



Electronic structure and its effects on beta decay

Presented by A. Andoche, in collaboration
with X. Mougeot, P. A. Hervieux and Charlie
Jeudy

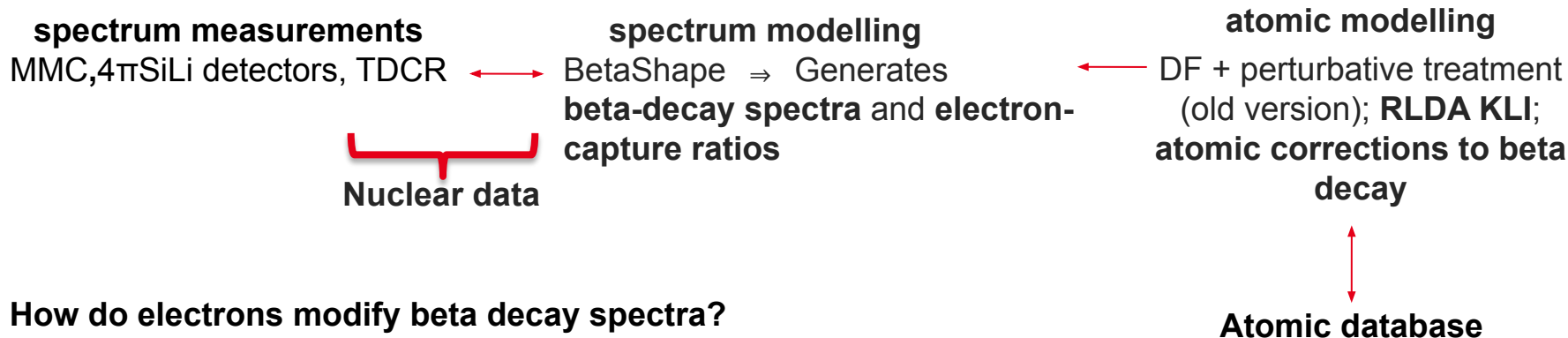


Henri Becquerel National Laboratory (LNHB)

■ The laboratory

LNHB is the French national laboratory for ionizing-radiation metrology. It is divided into two entities, MA and MD, dedicated to activity and dosimetry metrology. **LNHB-MA** develops activity standards, relying on source preparation techniques, experimental setups, and **evaluated atomic and nuclear data**.

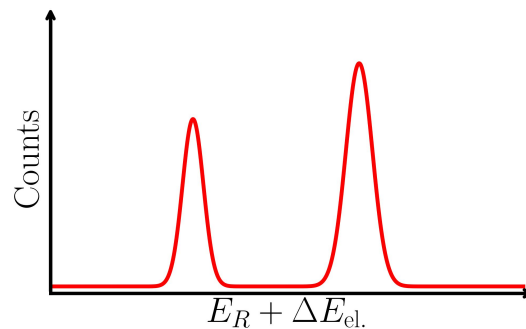
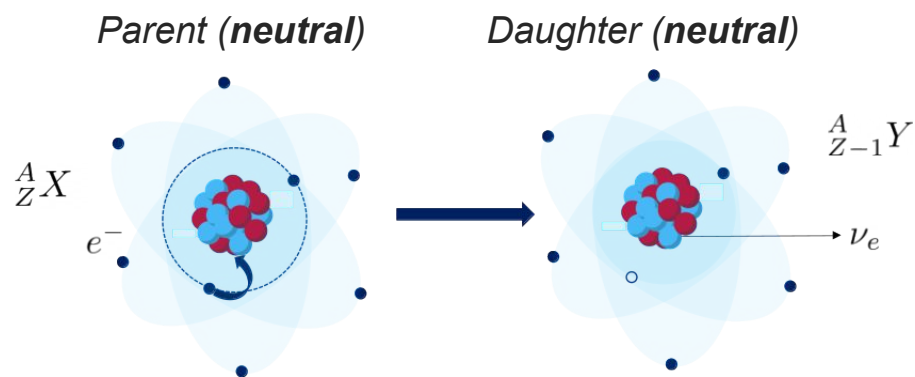
■ Atomic and nuclear data





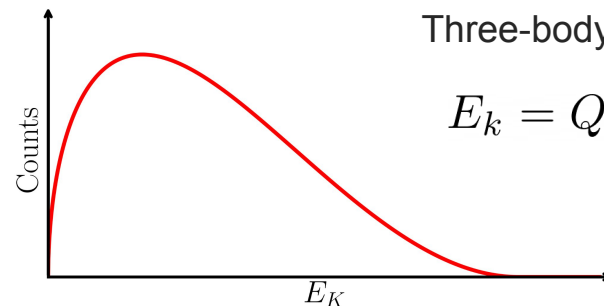
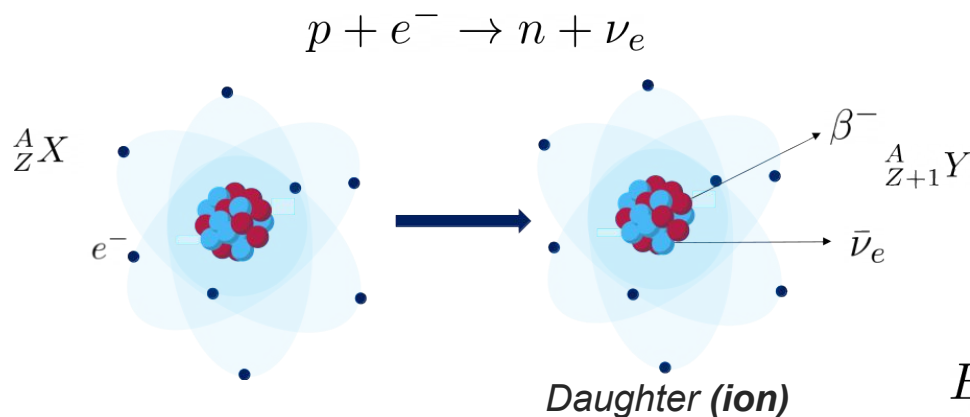
1 ■ Atomic effects on electron capture and beta decay

Beta and electron capture decay



Two-body process
 $\Rightarrow$ Sharp recoil peaks

$$E_R = Q_\epsilon - E_{\nu_e}$$



Three-body decay process

$$E_k = Q_\beta - E_R - E_{\bar{\nu}_e}$$

$$n \rightarrow p + \beta^- + \bar{\nu}_e$$

E_k Kinetic energy of the β particle

E_R recoil energy
 Q_ϵ reaction energy

$E_{\nu_e}/\bar{\nu}_e$ Neutrino/antineutrino energy

Beta decay and nuclear structure

“V-A” modelling of beta decay

$$\mathcal{H}_W = \frac{G_F V_{ud}}{\sqrt{2}} J_{\text{had}}^\mu J_\mu^{\text{lep}} + \text{h.c.}$$

$$J_\mu^{\text{lep}} = \bar{\psi}_e \gamma_\mu (1 - \gamma^5) \psi_\nu$$

$$J_{\text{had}}^\mu = \bar{\psi}_p \gamma^\mu (g_V - g_A \gamma^5) \psi_n$$

Accurate atomic and nuclear modelling is required to extract g_V and g_A , G_F , V_{ud} from measured spectra while reducing uncertainties and systematic biases.

