The search for «missing links» of nuclear quadrupole moments – A status report

Heinz Haas, University of Aveiro, Portugal, and EP-Division, CERN,

for the IS640, IS673, and IS703 teams

Nuclear moments are determined by measuring their interaction energy/frequency with an external field:

For magnetic moments μ the Larmor frequency
$\nu_L = B \cdot \mu / (I \cdot h)$
with B the external magnetic field, generally well known,
it requires $I > 0$

For quadrupole moments Q the quadrupole coupling constant
$\nu_Q = V_{zz} \cdot Q \cdot e / h$
with V_{zz} the electric field gradient (efg) at the nucleus, to be obtained by theory,
it requires $I > 1/2$

Methods for quadrupole moment determination

For long-lived states >10 ms
Atomic spectroscopy
For stable isotopes also:
Molecular spectroscopy

in
atoms or simple molecules

Accuracy of efg theory
1% in most cases

For short-lived states < 1 ms
Mössbauer spectroscopy ME
Perturbed angular correlation PAC
Perturbed angular distribution PAD

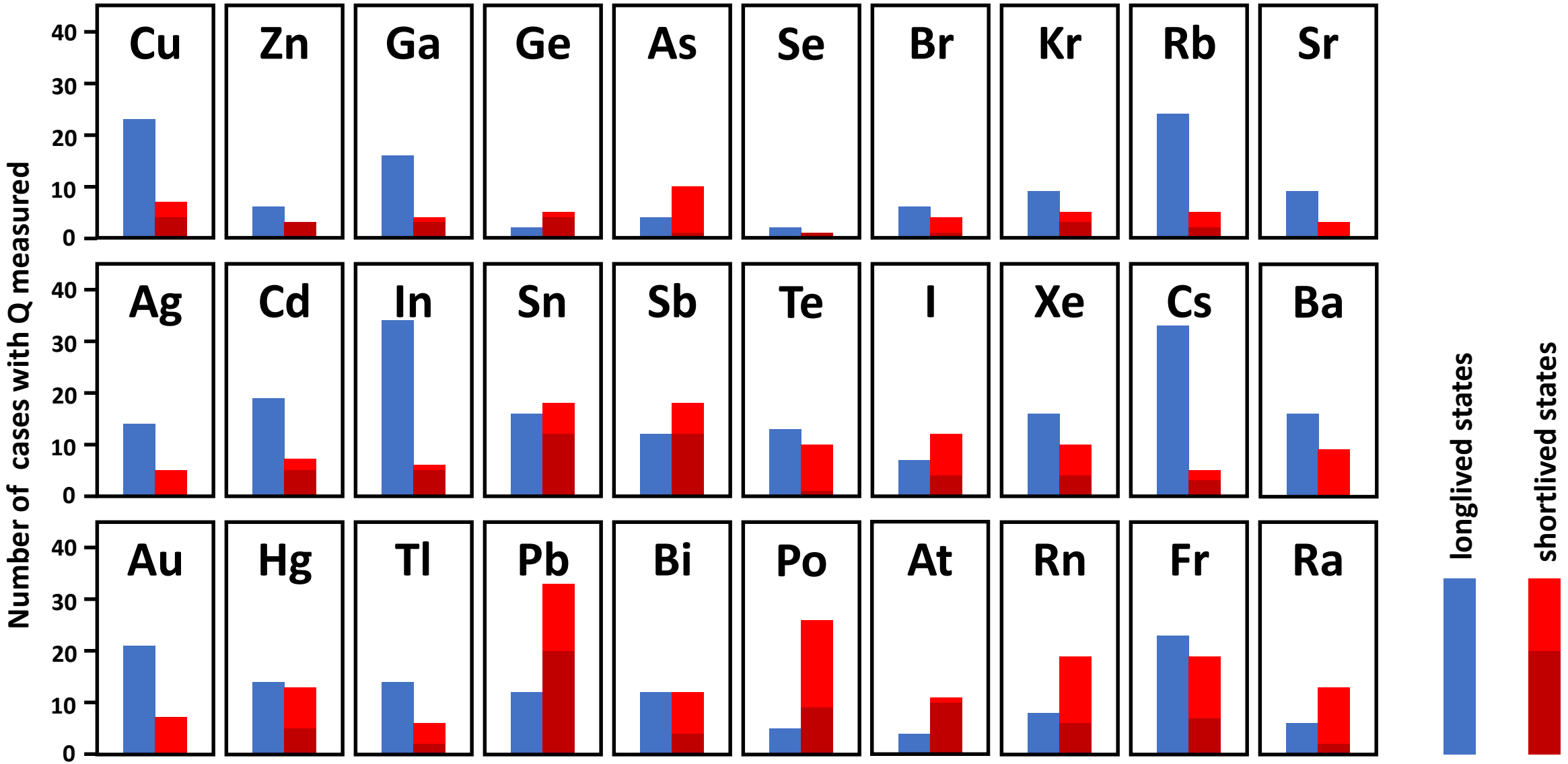
in
condensed matter, generally metals

Accuracy of efg theory
10-20%

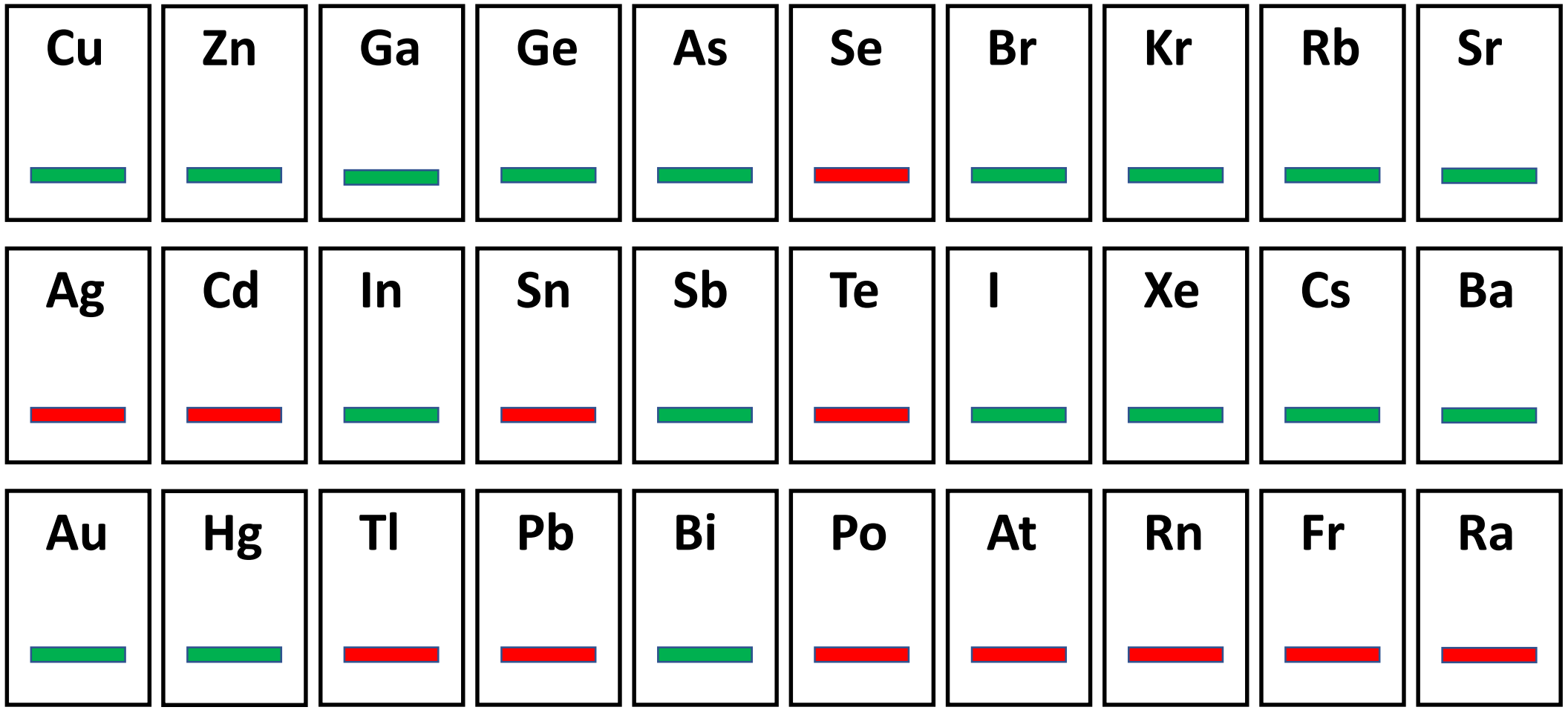
Linking the two areas by
Nuclear magnetic resonance NMR
Nuclear quadrupole resonance NQR
Mössbauer spectroscopy ME

Only possible for stable isotopes !!

