

# AMO in the keV regime

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**In many ways, this meeting, along with many others, is a celebration of the tremendously successful employment of the interaction of EM waves with atoms.**

**Many precision measurements in physics and technological breakthroughs have derived from it.**

***We want to explore the relatively undeveloped field of EM waves interaction with nuclei.***

***Why?***

***“Because it’s there” and is potentially very different (from the atomic case) may actually be a good answer.***

**But one can already identify at least three reasons why this may lead to substantial progress:**

- Nuclei are electromagnetically isolated from the external environment:  
(more later), yet, they are sensitive to other external interactions, e.g. gravitational.
- One can access nuclear interactions (“Strong” and “Weak” interactions) that are not commonly exploited in technology (except for neutron scattering or nuclear energy).
- There is a fantastic range of lifetimes and line widths that become available, down to relative line widths  $<10^{-25}$ .

**So, why is this area underdeveloped? Mainly because:**

- Cross sections are very small.
- Energies are in the  $>keV$  regime\*, so Doppler shift from recoils is large and high resolution only possible with the Mossbauer technique.
- Energies are in the  $>keV$  regime\*, hence somewhat awkward. However, this is changing rapidly with the advent of synchrotron light sources, and, anyway, there is a hint of a circular argument here!  
Maybe more development in the area will provide handles to deal with keV photons.

\* With some notable exceptions, e.g.  $^{229m}\text{Th}$  transition that is investigated for clocks

## Recoils in atomic and nuclear spectroscopy

In atomic physics, we are used to the fact that a photon emitted by an atom can be re-absorbed by another atom of the same species:



This only works if the relative linewidth  $\frac{\Gamma}{E_\gamma} > \frac{\Delta E}{Mc^2}$  ( $M$  atomic mass)

...which is generally the case in atomic physics but not in nuclear physics.

However, recoilless, ultra-sharp lines are observed in the Mössbauer effect

→ the entire crystal recoils coherently, effectively providing a very large value of  $M$ .

# Mössbauer spectroscopy

Exceedingly narrow linewidths are observed.

Nuclide	$E$ (eV)	$T_{1/2}$	$\Gamma$ (eV)	$\Gamma/E$
$^{57}_{26}\text{Fe}$	14,413	98.3 ns	$4.7 \times 10^{-9}$	$6.4 \times 10^{-13}$
$^{73}_{32}\text{Ge}$	13,328	2.92 $\mu\text{s}$	$1.6 \times 10^{-10}$	$1.2 \times 10^{-14}$
$^{181}_{73}\text{Ta}$	6,237	6.05 $\mu\text{s}$	$7.5 \times 10^{-11}$	$1.2 \times 10^{-14}$
$^{67}_{30}\text{Zn}$	93,300	9.07 $\mu\text{s}$	$5.0 \times 10^{-11}$	$5.4 \times 10^{-16}$
$^{45}_{21}\text{Sc}$	12,400	318 ms	$1.4 \times 10^{-15}$	$1.13 \times 10^{-19}$
$^{107}_{47}\text{Ag}$	93,125	44.3 s	$1.03 \times 10^{-17}$	$1.1 \times 10^{-22}$
$^{103}_{45}\text{Rh}$	39,753	56.1 min	$1.36 \times 10^{-19}$	$3.4 \times 10^{-24}$
$^{189}_{76}\text{Os}$	30,814	5.8 hr	$2.2 \times 10^{-20}$	$7.0 \times 10^{-25}$

$f(T)$  Lamb-Mössbauer factor: the fraction of gammas emitted or absorbed recoillessly

At high temperature ( $T > \theta$ ,  $\theta$  is the Debye temperature)

$$f(T) = \exp\left[-\frac{3E_{\gamma}^2 T}{k_B \theta^2 M c^2}\right]$$

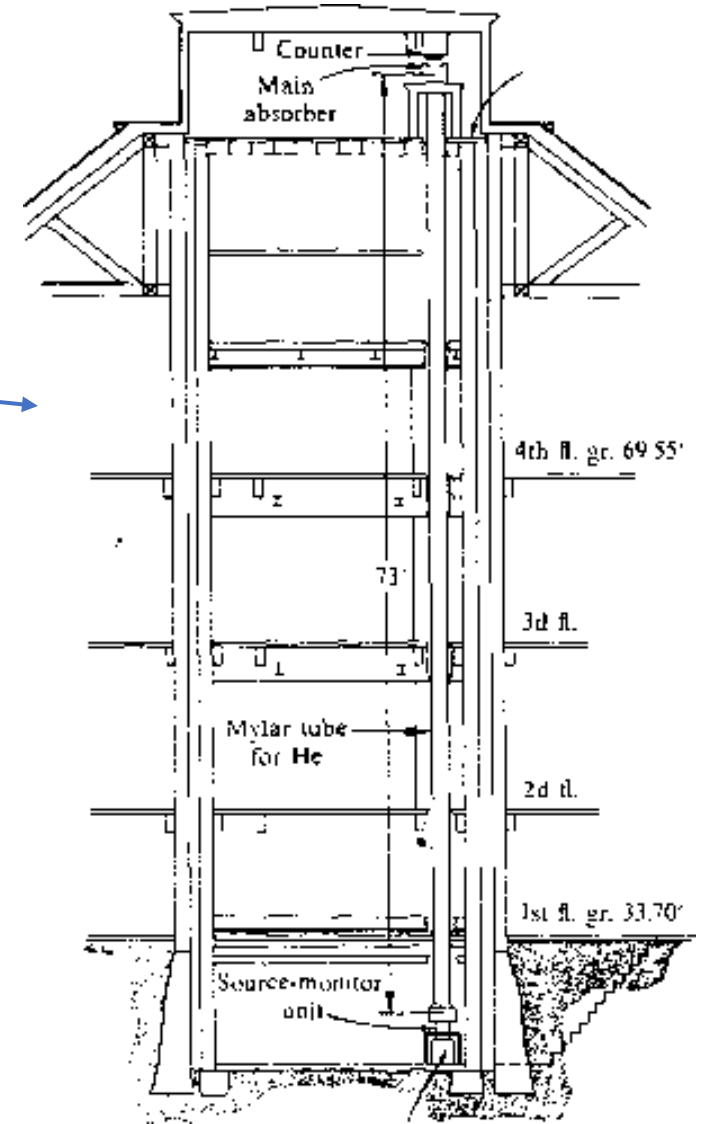
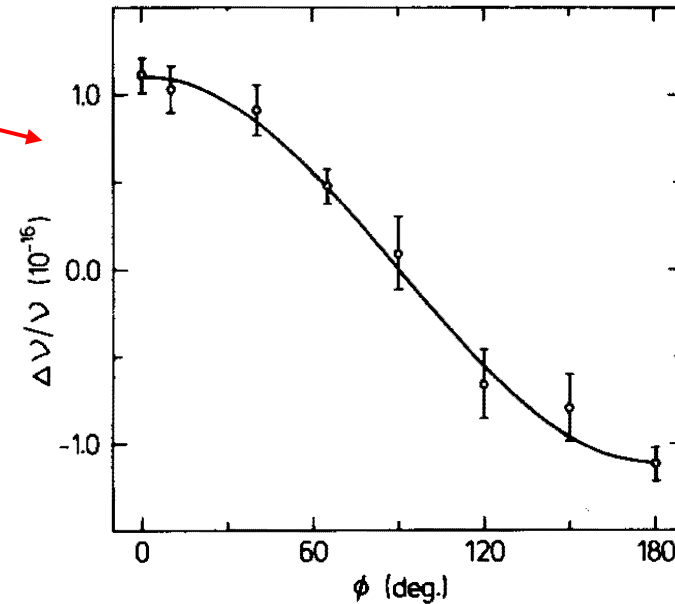
so you want low  $T$ , large  $\theta$  and small  $E_{\gamma}$ .