In the simple framework of allowed and allowed-like beta and EC decay

Decay rate for electron capture

$$\lambda_h \propto C_h q_h^2 n_h \beta_h^2$$

q_h^2 momentum of the ejected neutrino

n_h **relative occupation number of subshell h**

C_h nuclear matrix elements

β_h^2 **Coulomb Amplitude**

β_h dominant contribution for ns and $np_{1/2}$ states

Beta decay spectra

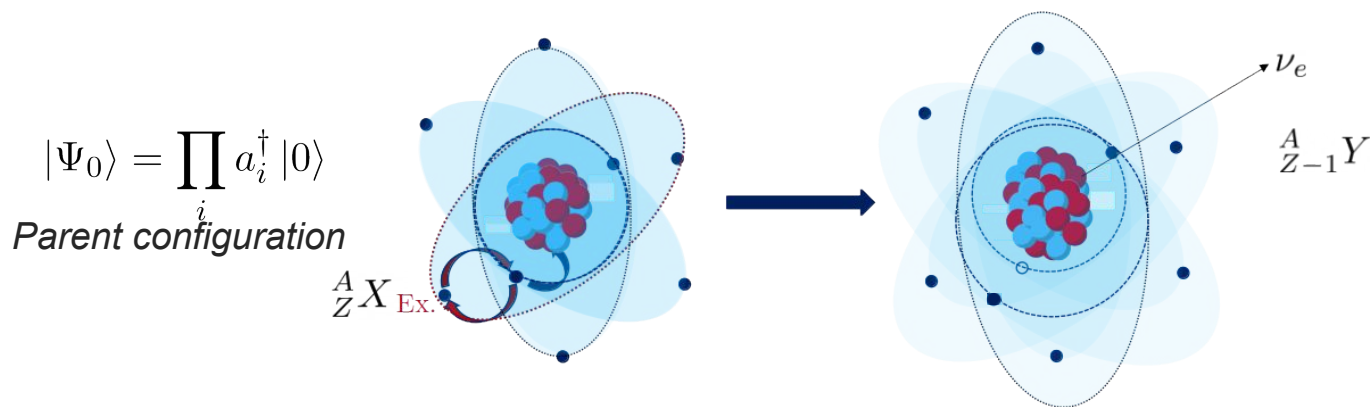
$$\frac{dN}{dW} \propto p(W) W q^2(W) F(Z, W) C(W)$$

$F(Z, W)$ **Fermi function**

W Energy of the β particle $1 \leq W \leq W_0$

Are there any additional effects of the electrons ?

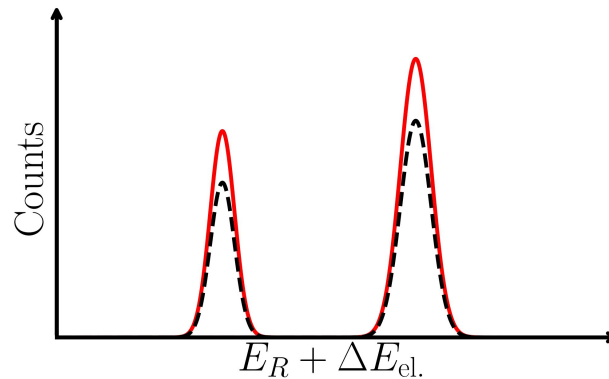
Exchange and overlap effects



$$|D_f^{(h)}\rangle = \prod_{j \neq h} \tilde{a}_j^\dagger |0\rangle$$

Daughter configuration

Exchange and overlap correction





The exchange and overlap corrections modelisation

1. **Bahcall (B)**: only $\{ns\}$ electrons, without explicit consideration of the ns state degeneracy

Bahcall, *Nuclear Physics* **71**, 1965

$$B_h^{(B)} = \left| \prod_{i \in ns, i \neq h} \langle (i_h)' | (i) \rangle - \sum_{j \in ns, j \neq h} \left(\frac{\beta_j}{\beta_h} \right) \langle (j_h)' | (h) \rangle \prod_{i \in ns, i \neq j, h} \langle (i_h)' | (i) \rangle \right|^2$$

2. **Vatai (V)**: considers the remaining N electrons, using an approximation

Vatai, *Nuclear Physics A* **156**, 1970

$$B_h^{(V)} = \left| \prod_{i \in N, i \neq h} \langle (i_h)' | (i) \rangle - \sum_{j \in N, j \neq h} \left(\frac{\beta_j}{\beta_h} \right) \langle (j_h)' | (h) \rangle \prod_{i \in N, i \neq j, h} \langle (i_h)' | (i) \rangle \right|^2$$

3. **Faessler (F)**:

Z trends (HF calculation)

$$B_h^{(F)} = \left| \sum_{\sigma \in S_N} (-1)^{\epsilon_\sigma} \frac{\beta_{\sigma(h)}^2}{\beta_h^2} \langle \Phi_h' | \Phi^\sigma \rangle \right|^2$$

Faessler, *J. Phys. G* **42**, 2015

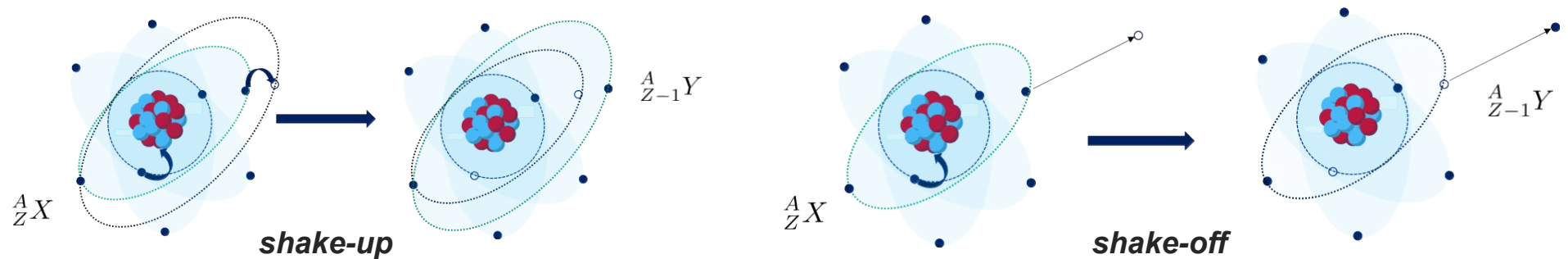
Multielectron overlaps

$$\langle \Phi_h' | \Phi^\sigma \rangle$$

Element	Bahcall B_K	Vatai B_K	Faessler B_K	Bahcall B_L	Vatai B_L	Faessler B_L
⁷ Be	0.913	0.838	0.871	3.203	2.345	2.402
³⁷ Ar	0.955	0.784	0.784	1.106	0.805	0.804
⁴⁰ Ca	0.980	0.977	0.977	1.140	1.111	1.113

contains individual electronic overlap $\langle (i_h)' | (j) \rangle$

Shake-up and shake-off processes



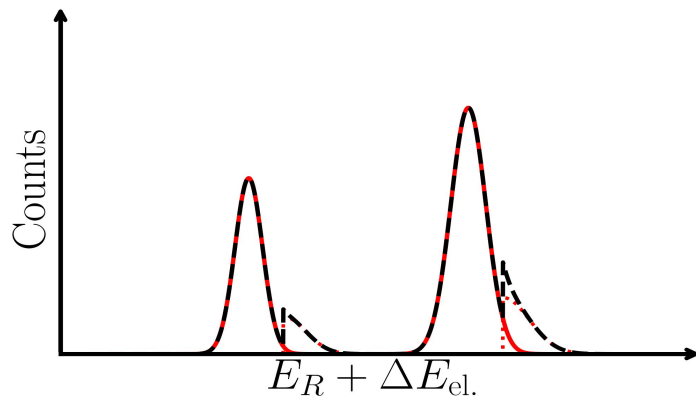
Shaking probability for electron (i), conditional on electron capture from subshell (h)

$$P_h^i = 1 - \langle (i_h)' | (i) \rangle^{2N_i} - \frac{N_i}{2k_i} \sum_{j \neq i} N_h'^j \langle (j_h) | (i) \rangle^2$$

