PAC studies of isolated small molecules: The Pb nuclear quadrupole moments and further cases

H. Haas[1,2], J. Röder[1,2], L. Hemmingsen[3], S.P.A. Sauer[3], J.G. Correia[2,4], J. Schell[2,5], V. Amaral[1], D.C. Lupascu[5], R. Vianden[6], V. Kellö[7]

[1] Department of Physics and CICECO, University of Aveiro, 3810-193 Aveiro, Portugal

[2] EP Division CERN, 1211Geneve-23, Switzerland

[3] Department of Chemistry, University of Copenhagen, 2100 Copenhagen, Denmark

[2] C2TN, DECN, Instituto Superior Técnico, Universidade de Lisboa, Portugal

[5] Institute for Materials Science and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 45141 Essen, Germany

[6] Helmholz-Institut für Strahlen- und Kernphysik, Universität Bonn, Nussallee 14-16, Bonn, Germany

[7] Department of Physical and Theoretical Chemistry, Comenius University, 84215 Bratislava, Slovakia

Follow-up of project IS640

Determination of nuclear quadrupole moments

Methods for long-lived (stable) nuclei:
Atomic spectroscopy
Molecular spectroscopy
Muonic / pionic spectroscopy

Methods for short-lived (excited) states:
PAC / PAD spectroscopy in solids
Moessbauer spectroscopy in solids
Level mixing spectroscopy in solids

Experimentally measured: $\nu_Q = eQV_{zz}/h$ **Problem:**

V_{zz} (EFG) has to come from theory!

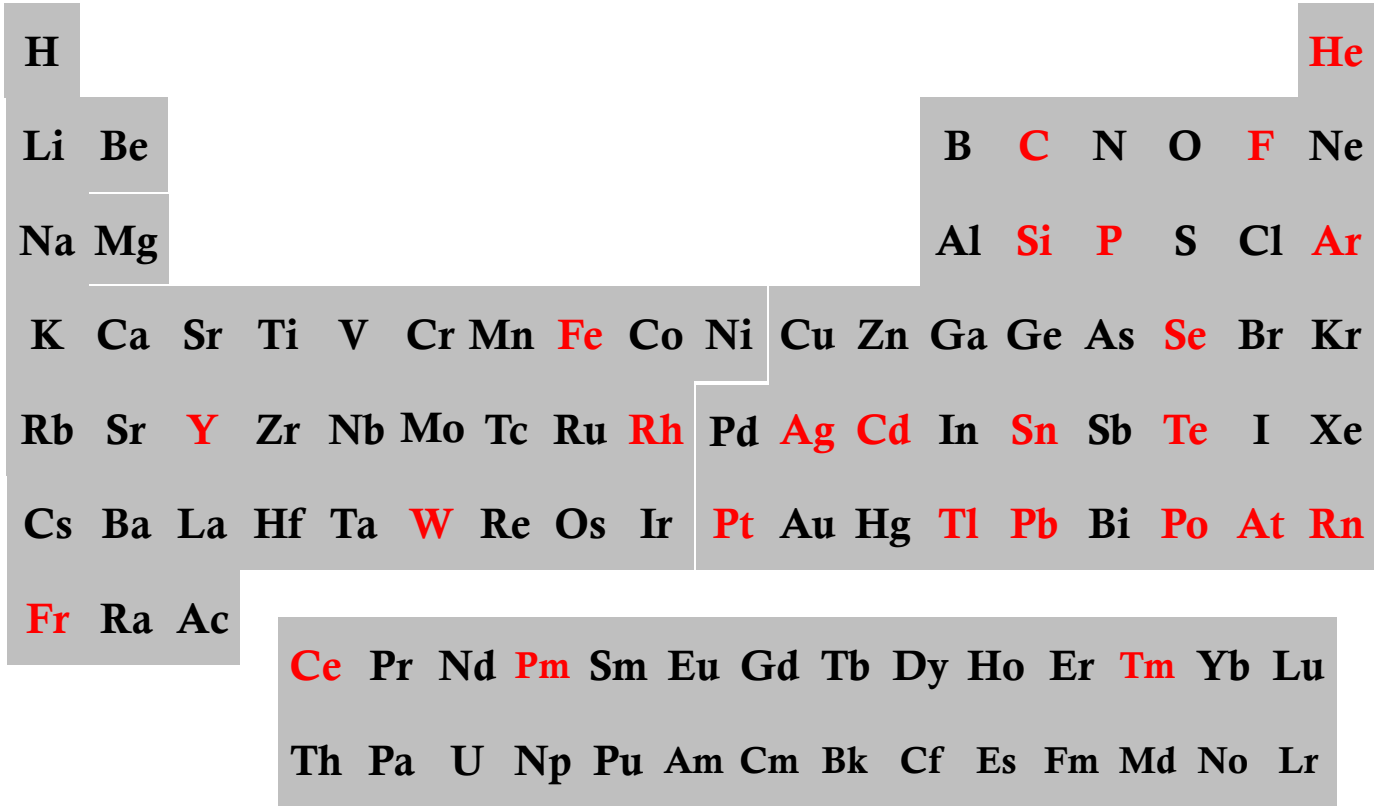
1-2% **Theoretical accuracy** **10-20%**

Link via NMR to get V_{zz} in solids (for stable isotopes with I > 1/2)

but
impossible for several elements:

«missing link peoblem»

The “missing link” cases of nuclear quadrupole moments



What we have done in IS640:

- Measure the quadrupole interaction in some free Cd (and Hg) molecules in the gas state by PAC
- Basic idea: In a linear molecule the EFG (V_{zz}^{mol}) is along the molecular axis
- The rotation axis J is always perpendicular to the molecular axis
- The EFG along J is then, independent of J:

$$V_{zz}^{rot} = -1/2\ V_{zz}^{mol}$$

- For large J the quantization should be fully along J, leading to a splitting frequency independent of J !

Quantum mechanics

- No J: $\hat{H} = eQV_{zz}\left\{\frac{3\boldsymbol{I}_z^{\,2} - I(I+1)}{4I(2I-1)}\right\}$
- $R(t) = a_0 + a_1\cos(w_0 t) + a_2\cos(2w_0 t) + a_3\cos(3w_0 t)$
- $$w_0 = \frac{6}{4I(2I-1)}\frac{eQV_{zz}}{h}$$