The extend of the problem



Elements with / without stable I > 1/2 isotopes



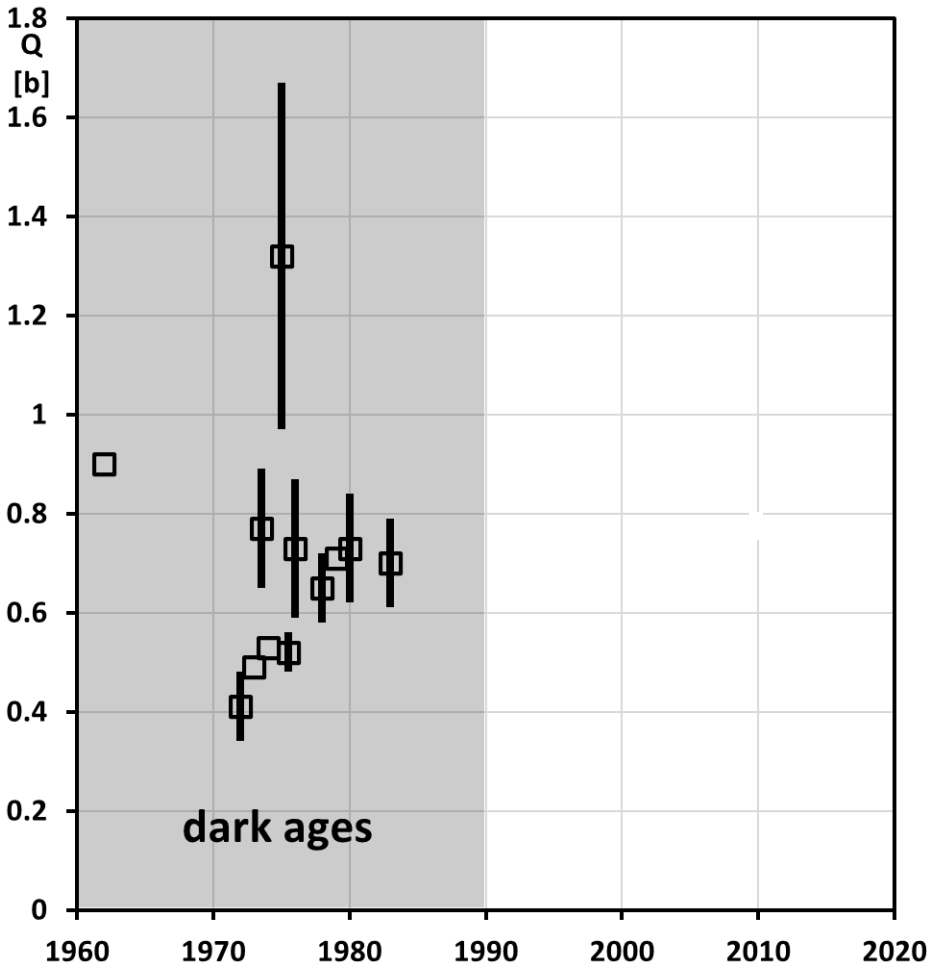
The «easy» cases

Cu NQR	Zn ME	Ga NQR	Ge ?	As NQR	Se	Br NQR	Kr ME	Rb NMR	Sr ?
Ag	Cd	In NQR	Sn	Sb NQR	Te	I NQR	Xe ?	Cs NMR	Ba ?
Au ME	Hg NQR	Tl	Pb	Bi NQR	Po	At	Rn	Fr	Ra

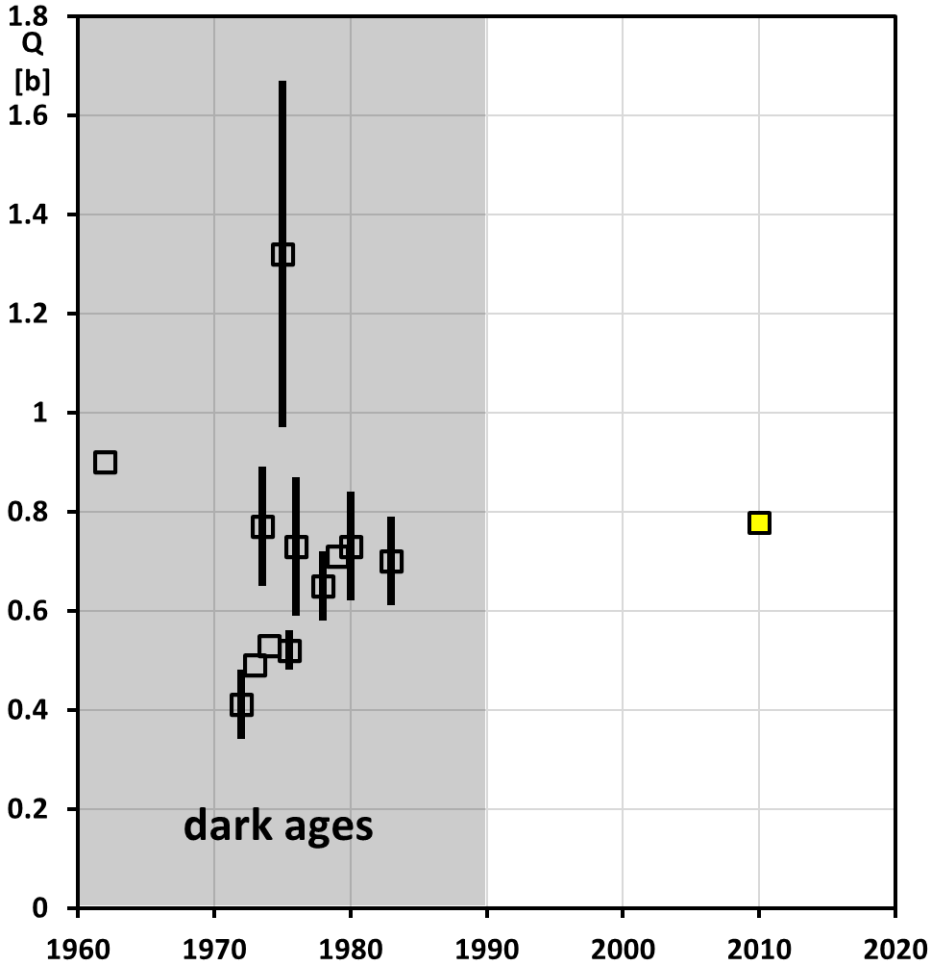
The problem cases

Cu	Zn IS640	Ga	Ge	As	Se	Br	Kr	Rb	Sr
Ag	Cd IS640	In	Sn IS673	Sb	Te	I	Xe	Cs	Ba
Au	Hg IS640	Tl	Pb IS703	Bi	Po	At	Rn	Fr	Ra