In practice, for metals,  $f(T) \approx 70\%$  at room temperature if  $E_{\gamma} < 20\text{keV}$ .

Somehow chemists and material scientists appropriated the technique and ran with it!

Only application to precision measurements:  
→ Pound & Rebka's very elegant gravitational red/blue shift with  $^{57}\text{Co}$  -  $^{57}\text{Fe}$ .

Even more impressive,  
Katila & Riski,  
Phys. Lett. 83A (1981) 51:  
→ Gravitational red/blue shift in a true tabletop (1 meter size) experiment with  $^{67}\text{Ga}$  -  $^{67}\text{Zn}$ .



Pound & Rebka Phys. Rev. Lett. 4 (1960) 337

**We plan to start by tackling one topic in sensing and one in keV optics:**

- 1) Sensing: search for new interactions at the sub-micron scale in a classic Mossbauer experiment (initially) using  $^{57}\text{Fe}$ .**
  - 2) keV optics: develop a technique to increase the efficiency of synchrotron light excitation of Mossbauer transitions.**
- 2) May eventually help 1), but the idea is to make some overall progress in this field.***

The search for new, long-range interactions (or deviation from  $1/R^2$  of gravity) at the micrometer scale is central in modern physics, both from an empirical standpoint and because of plenty of specific theoretical ideas.

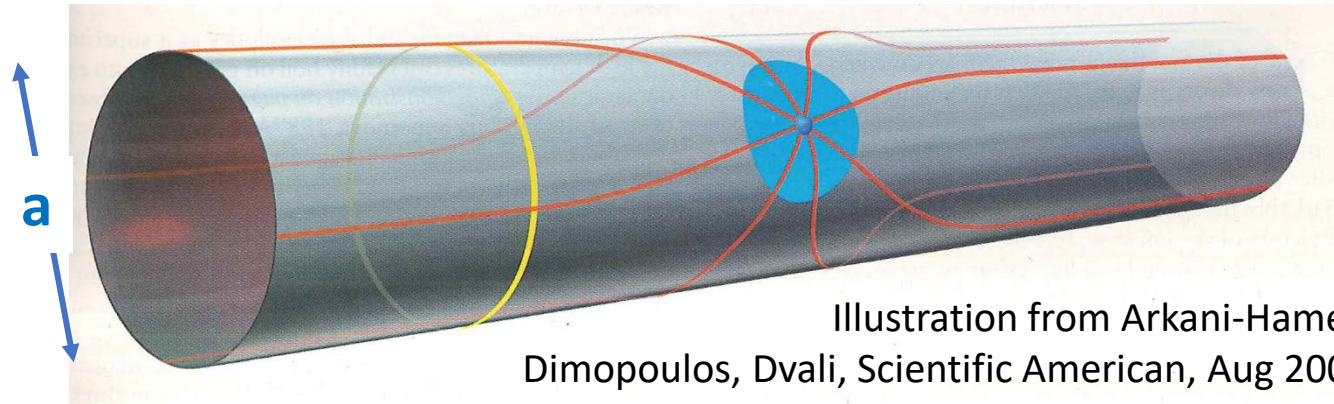


Illustration from Arkani-Hamed, Dimopoulos, Dvali, Scientific American, Aug 2000

**After all, it is quite arrogant to assume that gravity behaves as we know it, all the way down to the Planck scale!**



# Experimental challenges

- Since  $F = G \frac{M_1 M_2}{R^2} = G \frac{\rho_1 V_1 \rho_2 V_2}{R^2}$   
for materials we have access to (we can't use Neutron Stars!)  
 $\rho_1 \sim \rho_2 < 20 \text{ g/cm}^3$ , there is no silver bullet.  
In addition the volume  $V \sim R^3$ , so  $F \sim G \frac{\rho^2 R^6}{R^2}$  and it is clear that  
measurements at short distance become exceedingly difficult.  
Often the measured quantity is the acceleration of the test  
mass,  $a \sim G \frac{\rho R^3}{R^2} \sim G \rho R$ , but as  $R$  decreases this is still challenging
- At distances  $< 100 \mu\text{m}$  even neutral matter results in residual E&M interaction  
that are a dangerous background for these measurements
- Experiments should have discovery potential! So, only being able to set  
limits at the background level is not interesting.