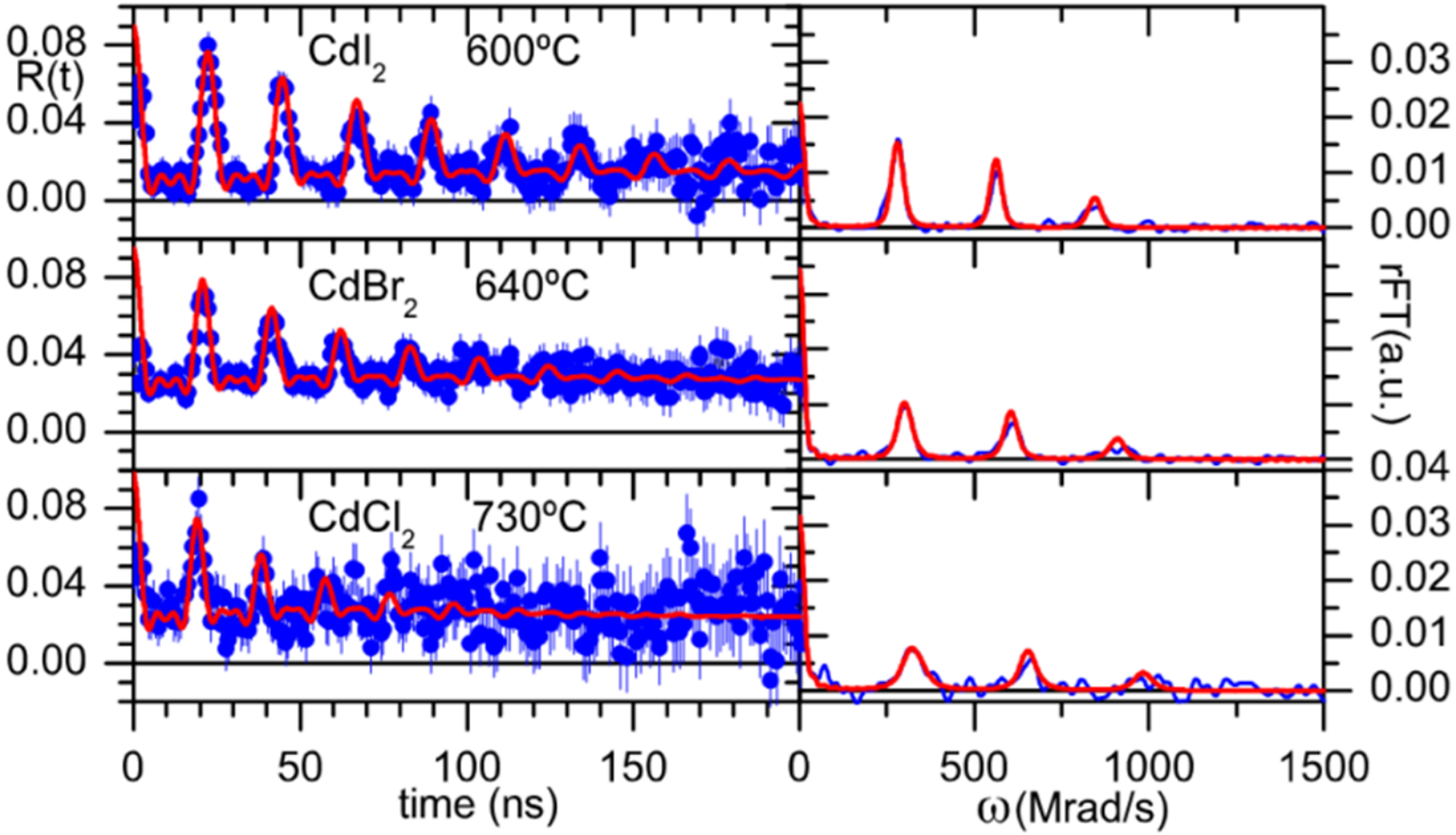
Quantum mechanics

- No J: $\hat{H} = eQV_{zz}\left\{\frac{3\boldsymbol{I}_z^{\,2} - I(I+1)}{4I(2I-1)}\right\}$
- $R(t) = a_0 + a_1\cos(w_0 t) + a_2\cos(2w_0 t) + a_3\cos(3w_0 t)$
- $$w_0 = \frac{6}{4I(2I-1)}\frac{eQV_{zz}}{h}$$

- With J: $\hat{H} = eQVzz\left\{\frac{3(\boldsymbol{I}.\boldsymbol{J})^2 + 1.5(\boldsymbol{I}.\boldsymbol{J}) - J(J+1)I(I+1)}{J(2J-1)2I(2I-1)}\right\}$
- Casimir formula (1933)

Free Molecule Studies by Perturbed γ-γ Angular Correlation: A New Path to Accurate Nuclear Quadrupole Moments

Heinz Haas,[1,2] Jens Röder,[1,2] Joao G. Correia,[3,2] J. Schell,[4,2] Abel S. Fenta,[1] Reiner Vianden,[5] Emil M. H. Larsen,[6] Patrick A. Aggelund,[6] Rasmus Fromsejer,[6] Lars B. S. Hemmingsen,[6] Stephan P. A. Sauer,[6] Doru C. Lupascu,[4] and Vitor S. Amaral[1]



Calibrating the Pb quadrupole moments

Present status:
Q from some 20 PAD / LEMS experiments and
V_{zz} from some 25 PAC measurements in solids

Q of PAC state

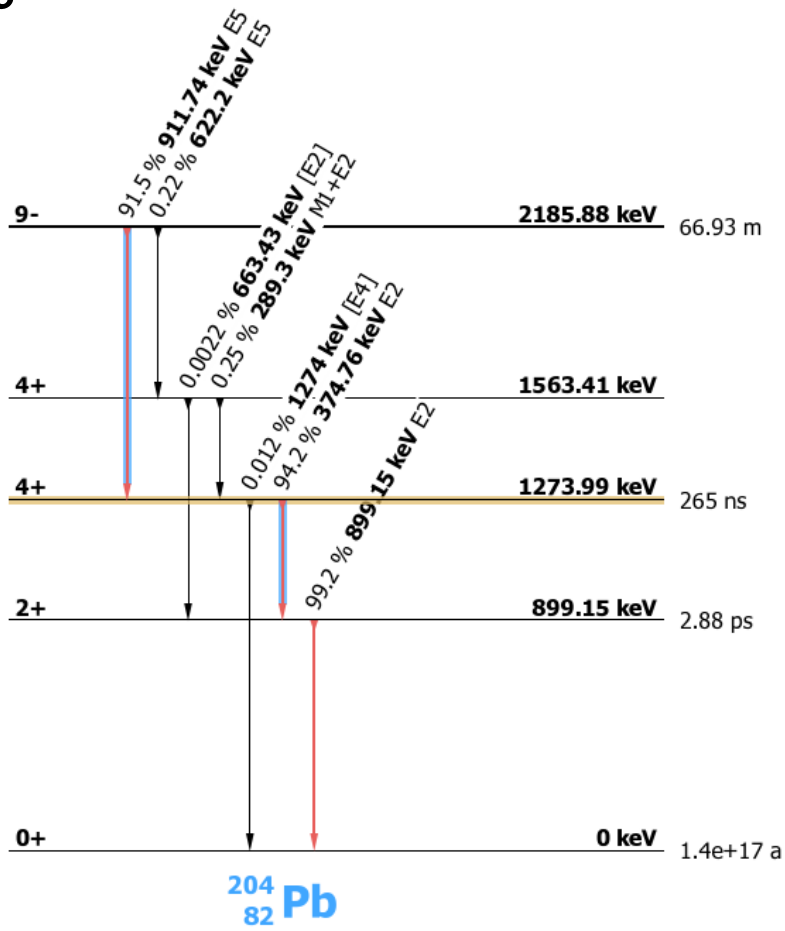
tied to

Solution:

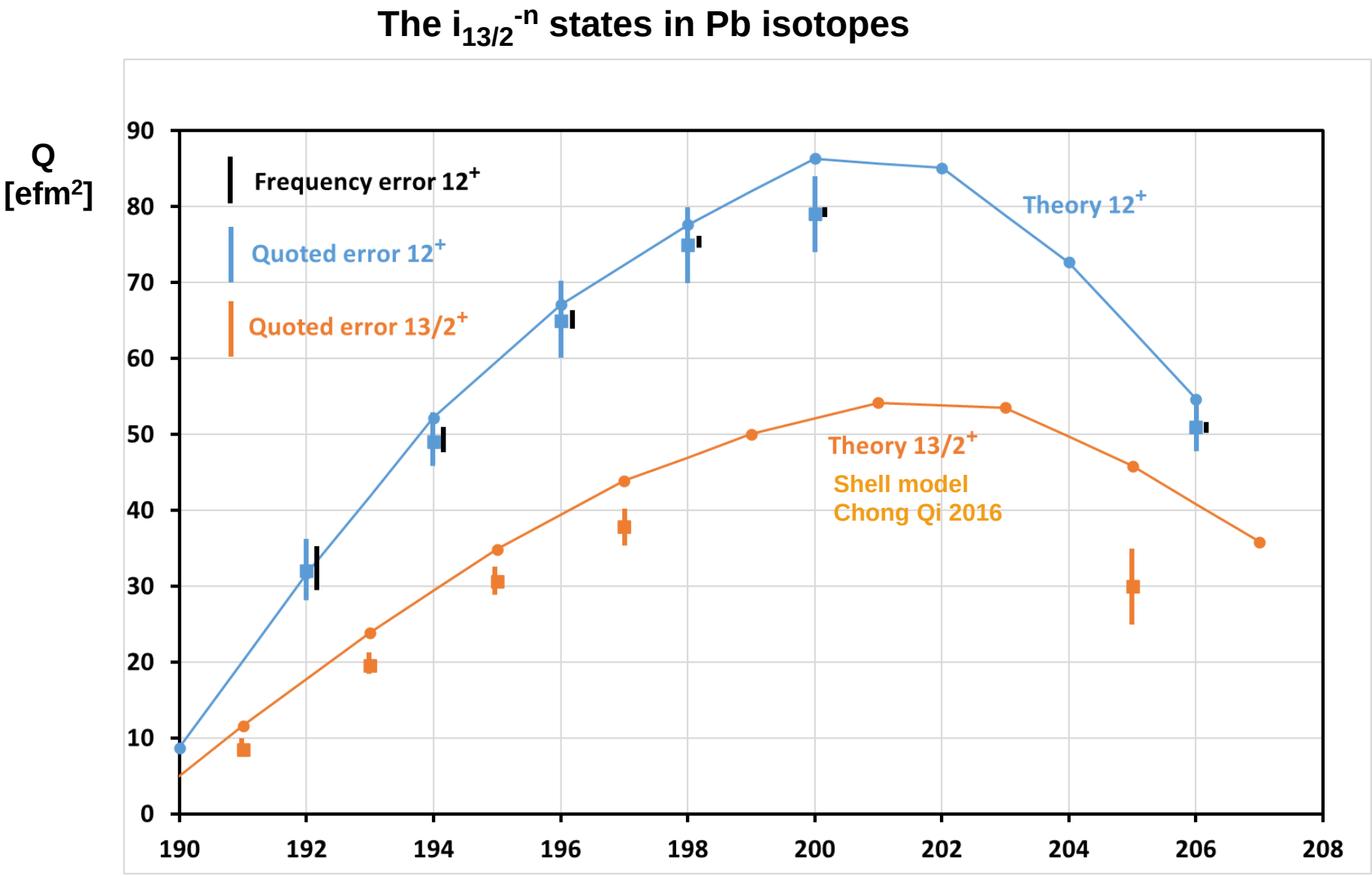
Measure ν_Q for some simple Pb molecules (PbTe,…) and apply quantum chemistry methods for V_{zz}

Problems:

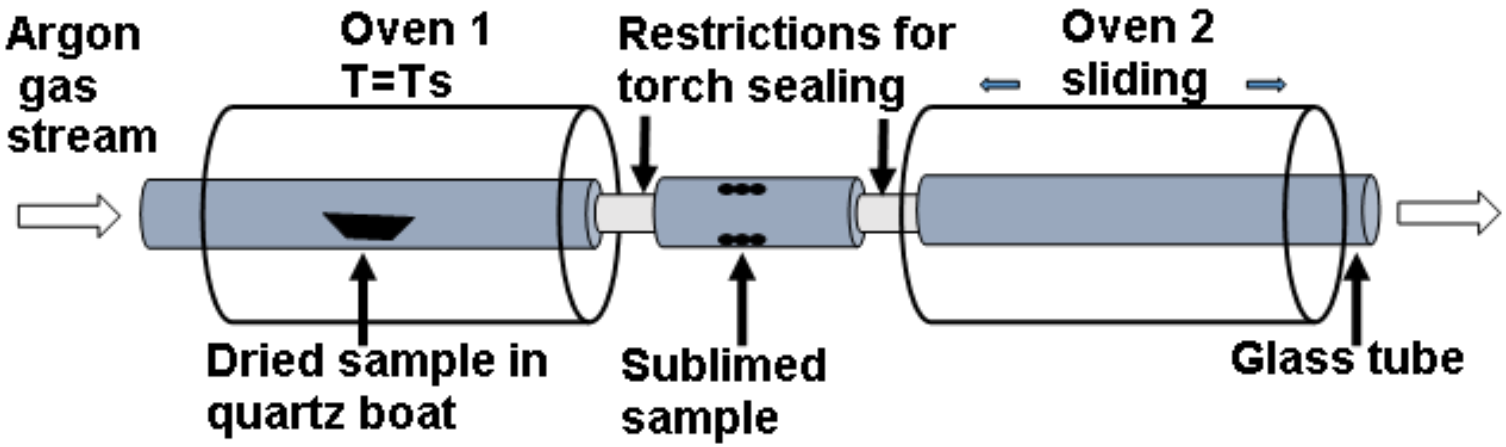
high temperatures needed
sample production
counting statistics
higher state spin