History of Q for ^{111}Cd 5/2^{+}



History of Q for ^{111}Cd 5/2^{+}



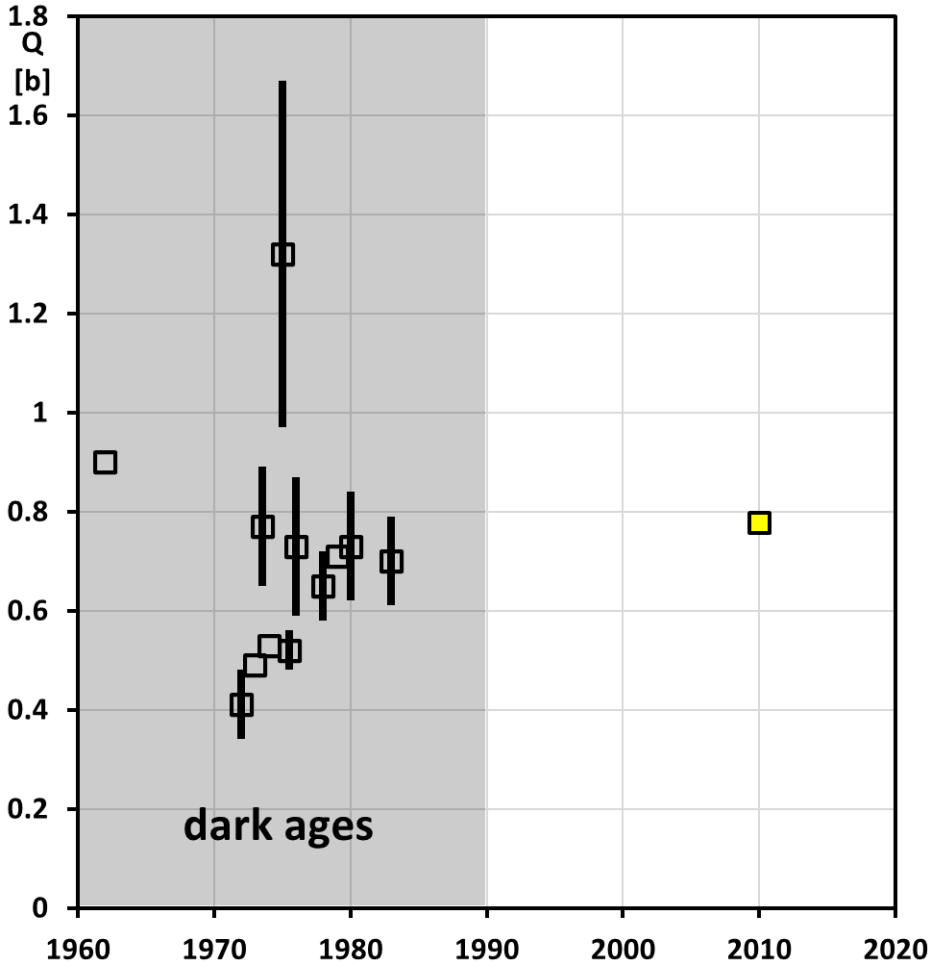
Hyperfine Interact (2010) 198:133–137

The quadrupole moments of Zn and Cd isotopes—an update

H. Haas · J. G. Correia

Standard DFT

History of Q for ^{111}Cd 5/2^{+}



Hyperfine Interact (2010) 198:133–137

The quadrupole moments of Zn and Cd isotopes—an update

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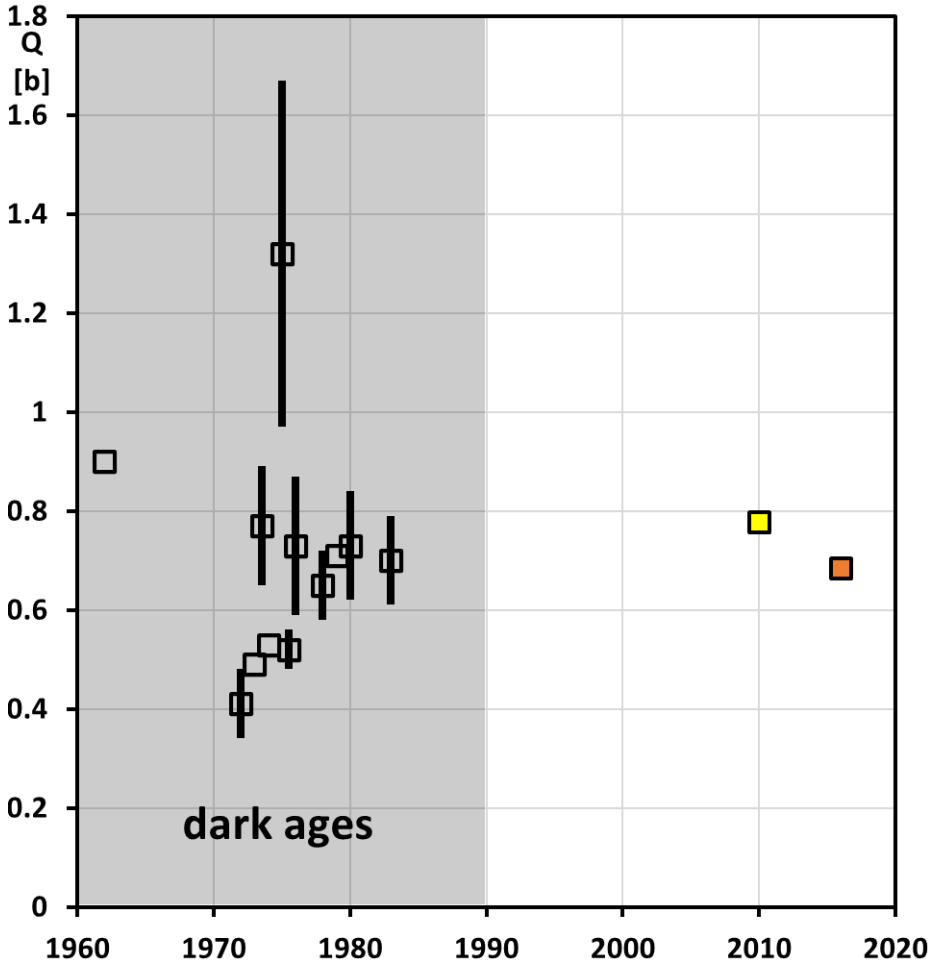
Standard DFT

But: DFT is not really a theory,
just a technique, so try another one!

History of Q for ^{111}Cd 5/2^{+}



Hyperfine Interact (2010) 198:133–137

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H. Haas · J. G. Correia

Standard DFT

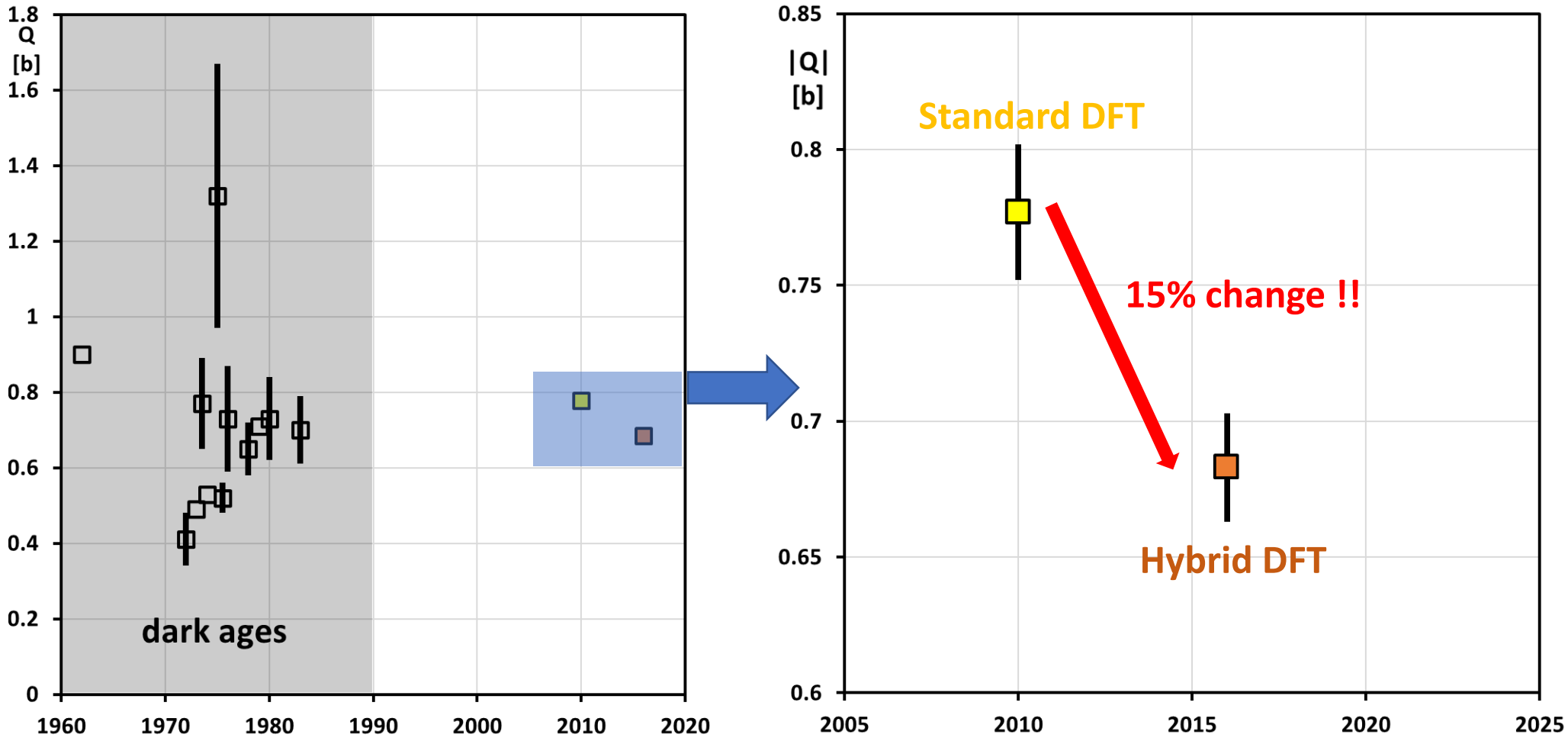
Hyperfine Interact (2016) 237:115

The quadrupole moments of Cd and Zn isotopes - an apology

H. Haas[1,4] · M. B. Barbosa[2] · J. G. Correia[3,4]

Hybrid DFT

Detail of Q for ^{111}Cd 5/2^{+}



What can we do better?

