# Low-Energy Nuclear Transitions in Subrelativistic Laser-Generated Plasmas

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## Syllabus:

- **Motivation for investigation of nuclear phenomena in laser-produced plasmas**
- **Selection of nuclei for low-energy nuclear excitations**
- **Experiments at medium-size high-power laser PALS:**
  **under way study of $^{181}$Ta activation - test bed for**
    **experimental strategy**
    **novel or modified instrumentation**
- **Conclusions and future work**


**Acknowledgments**

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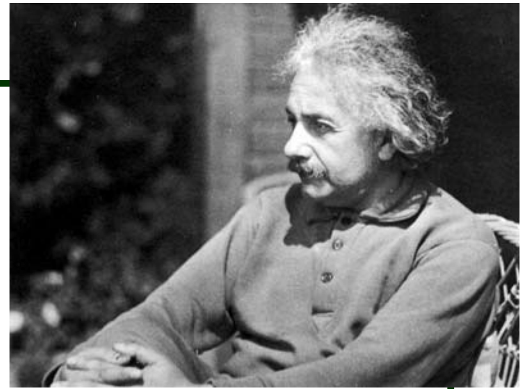
# Motivation: History Dates Back to 1920s



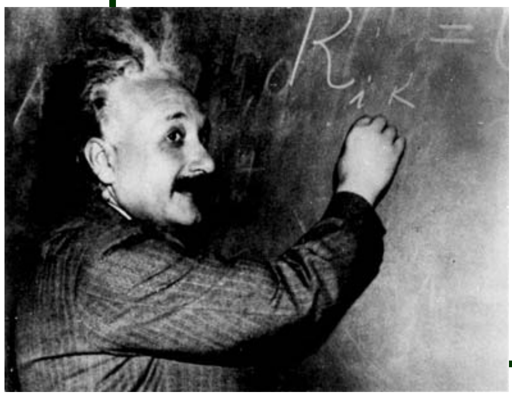
**National Academy of Science of Argentina, April 16, 1925**

Session to award A. Einstein the Academy's honorary diploma.

Q: Is it possible to obtain **induced radioactivity by bombardment of matter with quanta of light?**

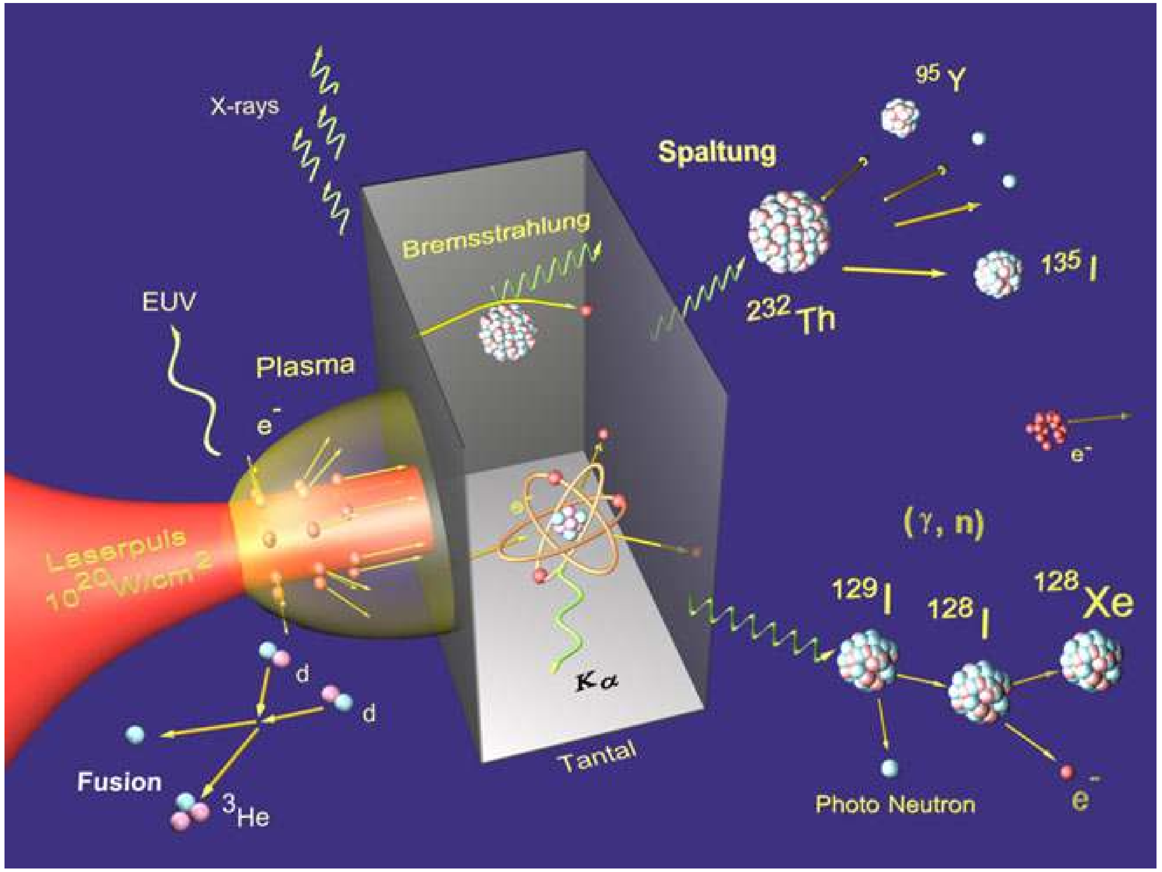
A: Probably, there exists radioactivity of matter induced by the action of the light quanta; **the difficulty of the observation** of such phenomenon, **if it exists**, **is that the effect which has to be observed is very small**.