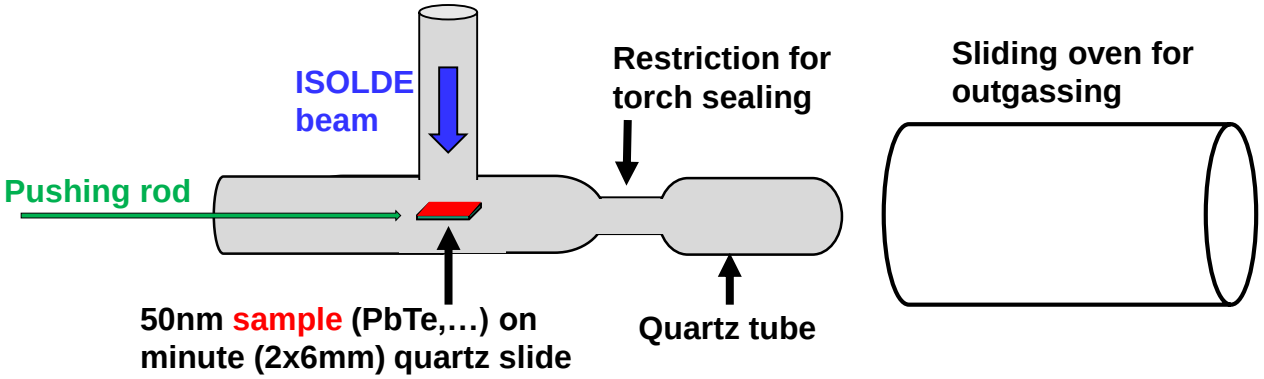
Typical example demonstrating need for accurate Q calibration

