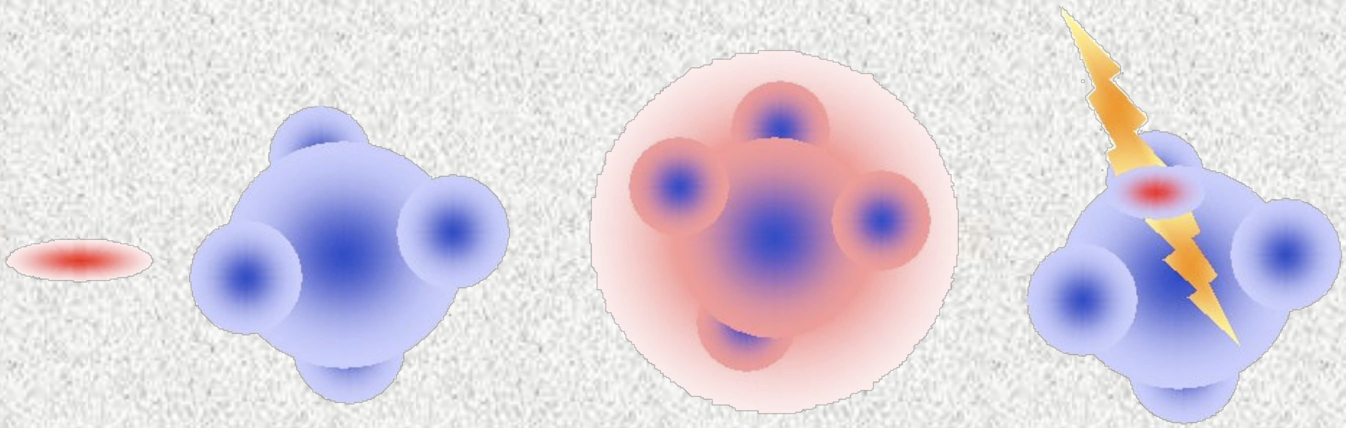
Resonant Positron Annihilation on Molecules and Relevance to Astrophysics



Cliff Surko[+]