**The effect confirmation is hard but possible.**



A. Einstein, Collected Scientific Works 4, Nauka, Moscow, 1967 (cited after S. Matinyan, Phys. Rep. 298 (1998) 199)

# Artist's (Researcher's) View of Relativistic Laser-Matter Interactions




**Laser relativistic limit: $I\lambda^2 \approx 10^{18}$ Wcm$^{-2}\mu$m$^2$**

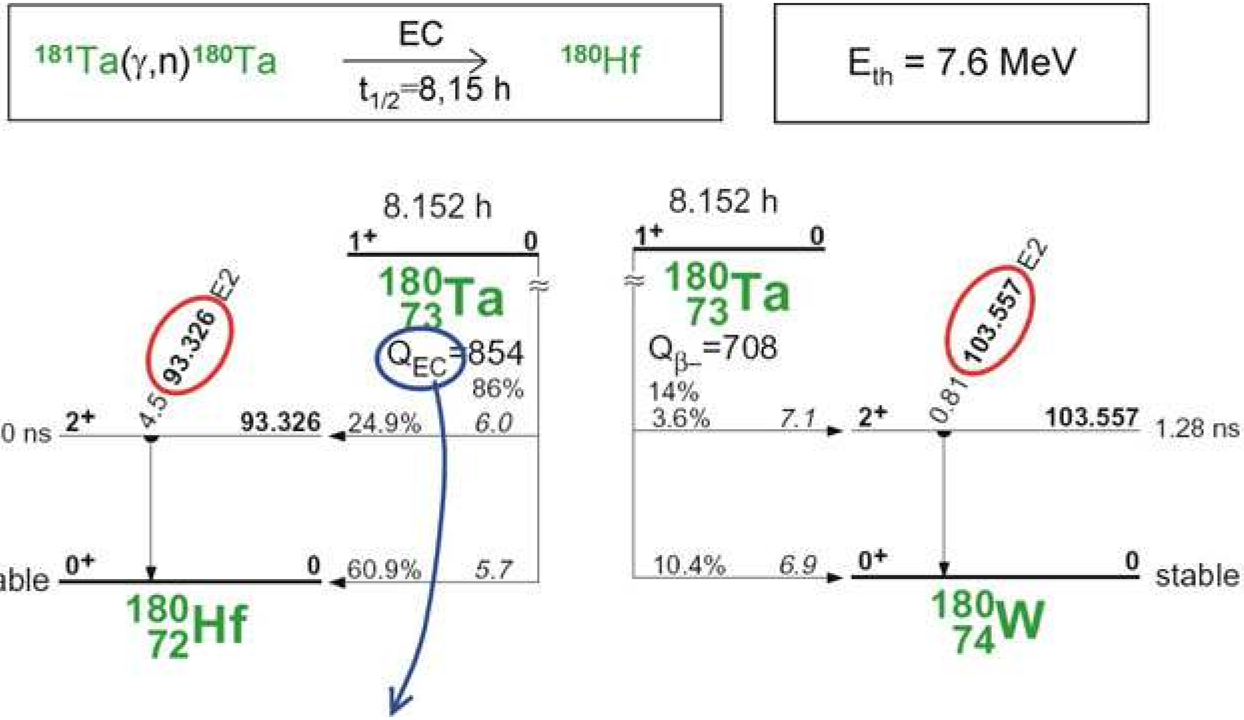
**Photon yield:**
**IOQ Jena**
**fs laser JETI**
**0.1 ps, 0.3 J**

**Si, $10^{17}$ W/cm$^2$**
**$10^9$ quanta/4π**
**X-rays Si Kα**
**(E~2 keV)**

**Ta, $10^{19}$ W/cm$^2$**
**$10^5$ quanta/4π**
**γ-radiation**
**(E>8 MeV)**

Picture by courtesy of colleagues from IOQ FSU Jena

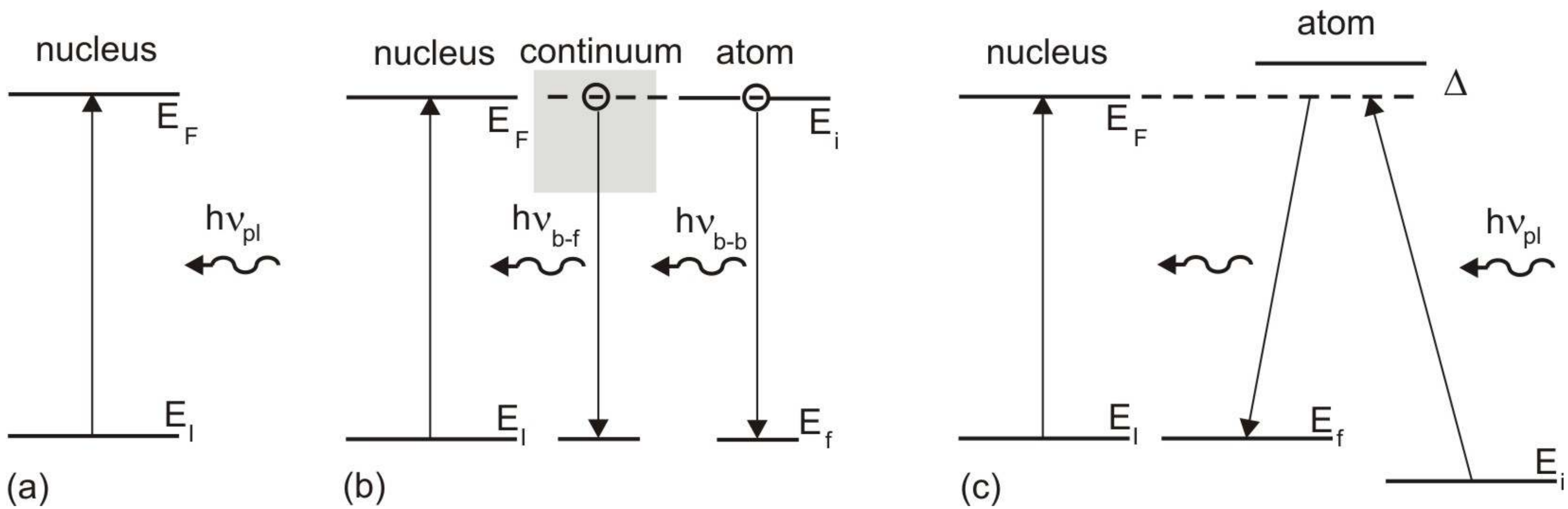
# Identification of Nuclear Processes Induced by fs-Lasers




**Transmutation of $^{181}$Ta (γ, n) to $^{180}$Ta and further to stable $^{180}$Hf (probability 86%) or $^{180}$W (probability 14%) accompanied by weak γ emission at 93.3 keV (Hf) or 103.5 keV (W) and strong x-ray Hf Kα$_1$ (55.8 keV), Kα$_2$ (54.6 keV) and Kβ (63.2 keV) emission due to numerous holes in K-shell of Hf → identification of Ta decay**

**Data by courtesy of colleagues from IOQ FSU Jena**

# Subrelativistic Laser-Matter Interaction: Excitation of Low-Energy Nuclear Transitions




**$T_e$ close to excitation energy → resonance mechanisms**
**photoexcitation, IIEC, & IEB**
**dominate in excitation of low-lying nuclear levels in plasmas**

# Relativistic vs. Subrelativistic Laser Plasmas

## Sub-relativistic nuclear excitation

- direct photoexcitation by plasma radiation
- nuclear excitation by electron transition (NEET)
- inverse internal electron conversion (IIEC)
- nuclear excitation by electron capture (NEEC)
- excitation upon collective atomic shell transition
- inelastic electron scattering

## de–excitation

- gamma–ray emission
- internal electron conversion (IEC)

**high–energy (>100 keV) photonuclear reactions well established:**

- ($\gamma$,n)
- ($\gamma$,$\alpha$)
- ($\gamma$,p)
- ($\gamma$, $\gamma$‘)

**low–energy nuclear excitation (1–10 keV)**

- shielding of nuclei by atomic electron shell
- competition between IEC and $\gamma$–decay
- effect of ionization (on excitation and decay)
- energy level broadening in ionized systems
- stimulated isomeric nuclei de-excitation