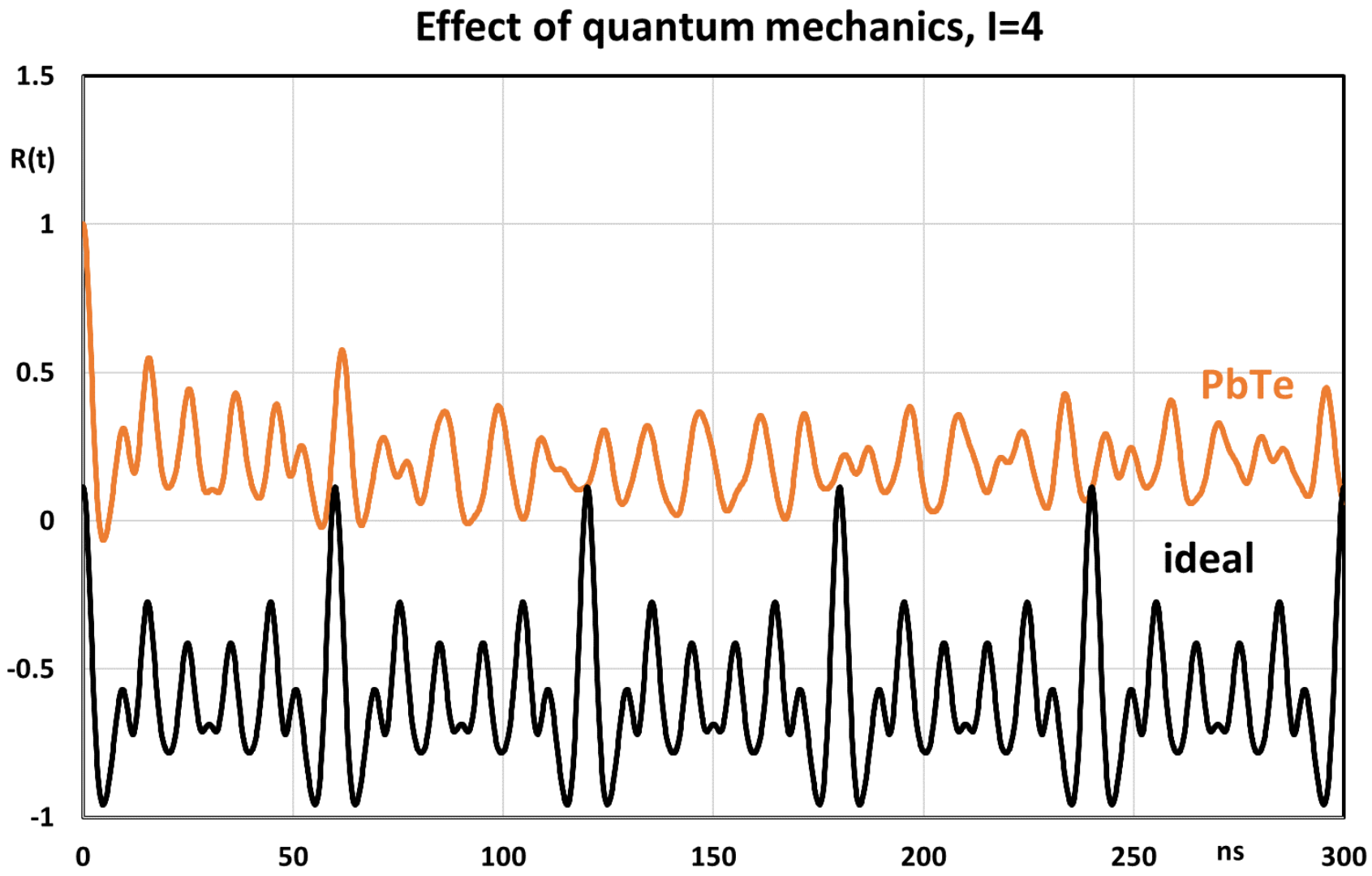


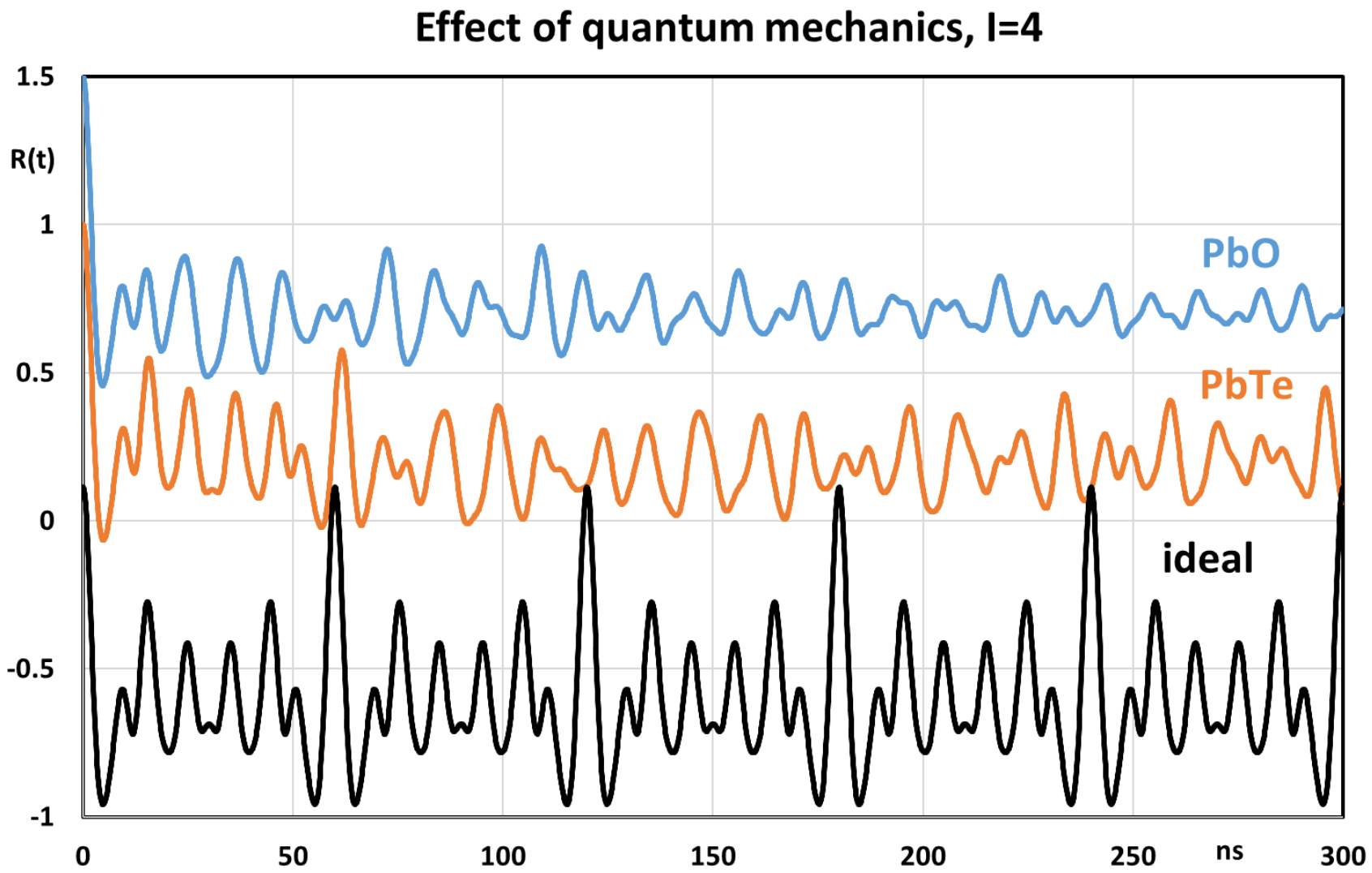
The proven technique



The new high temperature technique



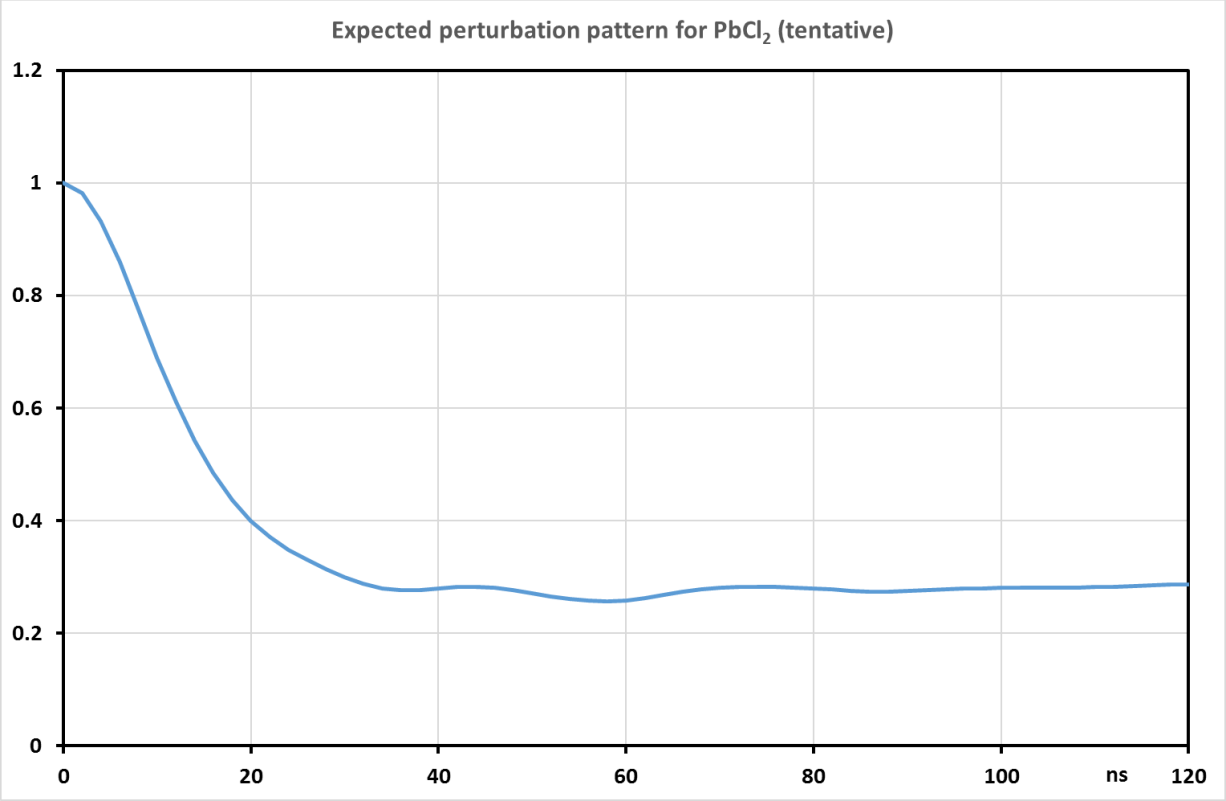

Effect of quantum mechanics, l=4
1.5
R(t)
1
0.5
0
-0.5
-1
0
50
100
150
200
250
ns
300
PbTe
ideal


Effect of quantum mechanics, l=4
1.5
R(t)
1
0.5
0
-0.5
-1
0
50
100
150
200
250
ns
300
PbO
PbTe
ideal

Test case 1: Non-linear molecule $^{204}PbCl_2$

Special interest: **Testing possible application to more cases,**
Checking complex treatment of perturbation function