Calculate the efg in a simple molecule $Cd(CH_3)_2$ with quantum chemistry techniques and extract Q for ^{111}Cd $5/2^+$, using a 50 years old PAC result (in molecular solid)

EPL, **117** (2017) 62001

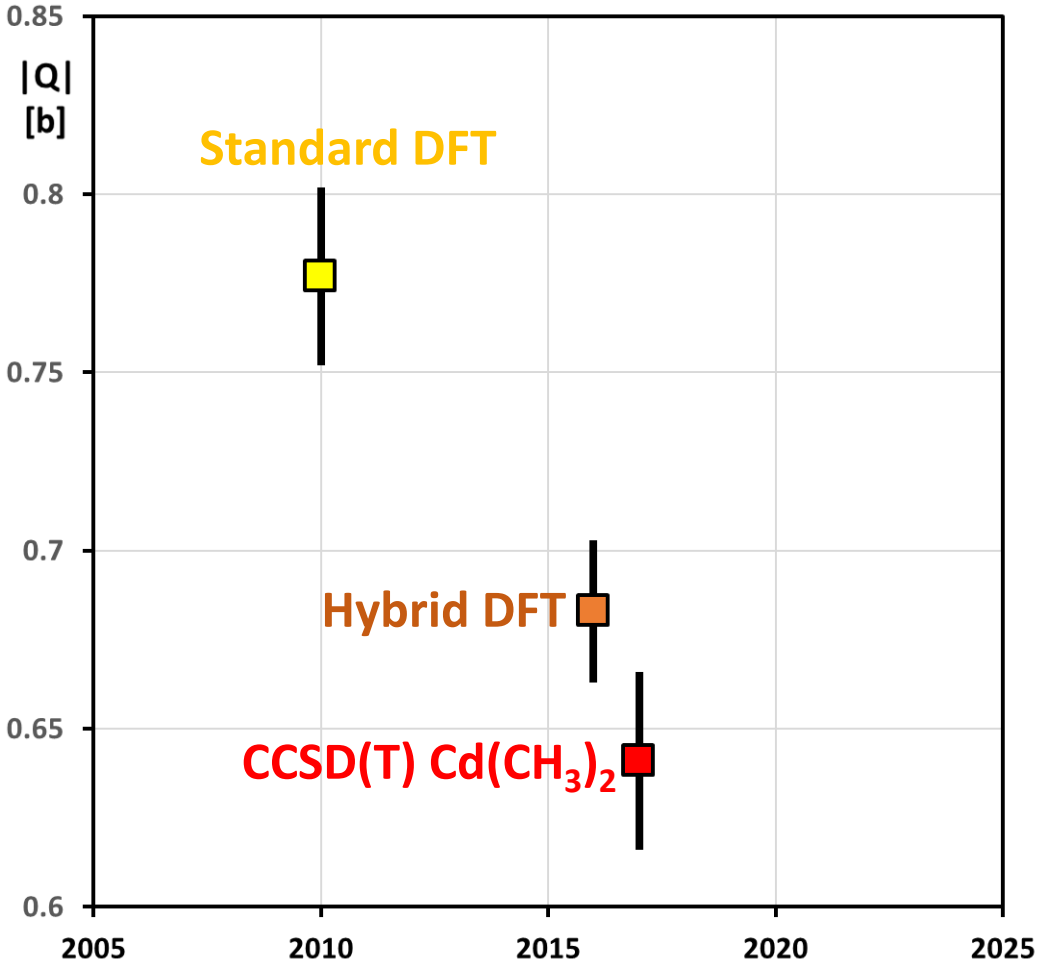
Quadrupole moments of Cd and Zn nuclei: When solid-state, molecular, atomic, and nuclear theory meet

H. HAAS[1,5], S. P. A. SAUER[2], L. HEMMINGSEN[2], V. KELLÖ[3] and P. W. ZHAO[4]

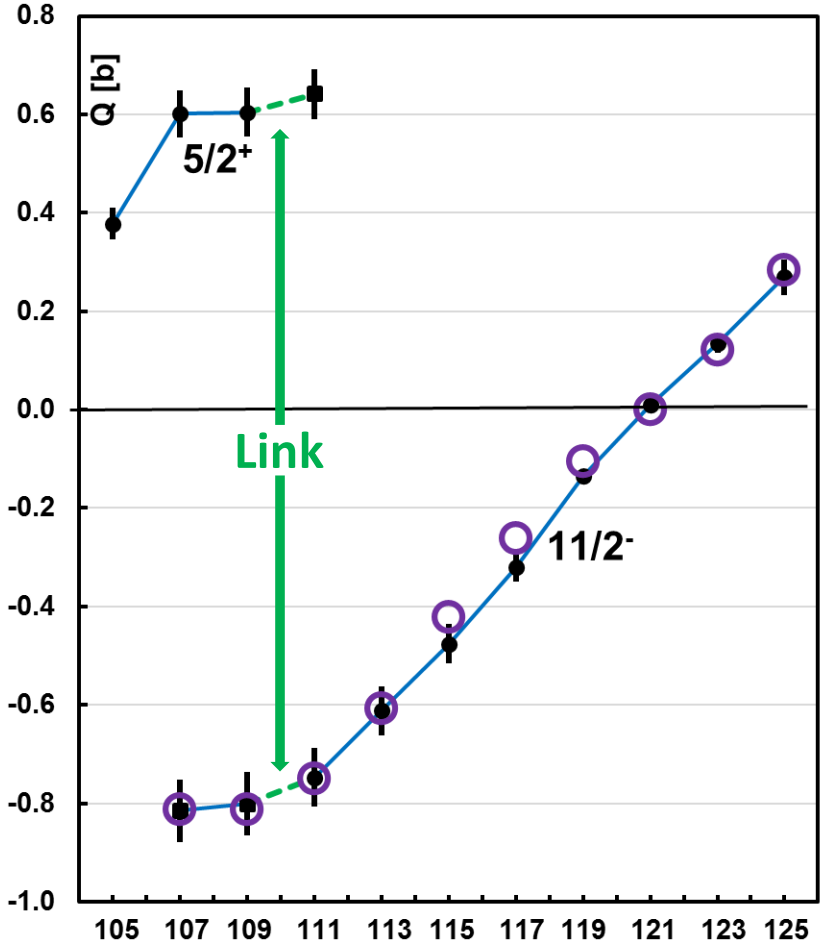
Extended the series of Q for $11/2^-$ states in Cd by combining ISOLDE laser spectroscopy results with old PAC and PAD data

Confirmed the saturation of Q at ^{109}Cd predicted by nuclear covariant density functional calculations

Result: Q for ^{111}Cd $5/2^+$



Nuclear theory comparison



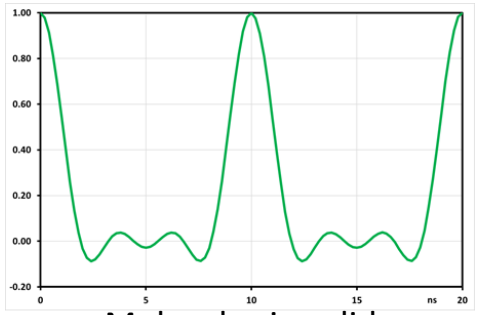
Can we do better yet? Project IS640

- Measure the quadrupole interaction in some free Cd (and Hg) molecules in the gas state by PAC
- Basic concept: 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 component 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 !
- Our 50 years old idea, but early experiments (Berkeley, Bonn) in the 1970s have failed

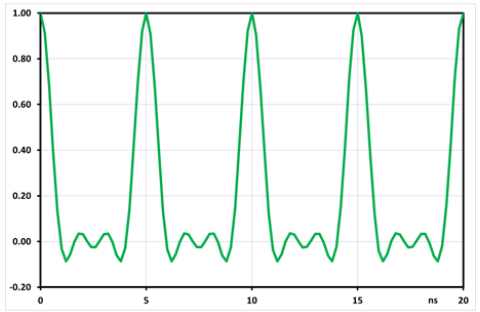
Test of concept with ^{199}Hg 5/2^{-}

What R(t) could we get ?

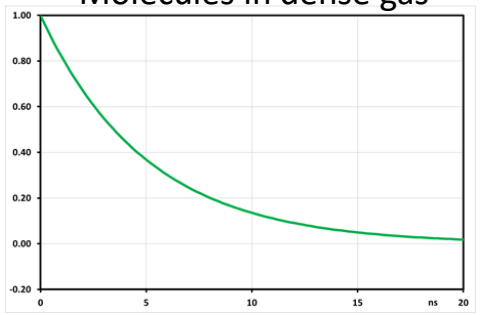
Free molecules in gas



Molecules in solid



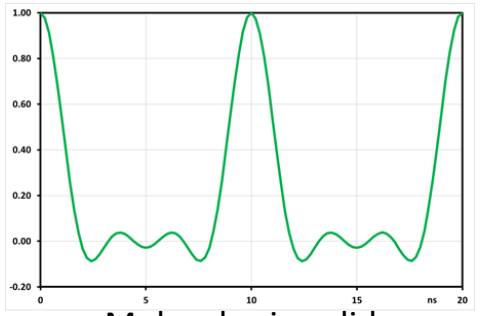
Molecules in dense gas



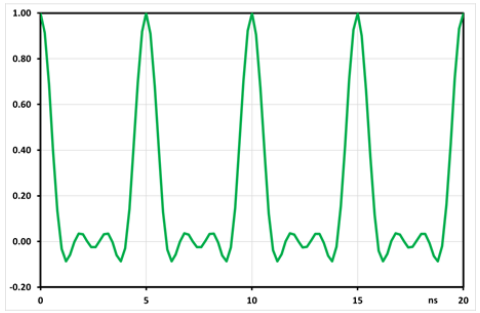
Test of concept with ^{199}Hg 5/2^{-}

What R(t) could we get ?