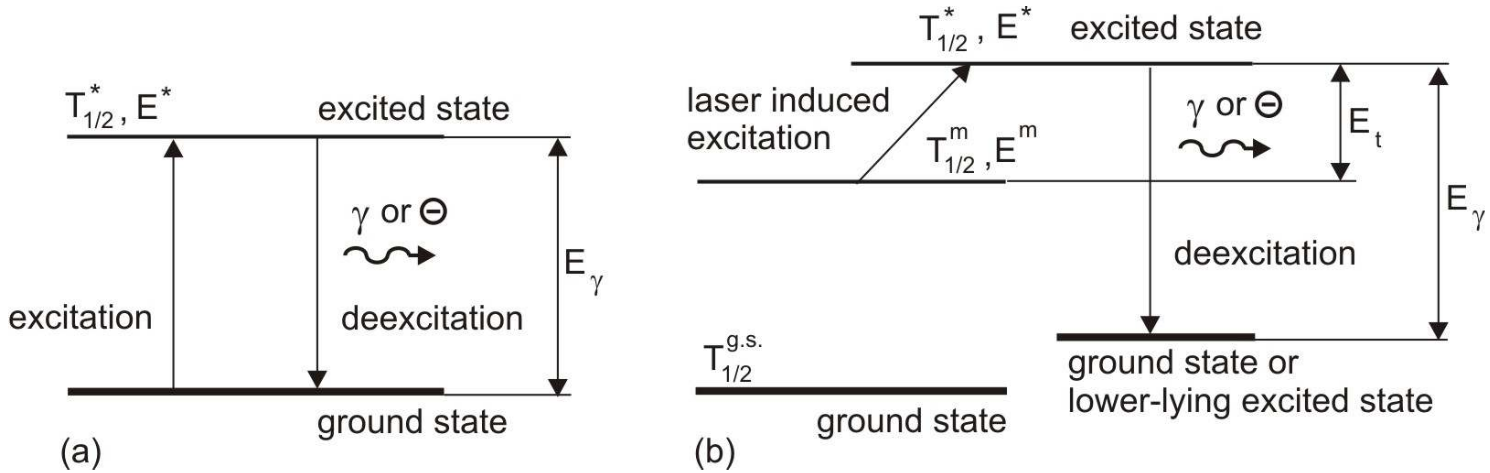
**Interesting physics of low–energy nuclear excitations**
**Excitation & modification of nuclear levels - energy storage & release**

Schwoerer H., et al. (Eds.), Lasers and Nuclei, Springer, Berlin, (2006)

## Strategy of Experiments Directed to Identification of Low-Lying Nuclear Transitions

- selection of isotopes and nuclear transitions to be studied, modeling of relevant processes
- optimization, production, and handling the nuclear (especially isomeric) targets
- generation of the intense plasma sources, characterization of nuclei surroundings and competing processes
- extraction of energy- and time-resolved signals accompanying the excited-state decay from high-level background
- interpretation of the obtained results with underlying theories

# Selection of Stable (Long Lived) & Isomeric Nuclei with Low-Lying Excited Levels




**Screening criteria:**

**energy and half-life of the excited nuclear levels**
**transition multipolarity (*E*1–*E*3, *M*1–*M*3) , IEC coefficient α**
**abundance, production and/or enrichment procedures**
**accumulation, transport and handling of the target materials (safety!)**
**effective excitation cross-sections not considered (lack of reliable data)**

BNL Nuclear Structure and Decay Data, Nuclear Data Sheets

# Candidates for Observation of Low-Lying Transitions in Stable or Long-Lived Nuclei

Table 1: Stable nuclei (upper part) and long–lived (> 1 h) nuclei (bottom part) having excited states $E^*$ within 10 eV – 15 keV and half–life of excited state $T^*_{1/2}$ longer than 1 ns. Nuclei are arranged according to isotope abundance $\eta$ (stable nuclei) or ground state half–life $T^{g.s.}_{1/2}$ (long–lived nuclei). Included are the induced transition multipolarity ($XL$) and, if known [22], the most intense depopulating $\gamma$ rays with corresponding internal conversion coefficients ($\alpha$).

| nucleus | $\eta$[%] or $T^{g.s.}_{\frac{1}{2}}$ | $XL^{\#}$ | $E^*$[keV] | $T^*_{\frac{1}{2}}$ | $E^{\%}_{\gamma}$[keV] | $\alpha$ | Ref. |
|---|---|---|---|---|---|---|---|
| $^{45}$Sc | 100 | $M2$ | 12.4(2) | 318(7) ms | 12.4(2) | 632(71) | [2] |
| $^{169}$Tm | 100 | $M1+E2$ | 8.41031(19) | 4.08(8) ns | 8.41031(19) | 285 | [23] |
| $^{181}$Ta | 99.988(2) | $E1$ | 6.238(20) | 6.05(12)$\mu$s | 6.238(20) | 70.5(25) | [3],[11],[29],[30] |
| $^{201}$Hg | 13.18(9) | $M1$ | 1.556(15) | | 1.565(6) | $4.7(7) \times 10^4$ | [2],[3],[29] |
| | | $M1$ | 32.138(16) | 1ns | 32.19(3) | 41.9(2) | |
| $^{83}$Kr | 11.49(6) | $M1+E2$ | 9.4053(8) | 154.4(11) ns | 9.4051(8) | 17.09(5) | |
| $^{73}$Ge | 7.76(8) | $E2$ | 13.2845(15) | 2.92(3)$\mu$s | 13.2845(15) | $1.2 \times 10^3$ | [3] |
| $^{57}$Fe | 2.119(10) | $M1+E2$ | 14.4129(6) | 98.3(3) ns | 14.4129(6) | 8.56(26) | |
| $^{187}$Os | 1.6(3) | $M1+E2$ | 9.746(24) | 2.38(18) ns | 9.746(24) | 264(33) | |
| $^{235}$U | $7.04(1) \times 10^8$ y | $7/2^- \to 1/2^+$ | 0.0765(4) | 26 min | 0.0765(4) | $> 1.0 \times 10^{10}$ | [1],[2],[3],[10],[29],[31],[32],[33] |
| | | $M2$ | 13.0400(21) | 0.50(3) ns | 12.975(10) | $1.0 \times 10^3$ | |
| $^{205}$Pb | $1.53(7) \times 10^7$ y | $E2$ | 2.329(7) | 24.2(4) $\mu$s | 2.328(7) | $4.4 \times 10^8$ | [3] |
| $^{151}$Sm | 90(8) y | $M1$ | 4.821(3) | 35(8) ns | 4.821(3) | $9.2(12) \times 10^2$ | |
| $^{193}$Pt | 50(6) y | $M1$ | 1.642(2) | 9.7(3) ns | 1.642(2) | $1.2 \times 10^4$ | |
| | | $E2$ | 14.276(8) | 2.52(5) ns | 12.634(8) | 151(3) | |
| $^{133}$Ba | 10.51(5) y | $M1$ | 12.322(5) | 7.0(3) ns | 12.327(6) | 70.3 | |
| $^{171}$Tm | 1.92(1) y | $M1$ | 5.0361(11) | 4.77(8) ns | 5.025(6) | 1408(55) | |
| $^{182}$Ta | 114.43(3) d | $E3$ | 16.263(3) | 283(3) ms | 16.263(3) | 45200 | |
| $^{124}$Sb | 60.20(3) d | $E3$ | 10.8627(8) | 93(5) s | 10.8630(11) | $2.19 \times 10^4$ | |
| $^{126}$Sb | 12.35(6) d | ($E3$) | 17.7(3) | 19.15 min | 17.7(3) | 718 | |
| $^{167}$Tm | 9.25(2) d | $M1$ | 10.400(19) | 0.95 ns | 10.45(5) | 650 | |
| $^{142}$Pr | 19.12(4) h | $M3$ | 3.694(3) | 14.6 min | 3.683(4) | $1.17 \times 10^{1}0$ | |
| $^{129}$Ba | 2.23(11) h | $M3$ | 8.42(6) | 2.16(2) h | 8.4(2) | $1.084 \times 10^8$ | |
| $^{104}$Ag | 69.2 9(10) min | $M3$ | 6.9(4) | 33.5(20) min | 6.9(4) | $1.0(4) \times 10^8$ | |

\# For $L > 3$, the spin and parity of the involved levels are given.
% Most intense $\gamma$ ray depopulating the given level.

