In-Source Photoionization Spectroscopy: Methods of data analysis

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Resonant photoionization in-source spectroscopy

Isotope shift $\delta\nu_{A,A'}$

$$\delta\nu_{A,A'} = F\,\lambda_{A,A'} + MS$$

Rms charge radius

$$\lambda_{A,A'} = \delta\langle r^2\rangle_{A,A'} + C_2\delta\langle r^4\rangle_{A,A'} + \ldots = 0.93\ \delta\langle r^2\rangle_{A,A'}$$

Relative line position → hyperfine constants *A* & *B* → m_I, Q_S

Doppler limitation:

$$\delta\omega_D = \frac{\omega_0}{c}\sqrt{8kT\frac{\ln 2}{m}}$$

For ^{208}Pb at T_{LIS} = 2000K:

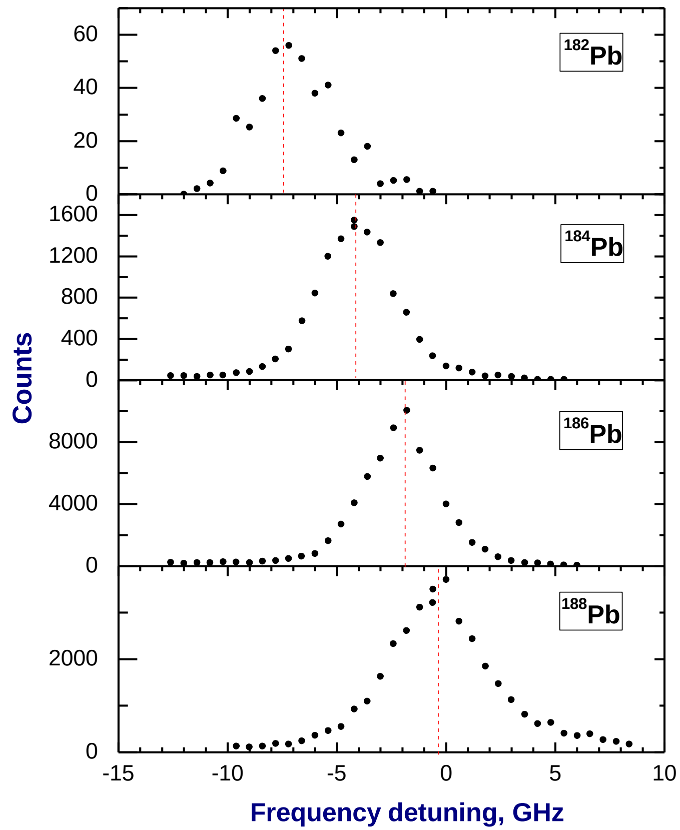
$\delta\omega_D$ = 2.4 GHz (FWHM)

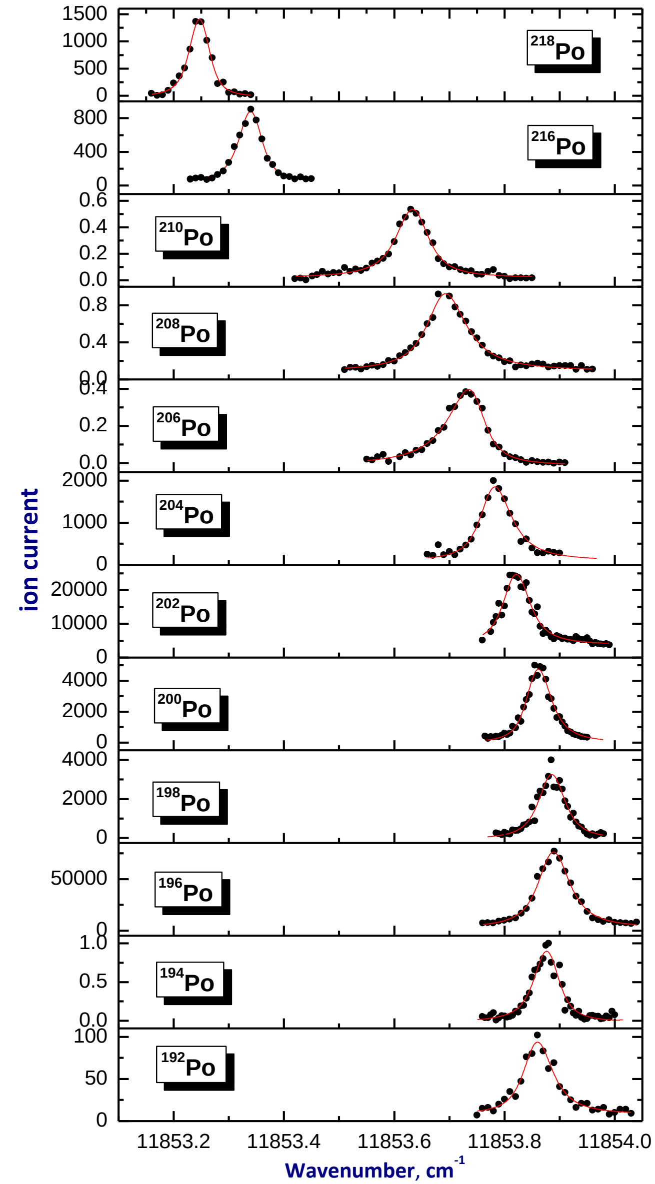
Laser linewidth about 1 GHz

For Pb isotope chain:

$\delta\nu_{A,A+1}$ = 1 GHz

Resonant photoionization in-source spectroscopy





Spectra of even-*A* isotopes of Pb and Po

Fitting function

The simplest case:

$$N_i(v) = C_1 \int N_0^G(v')\, I^L(v - v')\, dv' + C_0$$

(Voigt profile)

General form:

$$N_i(v) = C_1 \int N_0^G(v')\, P_i\left(I^{L'}(v - v')\right) dv' + C_0$$

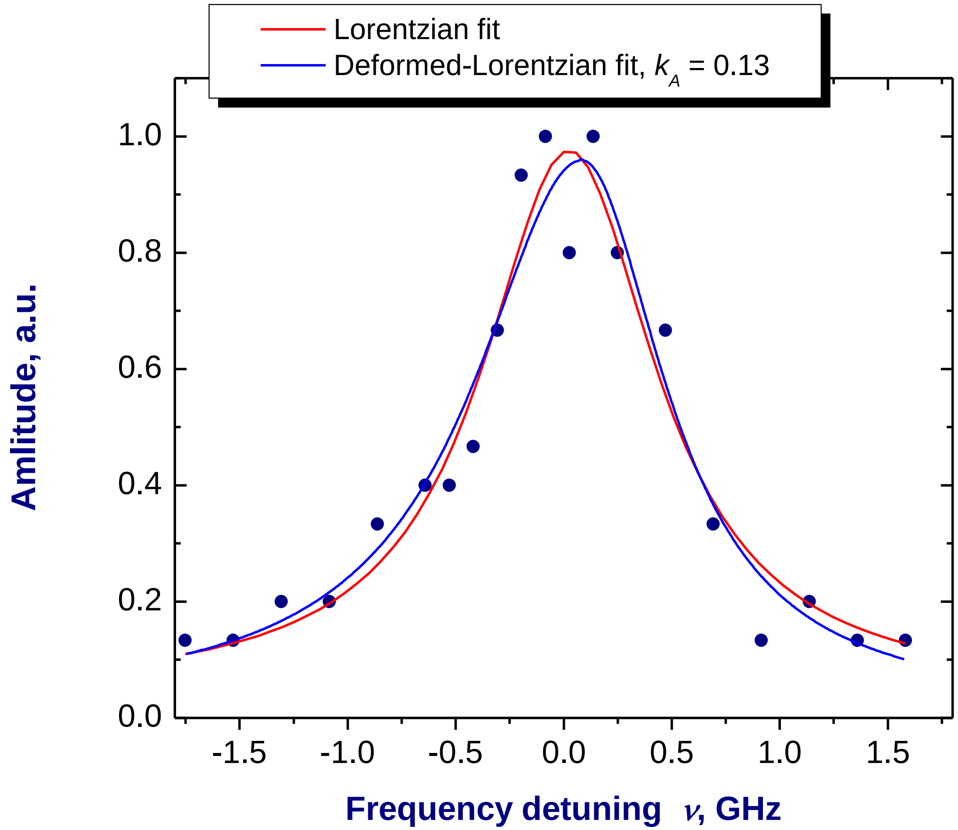
$$P_i\left(I^{L'}(v - v')\right) \neq k\, I^{L'}(v - v')$$