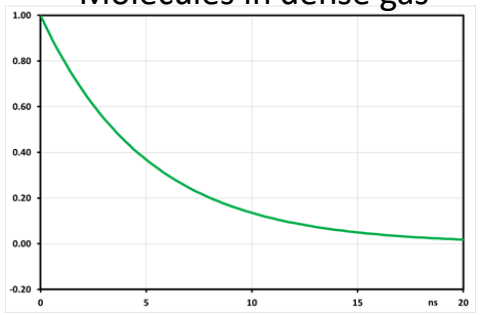
Free molecules in gas



Molecules in solid

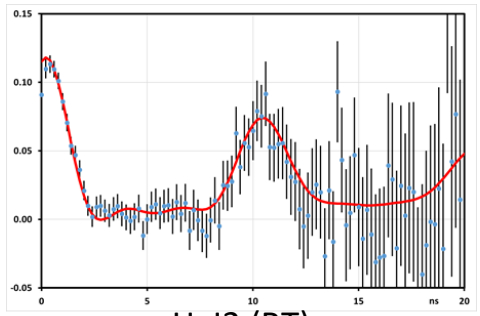


Molecules in dense gas

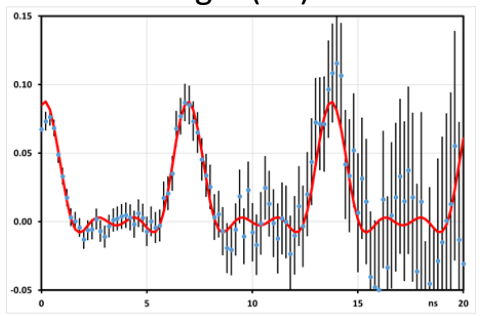


What R(t) did we see ?

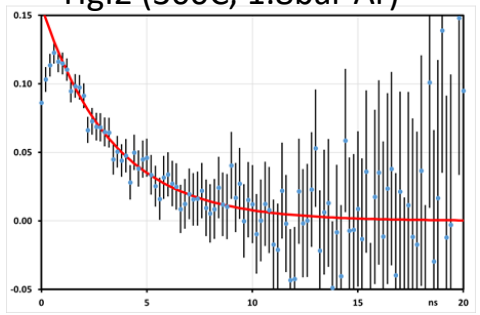
HgI2 (300C)



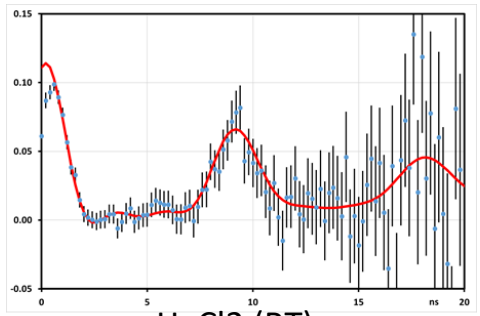
HgI2 (RT)



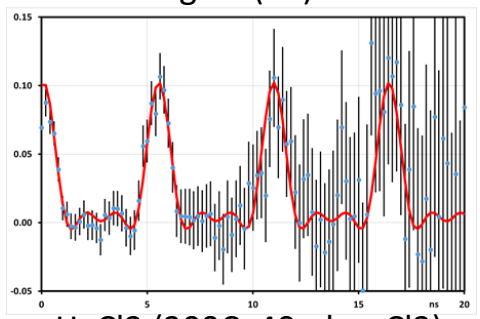
HgI2 (300C, 1.8bar Ar)



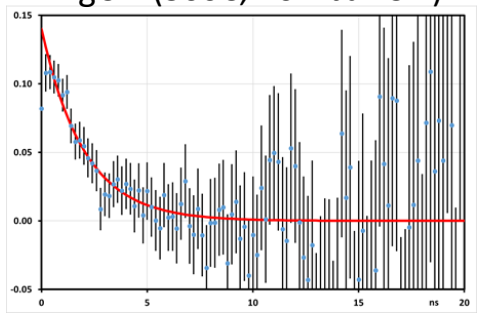
HgCl2 (300C, 2mbar Cl2)



HgCl2 (RT)



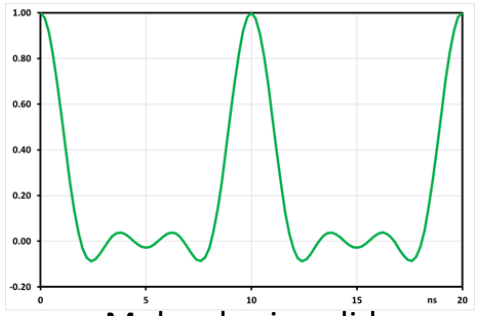
HgCl2 (300C, 40mbar Cl2)



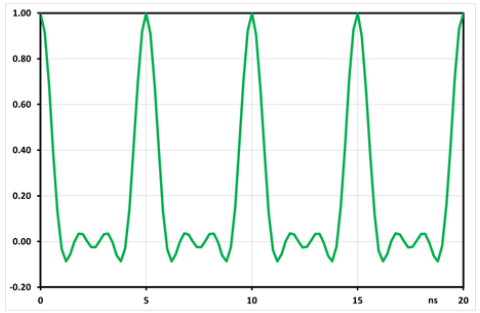
Test of concept with ^{199}Hg $5/2^-$

What R(t) could we get ?

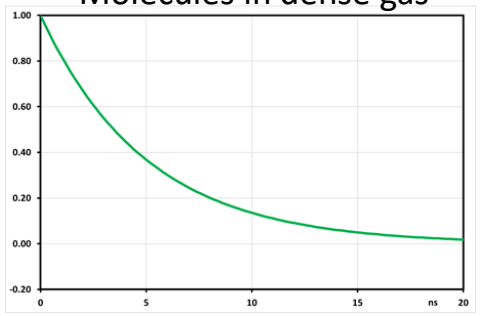
Free molecules in gas



Molecules in solid



Molecules in dense gas



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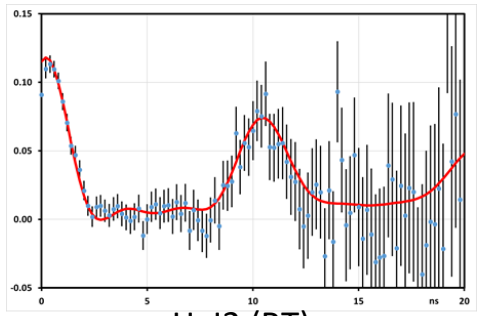
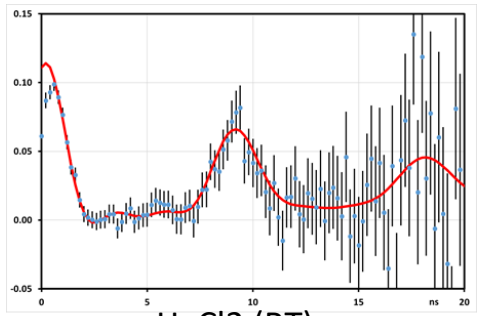
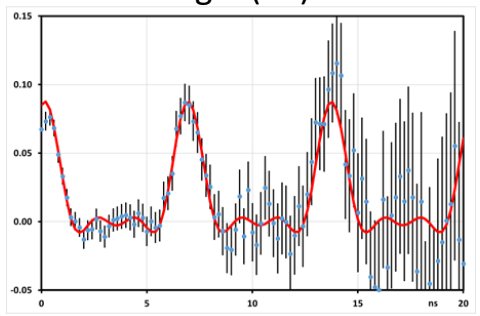


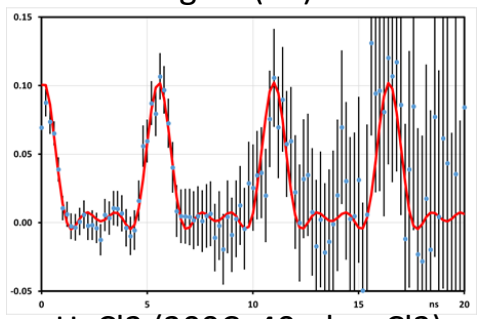
HgCl2 (300C, 2mbar Cl2)