C. Granja et. al., Nuclear Physics A (2007), in print

# Nuclei Considered for Medium-Size Lasers Experiments

| Nucleus | $E^m$ [keV] | $T^m_{1/2}$ | $E_t$ [keV] | $T^*_{1/2}$ | $E_\gamma$ [keV] | XL | α |
|---|---|---|---|---|---|---|---|
| $^{45}$Sc | — | — | 12.4(2) | 318(7) ms | 12.4(2) | *M2* | 632(71) |
| $^{181}$Ta | — | — | 6.238(20) | 6.05(12) μs | 6.238(20) | *E1* | 70.5(25) |
| $^{205}$Pb | — | $1.53\times10^7$ y | 2.329(7) | 24.2(4) μs | 2.328(7) | *E2* | $4.4\times10^8$ |
| $^{192}$Ir | 168.14(12) | 241(9) y | 5.06/9.86 | | | | |
| $^{242}$Am | 48.60(5) | 141(2) y | 4.10 | | 52.77 | *E2* | 366.8 |
| $^{110}$Ag | 117.59(5) | 249.76(4) d | 1.13 | 36.7(7) ns | 117.607 | *M3* | 168 |
| $^{84}$Rb | 463.62(9) | 20.26 min | 3.48 | 9(2) ns | 219.0 | (*M3*) | |

**$^{181}$Ta:** **~100% isotopic abundance, transition energy of 6.2 keV, *E*1 multipolarity, lifetime of the excited level above 6 μs BUT high internal electron conversion coefficient $\alpha = 70.5$ narrow level widths ($\Gamma_\gamma = 1.3\times10^{-12}$ eV, $\Gamma_T = 1.6\times10^{-10}$ eV) → small photoabsorption cross section $\sigma_\gamma \approx 4.0\times10^{-13}$ barn**

**Previous experiments:**

**positive results** Andreev A.V., et al., JETP 91 (2000) 1163 **questioned** Aleonard, MM., Gobet, F., Robson, L., et al., 7th AFSOR Workshop (2005) Fedosejefs, R., et al, Proc. Conf. Plasma Phys. Tarragona, ECA 29C (2005) P1.152

# Motivation for Redo Experiments at PALS

**Andreev's results indicate an enormous broadening of nuclear level widths in plasma; if confirmed, this result would be of primary importance for prospective energy storage in nuclei**

**relatively large amount of experimental data (albeit negative) on $^{181}$Ta excitation was gathered → proper test bed for checking experimental strategy & instrumentation used**

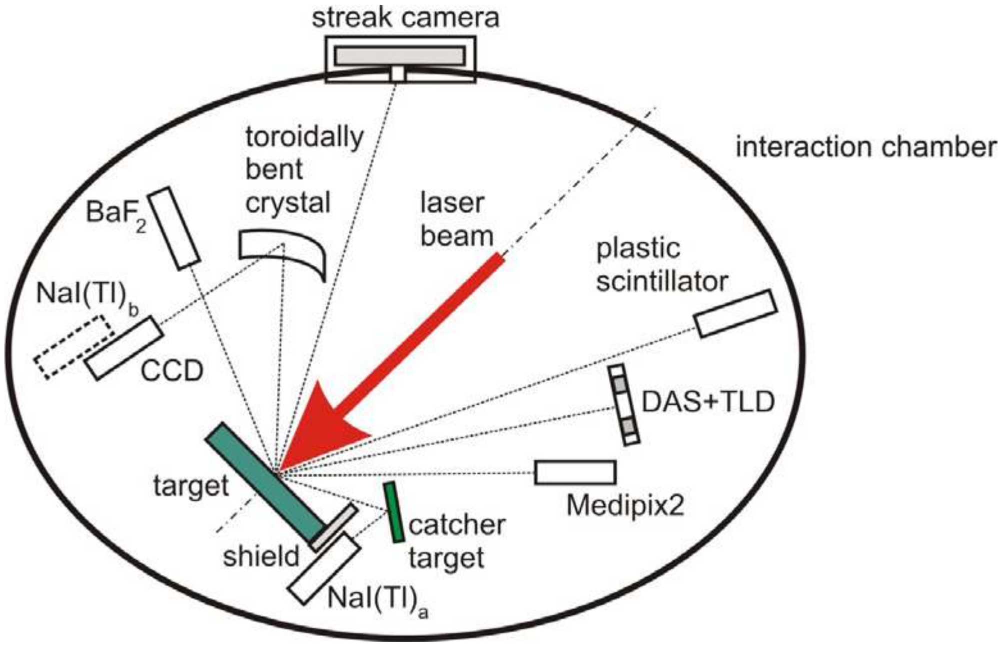
**Characteristic aspects of the experimental approach adopted:**

- **PALS (nominal parameters: 1 kJ, $\tau \sim 300$ ps, $7\times10^{16}$ Wcm$^{-2}$) –**
  - **large plasma volumes, duration and radiative fluxes**
- **complex characterization of the plasma environment**
- **application of advanced spectroscopic methods and detectors**

**Research objectives of pilot experiments:**

- **evidence for feasibility of nuclear experiments at ns systems**
- **definition of upper limits for excitation and decay of $^{181}$Ta**