## A new idea

*(GG, D.E. Kaplan, S. Rajendran, Phys Rev D 102 (2020) 115031)*

### Nuclei are well protected affairs

*(the fact that very long half lives, even for EM transitions, are readily observed, is witness to this statement)*

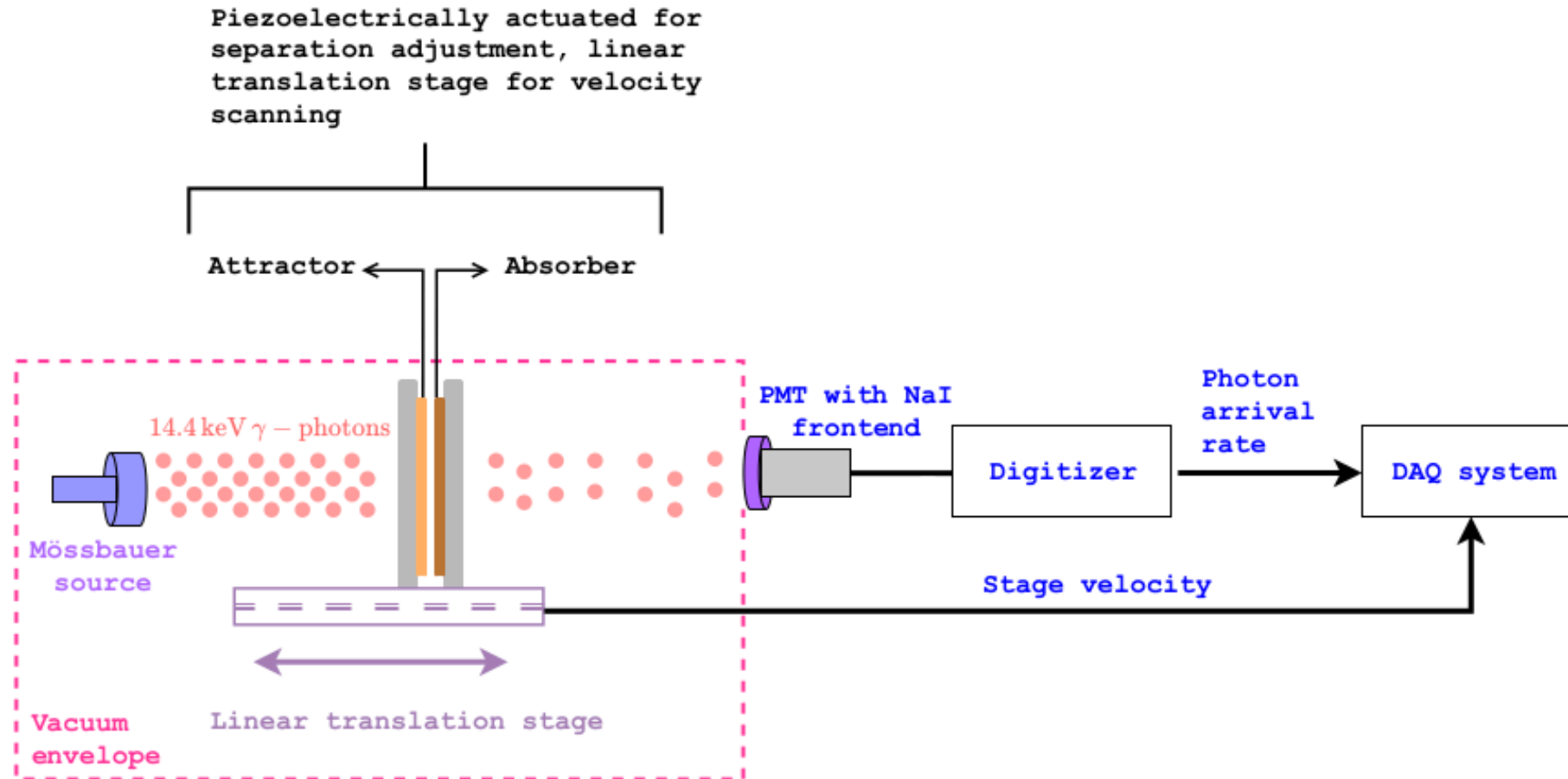
They have electric charge, but that is screened by the electron cloud and so has little coupling to external E&M disturbances.

In addition, nuclear level shifts due to E&M coupling occur through coupling to multipole (mainly dipole) moments and these are suppressed by the size of the nucleus.

And, this is further suppressed, in the case of unpolarized nuclei, by  $\sqrt{N}$ , when looking for the shift of a spectroscopy line that is measured by  $N$  events.

**Mossbauer spectroscopy can be used to investigate new forces**

The experiment consists in monitoring the resonant absorption of a Mossbauer line, while an “attractor” is positioned as variable distance from the absorber.



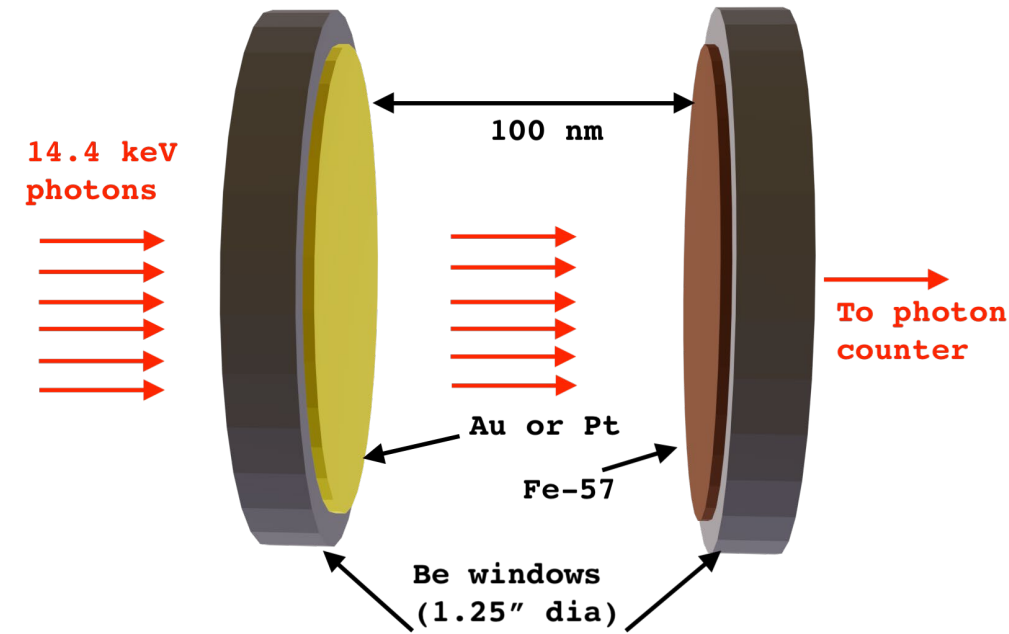
The new, hypothetical interaction, perturbs the isomeric state, slightly changing the position of the line.

By design, the experiment is insensitive to vector interactions, highly suppressing EM backgrounds.

Note that we search for the new interaction by measuring the effect of the field on a bound state, and not by sensing a force!