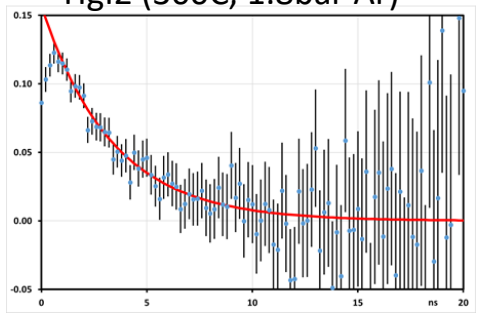
HgI2 (RT)



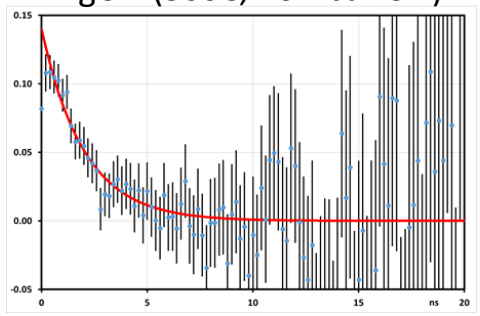
HgCl2 (RT)



HgI2 (300C, 1.8bar Ar)



HgCl2 (300C, 40mbar Cl2)



Results:

Idea confirmed

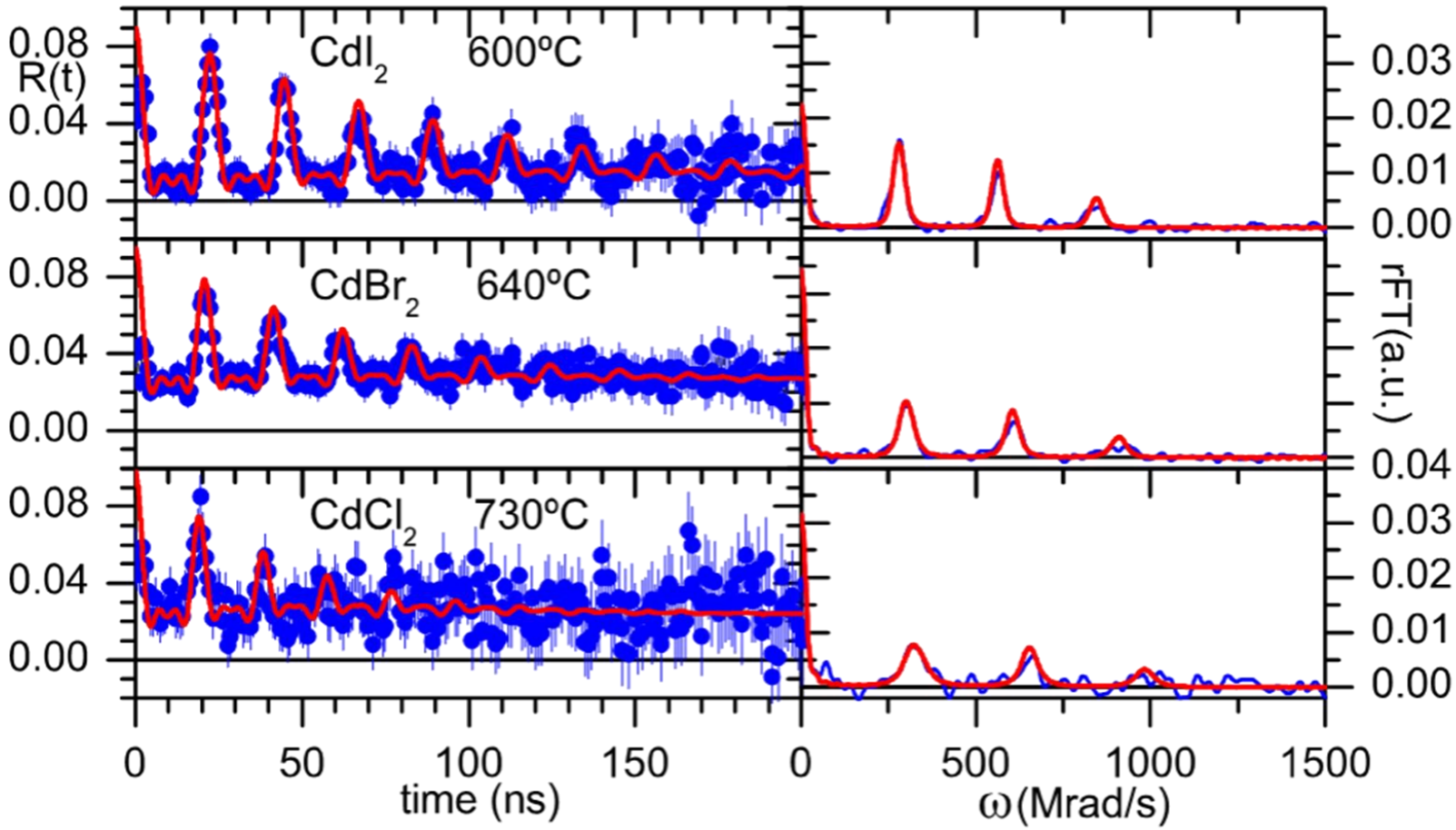
Known Q redetermined

Effect of inter-molecular bond measured

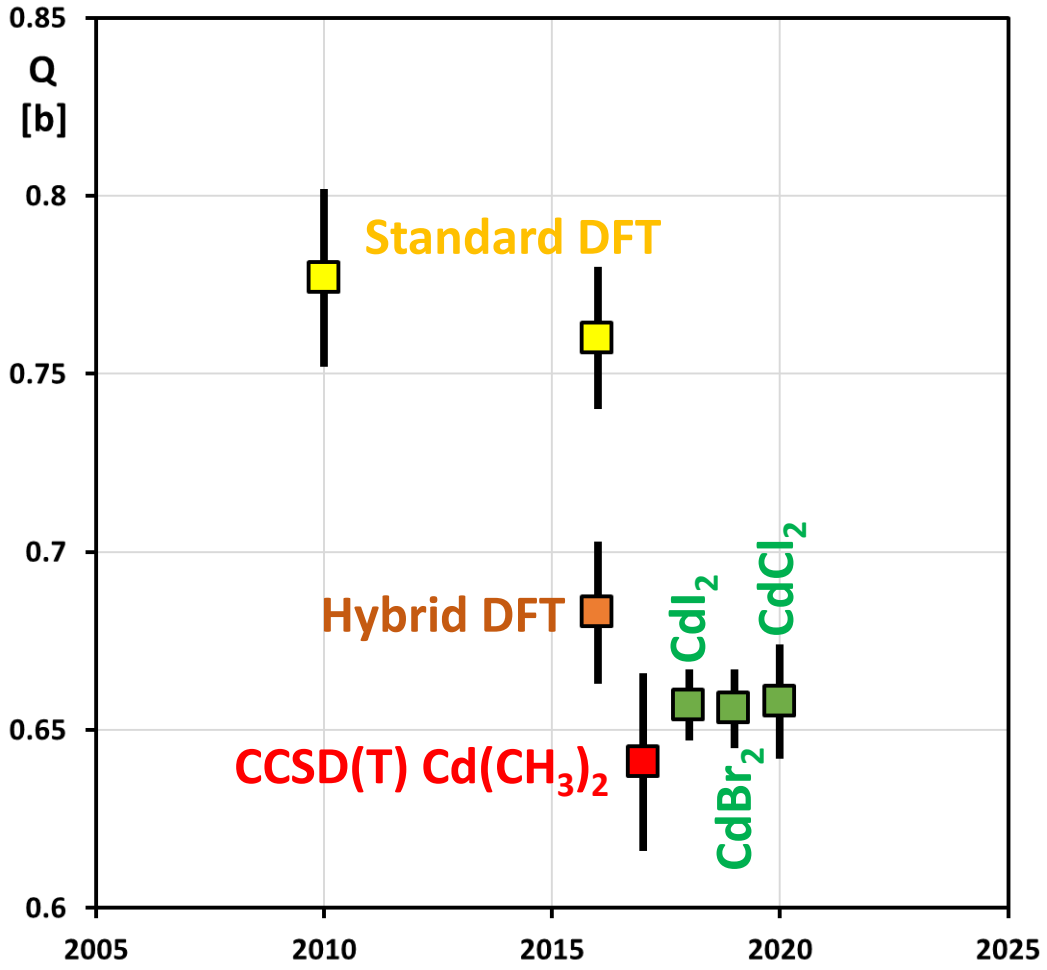
PHYSICAL REVIEW LETTERS **126,** 103001 (2021)

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]