T. A. Carlson and C. W. Nestor, *Phys. Rev. A*, 1973

Total shaking probability, conditional on electron capture of electron h

$$S_h = \sum_i \left[\sum_j P_h^{i \rightarrow j} + \int_E P_h^{i \rightarrow E} dE \right] = \sum_i P_h^i$$



Shake-off $\Rightarrow$ broadening of the electron-capture spectrum

Electron capture decay rate per subshell



Behrens and Buhning, *Nuclear Phys.* **A162**, 1971
N. C. Pyper, *Mathematical and Physical Sciences* **420**, 1988

$$\lambda_h \propto C_h q_h^2 n_h \beta_h^2 B_h (1 + S_h)$$

**Nuclear matrix element
and neutrino dependence**

q_h^2 momentum of the ejected
neutrino

C_h nuclear shape factor

**Capture state
dependency**

n_h relative occupation number
of electron in shell h

β_h **Coulomb amplitude**

**Many-body
corrections**

B_h **Exchange and
overlap corrections**

S_h **Shaking corrections**

All these **quantities** rely on approximations and accurate modelling. The nuclear shape factor is nearly independent of the capture channel for the transitions considered;

The dominant capture **channels are those involving** (ns and $np_{1/2}$) **subshells** for which the Coulomb amplitude reduce to a radial component of the capture wave function

Exchange and overlap corrections: impact on beta decay

Reminder

$$\frac{dN}{dW} \propto p(W) W q^2(W) F(Z, W) C(W)$$

Extension of the shape factor to L-th forbidden unique transition

$$C(W) = \left\{ \left[{}^V F_{000}^{(0)} \right]^2 \delta_{L,1} + \left[{}^A F_{L,L-1,1}^{(0)} \right]^2 \right\} \frac{R^{2(L-1)}}{[(2L-1)!!]^2 (2L-1)!} C_{ov} \sum_{k=1}^L \lambda_k \frac{p^{2(k-1)} q^{2(L-k)}}{(2k-1)! [2(L-k)+1]!} [1 + \eta_{-k} + \eta_{+k}]$$

$k = |\kappa|$

X. Mougeot, *Applied Radiation and Isotopes*, **201**, 111018, 2023

Setting $\eta_{\pm 1} = 0$, and $C_{ov} = 1$ □ turns off the exchange and overlap corrections.

AQE corrections (Atomic Quantum Exchange) refer to the η_{κ} individual exchange channels' contribution.

How are those corrections computed?

RLDA atomic modelling

Modèle RLDA-KLI-OEP⁴

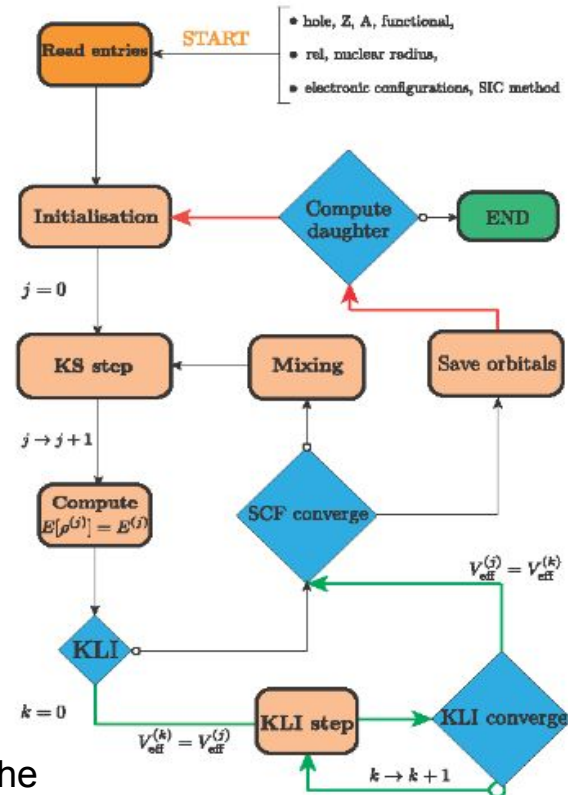
Relativistic Local Density Approximation: VWN default mode and other local exchange and correlation functionals; The exchange functional is the Slater with relativistic corrections; finite charge distribution (Fermi-like); Tong et. al., *Phys. Rev. A*, **57**, 855 - 1998

Treatment of self-interaction with the KLI potential → correct asymptotic behaviour of the effective potential

$$V_{\text{eff}}^{\text{KLI}}(r) = V_{\text{eff}}[\rho](r) + \frac{N_{\text{Norb}} \rho_{\text{Norb}}(r)}{\rho(r)} \{-V_{\text{H}}[\rho_{\text{Norb}}](r) - V_{\text{xc}}[\rho_{\text{Norb}}](r)\} + \sum_{i=1}^{N_{\text{orb}}-1} \frac{N_i \rho_i(r)}{\rho(r)} \times \left\{ -V_{\text{H}}[\rho_i](r) - V_{\text{xc}}[\rho_i](r) - \int_0^\infty \rho_i(r) \{-V_{\text{xc}}[\rho_i](r) - V_{\text{H}}[\rho_i](r)\} r^2 dr + \int_0^\infty \rho_i(r) V_{\text{eff}}^{\text{KLI}}(r) r^2 dr \right\}$$

Possible improvements

Implementation of Breit and retardation terms; Add an effective potential for the vacuum polarisation and self-energy terms compatible with RLDA.

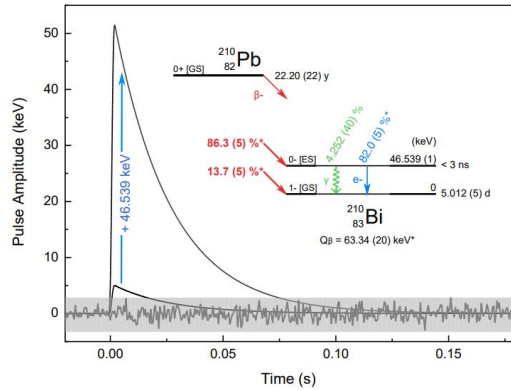


RLDA-KLI flowchart



2. ■ Electron capture spectra and beta-decay spectra with RLDA

Measurement and modelling of the ^{210}Pb β spectrum



Threshold-free ^{210}Pb beta spectrum

Low-energy $\Rightarrow$ test of the exchange and overlap

First forbidden non-unique transition

Two nuclear decay channels
main branch goes to a short-life excited state $\square$ shifted beta decay spectrum

Precision measurement and modelling of the threshold-free ^{210}Pb β spectrum

Shuo Zhang^{1,2}, Hao-Ran Liu³, Ke Han^{4,5*}, Xavier Mougeot^{6*}, Paul-Antoine Hervieux^{7*}, Tao Sun⁸, Wen-Tao Wu⁸, Robin Cantor⁹, Jing-Kai Xia², Zhi Liu², Jun-Cheng Liang³, Fu-You Fan³, Le Zhang¹⁰, Ming-Yu Ge¹¹, Xiao-Peng Zhou¹², Adrien Andoche⁷