Saturation is possible

$$I^{L'}(v - v')$$

Laser lineshape can be an asymmetrical function

Asymmetry of the fitting function



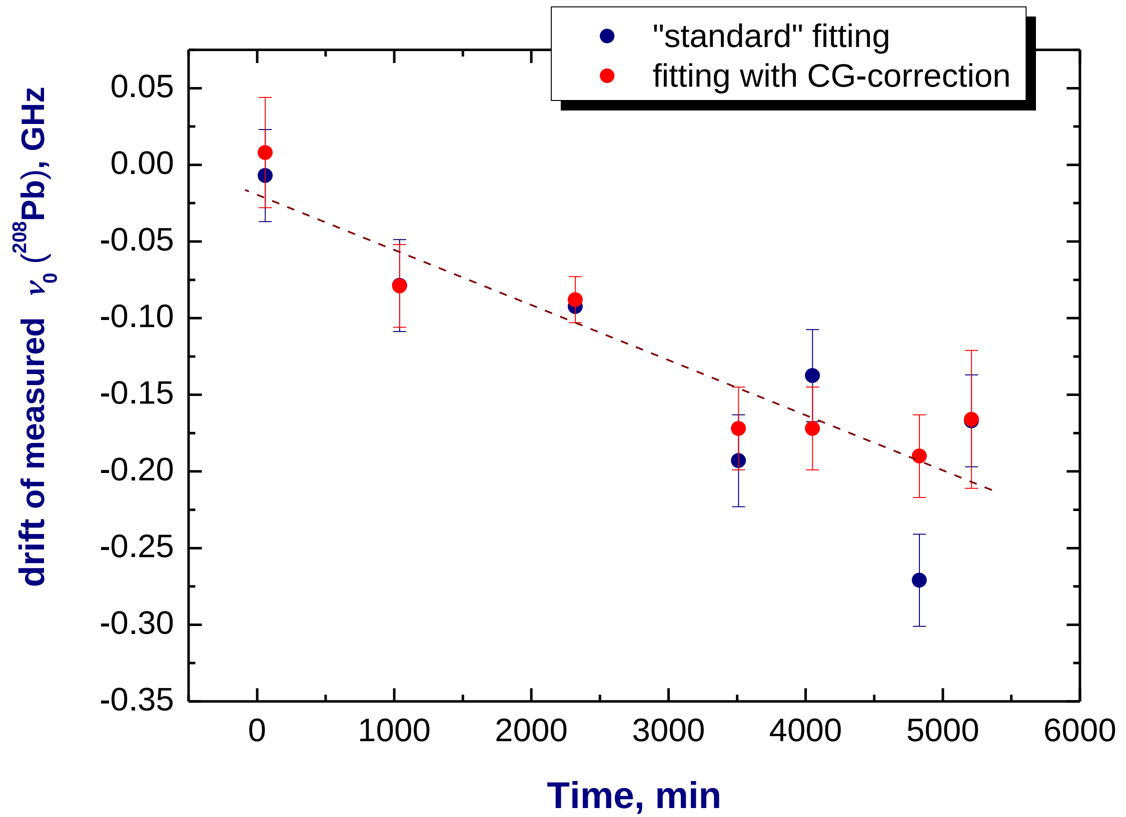
"standard" Lorentzian:

$$L(\nu) = \frac{\gamma / 2\pi}{(\nu - \nu_0)^2 + (\gamma / 2)^2}$$

"deformed" Lorentzian:

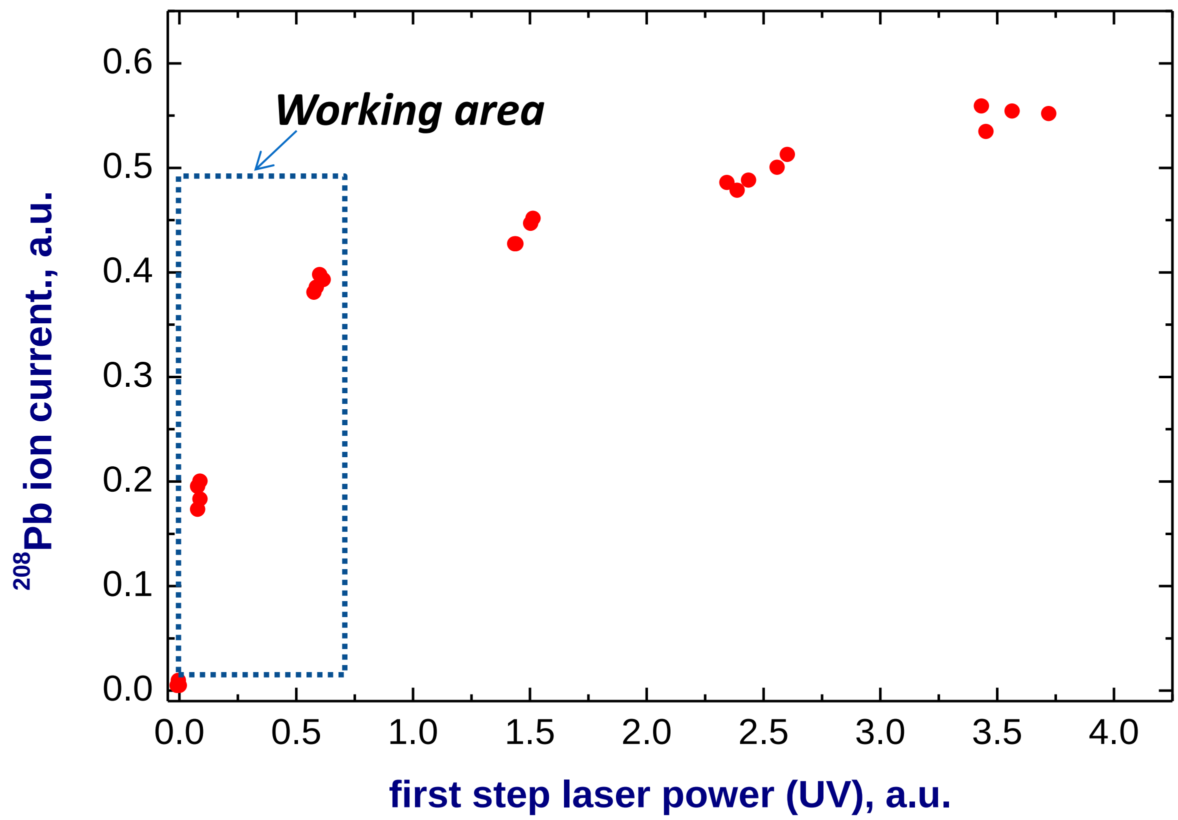
$$L(\nu) = L(\nu'), \begin{cases} \nu' = \nu(1 + k_a), \nu' > \nu_0 \\ \nu' = \nu(1 - k_a), \nu' < \nu_0 \end{cases}$$