*This is a rather new concept of measurement.*

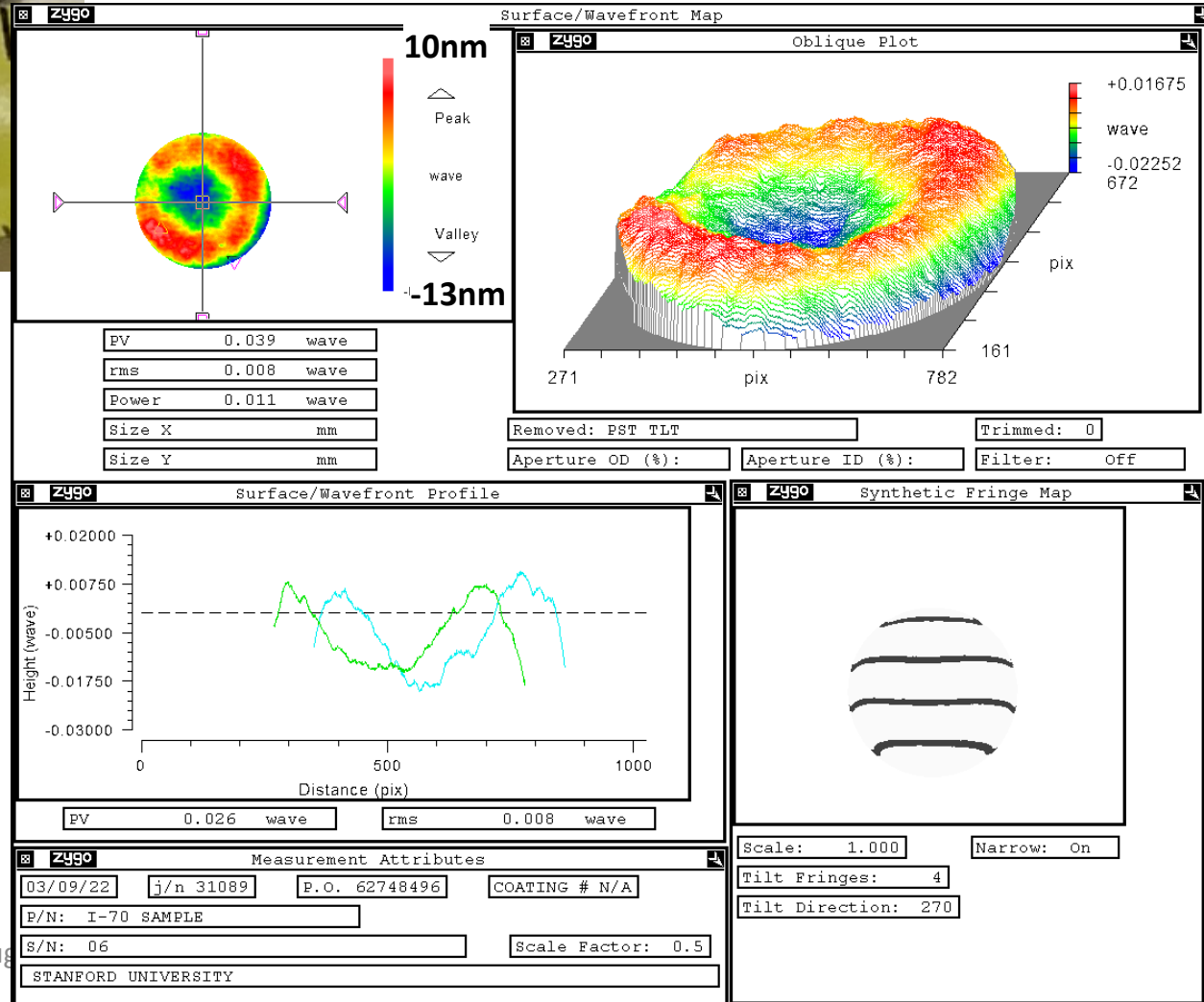
- The attractor can be something heavy, say a Pt coating.
- Both attractor and absorber have to be deposited on rigid substrates, flat enough so that they can be brought together to distances under test. The substrates also need to be transparent for the  $\gamma$ s used. → Beryllium substrates.
- In some future, may be able to simply freeze Xenon layers on the absorber.