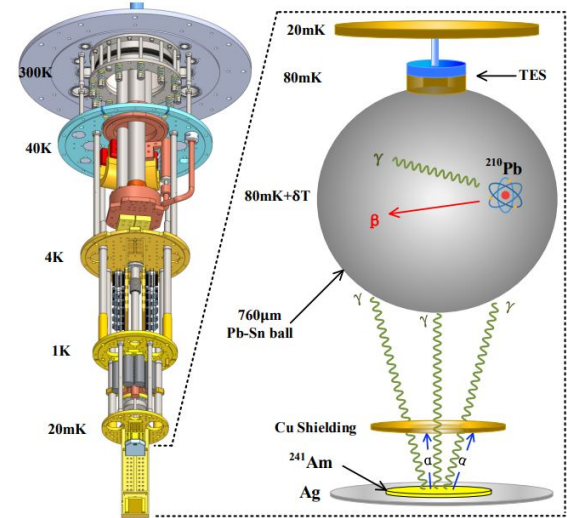


Fig. 2 Schematic drawing of the experimental setup. The micro-calorimeter is operated in the dilution refrigerator at a base temperature of 20 mK. The Pb-Sn ball acts both as the ^{210}Pb source and the energy absorber for the TES micro-calorimeter. An ^{241}Am γ source is placed 22 cm from the absorber with a copper shielding in between to stop α particles from ^{241}Am .

TES measurement

S. Zhang, H. Liu et al., [arxiv:2509.26390](https://arxiv.org/abs/2509.26390), 2025

Effects of the exchange channel on the shape of the ^{210}Pb β spectrum

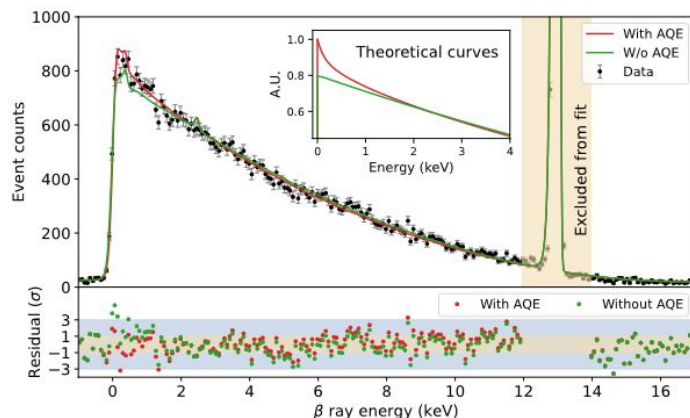


Fig. 5 The measured β -ES spectrum after shifting back to 0 keV starting point and theoretical β -ES calculations with (red curve) and without (green) AQE effect considered. The spectrum bin width is 80 eV. The fit curves include the β -ES signal and backgrounds, detailed in the appendix. The bottom panel shows residuals between theories and data. For each bin, the residual is calculated as (data-theory)/ $\sqrt{\text{data}}$.

S. Zhang, H. Liu et al., *arxiv:2509.26390*, 2025

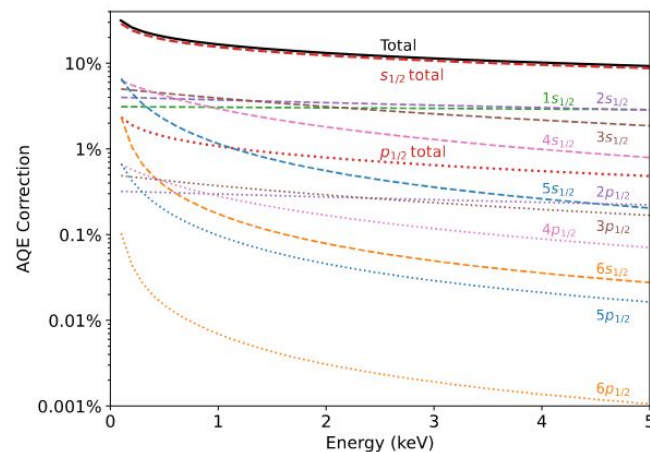
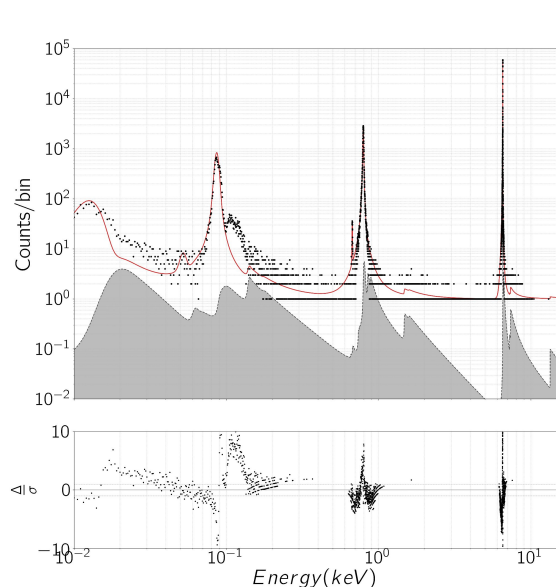


Fig. 7 Total atomic quantum exchange (AQE) correction (in percent), the total contributions of the $s_{1/2}$ and $p_{1/2}$ orbitals, and the contributions of each subshell. The $s_{1/2}$ orbitals predominantly contribute the total AQE correction.

The **dominant** AQE contribution is from the ***ns*** states

Valence-state contributions to the AQE are localised in the low-energy region (0–2 keV)

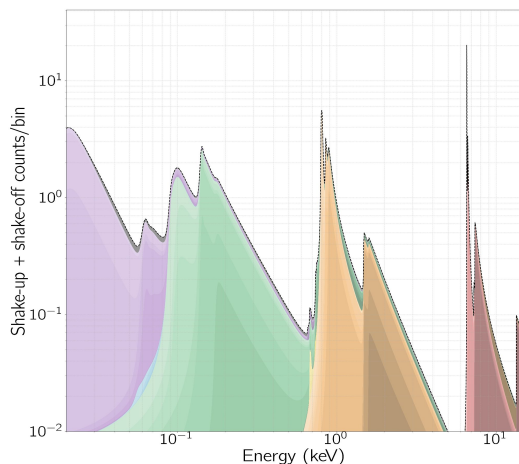
Bolometric spectrum of ^{55}Fe electron capture (preliminary)



Semi-empirical spectra

$$\frac{dN}{dE_f} \propto (Q - E_c)^2 \sum_f \lambda_0 B_f \mathcal{L}(E_c; \bar{E}_f, \Gamma_f)$$

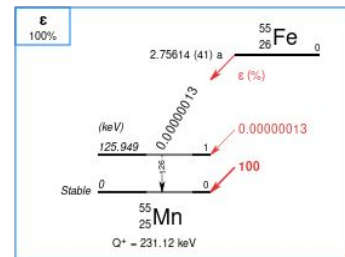
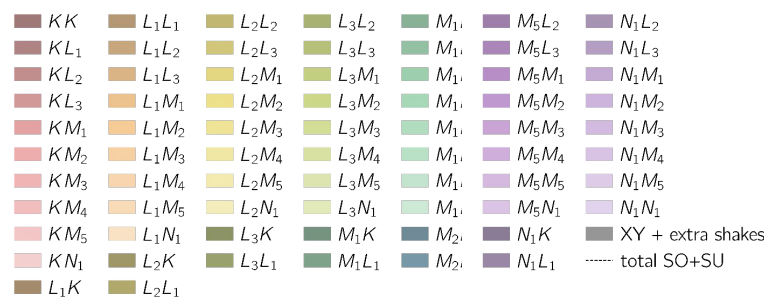
A. Faessler et al. , *Phys. Rev. C*, **95**, 045502, 2017