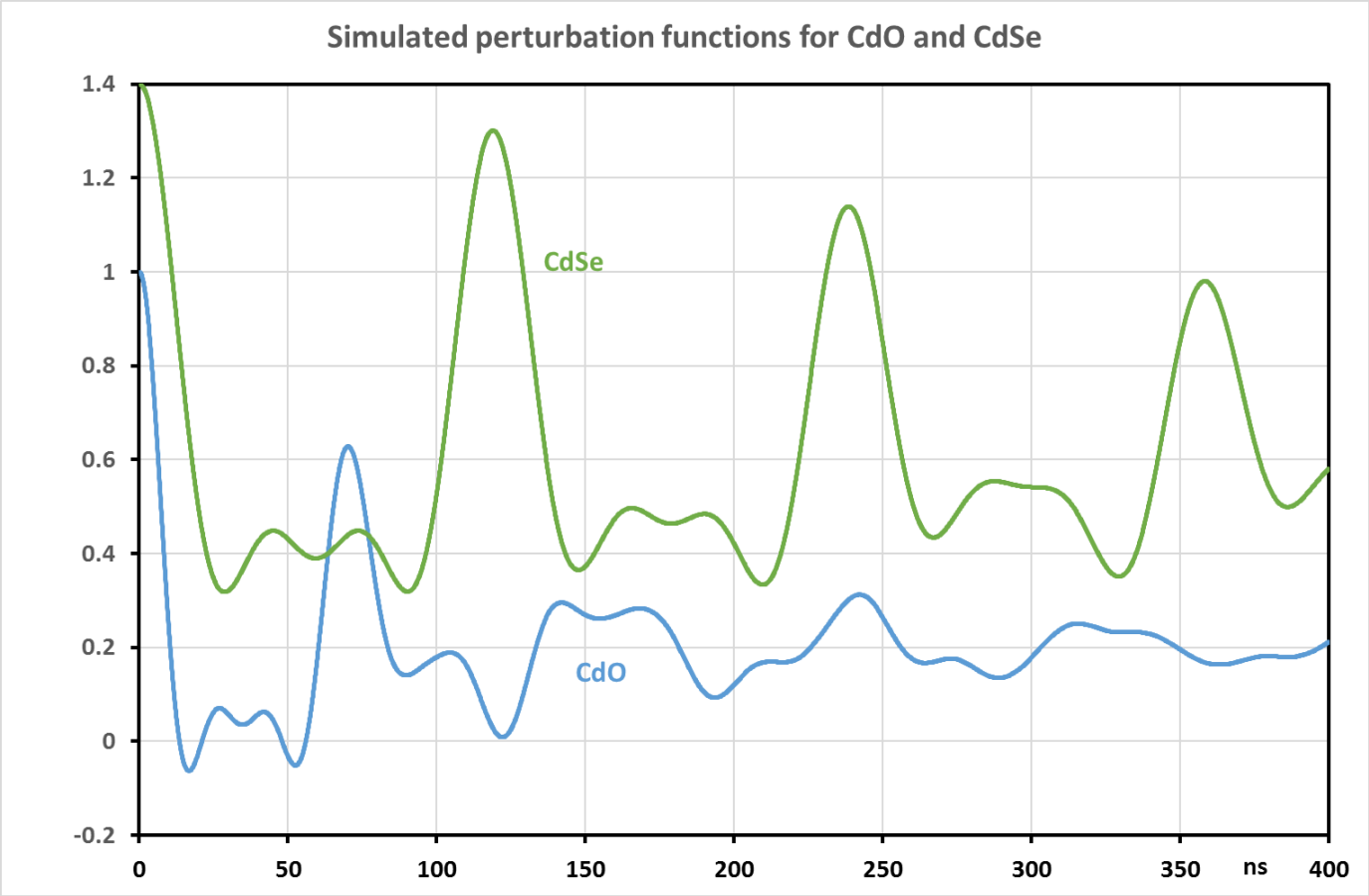
Difficulty: **Absence of periodic structure,**
Obtaining sufficient statistical accuracy



Test case 2: Weakly bound molecule ^{111}CdSe

Special interest: Checking quantum chemistry calculation for small EFG, Testing new sample production method

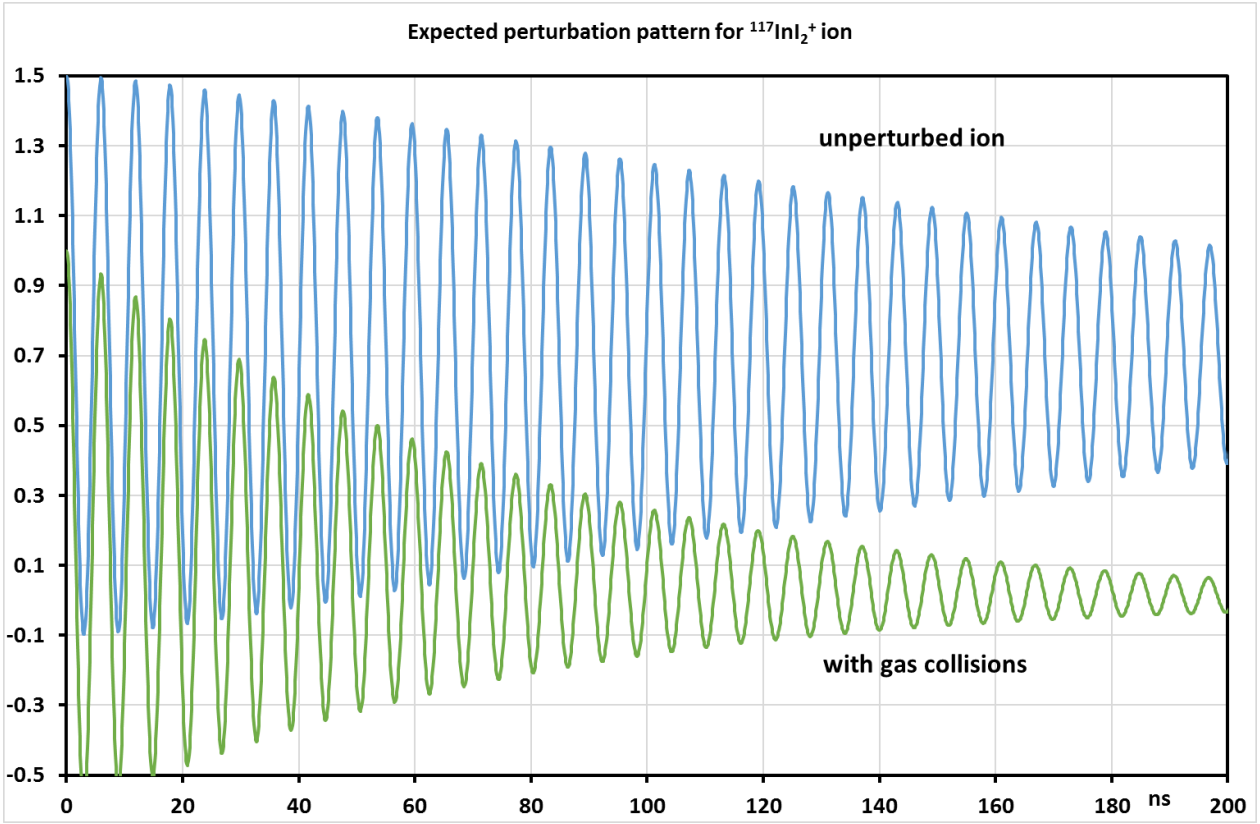
Difficulty: High measuring temperature, low molecule stability



Test case 3: Molecular ion $^{117}InI_2^+$

Special interest: Checking use for β^- produced ionized molecules, Extending applications to further cases (Sn «missing link»?)