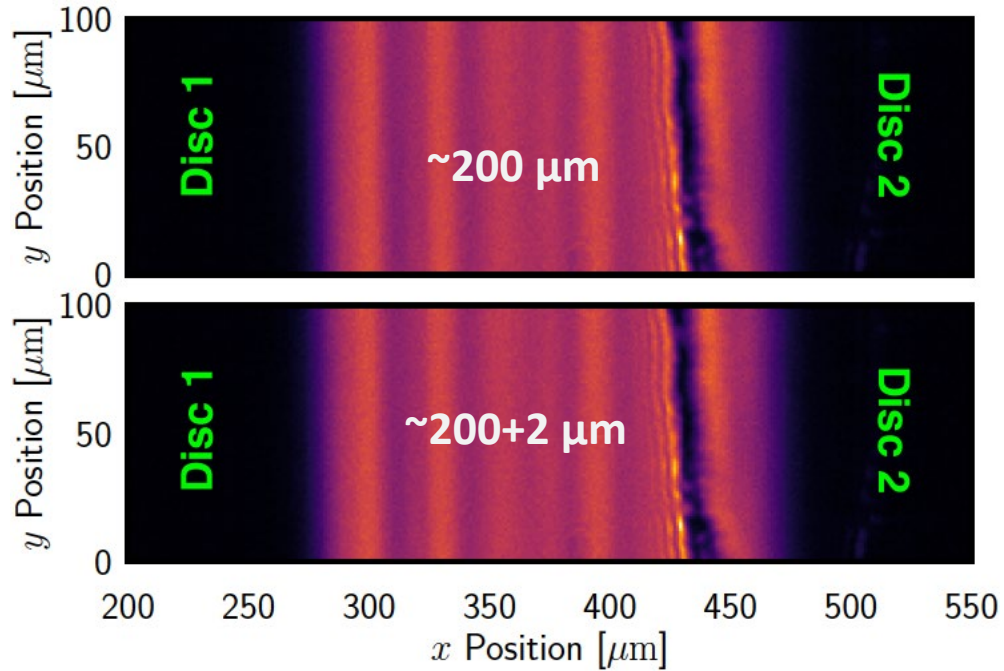


Ultra polished Be substrates are in hand

Flatness better than  $\pm 15$  nm achieved

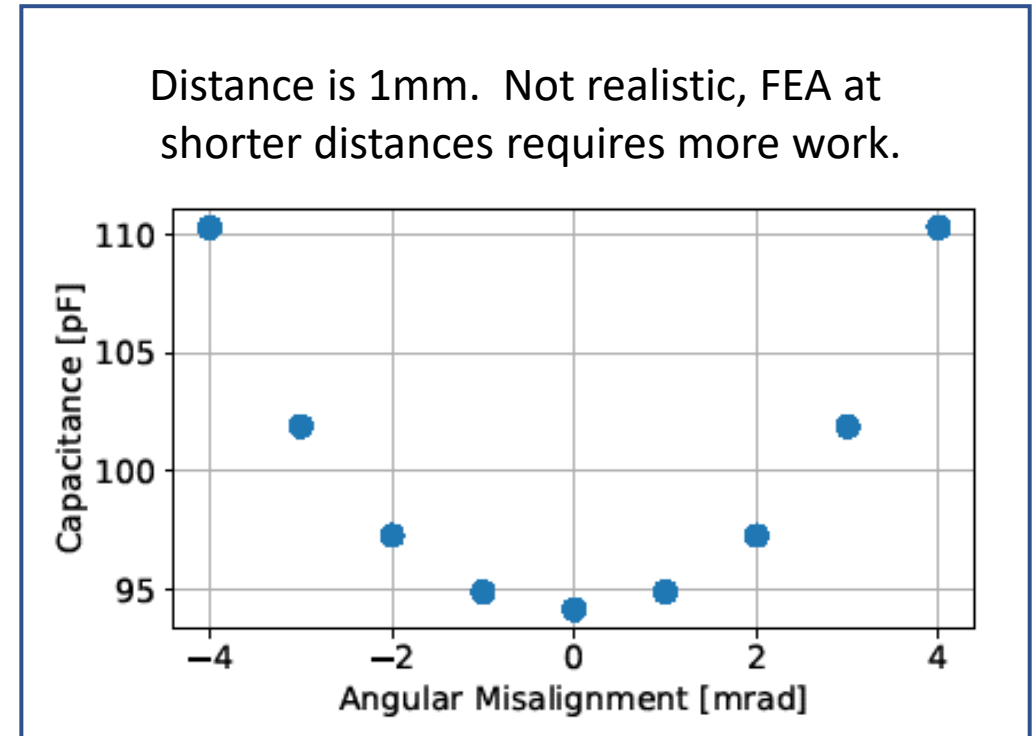


# Now working on piezo system to bring two substrates together in a controlled fashion

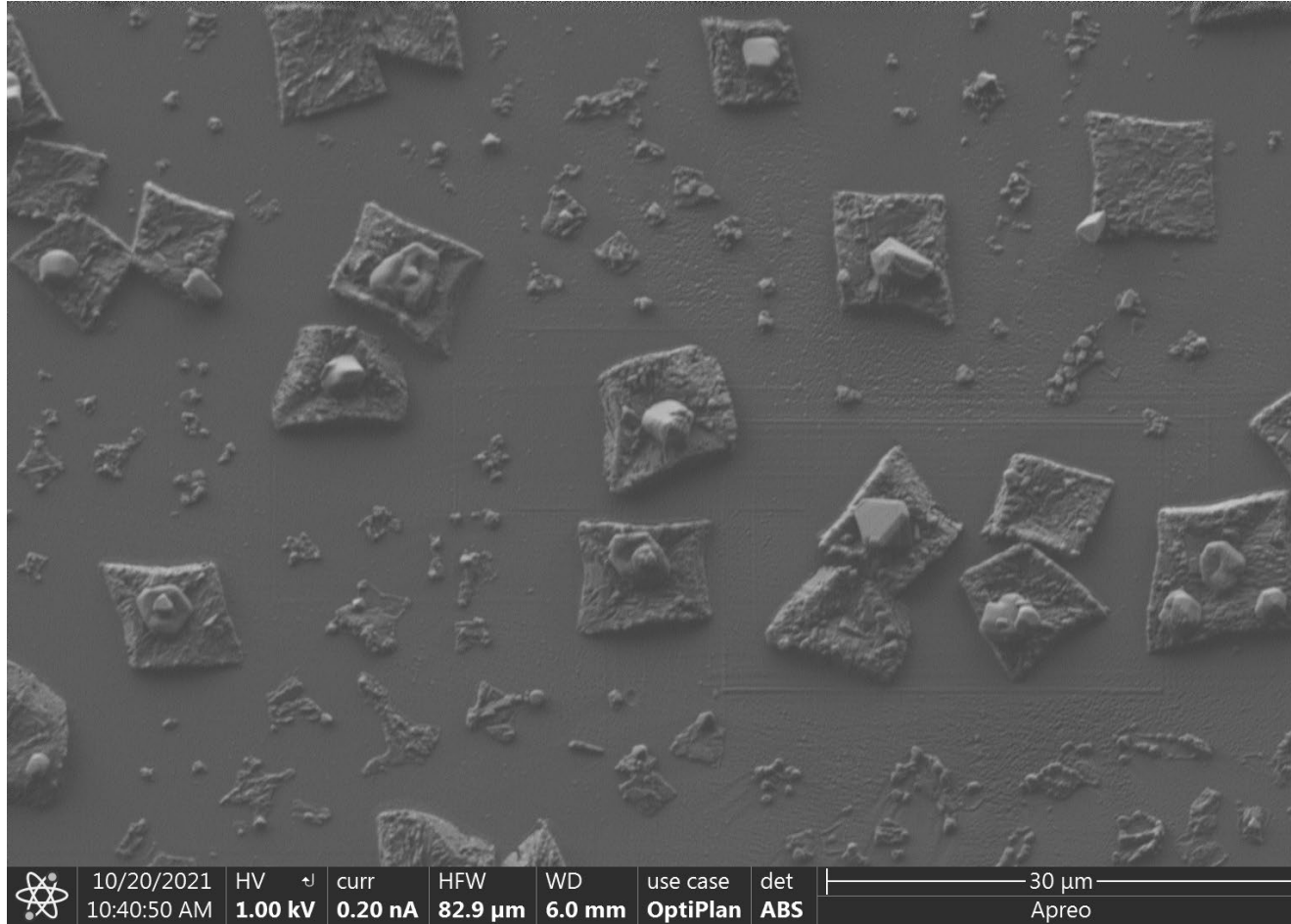


Learning to measure small shifts and misalignments by image analysis.

Absolutely calibrated piezos and capacitive measurements will then be used to bring the substrates together.



