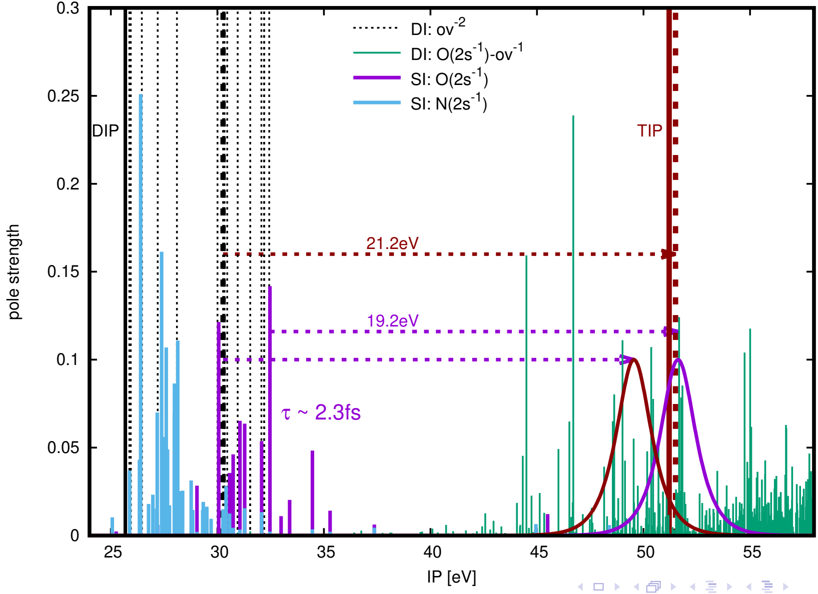

# Double Auger decay – rate equations

$$\frac{dN_{\mathrm{X}_c^+}}{dt} = -N_{\mathrm{X}_c^+}(k_{SA}^n + k_{DA}^n + R\mathcal{E}(t-\tau))$$

$$\frac{dN_{\mathrm{X}_{cv}^{2+}}}{dt} = N_{\mathrm{X}_c^+} R\mathcal{E}(t-\tau) - N_{\mathrm{X}_{cv}^{2+}} k^{n-1}$$

$$\frac{dN_{\mathrm{X}_{vv}^{2+}}}{dt} = N_{\mathrm{X}_c^+} k_{SA}^n$$

$$\frac{dN_{\mathrm{X}^{3+}}}{dt} = N_{\mathrm{X}_c^+} k_{DA}^n + N_{\mathrm{X}_{cv}^{2+}} k_{SA}^{n-1}$$

$$\frac{dN_{\mathrm{X}^{4+}}}{dt} = N_{\mathrm{X}_{cv}^{2+}} k_{DA}^{n-1}$$

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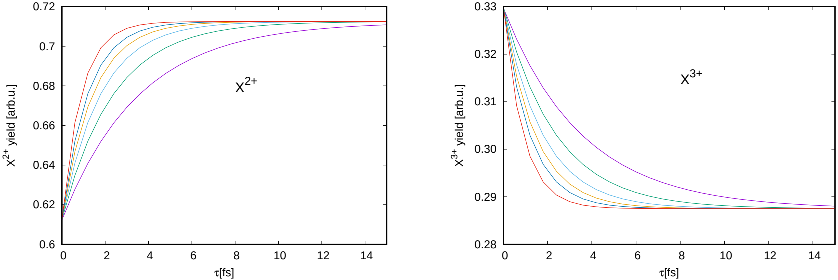


$$N_{\mathrm{X}^{2+}}(t \to \infty) \propto \frac{k_{SA}^n}{k_{\mathrm{tot}}^n}\left(1 - R\exp(-k_{\mathrm{tot}}^n \tau)\right)$$

# Conclusions

1. attosecond pump-probe scheme for Auger-active states
2. decay dynamics encoded in $X^{2+}$ ion signal
    - for inner-valence vacancies also in $X^{3+}$ ion signals
3. background-free measurement unlikely
    - $X^{2+}$ signal-to-background ratio controllable by the probe intensity
    - $X^{3+}$ background depends also on the double-to-single Auger ratio

Thank you!

# Other systems – probe pulse limits

| System | Lower limit | Upper limit | Gap |
|---|---|---|---|
| $ClF^+$ (F $2s^{-1}$) | 33.14 | 33.58 | 0.44 |
| $CH_3F^+$ (F $2s^{-1}$) | 32.36 | 32.98 | 0.61 |
| $C_2H_5F^+$ (F $2s^{-1}$) | 24.18 | 26.40 | 2.22 |
| COOH (O $2s^{-1}$) | 13.18 | 20.36 | 7.18 |
| glycine$^+$ (O $2s^{-1}$) | 15.52 | 20.31 | 4.79 |
| glycine$^+$ (N $2s^{-1}$) | 23.15 | 25.34 | 2.19 |
| urea$^+$ (O $2s^{-1}$) | 18.73 | 21.23 | 2.50 |
| furan$^+$ (O $2s^{-1}$) | 17.42 | 21.91 | 4.49 |
| $Xe^+$ ($4d^{-1}$) | 22.70 | 29.13 | 6.43 |
| $Ar^+$ ($2s^{-1}$) | 30.84 | 39.01 | 8.17 |
| $Ne^+$ ($1s^{-1}$) | 50.04 | 62.13 | 12.09 |
| $CO^+$ (C $1s^{-1}$) | 28.86 | 35.04 | 6.18 |
| $CO^+$ (O $1s^{-1}$) | 26.80 | 35.04 | 8.24 |
| $N_2^+$ (N $1s^{-1}$) | 31.43 | 39.05 | 7.62 |