Black: MMC measurement PTB (PRIMA-LTD), 1-2Bq of ion
implante ^{55}Fe source per absorber, 8 gold absorbers of

Red: computed spectra

Shaded area: shake-off/up contributions

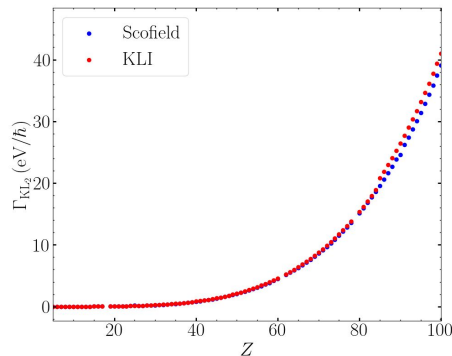


See M. Rodrigues
presentation
for more information
on MMC

Technical details

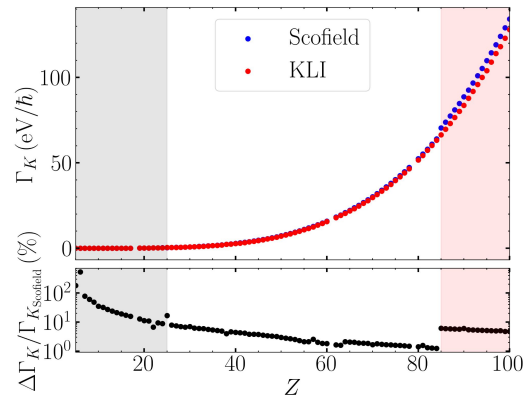
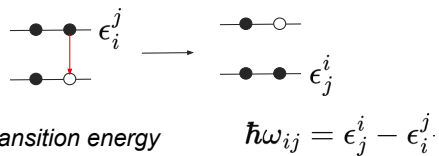
- ❖ Hole linewidths Γ_f are taken from the EADL Dirac–Fock tables.
- ❖ All atomic electrons are included in the calculation.
- ❖ Shake-off is calculated using the daughter atom's effective KLI potential.

Radiative and non-radiative rates



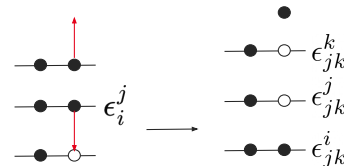
Example: KL₂ J. H. Scofield, *PR* **179**, 9, 1969

Radiative transitions



Total radiative rates $\Gamma_i = \sum_j n_o(j) \Gamma_{ij}$

Non radiative transitions



Auger energy $\hbar\omega_{ijk}^{kj} = \epsilon_j^k + \epsilon_j^j - \epsilon_j^i$

- All K, L, M, N, O and P X-Ray lines have been determined individually (~40k transitions)
- Auger lines will soon be compared with EADL for Z=2-100 using Coulomb-Breit, jj coupling scheme.
- Significant deviations at low Z (shaded area) $\Rightarrow$ numerical precision, and/or overestimation of correlation effects by the KLI model.



3 ■

**Effect of the chemical
environment on the
electron-capture ratio
of ^7Be**

Failure of the atomic descriptions for Be, EC Decay

■ Publication of P_L/P_K capture ratios and shaking probabilities

Benchmarking of RLDA-KLI

Element	Kotochigova	KLI	MCDF	Experimental
⁷ Be	-3.856	^{1s_{1/2}} -4.815	-4.733 [-4.753] ^a	-4.384
³⁷ Ar	-114.08	-118.14	-119.05	-117.704 (11)
⁴⁴ Ca	-144.40	-148.93	-150.05	-148.57 (7)
⁵⁴ Mn	-235.08	-240.81		-240.70 (6)
⁵⁵ Fe	-255.90	-261.88		-261.802 (37)
¹⁰⁹ Cd	-968.69	-981.42	-985.50	-981.619 (11)
¹²⁵ I	-1204.13	-1218.71		-1218.953 (15)
¹³⁸ La	-1414.24	-1430.37		-1430.453 (15)
⁷ Be	-0.206	^{2s_{1/2}} -0.377	-0.309 [-0.349] ^a	-0.343
³⁷ Ar	-10.86	-11.74	-12.41	-11.9803 (33)
⁴⁴ Ca	-15.16	-16.14	-16.26 (7)	-16.26 (7)
⁵⁴ Mn	-27.22	-28.38	-28.72 (6)	-28.72 (6)
⁵⁵ Fe	-29.99	-31.20	-31.49 (7)	-31.49 (7)
¹⁰⁹ Cd	-143.97	-146.64	-147.659 (11)	-147.659 (11)
¹²⁵ I	-186.24	-189.33	-190.659 (11)	-190.659 (11)
¹³⁸ La	-225.36	-228.79	-230.282 (18)	-230.282 (18)
³⁷ Ar	-8.50	^{2p_{1/2}} -9.40	-9.62	-9.2075 (26)
⁴⁴ Ca	-12.375	-13.394	-13.721	-13.252 (29)
⁵⁴ Mn	-23.368	-24.606		-24.357 (37)
⁵⁵ Fe	-25.92	-27.21		-26.937 (37)
¹⁰⁹ Cd	-134.23	-137.12	-138.71	-136.965 (11)
¹²⁵ I	-174.97	-178.32		-178.311 (11)
¹³⁸ La	-212.81	-216.51		-216.476 (15)

^aWith correlations.

TABLE I. Binding energies (in atomic units) of the innermost orbitals for Kotochigova [26,27] (RLDA approximation, neutral atoms), KLI, and MCDF models. Experimental values are from Ref. [53] for ⁷Be to ⁵⁵Fe atoms (in gaseous or vapor state) and from Ref. [54] for ¹⁰⁹Cd to ¹³⁸La atoms (in elemental or oxide state).

TABLE II. Comparison of calculated and measured capture probabilities for different isotopes considered in the present work. The three models and the experimental values are described in the text.

Isotope	Energy (keV)	Quantity	Experimental	BetaShape	KLI	MCDF
⁷ Be	861.89 (7)	P_L/P_K	0.070 (7)	0.105 (8)	0.168 (12)	0.131 (10) [0.127 (11)] ^b
³⁷ Ar	813.87 (20)	P_L/P_K	0.09750 (46)	0.09260 (45)	0.0991 (9)	0.0922 (26)
⁴⁴ Ca	421.64 (14)	P_L/P_K		0.09800 (40)	0.1053 (8)	0.1064 (9)
⁵⁴ Mn	542.3 (10)	P_L/P_K	0.1044 (27)	0.11219 (31)	0.1088 (8)	
		P_K	0.901 (6)	0.88419 (34)	0.8849 (10)	
⁵⁵ Fe	231.12 (18)	P_L/P_K	0.1165 (9)	0.11629 (31)	0.1134 (8)	
		P_M/P_K	0.01786 (29)	0.01824 (12)	0.01991 (36)	
		P_M/P_L	0.1556 (26)	0.1568 (11)	0.1756 (34)	
¹⁰⁹ Cd	127.1 (18)	P_{LMNO}/P_K	0.2279 (21)	0.2273 (15)	0.2384 (17)	0.2374 (14)
		P_K	0.815 (2)	0.8148 (14)	0.8075 (16)	0.8081 (13)
¹²⁵ I	150.28 (6)	P_K	0.7971 (14)	0.79927 (41)	0.7959 (5)	
¹³⁸ La	312.59 (34)	P_L/P_K	0.391 (3)	0.3913 (26)	0.4163 (10)	
		P_M/P_K	0.102 (3)	0.0965 (10)	0.1018 (5)	
		P_M/P_L	0.261 (9)	0.2466 (30)	0.2445 (13)	

^aWith correlations.