## ...and the development of the appropriate thin film containing $^{57}\text{Fe}$



← Micron-scale crystals of  $\text{K}_4[\text{Fe}(\text{CN})_6] \cdot 3\text{H}_2\text{O}$  (potassium ferrocyanide) produced by spinning of water solution.

$\text{K}_4[\text{Fe}(\text{CN})_6] \cdot 3\text{H}_2\text{O}$  is considered interesting because there is no isomeric shift/splitting.

In the end it may be simpler to deposit metallic Fe and use tuned magnetic fields to remove the splitting.

➔ This needs to be studied.

## Preliminary summary of systematics

Express them as an estimated spurious shift,  $\delta E$ . Assume that the measurement is done with  $N_\gamma \approx 10^{13}$  decays

EM coupling to the attractor. For unpolarized nuclei these are all suppressed by  $\sqrt{N_\gamma}$

- Casimir interactions  $\delta E \approx 10^{-13}$  eV
- Patch potentials  $\delta E \approx 10^{-15}$  eV
- Magnetic domains  $\delta E \approx 10^{-10}$  eV

so, the worst of these is  $3 \cdot 10^{-17}$  eV

Second order EM effects: external EM fields shifts the electron clouds that change the overlap with the nuclei.

Those do not average down with  $\sqrt{N_\gamma}$  (but they are second order).

- Dominant is Casimir effect:  $\delta E \approx 3 \cdot 10^{-15} \left(\frac{10nm}{d}\right)^4$  eV



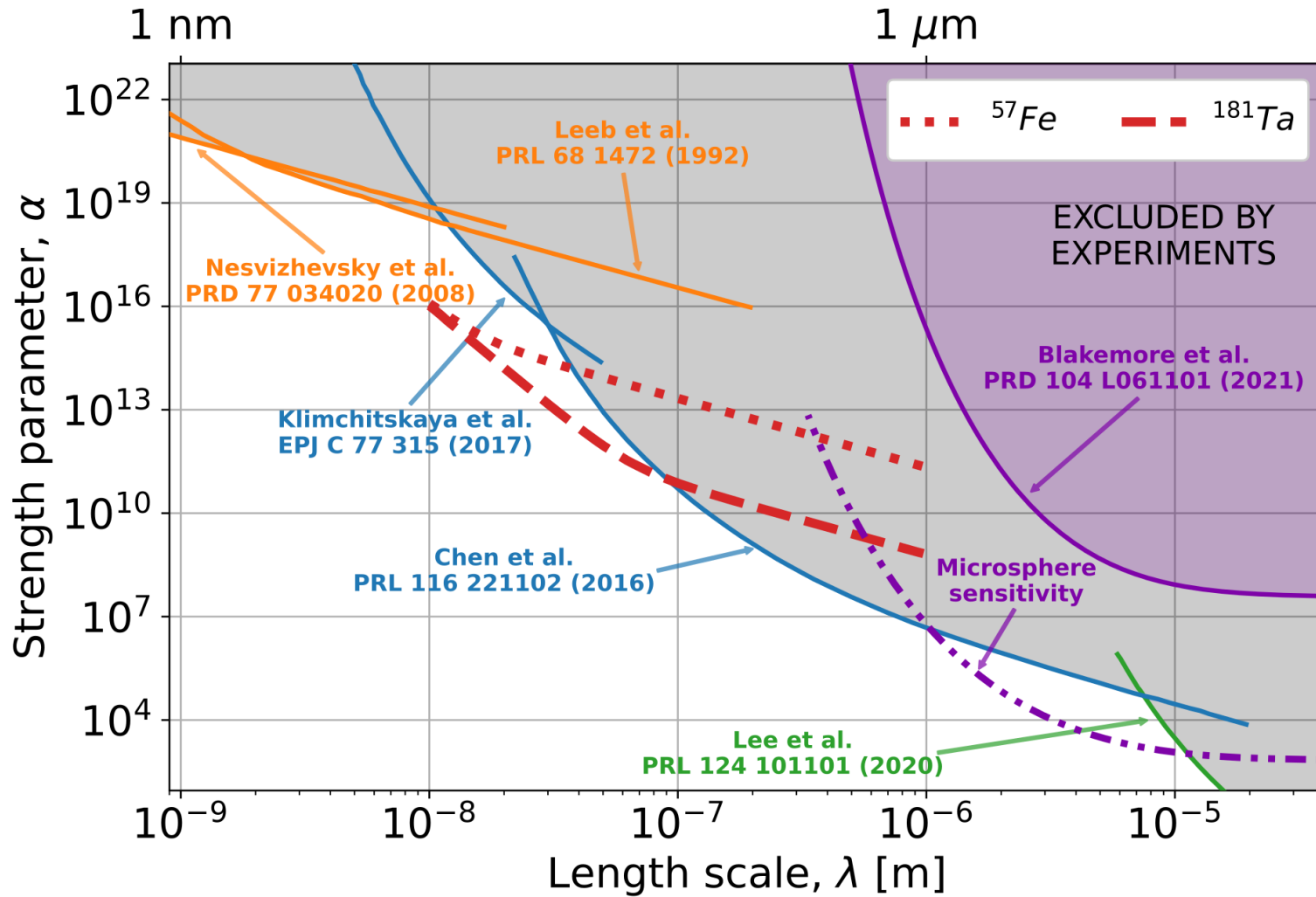
## Temperature effects.

- **Second order Doppler:** thermal motion of nuclei results in time dilatation and a line shift. Of course, it is not that the attractor changes the temperature of the source or absorber... The effects scales as  $\delta E \approx 10^{-11} \left(\frac{T}{300K}\right)^3$ , so e.g. at 30K one needs the temperature (of source and absorber) to be stable to better than 100mK, not hard.
- **The Lamb-Mossbauer factor** (the fraction of recoilless events) changes with temperature. This is not a line shift, but, we are crude and just measure the rate for  $E_\gamma \approx 10$  keV,  $\theta_D \approx 400$  K the temperature coefficient of the resonant rate is  $10^{-4} \text{ K}^{-1}$ , so one needs to keep the 30 K temperature constant to 3 mK between attractor in and attractor out. This is more challenging, but can be mitigated by measuring the resonance by 3 (or more) points (that hurts a bit the statistical power), or by bringing the attractor in a out many times (frozen Xe does not look that good for this).