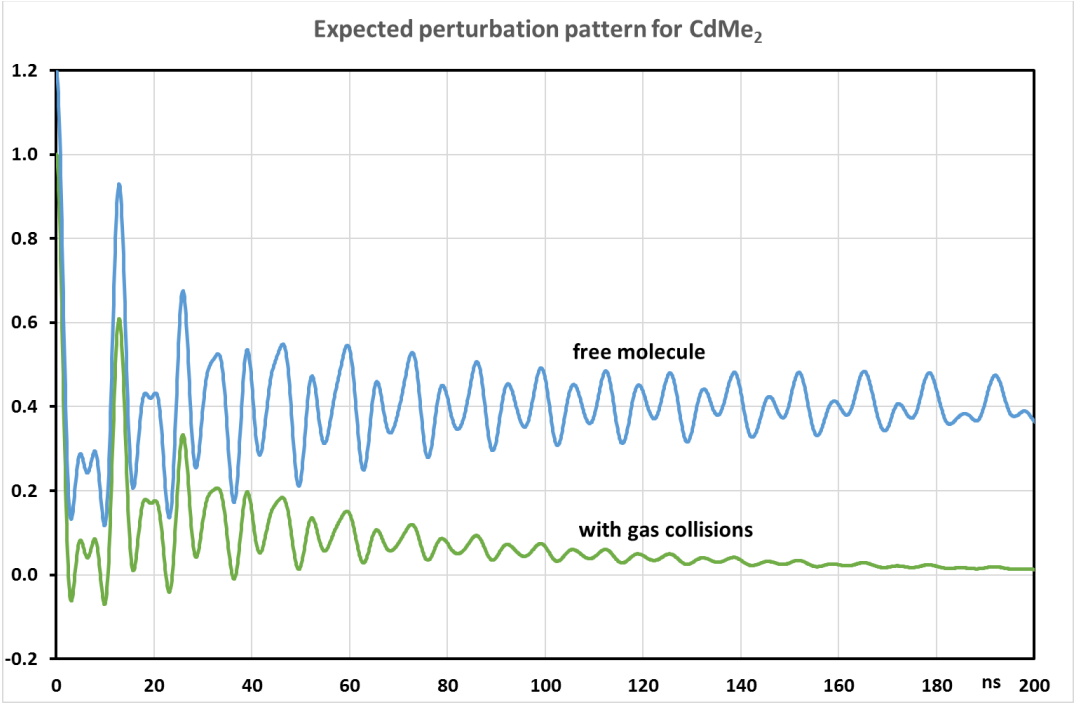
Difficulty: High expected interaction frequency, Treatment of beta-recoil in analysis



IS640 completion 1: Measuring ^{111}Cd-dimethyl molecule

Special interest: **Measuring EFG shift for weakly interacting molecules in solid, checking our published estimate**
Measuring quasi-linear molecule

Difficulty: **Sample production via chemistry, high interaction frequency**



IS640 completion 2: Measuring ^{199}Hg-dibromide molecule

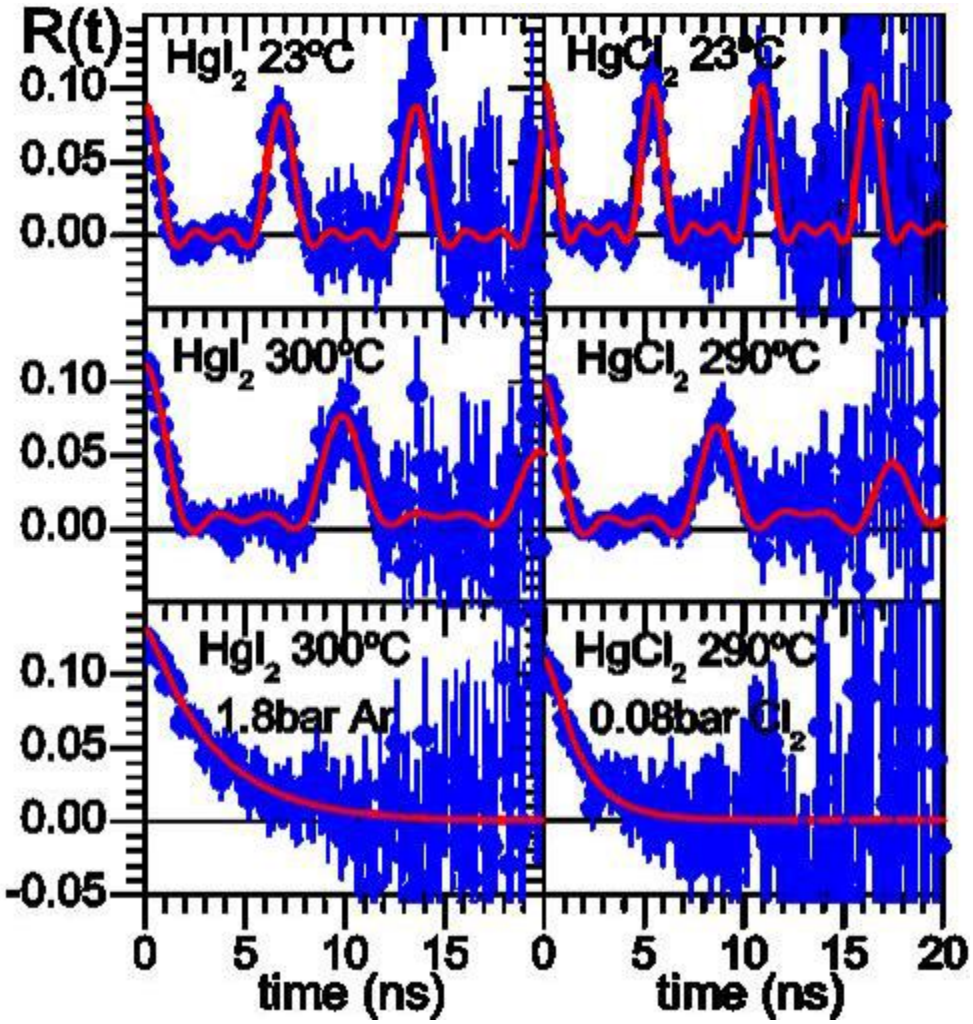
Special interest:

Obtain 2nd case (after $HgCl_2$) for shift of EFG in molecular solid

Testing DFT calculations

Difficulty:

Sample production for volatile molecule by first formation of metal deposit and then reaction with Br_2



Summary

We plan further PAC experiments on free molecules

Primary goal: A precision value for Q of the 4^+ state in ^{204}Pb

Test of novel applications of the technique:

- **to non-linear molecules ($^{204}PbCl_2$)**
- **to unstable molecules ($^{111}CdSe$)**
- **to charged molecules ($^{117}InI_2^+$)**

Complete IS640 studies of Cd and Hg cases:

- **Cd-dimethyl**
- **Hg-dibromide**

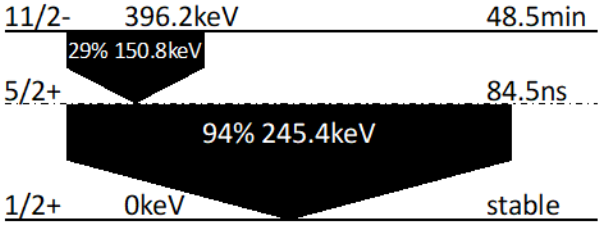
Beam Time Request

Required isotope	Implanted beam	Probe element	Type of experiment	Intensity [at/μC]	Target / Ion source	Atoms per sample	nº of shifts
^{204m}Pb (67 min)	^{204m}Pb	^{204}Pb	γ-γ PAC	10^8	UCx/ RILIS	2×10^{10}	1 x 4
^{111m}Cd (48 min)	^{111m}Cd	^{111}Cd	γ-γ PAC	10^9	Molten Sn/ VADIS	2×10^{10}	2 x 1
^{117}Cd (150 min)	^{117}Ag	^{117}In	γ-γ PAC	10^8	UCx/ RILIS	5×10^{10}	1 x 1
^{199m}Hg (43 min)	^{199m}Hg	^{199}Hg	γ-γ PAC	10^9	Molten Pb/ VADIS	2×10^{10}	1 x 1

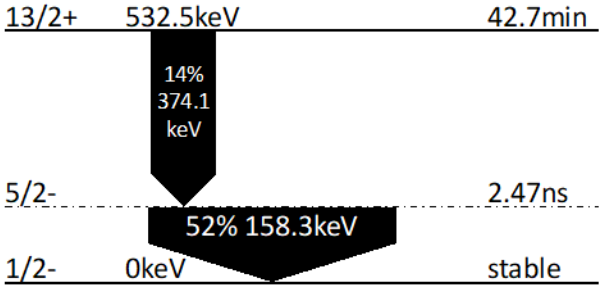
Slides for discussion

Problems with previous calibration of Q via B(E2)

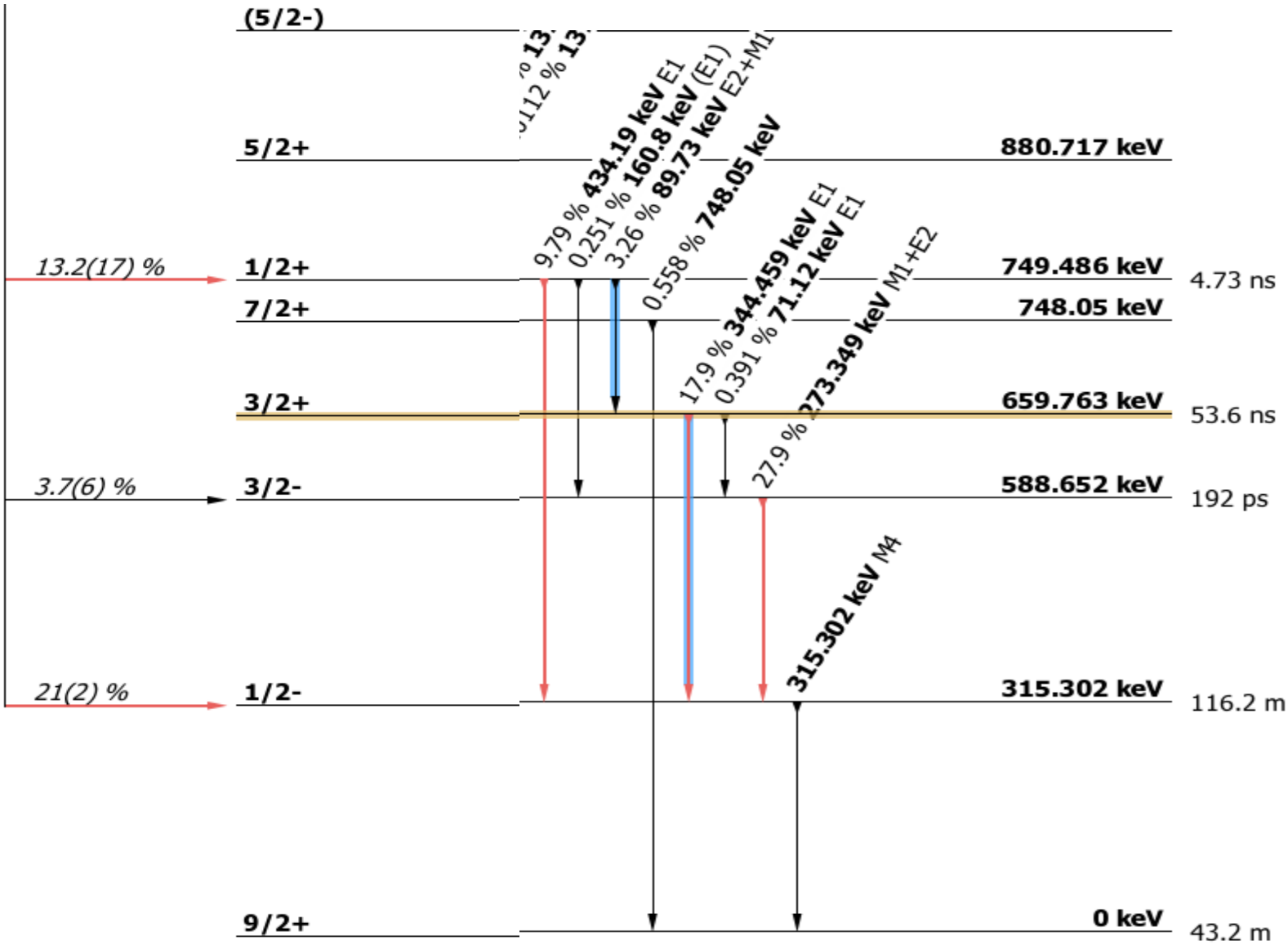
1. Assuming pure wavefunctions for 12^+ and 10^+
2. Accuracy of $t_{1/2}$
3. Correction for branch feeding


11/2-
396.2keV
48.5min
29% 150.8keV
5/2+
84.5ns
94% 245.4keV
1/2+
0keV
stable

^{111}Cd


13/2+
532.5keV
42.7min
14%
374.1
keV
5/2-
2.47ns
52% 158.3keV
1/2-
0keV
stable

^{199}Hg


(5/2-)
5/2+
880.717 keV
13.2(17) %
1/2+
749.486 keV
4.73 ns
7/2+
748.05 keV
3/2+
659.763 keV
53.6 ns
3.7(6) %
3/2-
588.652 keV
192 ps
21(2) %
1/2-
315.302 keV
116.2 m
9/2+
0 keV
43.2 m
9.79 % 434.19 keV E1
0.251 % 160.8 keV (E1)
3.26 % 89.73 keV E2+M1
0.558 % 748.05 keV
17.9 % 344.459 keV E1
0.391 % 71.12 keV E1
27.9 % 273.349 keV M1+E2
315.302 keV M4