# PALS Experimental Setup

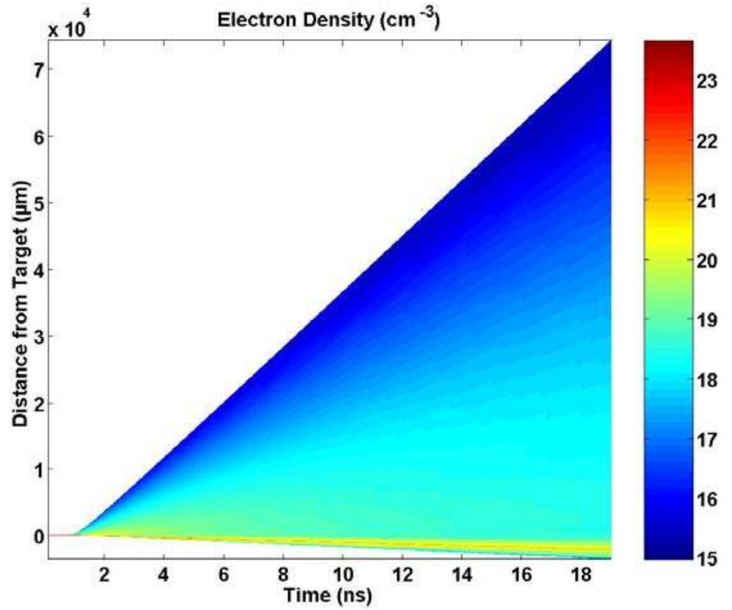


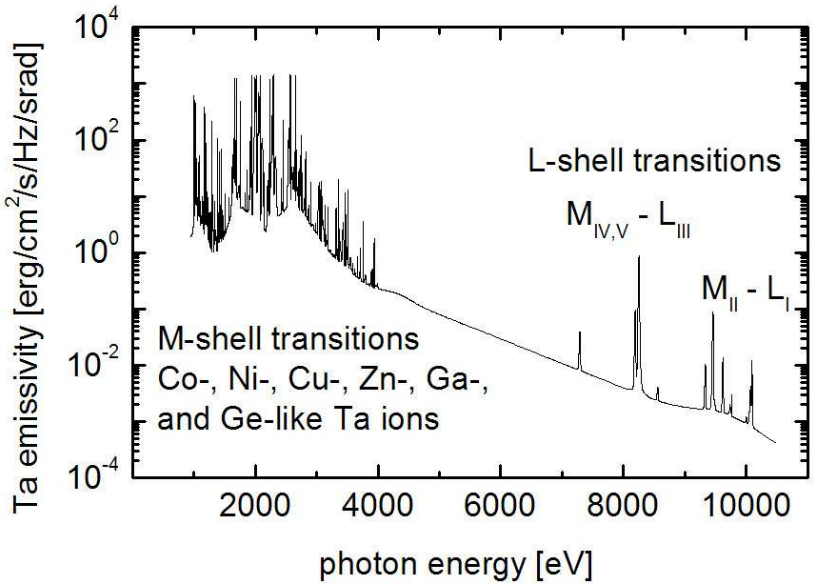

**Plasma diagnosis:**
**x-ray streak camera**
**TBS + CCD camera**
**DAS + array of TLDs**

**Time-resolving detectors**
**scintillators: plastic**
**$BaF_2$**
**NaI(Tl)**
**semiconductor (CMOS):**
**Medipix2**

**Prague Iodine Laser System PALS**
**single Gaussian-profile beam (1000 J/1ω, 1.315 μm, 0.3 ns, 80 μm, $7\times10^{16}$ $Wcm^{-2}$)**
**frequency-tripled radiation (300 J/3ω, 0.44 μm, pulse length 0.25-0.3 ns)**

# Plasma Simulations




**Hydrocodes**
**1D MEDUSA** basic characteristics (n, T) vs. time & distance from the target
**EHYBRID** similarly calculates the main plasma characteristics in 1.5D

Djaoui A, Rose SJ, J Phys **B25** (1992) 2745

J. Kuba et al., Fac Nucl Sci CTU, unpublished

**Ta spectra synthesis**
performed with code **FLYCHK**
for electron temperature $T_e$=1 keV and density $n_e$=1×$10^{21}$ cm-3
around the 6 keV energy range,
only continuum is emitted

Chung, H.-K., et al., HEDP **1** (2005) 3

# Theoretical Estimates of $^{181}$Ta Excitation Yield at PALS

**reaction yield:**

$$N = D\int_{Et}^{\infty} \sigma(E) I(E) I(E) d(E)$$

**resonant cross section:**

$$\sigma_{\gamma}(E_{\gamma}) = \frac{\pi}{2} g \lambda_{\gamma}^{2} \frac{\Gamma_{\gamma}\Gamma_{T}}{\left(E_{\gamma} - E^{*}\right) + \Gamma_{T}^{2}/4}$$

**x-ray flux from surface area *S*:**

$$\frac{dN_{\gamma}}{dtdE_{\gamma}dS} = \frac{1}{\lambda_{\gamma}^{2}} \frac{1}{\exp\left(E_{\gamma}/T_{e}\right) - 1}$$

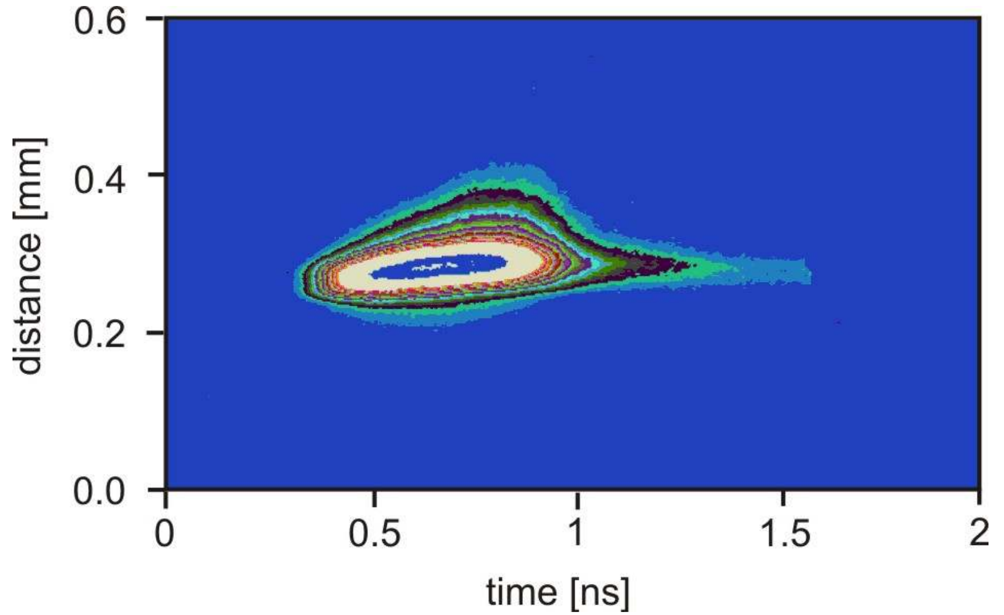
**total number of excited isomeric nuclei *N*:***

$$N^{*} = \frac{dN_{\gamma}}{dtdE_{\gamma}dS} \sigma_{\gamma} n_{i0} Sl \approx N_{i0} \frac{\Gamma_{\gamma}\tau_{p}}{\exp\left(E_{\gamma}/T_{e}\right) - 1} \approx 1.5 \times 10^{7}$$