## Estimated sensitivity

	Activity (mCi)	collimation	$\Delta E$ (eV)	Comment
$^{57}\text{Fe}$	100	$\pm 10^\circ$	$10^{-15}$	Conservative, Stat dominated
$^{181}\text{Ta}$	1000	$2\pi$	$10^{-17}$	Aggressive, Syst dominated

# Estimated sensitivity



It is customary to express potential deviations from the  $1/R^2$  law by modifying the potential with a Yukawa term, obtaining:

$$V(R) = G \frac{M_1 M_2}{R} (1 + \alpha e^{-R/\lambda})$$

$\alpha$ : magnitude of the effect  
 $\lambda$ : scale of the effect

## ***Beyond the first experiments***

***Synchrotron radiation can be used to directly excite the nuclear isomeric states.***

- Access a much broader set of isotopes/isomeric states, some way narrower.***
- Absorber and emitter can be the same species, great simplification.***
- Can also use the pulsed structure of the exciting beam to suppress backgrounds.***

***Already been used in a few cases to study material science.***

***→ Quantum optics with  $\gamma$ -rays!***

***Note that “free electron lasers” are “lasers” because of stimulated emission,  
but they are neither monochromatic, nor coherent.***

***→ It is clearly worthwhile to investigate avenues to produce  
monochromatic  $\gamma$ -rays with good coherence.***

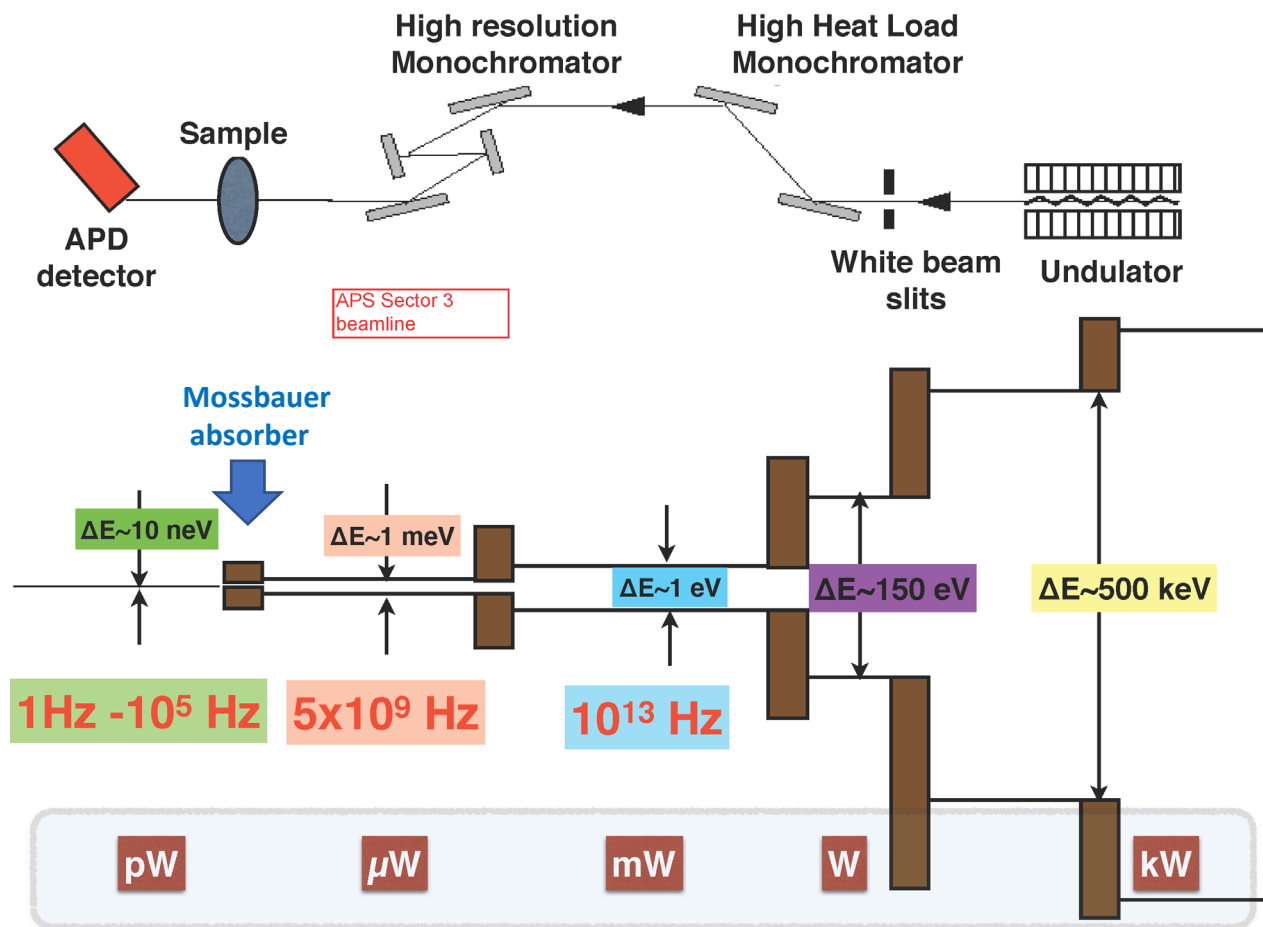


Figure courtesy of E.E. Alp, Argonne.  
 Similar parameters apply to Spring-8 and ESRF

## Main limitations:

- 1) Very limited spectral density, even for the most advanced synchrotron radiation sources.
- 2) Radiation damage issues.

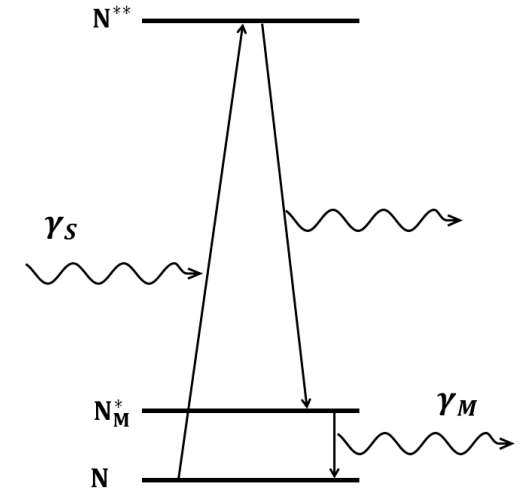
*No idea how to solve 2), but there are a number of ideas on how to address 1).*

## Use a 3-level scheme

Use synchrotron light to populate a (very short lived/broad) nuclear state with higher energy than the Mössbauer state.

This then de-excites into the long lived/ultra-narrow Mossbauer state.