Additional correction to asymmetry of the lineshape

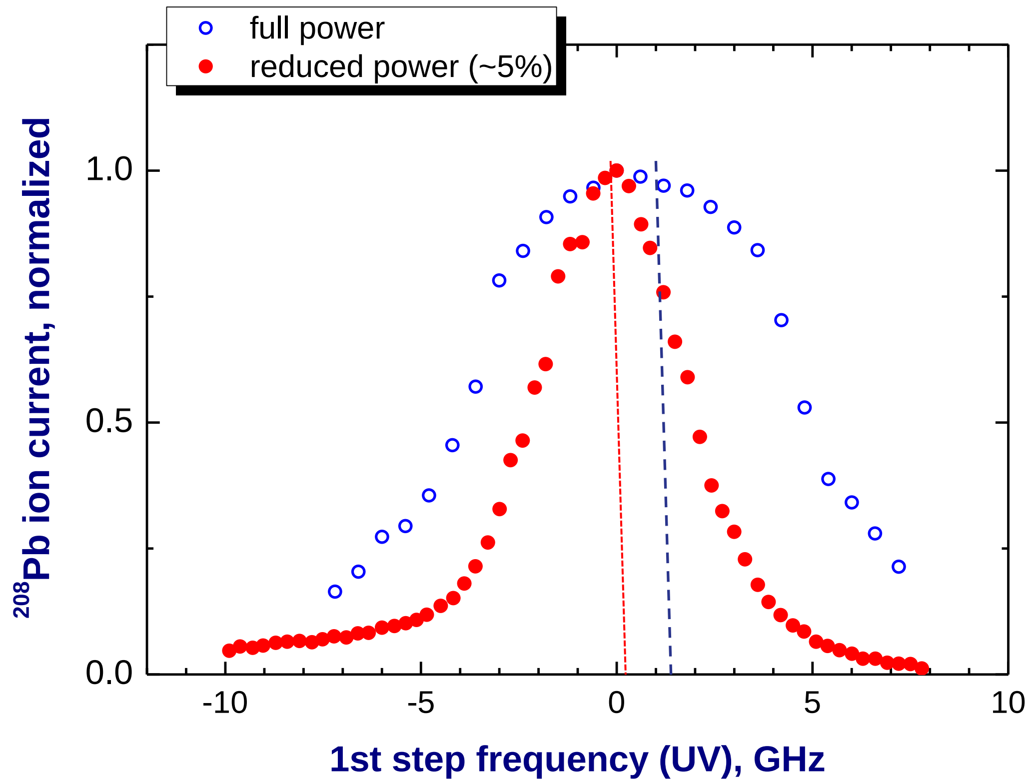


Correction to the laser power

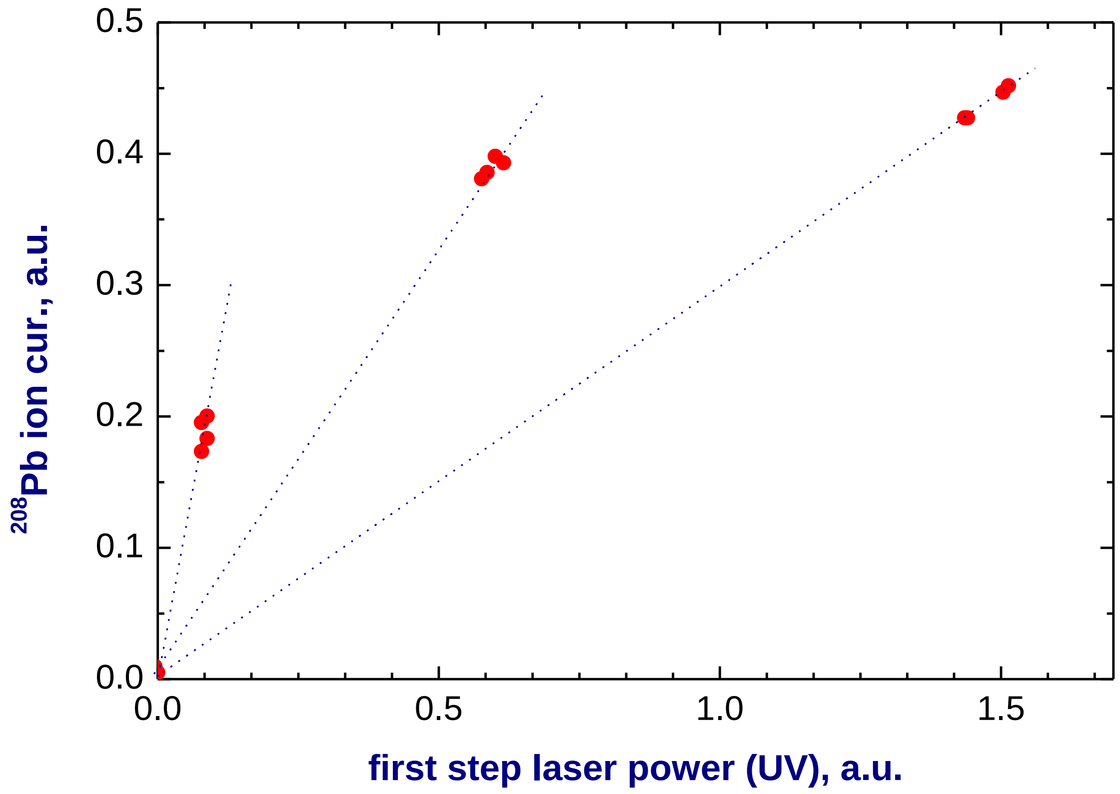
Optical transition saturation



Saturation broadening