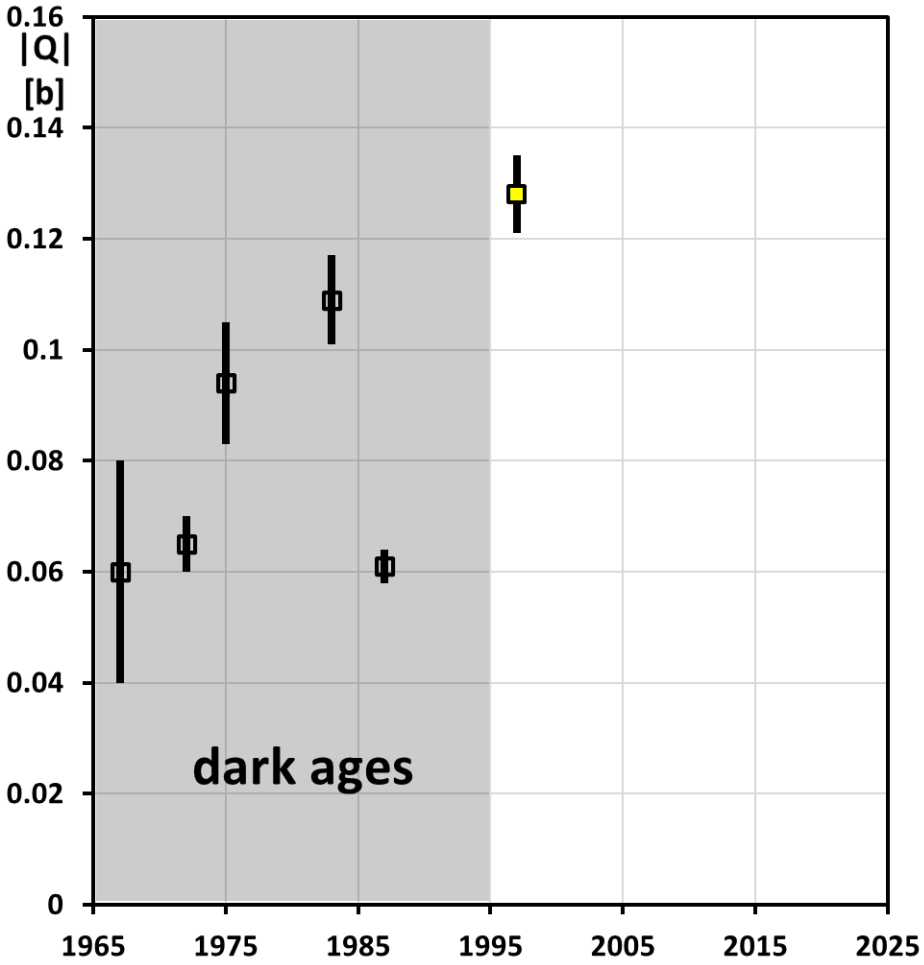


IS640: Final results for Q of ^{111}Cd 5/2^{+}

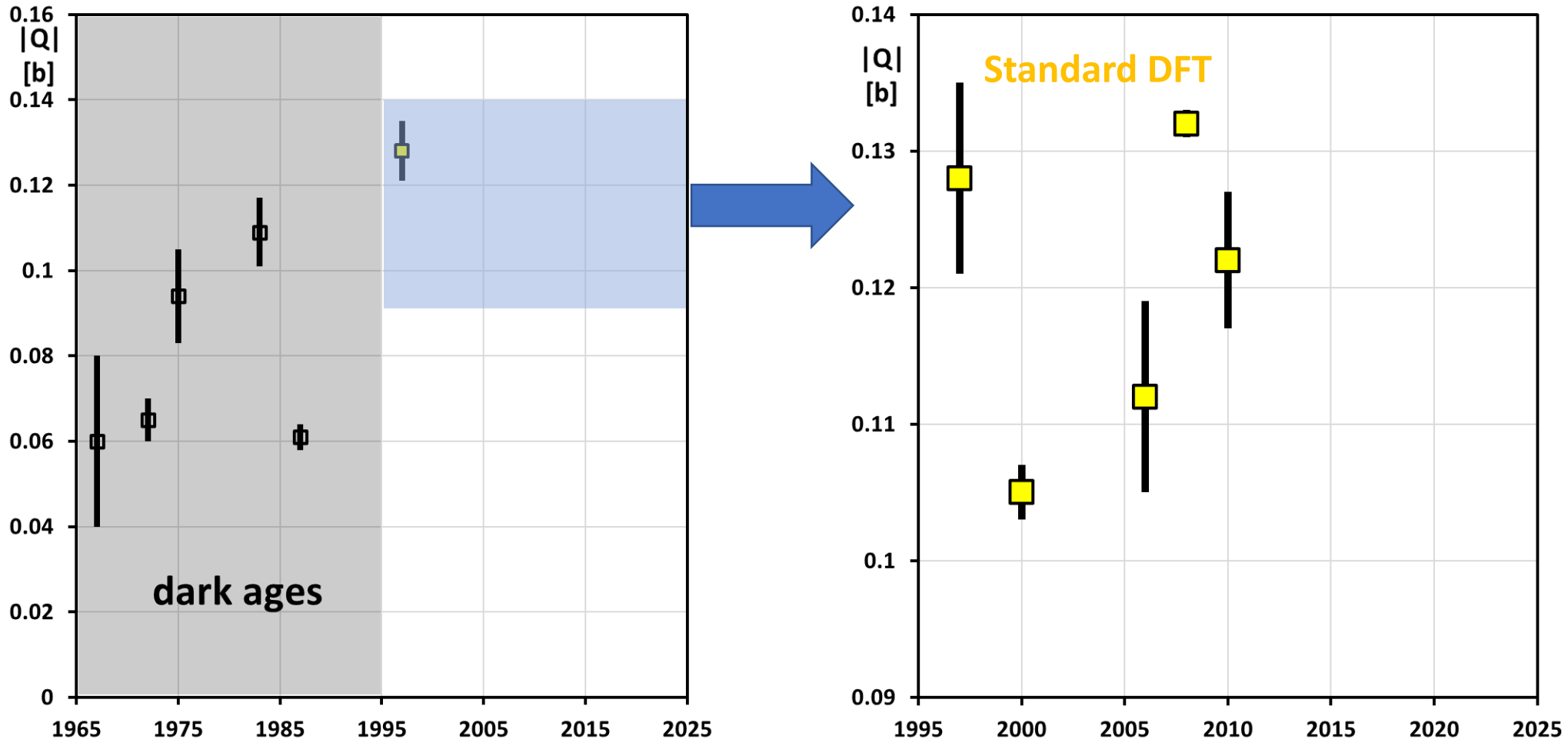


Accuracy achieved 1.2%

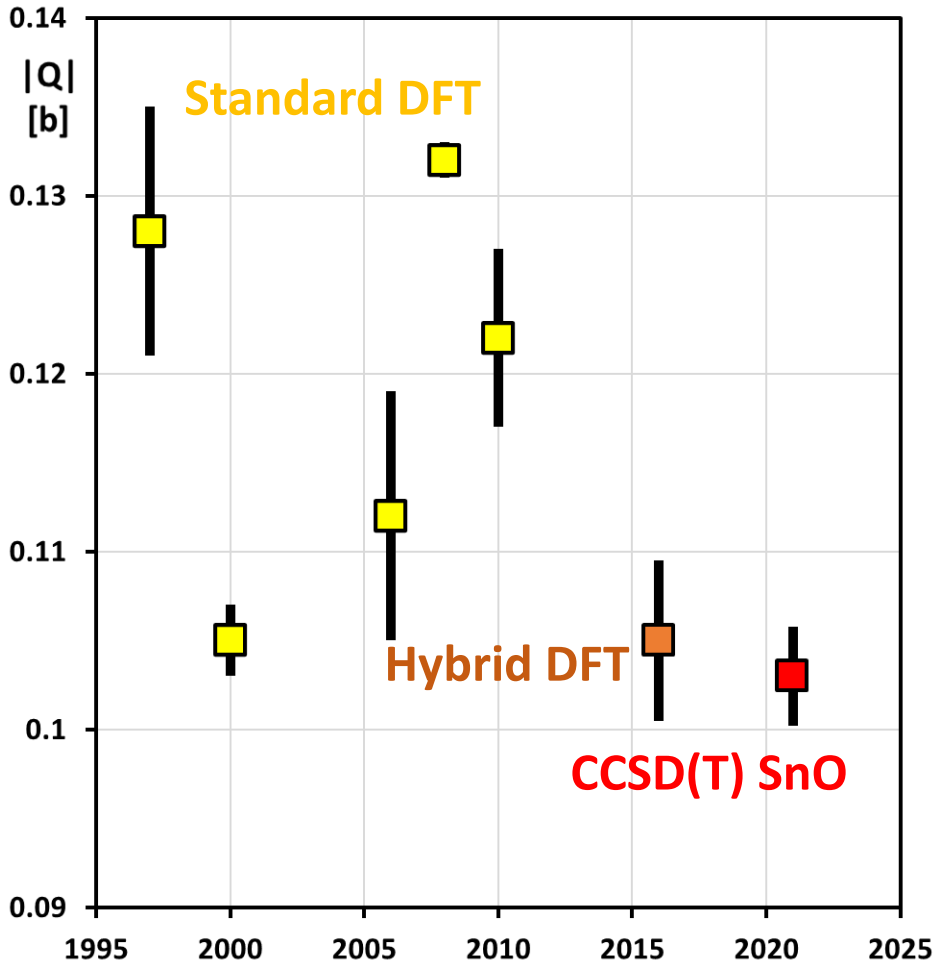
History of Q for ^{119}Sn 3/2^{+}



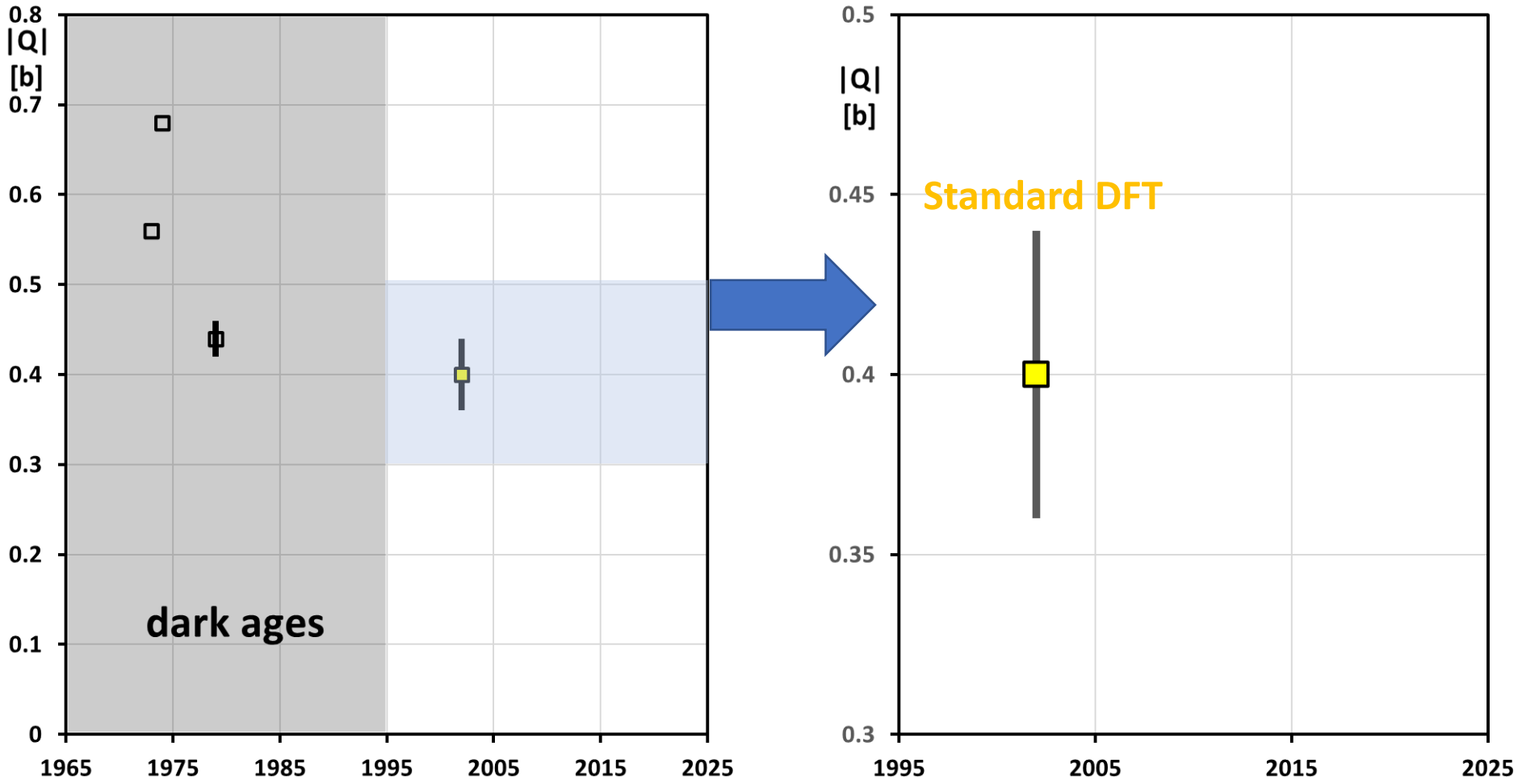