**$\alpha$ = 70.5 → number of nuclei decaying via x rays is about $2\times10^{5}$**

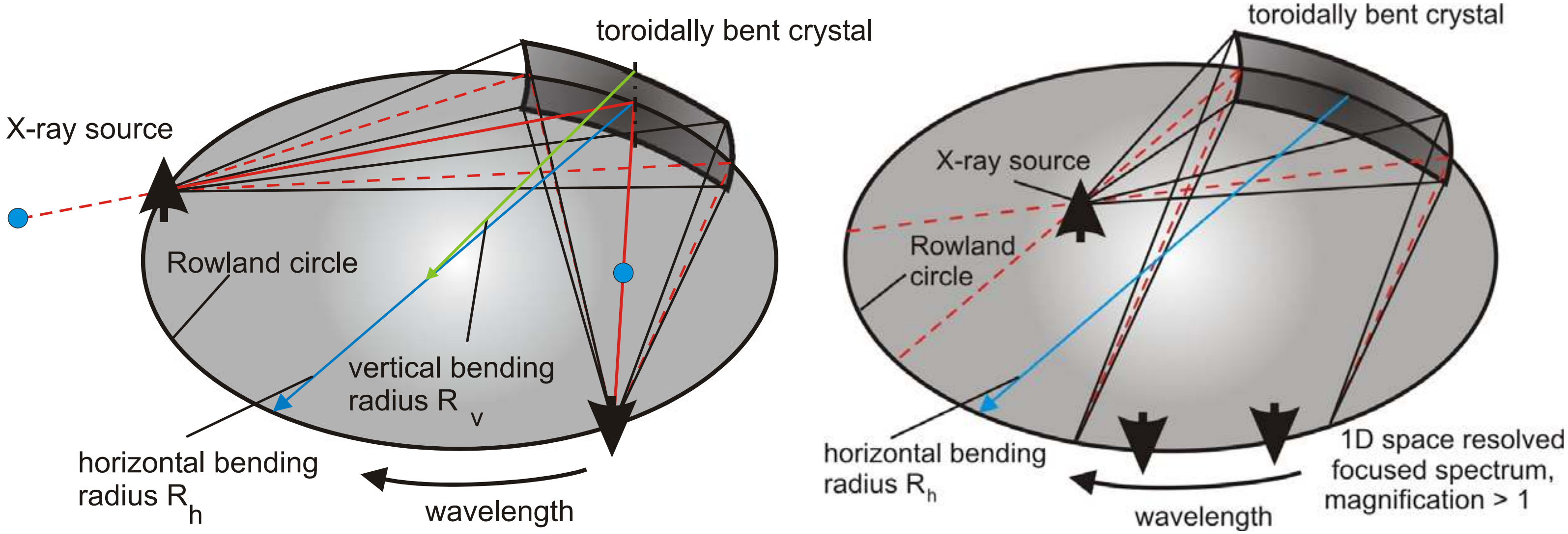
Andreev, A.A., private communication

# Selected Methods Used for X-ray Plasma Diagnosis: Imaging




**Pinhole + X-ray Streak Camera**
**1024x400 pixels, x: 3.3 ps/pixel, y: 2.0 μm/pixel**
**filters (1 mm Be, 8.5 μm aluminized mylar) → $E_{ph} > 2.5$ keV**
**3ω experiments (50-250 J, 0.44 μm, 250 ps, 0.4–2×$10^{16}$ $Wcm^{-2}$):**
**limited duration and spatial extent of the plasma emission**

# Toroidally Bent Crystal Spectrometer: Principle




**mirror equation:** $1/p + 1/q = 1/f_{h,v}$ **Focal lengths:** $f_h = (R_h \sin\theta)/2$

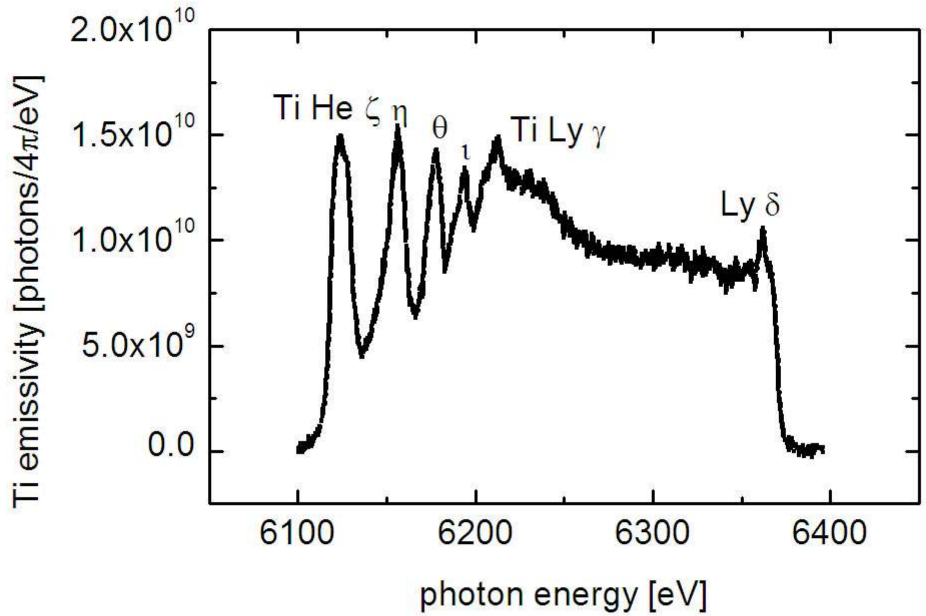
$f_v = R_v/(2 \sin\theta)$

**spectroscopic application: Δλ coverage requires source inside RC**

**spectra dispersed onto detector positioned at Rowland circle**

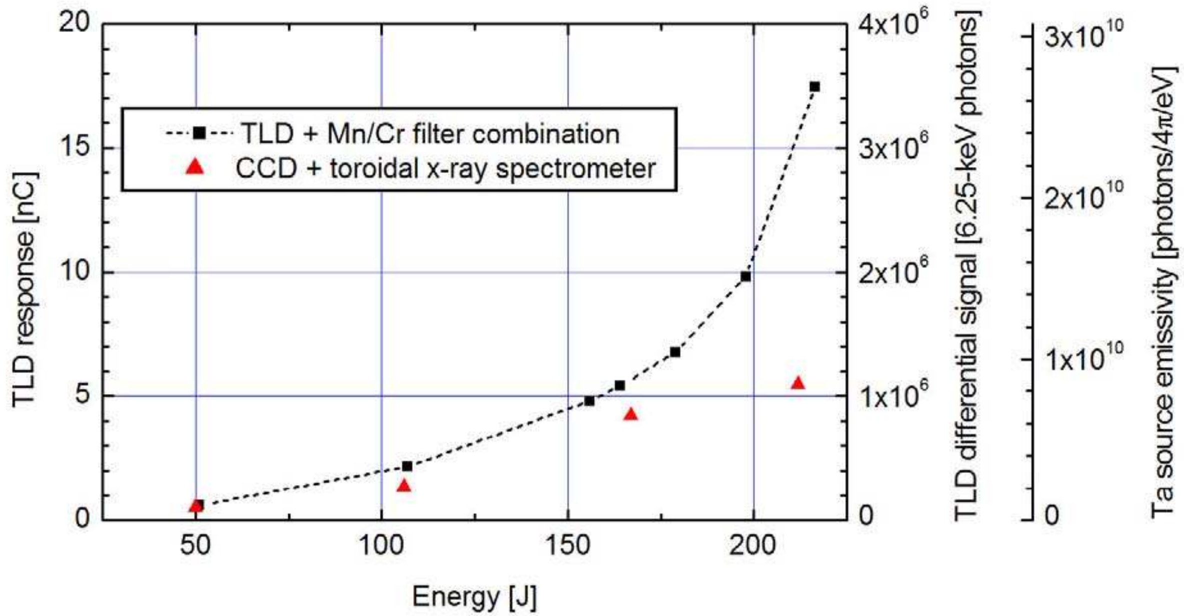
**combination of spectral and 1D spatial resolution**