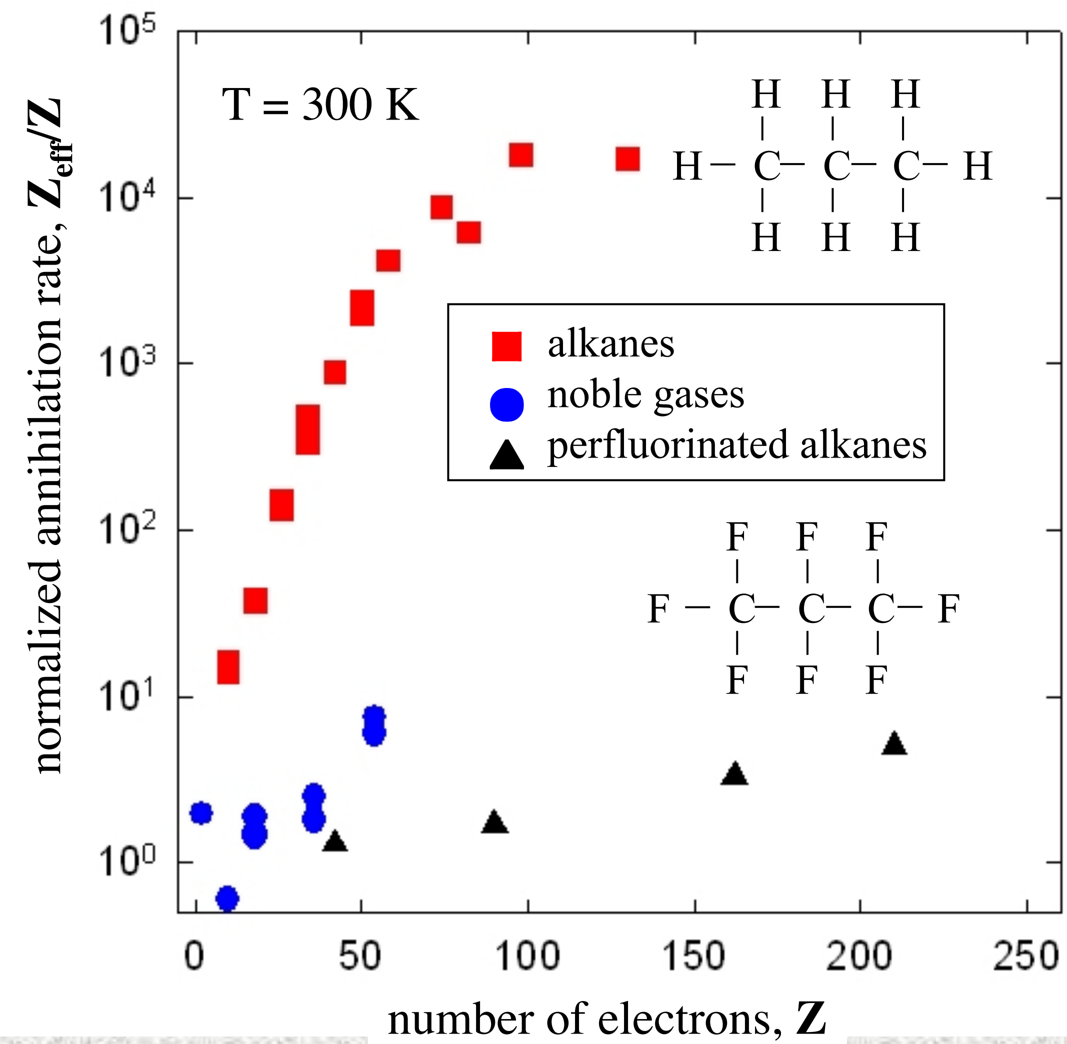
University of California, San Diego

Work supported by the U. S. National Science Foundation

[+]In collaboration with J. R. Danielson, A. C. L. Jones, and M. R. Natisin (UCSD); and Gleb Gribakin (Queens University, Belfast)

Positron Annihilation in Molecules

a mystery for many decades



Normalized annihilation rate, Z_{eff}:

$$Z_{eff} = \frac{\Gamma}{\pi r_0^2 c n_m}$$

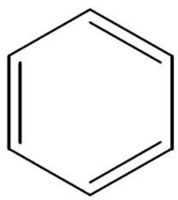
Expect $Z_{eff} \sim Z$

Find Z_{eff} >> Z

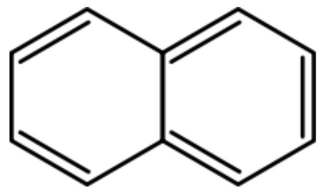
Iwata *et al*., PRA (1995)

C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

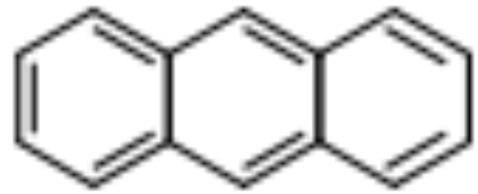
Benzene and
Polycyclic Aromatic Hydrocarbons (PAH molecules)



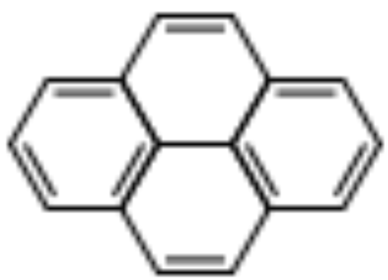
Benzene, C_6H_6
$Z_{eff} = 1.5 \times 10^4$



Naphthalene, $C_{10}H_8$
$Z_{eff} = 4.9 \times 10^5$



Anthracene, $C_{14}H_{10}$
$Z_{eff} = 4.3 \times 10^6$



Pyrene, $C_{16}H_{10}$

Coronene, $C_{24}H_{12}$

Z_{eff} for thermal positrons at 300 K

Astrophysical Consequences of Positron Annihilation on PAH Molecules

Iwata, Greaves, Surko, Can. J. Phys., 1996

Guessoum, Jean, Gillard, Astron. & Astrophys., 2005

Guessoum, Jean, Gillard, Applied Surface Science, 2006

Guessoum, Jean, Gillard, Mon. Not. R. Astron. Soc, 2010

What Determines Positron-Molecule Annihilation Rates?