Influence of atomic modeling on electron capture and shaking processes

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■ ⁷Be: a light nucleus of major interest

$P_L/P_K = 4.0(1)\%$ (HgTe matrices) P.A. Voytas et al., *Phys. Rev. L*, **57**, 2003

$P_L/P_K = 7.0(7)\%$ (Ta matrices phase III) S. Fretwell et al., *Phys. Rev. L*, **2020**

A. Andoche, L. Mouawad, et al., *Phys. Rev. A* **109**, 2024

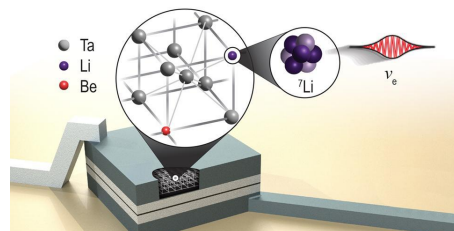
P. Indelicato and J. P. Desclaux, *Phys. Rev. A* **42**, 1990

The BeEST experiment

${}^7\text{Be}$ is a good candidate for Beyond Standard Model (BSM) tests

1. Lifetime of **53 days**
2. Two-body EC decay with a “**monochromatic**” recoil
3. Q-value (**811.815 keV**) < $2m_e c^2 \Rightarrow$ no competition with β^+ decay

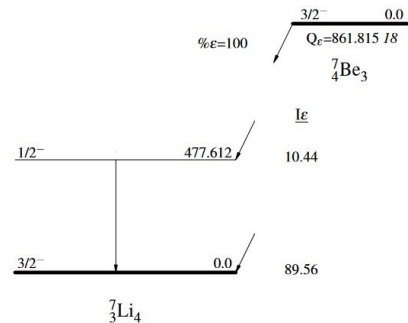
J. Smolsky, K. G. Leach, et al., *Nature* **638**, 2025



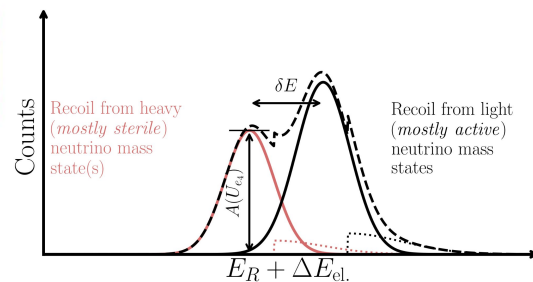
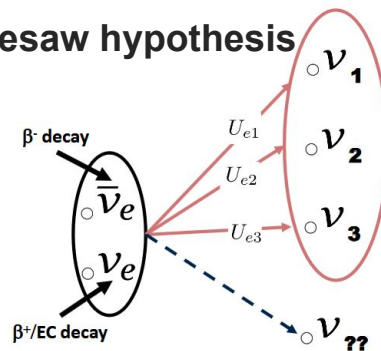
Direct experimental constraints on the spatial extent of a neutrino wavepacket

Joseph Smolsky, Kyle G. Leach, Ryan Abell, Pedro Amaro, Adrien Andoche, Keith Borbridge, Connor Bray, Robin Cantor, David Diercks, Spencer Fretwell, Stephan Friedrich, Abigail Gillespie, Mauro Guerra, Ad Hall, Cameron N. Harris, Jackson T. Harris, Leendert M. Hayen, Paul-Antoine Hervieux, Calvin Hinkle, Geon-Bo Kim, Inwook Kim, Amii Lamm, Annika Lennarz, Vincenzo Lordi, ... William K. Warburton

Nature **638**, 640–644 (2025) | [Cite this article](#)



Seesaw hypothesis



$$f(E_R) = (1 - A(U_{e4})) f_0(E_R) + A(U_{e4}) f_0(E_R - \delta E)$$

Determination of a set of $(U_{e4}, \delta E)$

The recoil spectra (1)

Before rescaling

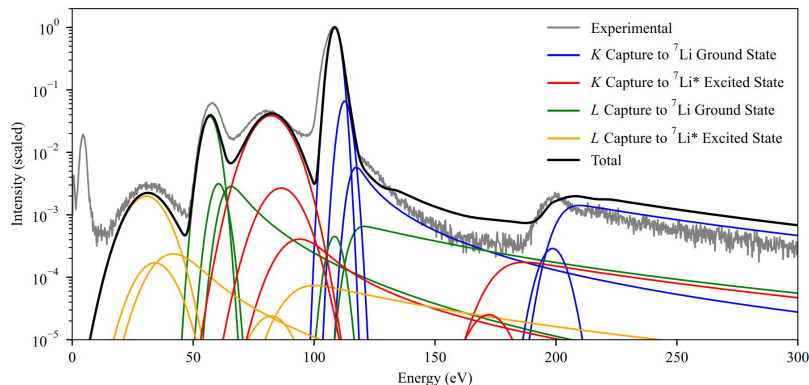


FIG. 8. Simulation of the full ${}^7\text{Be}$ electron capture spectrum in the 0- to 300-eV range, normalized to 1 and compared to a normalized one-day spectrum from phase-III of the BeEST experiment. All four primary peaks are accompanied by L shakeup, L shakeoff, K shakeup, and K shakeoff spectra. The atomic structure calculations were shifted in energy and convolved with Gaussian responses to match the experimental data. The L capture spectra were scaled by the L/K capture ratio of 0.070(7) [44] and the excited-state spectra by the branching ratio of 10.44% [41].

Theoretical L/K capture ratio doesn't match the measure one

Shake probabilities are high for this light nuclei

M. Guerra et al., *Phys. Rev. C* 2026

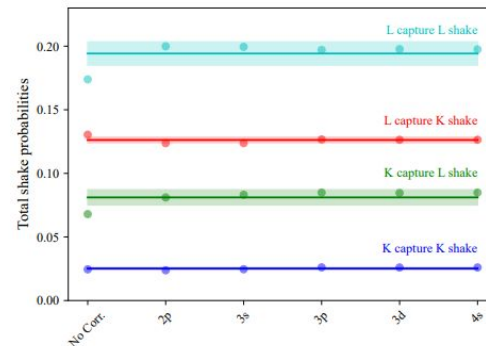
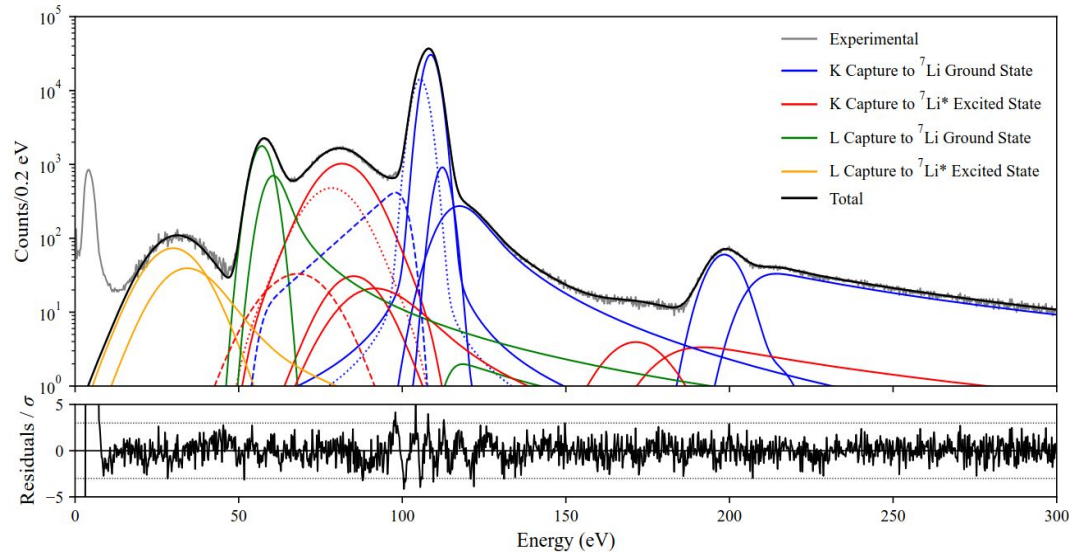