Saturation + desynchronization of the laser pulses

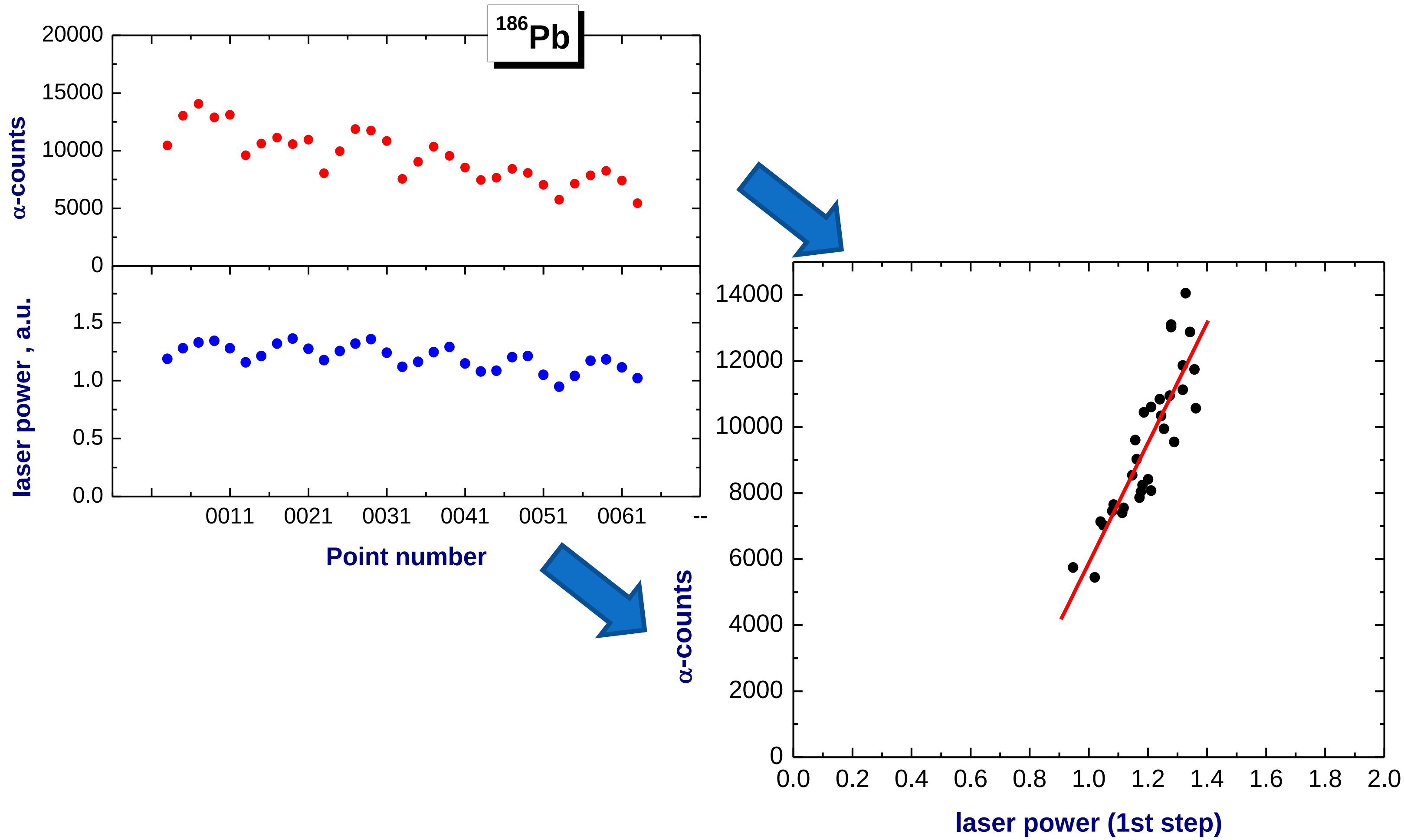


Dye lasers with CVL pumping

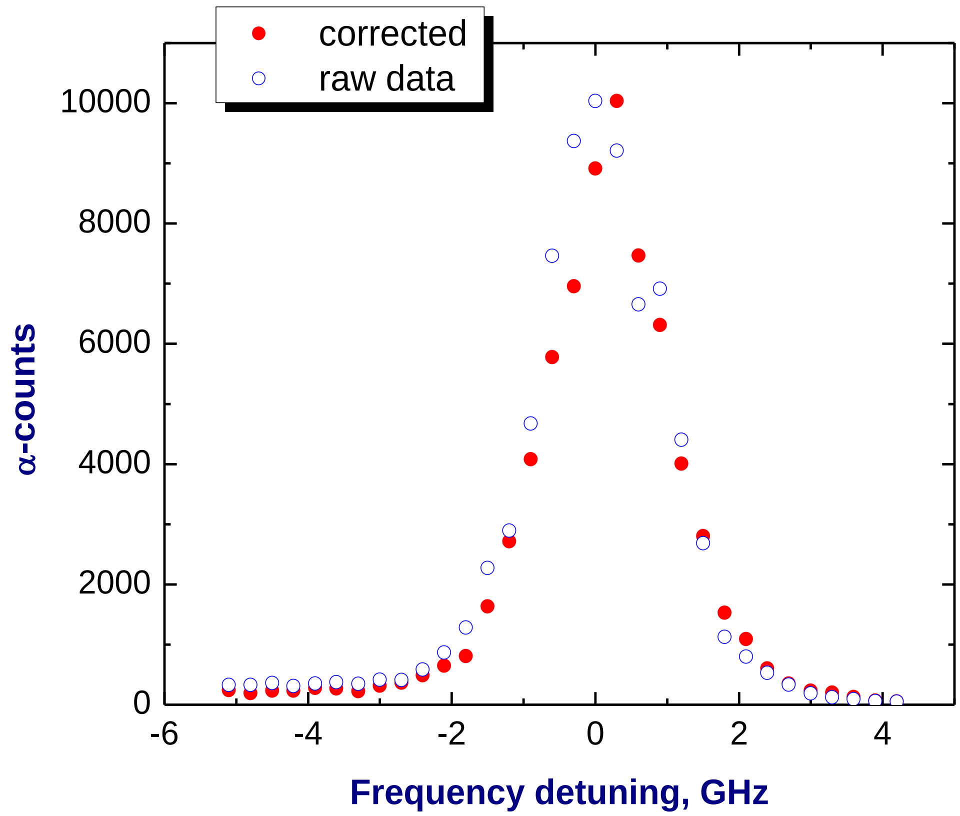
Dye lasers with Nd:YAG pumping: No timing problems