- Observe large enhancements in annihilation rates

  (e.g., up to 10^4 x simple collision models)

- Answer is vibrational resonances - we have a good theory of single-mode vibrational Feshbach resonances (VFR)

- There are also enhancements due to IVR (intramolecular vibrational energy redistribution), but there's more:

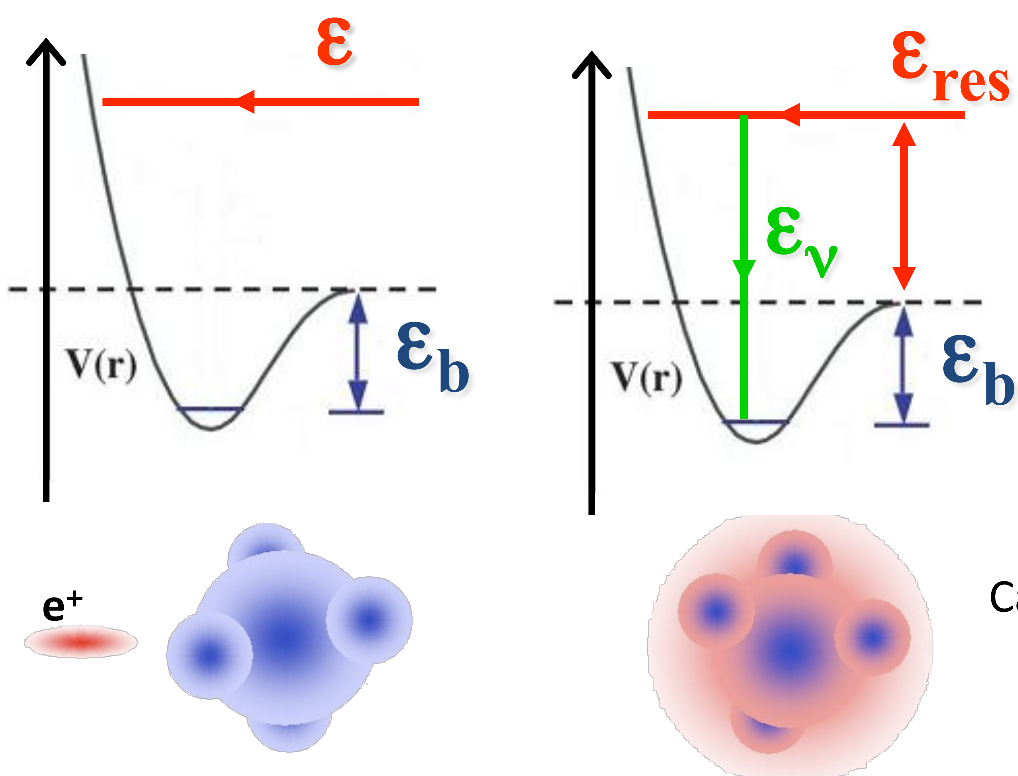
- A third, generic mechanism, Statistical Multimode Resonant Annihilation (SMRA) is present in most spectra and frequently dominates – directly relevant to PAH in the ISM

Vibrational Feshbach Resonances and Annihilation*

- positron with energy = ε
- bound state energy = ε_b
- molecular vibration energy = ε_ν