<sup>189</sup> Os		<sup>181</sup> Ta		<sup>73</sup> Ge		<sup>57</sup> Fe	
J <sup>P</sup>	E(keV)	J <sup>P</sup>	E(keV)	J <sup>P</sup>	E(keV)	J <sup>P</sup>	E(keV)
7/2 <sup>-</sup>	216.67	9/2 <sup>+</sup>	136.3	7/2 <sup>+</sup>	68.7	5/2 <sup>-</sup>	136.5
	$k^*=100$						
	$k_0=34.3$						
9/2 <sup>+</sup>	30.8	9/2 <sup>+</sup>	6.237	5/2 <sup>+</sup>	13.3	3/2 <sup>-</sup>	14.4
3/2 <sup>-</sup>		7/2 <sup>-</sup>		9/2 <sup>+</sup>		1/2 <sup>-</sup>	



The strengths of the coupling from  $N^{**}$  to  $N_M^*$  and  $N$  are only known for <sup>189</sup>Os and they are similar.

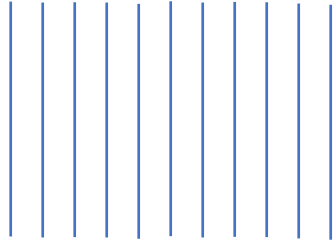
Synchrotron light sources do not like operating at 200 keV, still, for <sup>189</sup>Os a gain of  $\sim 3 \times 10^4$  appears possible.

Note that for <sup>189</sup>Os,  $\Gamma/E=7 \times 10^{-25}$  (for <sup>57</sup>Fe is  $6.4 \times 10^{-13}$ )!

# Magnetic bandwidth compression

Mossbauer absorber/emitter  
shown segmented for clarity

1 meV wide  
Synch light pulse



Spectrally narrow  
Mossbauer emission

Shifts in the absorption or emission lines can be produced with magnetic fields.

Hence a stack of absorbers in a pulsed gradient of magnetic field will detune the stack, so that each foil will absorb a slightly different energy from the synchrotron light pulse.

After the synchrotron pulse the field is quickly removed, so that all foils are tuned to the same energy.

**This may work best, to start, with  $^{181}\text{Ta}$  ( $\Gamma/E \sim 10^{-14}$ ).**

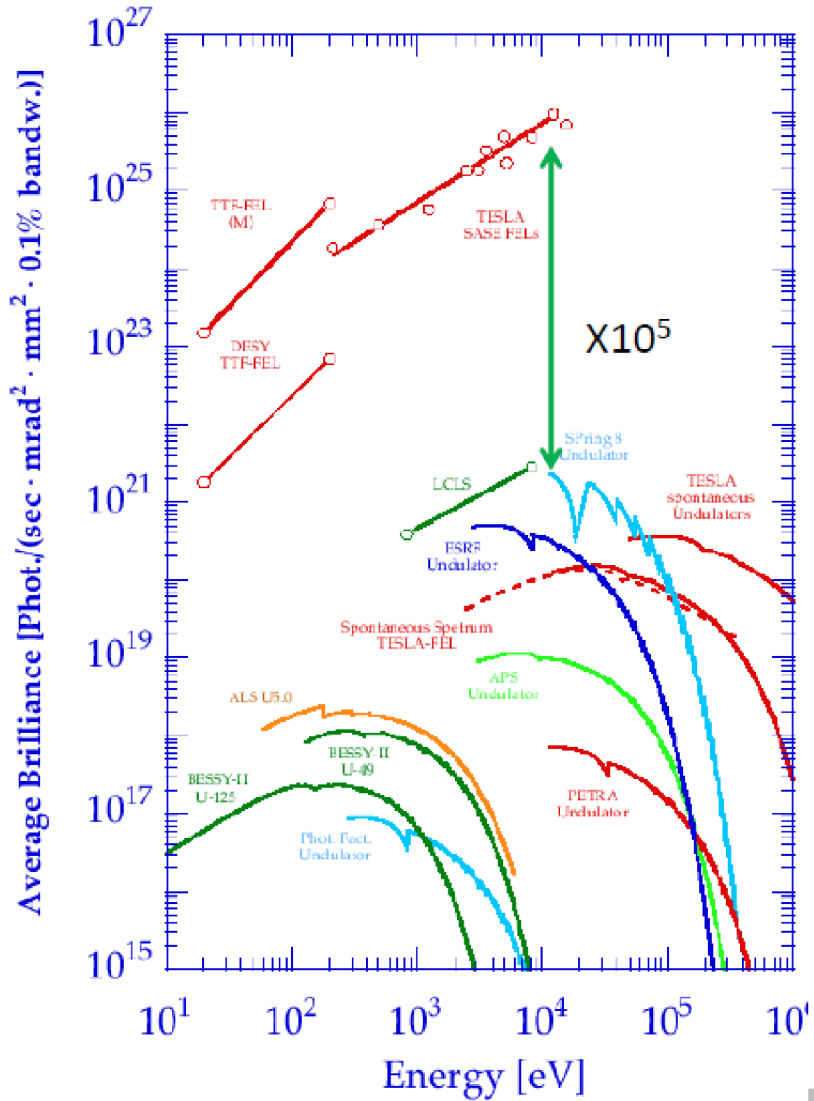
**The maximum gain should be  $\sim 30$ , limited by the incoherent photon absorption in the material.**

**The field required is 0.07 T, that should be feasible even in a fast pulsed magnet.**

**$^{57}\text{Fe}$  is expected to allow for gains up to  $\sim 500$ , but the maximum magnetic field required is  $\sim 75$  T**

# Conclusions

- We started exploring a new area at the interface between AMO, nuclear physics and synchrotron radiation, with applications to sensors and precision measurements.
- **Results may improve our knowledge of nature and its fundamental interactions.**
- They may also advance the technology towards the ultimate goal of fully coherent gamma-ray lasers.



Extracted from a talk of T. Shaftan, BNL

Trouble is, synchrotron light sources do not easily get >100keV

a figure of merit, with respect to direct excitation is

$$A \approx \frac{\Gamma^{**} \mathfrak{J}^{**}}{\Gamma} \frac{k^* k_0}{\mathfrak{J}_M^* (k^* + k_0)^2}$$

$$\mathfrak{J}_M^* \approx 5 \cdot 10^{13} \text{ s}^{-1} \text{ meV}^{-1}$$

$$\mathfrak{J}^* \approx 1.7 \cdot 10^4 \text{ s}^{-1} \text{ meV}^{-1}$$

For  $^{189}\text{Os}$ ,  $A \approx 3.4 \cdot 10^4$  so, substantial gains appear possible.