Ti:Sa – timing problems again

Corrections for desynchronization

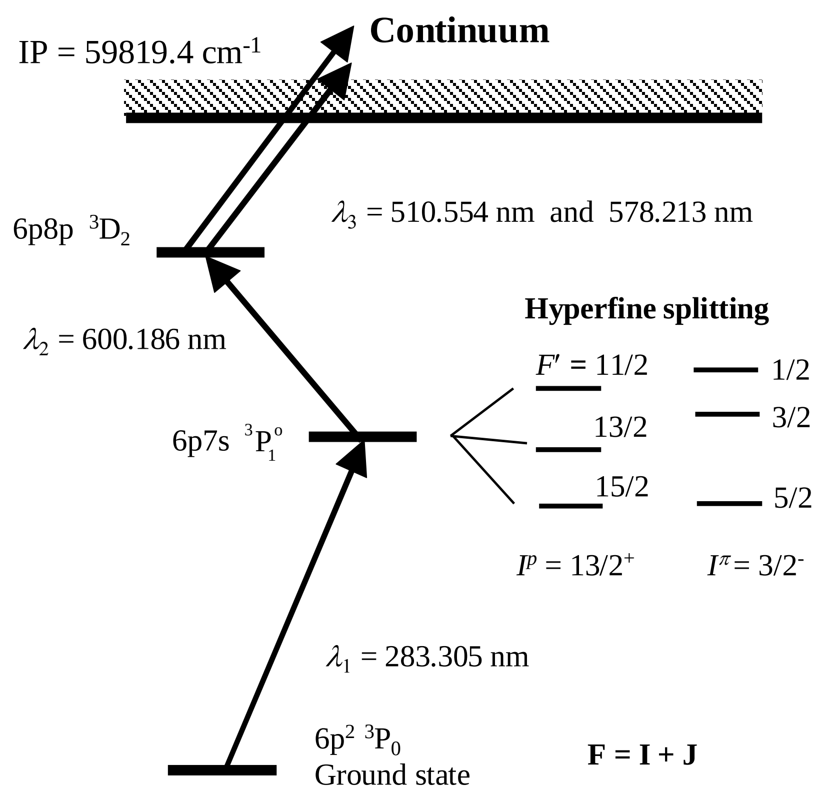


Corrections for desynchronization

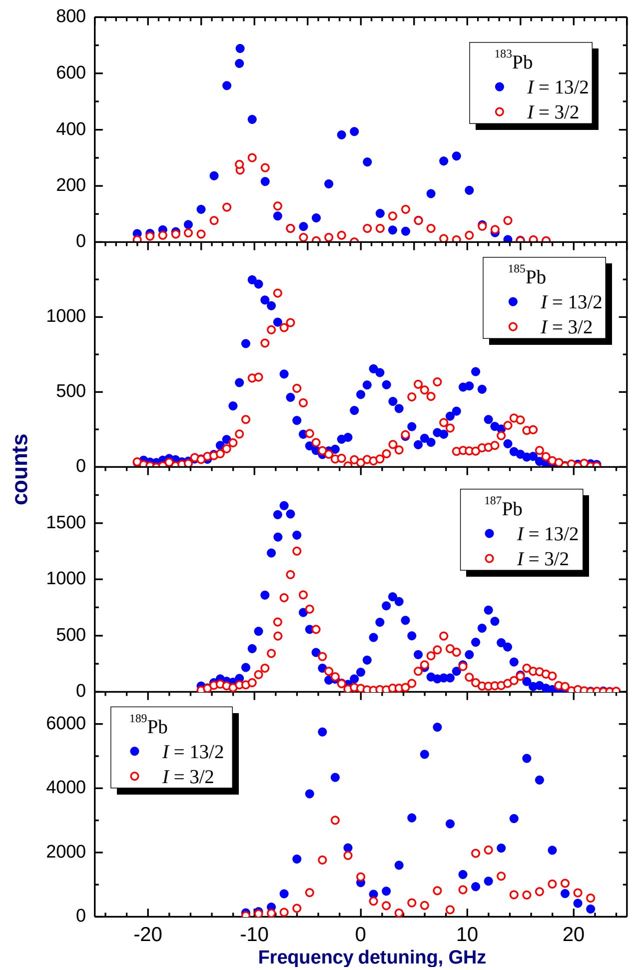


Hyperfine splitting (hfs)

Photoionization scheme for Pb

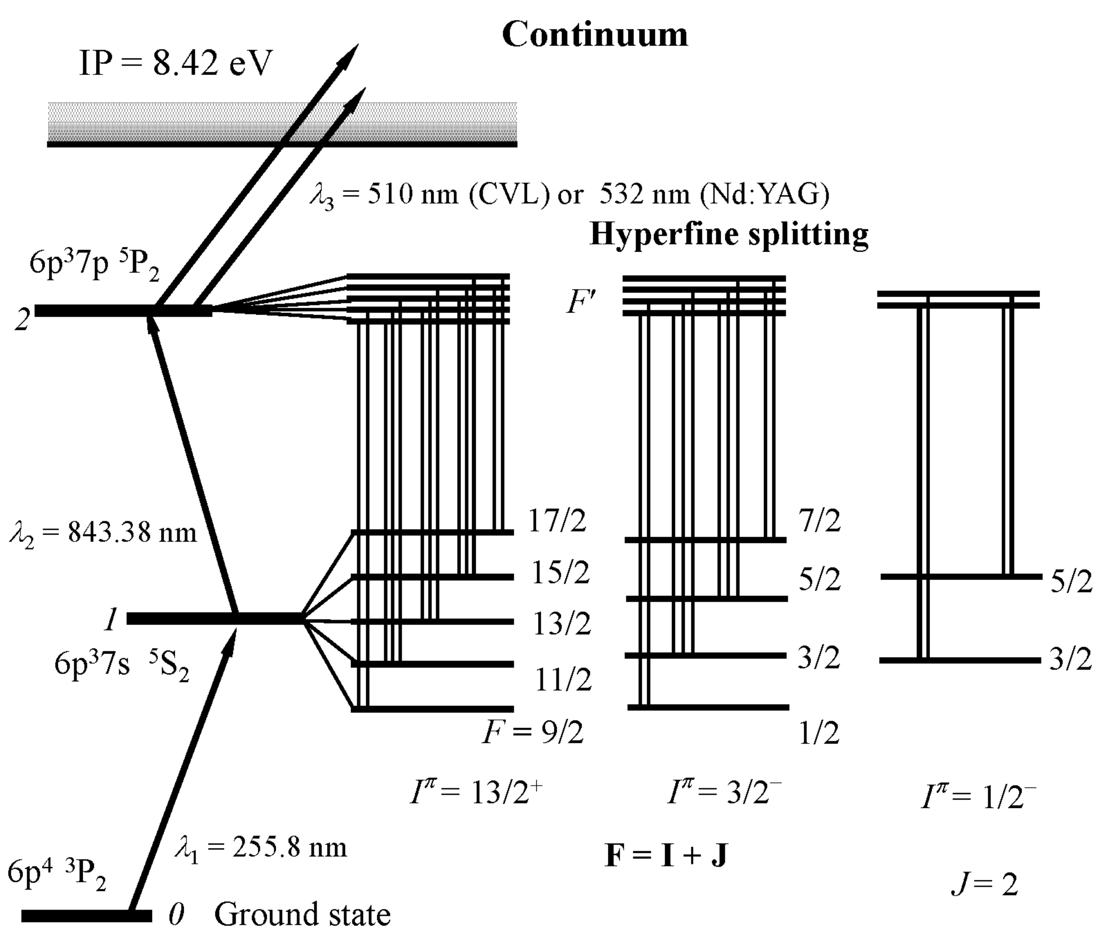


Hyperfine splitting (hfs)



Hyperfine splitting (hfs)

Photoionization scheme for Po



Hyperfine splitting (hfs)

Position of the hfs lines:

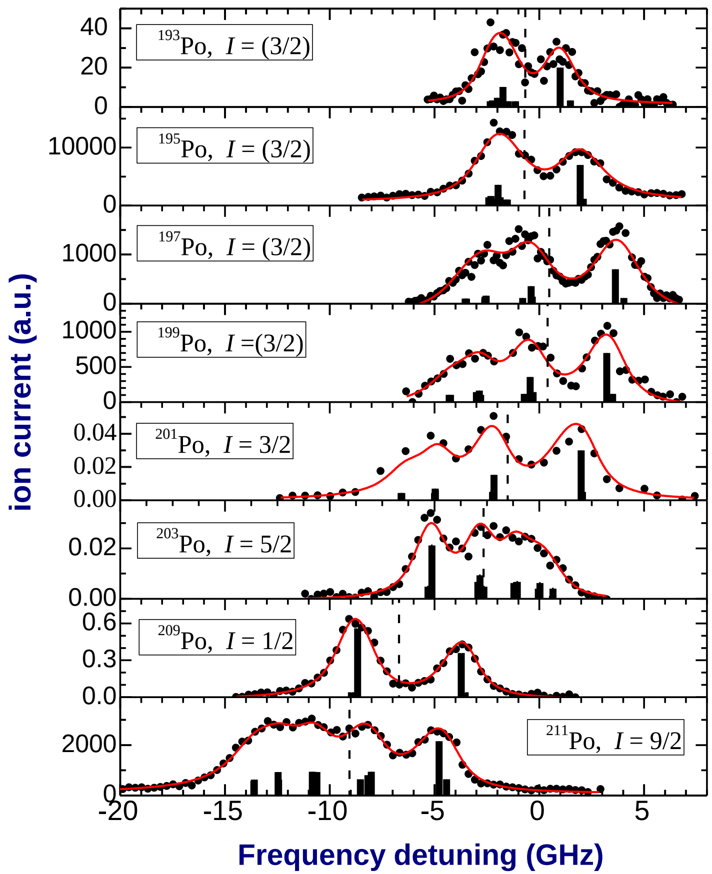
$$\nu_0^{F,F'} = \nu_0 - A\frac{K}{2} - B\frac{\frac{3}{4}K(K+1) - I(I+1)J(J+1)}{2(2I-1)(2J-1)IJ}$$
$$+ A'\frac{K'}{2} + B'\frac{\frac{3}{4}K'(K'+1) - I(I+1)J'(J'+1)}{2(2I-1)(2J'-1)IJ'}$$