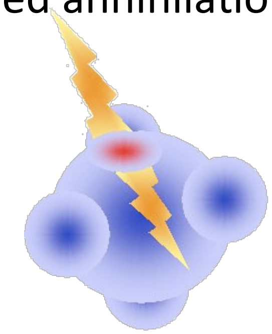
- Resonance condition:

$$\varepsilon_\nu = \varepsilon_{res} + \varepsilon_b$$



simple elastic scatter resonant scatter

Attached positron has greatly increased annihilation rate



Can measure ε_b by the shift:

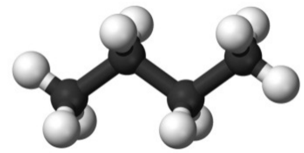
$$\varepsilon_b = \varepsilon_\nu - \varepsilon_{res}$$

C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

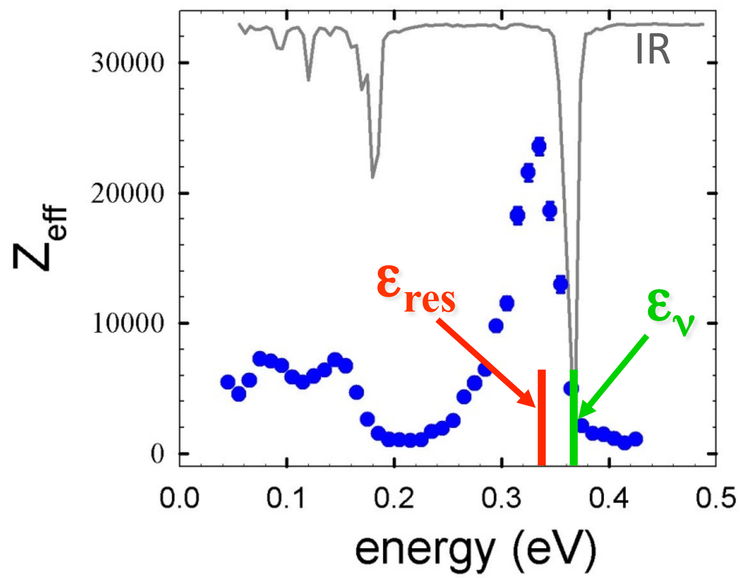
C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

Positron Binding to Butane

butane
C_4H_{10} (Z=34)



- Measured annihilation spectrum



$$\varepsilon_b = \varepsilon_\nu - \varepsilon_{res} = 35 \text{ meV}$$

Z_{eff} is annihilation rate normalized to the (Dirac) rate for a free electron gas

- Observe resonances with $Z_{eff} >> Z$

energy resolution +/− 10 meV

Z_{eff} statistical errors small; systematic errors ≤ 20%

- Experimentally, VFR's (and hence bound states) observed for a wide range of molecules

Gribakin-Lee (GL) Model for Single-mode Resonances*

Sum of vibrational Feshbach resonances (VFR):