# Spectroscopic Diagnostics Using TBS



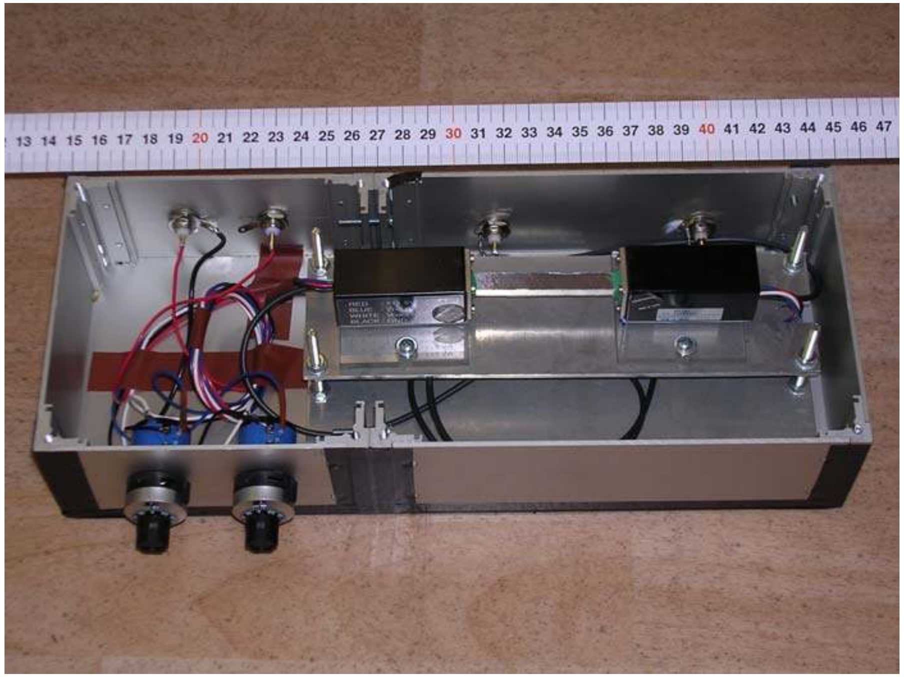

**GaAs (400) toroidally bent crystal, radii 450/305.9 mm (meridional/sagittal plane)**
**$R_{int}$ of GaAs (200) < 0.5 µrad, 20-µm-thick Al → suppression of 3 keV emission**
**spectral window of 250 eV centered around the photon energy 6243 eV**
**ray-tracing optimized transfer function: 1 photon emission → $10^{-5}$ photons/detector**
**CCD camera (PI-MTE, 1300×1340 pixels, pixel size 20×20 µm²)**
**alignment checked via overlap with spectral lines of Ti He *ζ-ι* and Ly *γ-δ***

# Absolute Calibration of the Ta Plasma Emissivity



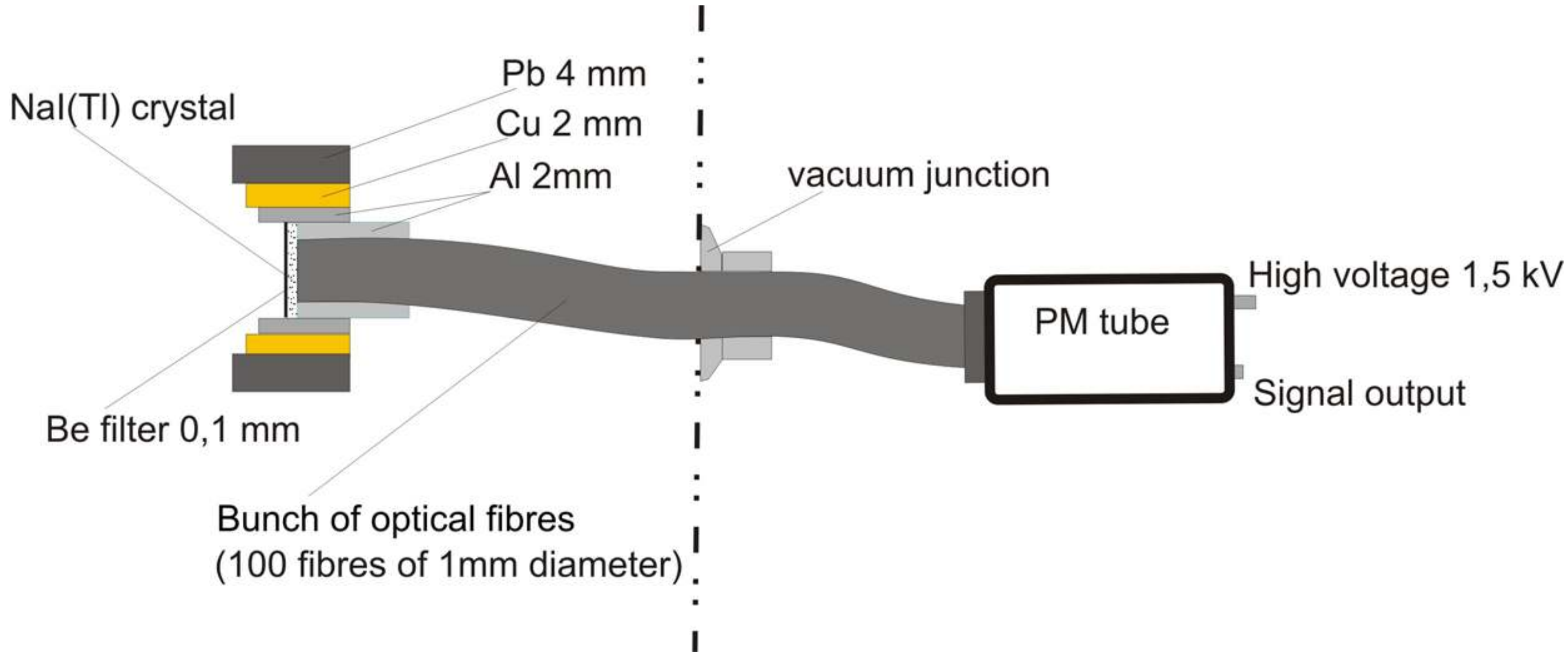

**TBS data vs. results of Differential Absorption Spectrometer**
**DAS: stack of filters (different thickness PMMA, Cr, Mn, Cu, Mo, Ag, and Ta foils)**
**LiF:Mg,Cu,P thermoluminescent detectors (TLD-600H, 3.2×3.2×0.9 mm$^3$)**
**filter combination Cr 30 µm and Mn 25 µm → spectral window 6.01-6.54 keV**
**calibrated with 5.9 keV radiation emitted from the $^{55}$Fe radionuclide**
**1.7×10$^{16}$ Wcm$^{-2}$: Ta emissivity at PALS by 5 orders of magnitude higher than that of Fedosejevs et al. (45 fs, 1 mJ, 1 kHz laser system)**