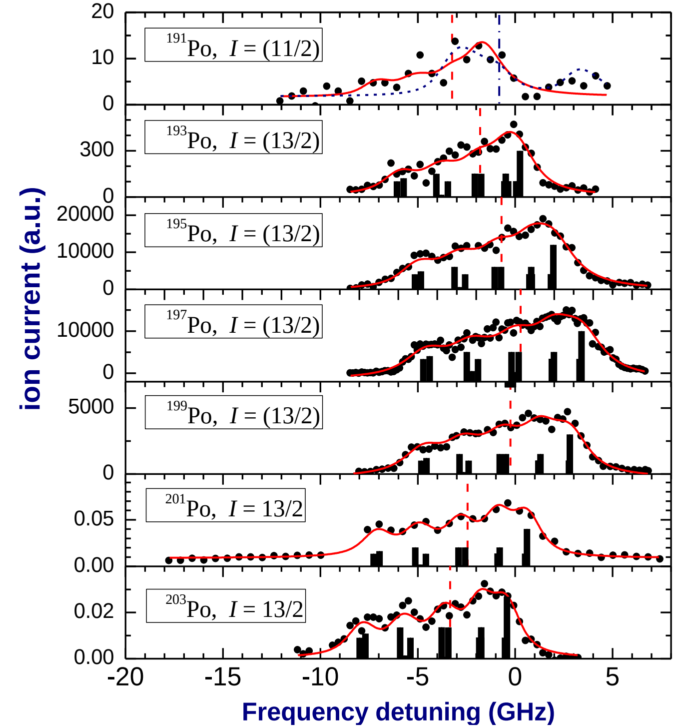
$$K = F(F+1) - I(I+1) - J(J+1)$$

Relative intensities (simplified form):

$$S_{FF'} \sim (2F+1)(2F'+1)\begin{Bmatrix} J' & F' & I \\ F & J & 1 \end{Bmatrix}^2$$

Hyperfine splitting (hfs)





Rate equations

$$N_i(\nu) = C_1 \int N_0^G(\nu') P_i\left(I^{L'}(\nu - \nu')\right) d\nu' + C_0$$

To take into account the saturation of transitions, pumping processes between hyperfine structure (hfs) components and a population redistribution of the hfs levels the number of photoions N_{ion} for each frequency step was calculated by solving the rate equations for the given photoionization scheme:

$$\begin{cases} \dfrac{dN_F}{dt} = \sum\limits_k W_{F'_k F} N_{F'_k} - \sum\limits_k W_{F F'_k} N_F - W_{F,ion} N_F \\ \vdots \\ \dfrac{dN_{ion}}{dt} = \sum\limits_k W_{F'_k, ion} N_{F'_k} \end{cases}$$

$$W_{FF'} \sim S^*_{FF'} I(\nu + \Delta\nu^{FF'} - \nu'), \quad S^*_{FF'} = S_{FF'}/(2F+1)$$

At $t = 0$: $N_F^0 \sim 2F + 1$

Rate equations

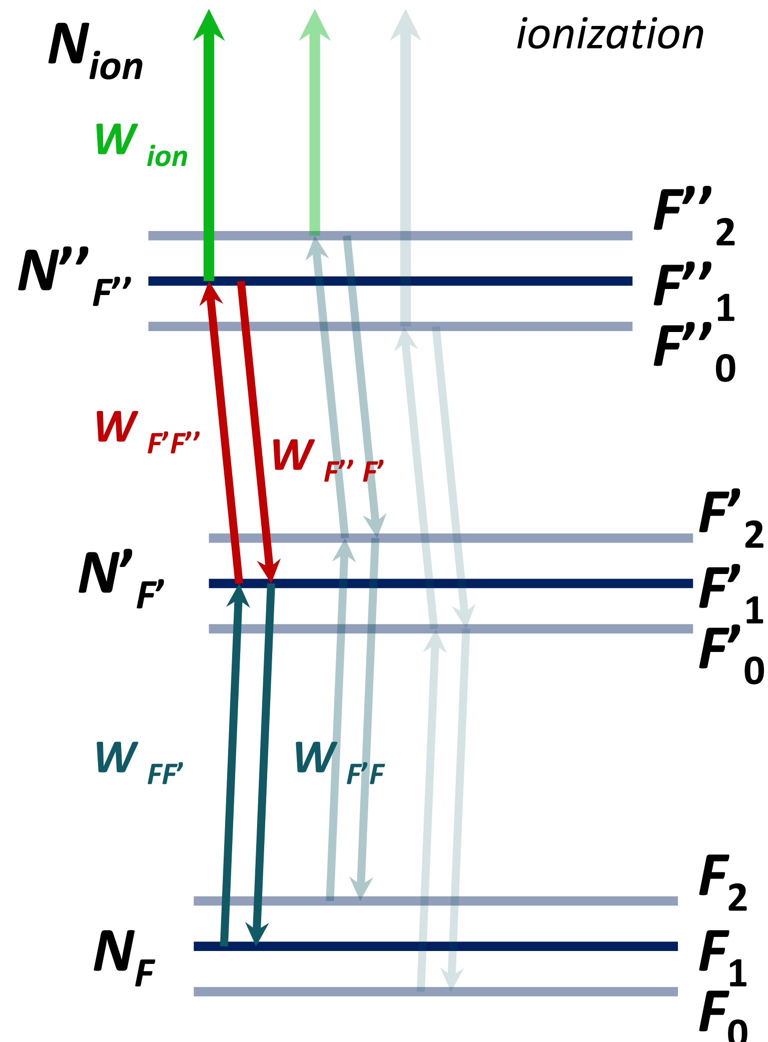
$$dN_{ion} = W_{ion} N''_{F''} dt + \dots.$$

$$dN''_{F''} = -N''_{F''} (W_{ion} + W_{F''F'} + \dots) dt + \\ + N'_{F'} W_{F'F''} dt + \dots$$

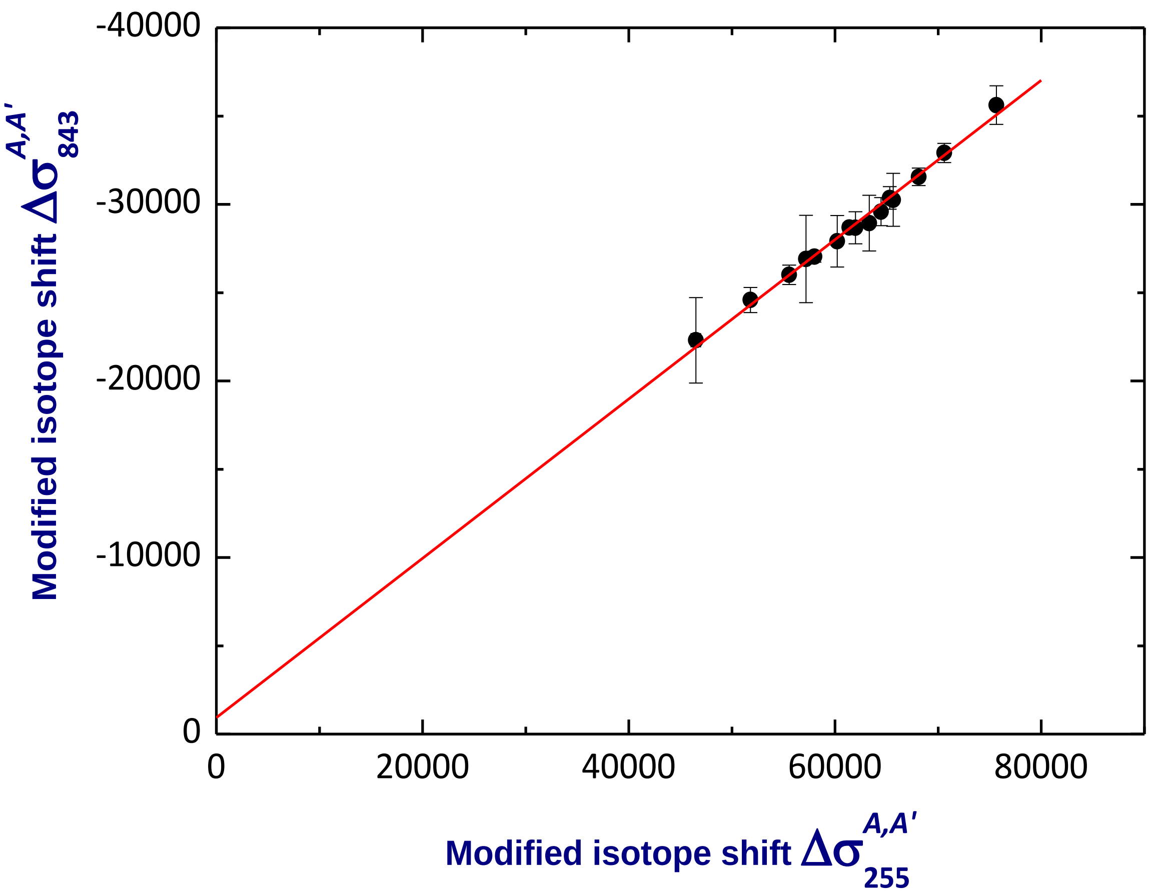
$$dN'_{F'} = -N'_{F'} (W_{F'F''} + W_{F'F} + \dots) dt + \\ + (N''_{F''} W_{F''F'} + N_F W_{FF'} + \dots) dt$$