Figure 3. Total shake probabilities according to Eq. (7) as a function of the size of the basis space. All the single and double excitations up to the $4s$ orbital are included. The colored bands represent the 1σ intervals.

The recoil spectra (2)

After rescaling



M. Guerra et al., *Phys. Rev. C* 2026

- Computed using the **MCDFGME** updated program to precisely compute ionised states of Li
- Measurements were performed at the **TRIUMF-ISAC** facility using Ta STJs with implanted Be sources.
- All peaks are clearly identified, including their positions and origins, allowing sterile-neutrino searches in the residual background.

However, the **capture-peak amplitudes are rescaled** to provide a new L/K ratio **0.0756(20)**

What is the chemical environment of the implanted Be ?

VASP calculations □ optimisation of the crystalline structure for different implantation types

Material Effects on Electron-Capture Decay in Cryogenic Sensors

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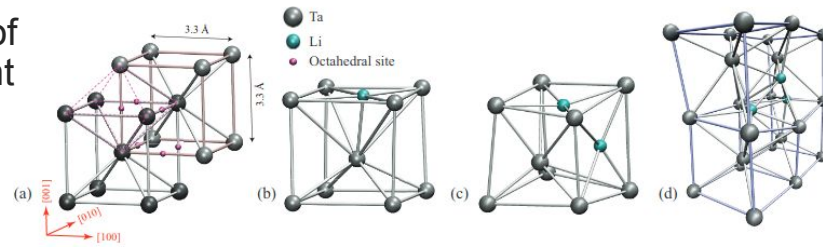
Several current searches for physics beyond the standard model are based on measuring the electron-capture (EC) decay of radionuclides implanted into cryogenic high-resolution sensors. The sensitivity of these experiments has already reached the level where systematic effects related to atomic state energy changes from the host material are a limiting factor. One example is a neutrino mass study based on the nuclear EC decay of ^7Be to ^7Li inside cryogenic Ta-based sensors. To understand the material effects at the required level, we use density-functional theory to model the electronic structure of lithium atoms in different atomic environments of the polycrystalline Ta absorber film. The calculations reveal that the Li 1s binding energies can vary by more than 2 eV due to insertion at different lattice sites, at grain boundaries, in disordered Ta, and in the vicinity of various impurities. However, the total range of Li 1s shifts does not exceed 4 eV, even for extreme amorphous disorder. Furthermore, when investigating the effects on the Li 2s levels, we find broadening of more than 5 eV due to hybridization with the Ta band structure. Material effects are shown to contribute significantly to peak broadening in Ta-based sensors that are used to search for physics beyond the standard model in the EC decay of ^7Be , but they do not explain the full extent of observed broadening. Understanding these in-medium effects will be required for current- and future-generation experiments that observe low-energy radiation from the EC decay of implanted isotopes to evaluate potential limitations on the measurement sensitivity.

Samanta *et al.*, *Phys. Rev. Applied* **19**, 2023

Calculation performed for Li@Ta for a density of state analysis (number of electrons at a given energy)

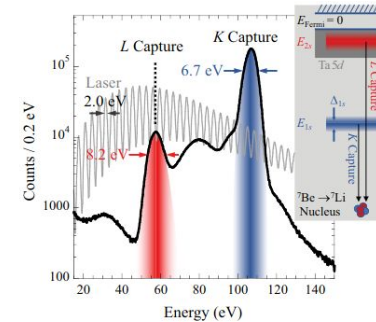
n_h occupation number $\longrightarrow$ density of states

How to compute the effect of the Neighbouring atoms on the decay (i.e. electronic sector)?



Five different configurations based on the lowest formation energies

Li site	1s Energy Shift, Δ_{1s} [eV]	Formation Energy, E_f [eV]	Defect Binding Energy per Atom, E_b [eV/Li]
Substitutional	0	1.23	—
Single interstitial	1.18	4.07	—
Two interstitials	1.58	7.63	-0.26
Three interstitials	1.30	11.23	-0.33
$\text{Li-Ta}-V_{\text{Ta}}$ complex	1.67	2.50	-1.53



Chemical environment approach

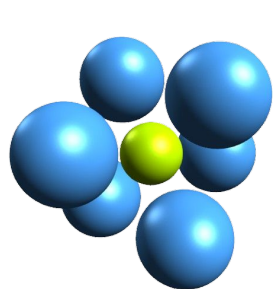


GAUSSIAN setup

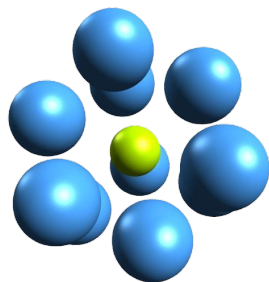
Geometry: extraction of the nearest neighbors from a frozen crystal geometry

Atomic model: Hartree–Fock with the def2-TZVPP basis set

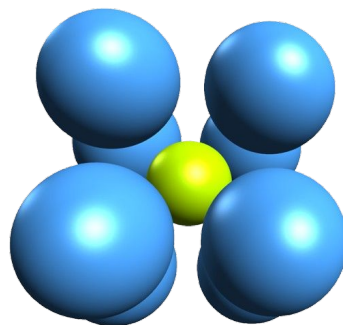
Blue spheres: ^{181}Ta
Green spheres: ^7Be



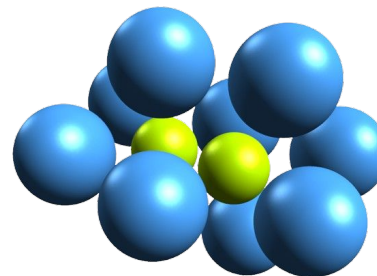
Octa



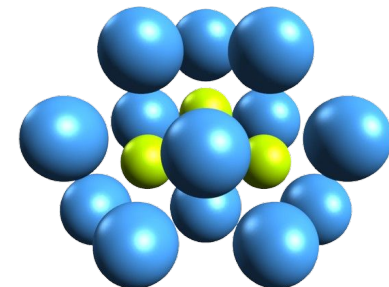
SubVac



Sub



2-Octa



3-Octa

M. J. Frisch *et al.*, **Gaussian 09, Revision E.01**, Gaussian, Inc., Wallingford CT, 2009.

Single Be implantation

Multiple Be implantation

Recentering of the molecular orbitals (MOs)