beam energy distribution

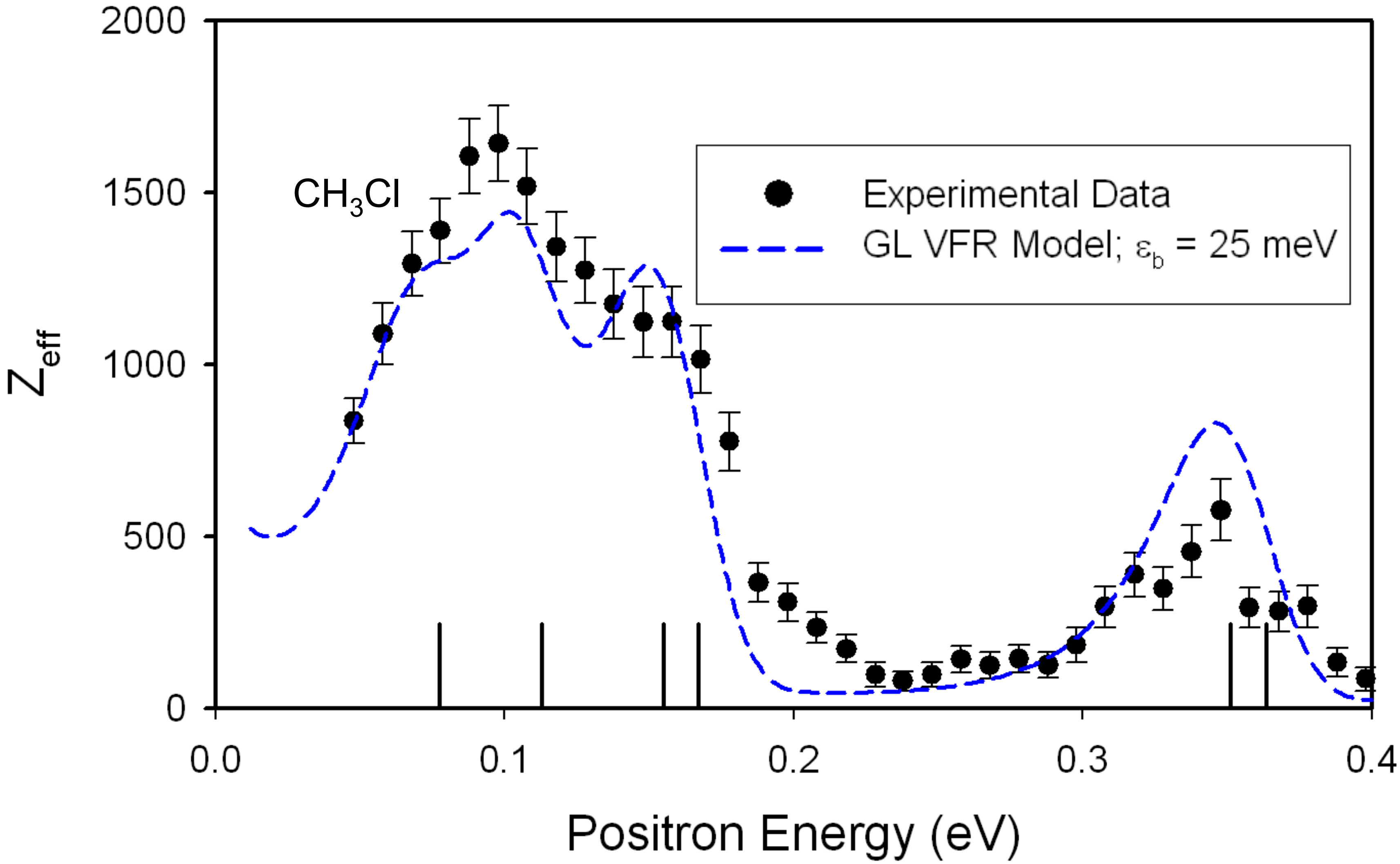
$$Z_{\mathrm{eff}}^{(\mathrm{res})}(\epsilon) \simeq \pi F \sum_{\nu} g_{\nu} \frac{\Gamma_{\nu}^{e}}{\Gamma_{\nu}} f(\epsilon) \quad \text{where } g_{\nu} = \sqrt{\frac{\epsilon_b}{\epsilon_{\nu} - \epsilon_b}}$$