$$dN_F = -N_F (W_{FF'} + \dots) dt + \\ + (N'_{F'} W_{F'F} + \dots) dt$$

$$\boxed{\frac{W_{FF'}}{W_{F'F}} = \frac{2F'+1}{2F+1}}$$



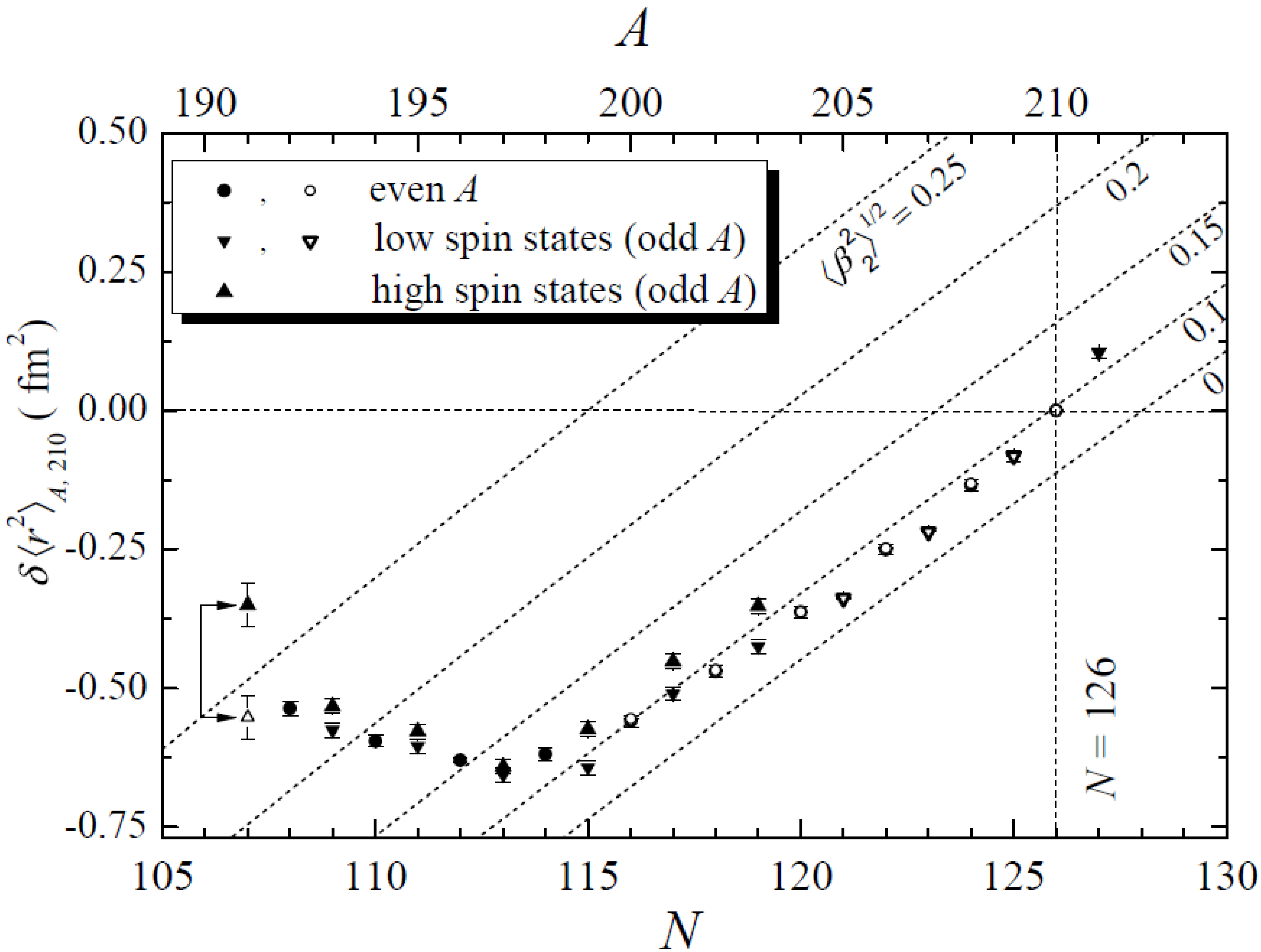
King plot for Po isotopes



$^{200–210}Po$

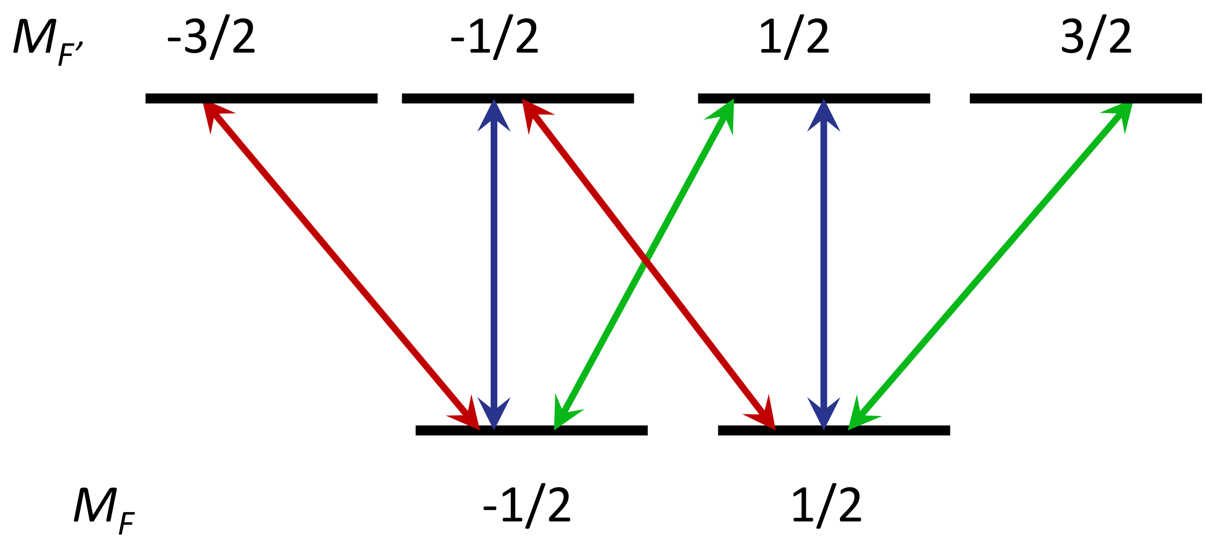
King plot for Po atomic transitions 843 nm (our data) and 255 nm.