Detail of Q for ^{119}Sn 3/2^{+}



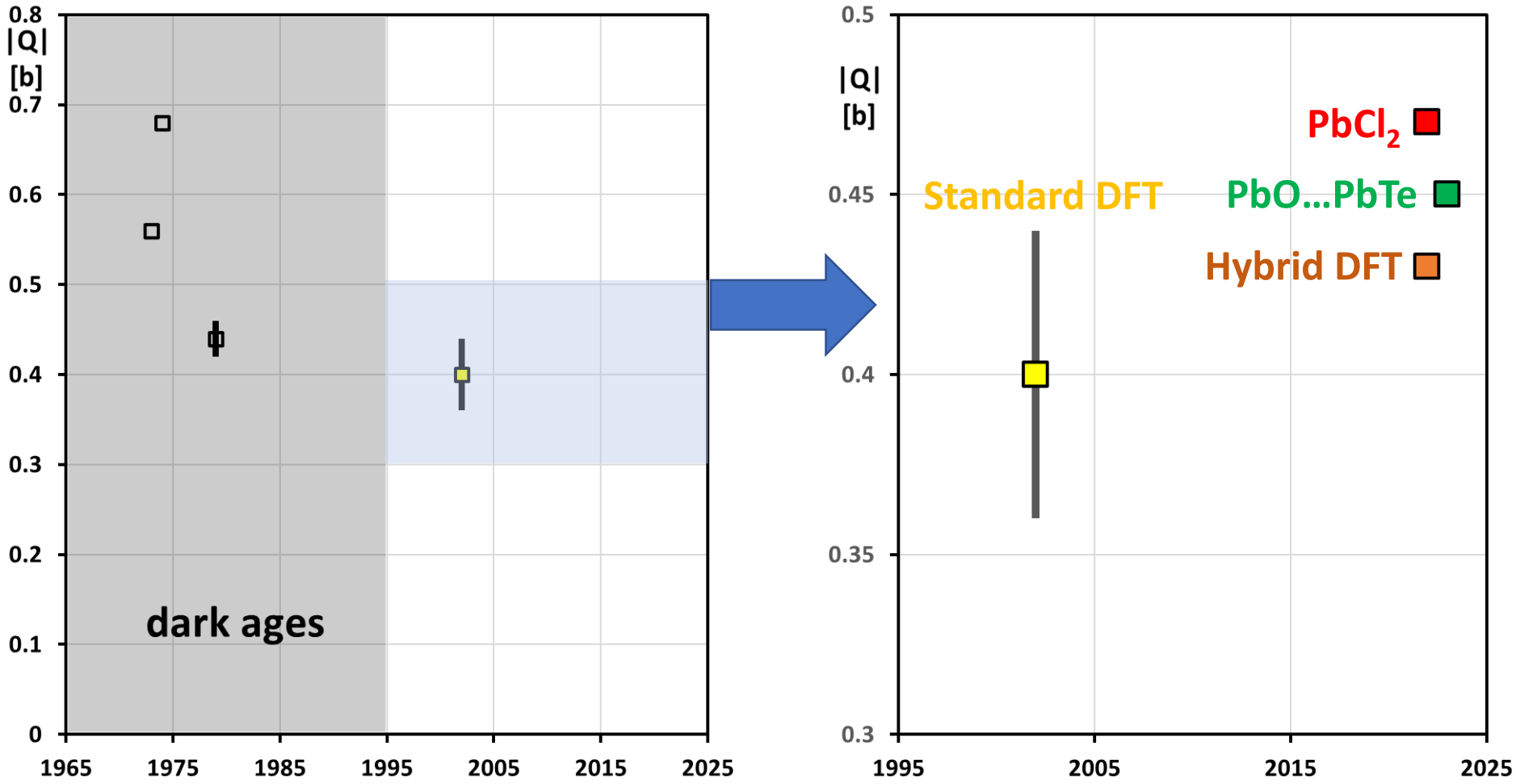
IS673: Results for Q of ^{119}Sn $3/2^+$ (preliminary)



History of Q for $^{204}Pb\ 4^{+}$



Project IS703 for Q of $^{204}Pb\ 4^{+}$



Project IS703 for Q of $^{204}Pb\ 4^{+}$