# Time-Resolving Detectors: Scintillators



plastic and $BaF_2$ scintillation crystals, active size 4×8×65 $mm^3$, $3\times10^{-3}$ srad
glued to two fast photomultipliers operating in a coincidence mode
LeCroy oscilloscope WavePro7000 (band width 1 GHz, sampling rate 1 GS/s)
duration of the fast signals shorter than 5 ns
calibrated using $^{55}$Fe (5.9 keV) and $^{241}$Am emitters (~60 keV)

# NaI(Tl) Scintillator




**NaI(Tl) scintillator (diameter 25 mm, thickness 1 mm)**
**protected by 0.15-mm-thick Be window, connected to a bunch of 100 optical fibers**
**outcoming signal amplified using the Hamamatsu H5783 photomultiplier**
**calibrated using $^{55}Fe$ radionuclide, fast optical switch designed**

# Semiconductor single photon pixel hybrid detector Medipix

- Single quanta counting position sensitive device
- Wide dynamic range & high count rate
  up to $10^{10}$ X–ray photons/cm$^2$/s
- X–rays: Direct photon detection
- Semiconductor (Si) high detection efficiency
  (100% for ≤ 10 keV x-rays)
- Room temperature & noiseless operation
  (≥ 5keV)
- Novel USB readout and power supply
  compact, portable, ease of use

- **Medipix2 - high spatial resolution CMOS pixel readout chip working in single particle counting mode**
- **combined with different semiconductor sensors converting ionizing particles directly into detectable electric signals**
- **High sensitivity, large dynamic range exceeds present charge integrating techniques**
- **Low sensitivity to dark currents allows long exposure times under very low intensity illumination (medical applications)**

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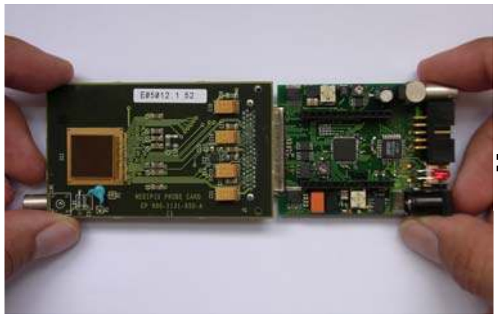

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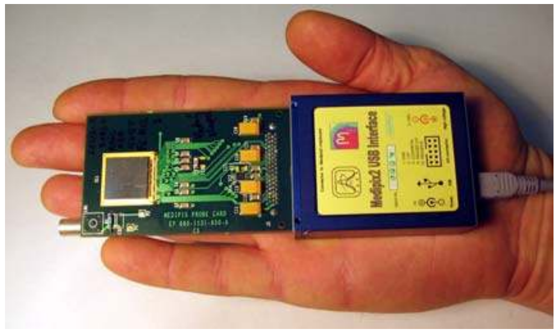
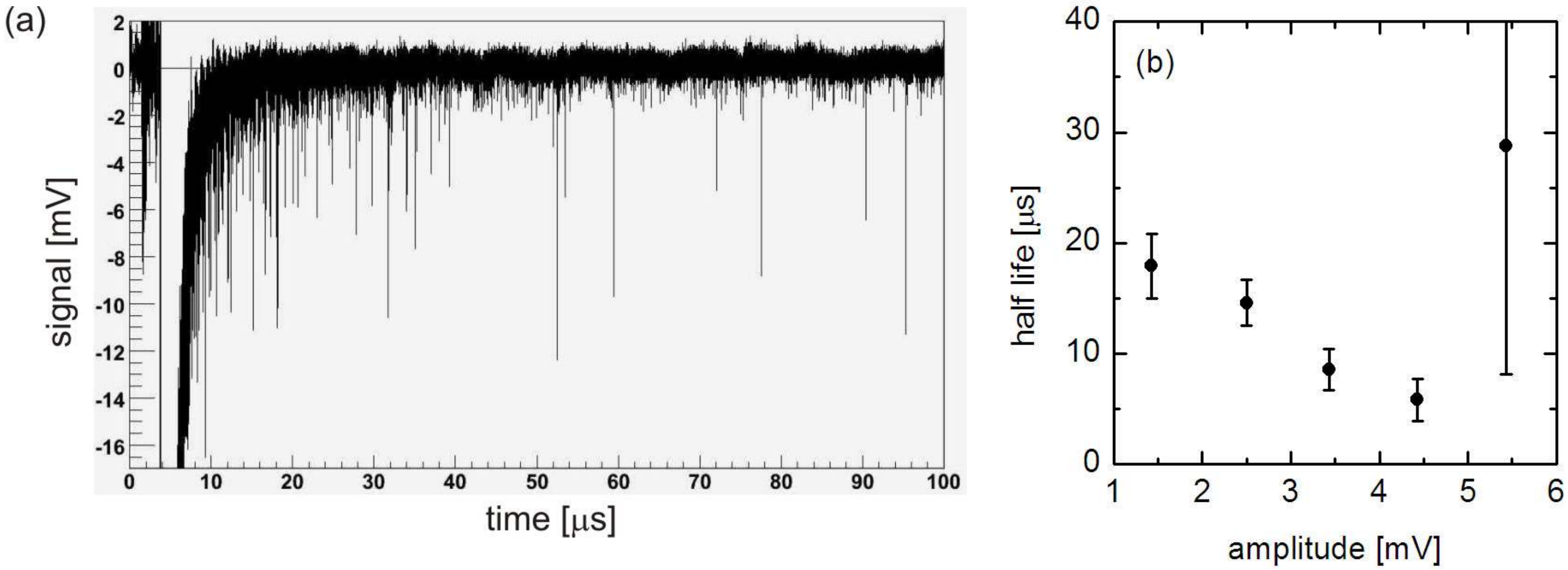



Vykydal, Z. , et. al., NIM A (2006)

# Example of Collected Data: $BaF_2$ Scintillator




**Detector calibration using the $^{55}Fe$ radionuclide → signal amplitudes corresponding to decay of nuclei $^{181}Ta$ expected at level of 1-2 mV**

**Alternate half-lives for different pulse amplitudes do not allow univocal interpretation of the measured signals**

**Collection of complex & reliable data should be subject of further research**

## Conclusions

- **Candidate nuclei for study of low-lying nuclear transitions were selected**
- **Pilot experiments directed to low-energy nuclear excitations at large-volume, long-duration plasmas were realized**
- **Feasibility of such measurements was indicated**
- **Hitherto experiments did not provide conclusive evidence for existence of low-lying nuclear transitions**
- **Experiments are difficult – giant radiative pulse precedes weak signals hidden in extremely noisy environment**
- **Attractive underlying physics justifies efforts connected with development of robust instrumentation**

**THANK YOU FOR YOUR ATTENTION!**