Charge radii of Po isotopes



Rate equations for the polarized light

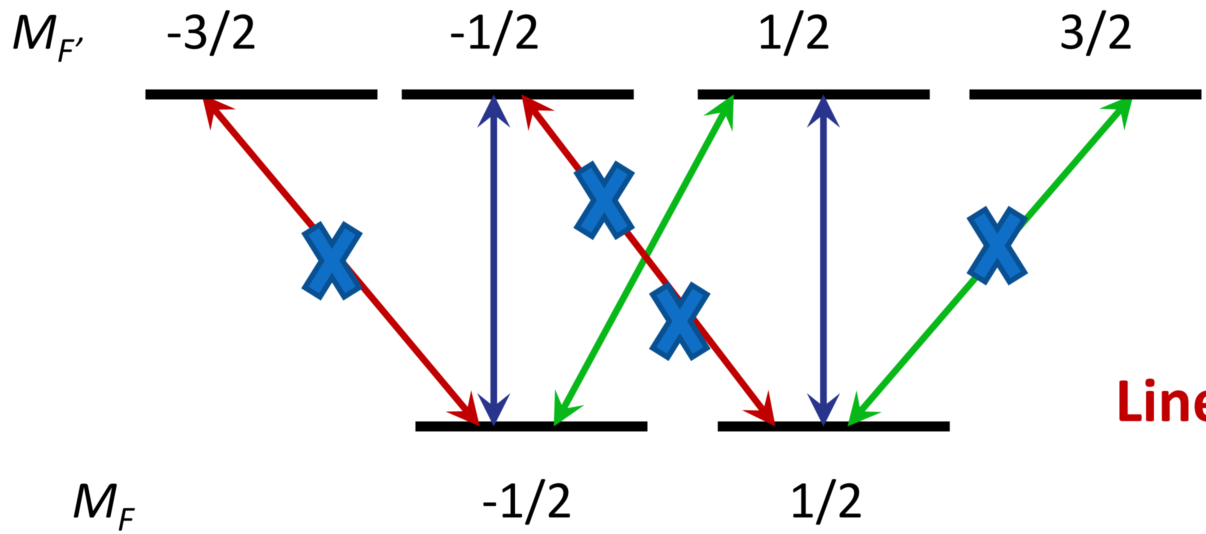
$F = 1/2 \quad \rightarrow \quad F' = 3/2$



$$(2F+1)(2F'+1)\begin{pmatrix} F & 1 & F' \\ -M_F & Q & M'_F \end{pmatrix}^2 \begin{Bmatrix} F & F' & 1 \\ J' & J & I \end{Bmatrix}^2, \qquad Q = M_F - M'_F$$

Rate equations for the polarized light

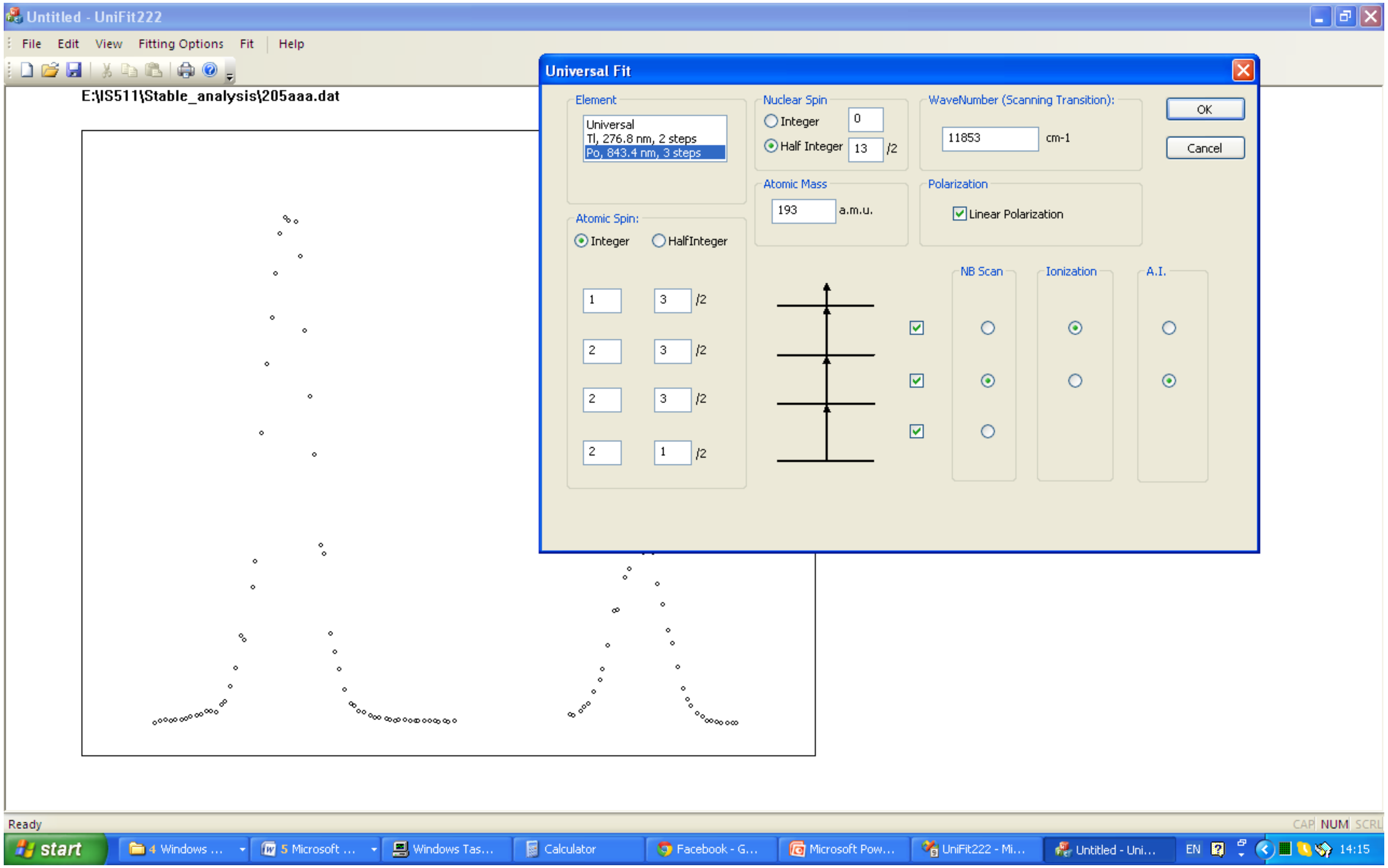
$F = 1/2 \;\rightarrow\; F' = 3/2$



Linear polarization

$$(2F+1)(2F'+1)\begin{pmatrix} F & 1 & F' \\ -M_F & Q & M'_F \end{pmatrix}^2 \begin{Bmatrix} F & F' & 1 \\ J' & J & I \end{Bmatrix}^2, \quad Q = M_F - M'_F$$

Universal fitting program



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Conclusions

The laser ion source is not only a very effective in its normal use as an element-selective tool for producing intense ion beams, but it can be used as a very powerful atomic-spectroscopy tool due to the resonance character of the photoionization (In-Source Laser Resonant Photoionization Spectroscopy). In contrast to other laser spectroscopic techniques, in that case the laser frequency scanning procedure is applied directly within the mass-separator ion source. The main advantage of this technique is its very high sensitivity, nevertheless its spectral resolution is Doppler-limited, therefore the accurate data analysis is of great importance.

Thanks for your attention!