1. Gaussian provides MOs expressed in an atomic basis set, itself expressed on spherical gaussian centered at each nuclear position $\mathbf{r}_j$

$$\psi_i(\mathbf{r}) = \sum_j A_i^j \phi(\mathbf{r}_j - \mathbf{r})$$

2. Recentering the molecular orbitals on the decaying nucleus

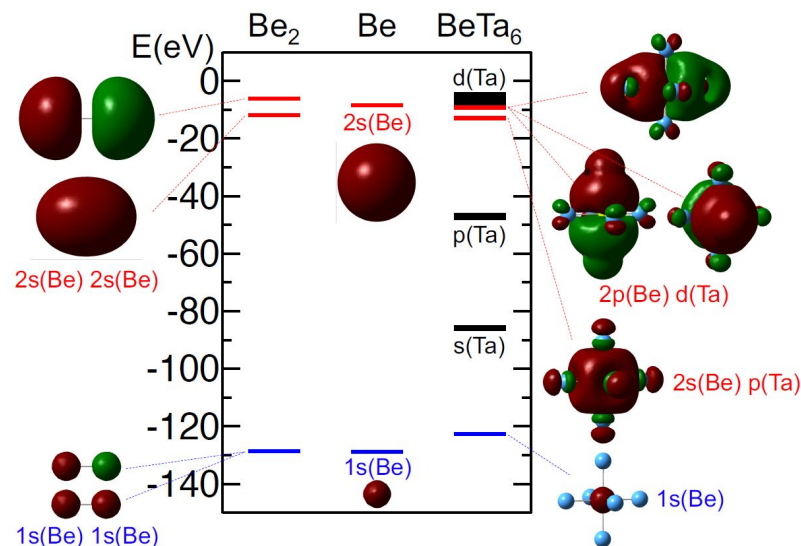
$$\psi_i(\mathbf{r}) = \sum_{\ell=0}^{\ell_{\max}} \sum_{m_{\ell}=-\ell}^{\ell} \tilde{R}_{\ell m_{\ell}}^i(r) Y_{\ell m_{\ell}}(\hat{\mathbf{r}})$$

3. Verification of the orthogonality relations

$$\langle \psi_i | \psi_j \rangle = \delta_j^i \pm \epsilon_j^i$$

Example (Octa):

$$\ell_{\max} = 12 \quad \epsilon_j^i \leq 2\%$$



Hybridisation and delocalisation □ L and K capture do not have the same physical meaning

Could we get anything from those states ?

Estimation of the Coulomb amplitude



The dominant contribution to the Coulomb amplitude

$$\tilde{R}_{\ell m_\ell}^i(R) \approx \tilde{R}_{00}^i(0) \approx \beta_i, \quad R \ll a_0$$

Ratios of Coulomb amplitude

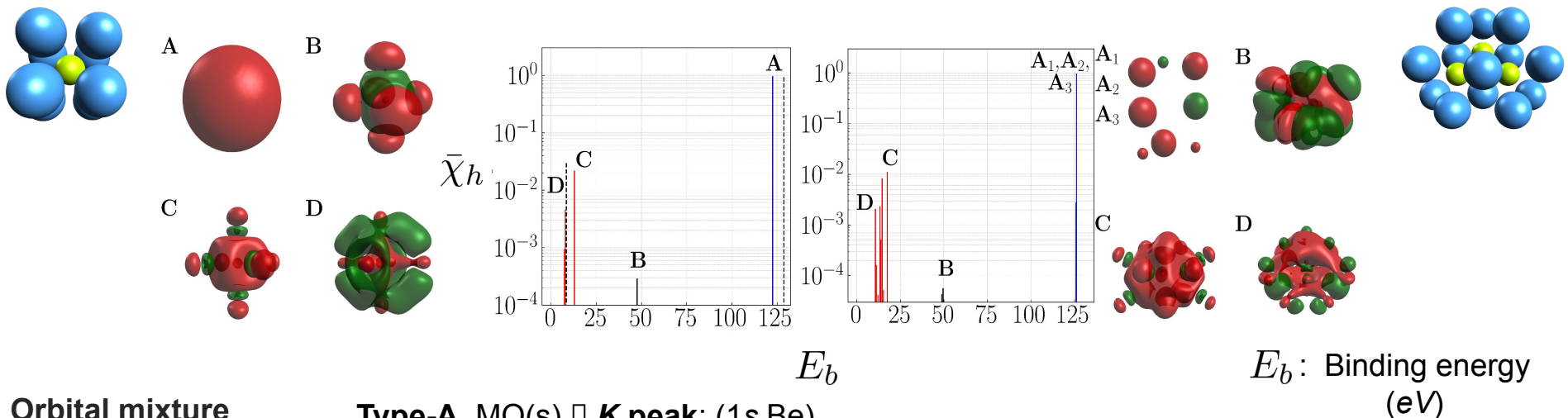
$$\beta_{i/j}^2 = \frac{|\beta_i|^2}{|\beta_j|^2} \approx \frac{|\tilde{R}_{00}^i(0)|^2}{|\tilde{R}_{00}^j(0)|^2}$$

Contribution to electron density at the nuclear center

One nuclear center $\bar{\chi}_h = \frac{\beta_h^2}{\sum_{j=1,N} \beta_j^2}$

Cluster with more than one site: $\bar{\chi}_h \rightarrow \bar{\chi}_h^{(b)}$ (one dedicated expansion for each site b)

MO composition: subspace discrimination



Estimation of the L/K capture ratio

Recall (in vacuum):

$$\frac{P_L}{P_K} \approx \beta_{L/K}^2 X_{L/K} \frac{(1 + S_L)}{(1 + S_K)} \xrightarrow{X_{L/K} \equiv \frac{B_L}{B_K}} \text{Ratio of Coulomb amplitude per Be atom in a cluster}$$

$$\beta_{L/K}^{2(b)} = \frac{\bar{\chi}_L^{(b)}}{\bar{\chi}_K^{(b)}}$$

Technical difficulties

1. Determination of the $X_{L/K}$ requires calculations of excited Li@Ta is computationally demanding. In vacuum (with HF)

Element	Bahcall $X_{L/K}$	Vatai $X_{L/K}$	Faessler $X_{L/K}$
⁷ Be	3.507	2.797	2.757

2. Loss of perfect orthogonality after recentering $\square$ incorrect overlap.
3. Ratio of probabilities is proportional to $\beta_{L/K}^2$ and ratio of exchange and overlap corrections $X_{L/K}$

Estimate of the L/K capture ratio for Be@Ta

After averaging over all Be sites

$$\langle \beta_{L/K}^2 \rangle = \frac{1}{N_{Be}} \sum_{b=1}^{N_{Be}} \beta_{L/K}^{2(b)}$$

Conf.	Vacuum	Octa	2-Octa	3-Octa	Sub	SubVac
$\langle \beta_{L/K}^2 \rangle$ (%)	3.32	2.80	2.47	2.98	2.36	1.81

Estimation of the L/K ratio

Configurations	Vacuum	Octa	2-Octa	3-Octa	Sub	SubVac	Experiment
P_L/P_K (%)	12.97	10.92	9.63	11.62	9.20	7.06	7.056(20)

NB: This estimate assumes that, within a given chemical environment, $X_{L/K}$ and $S_{L/K}$ corrections remain unchanged.

Element	Bahcall $X_{L/K}$	Vatai $X_{L/K}$	Faessler $X_{L/K}$
⁷ Be	3.507	2.797	2.757