VFR produce large peaks (**~ 10^3**) at positron energies

$$\varepsilon = \varepsilon_{\nu} - \varepsilon_{b}$$

* G. Gribakin & C. M. R. Lee, PRL '06

Example of GL Success – Methyl Chloride

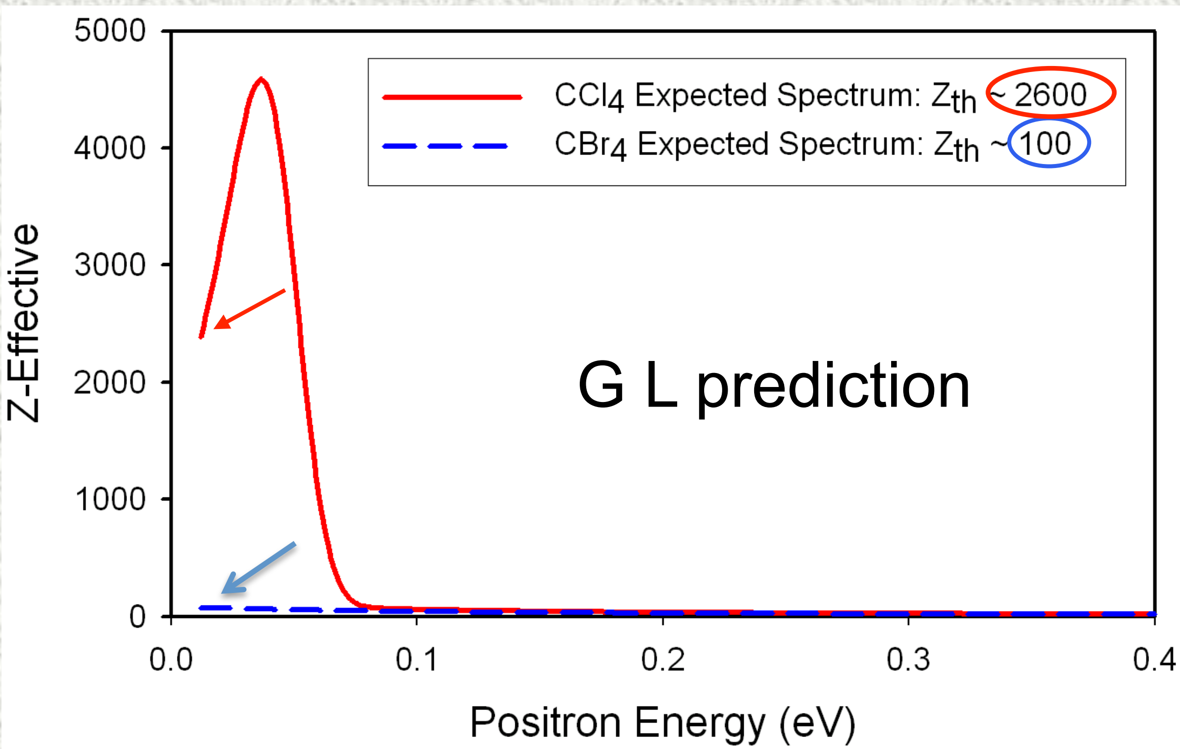


ε_b taken from experiment - no fitted parameters

But: Mystery of the Halogenated Methanes – Small Molecules, Large Z_{eff}?

Molecule	Measured thermal Z_{eff}*
CCl_4	**9,500**
CBr_4	**40,000**

Iwata et al., PRA '95



GL model fails badly?

Jones et al., PRL 2012

C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

A Paradigm Shift
Annihilation or Multimode States (SMRA)*

- Assume all multimode combinations can be excited
  ω_1, $\omega_1 + \omega_2$, $2\omega_1$, $\omega_1 + \omega_2 + \omega_3$, etc.,
  constrained only by energy conservation

- Assume coupling to positron continuum is the same for all,
  both in entrance and escape

- Z_{eff} depends on the number of vibrational final states, N,
  and the density of entrance channels $\rho(\varepsilon + \varepsilon_b + E_v)$,
  where E_v is the total molecular thermal energy

* Gribakin & Lee, Eur. Phys. J. D 51 (2009)

Statistical Multi-mode Resonant Annihilation (SMRA)*

Assume statistically complete limit, entrance and escape rates the same

density of multimode doorway states, *ρ(ε)*

$$Z_{\text{eff}}^{(\text{res})}(\epsilon) \simeq \pi F \sqrt{\frac{\epsilon_b}{\epsilon}} \frac{\rho(\epsilon + E_v + |\epsilon_b|)}{N(\epsilon + E_v)}$$

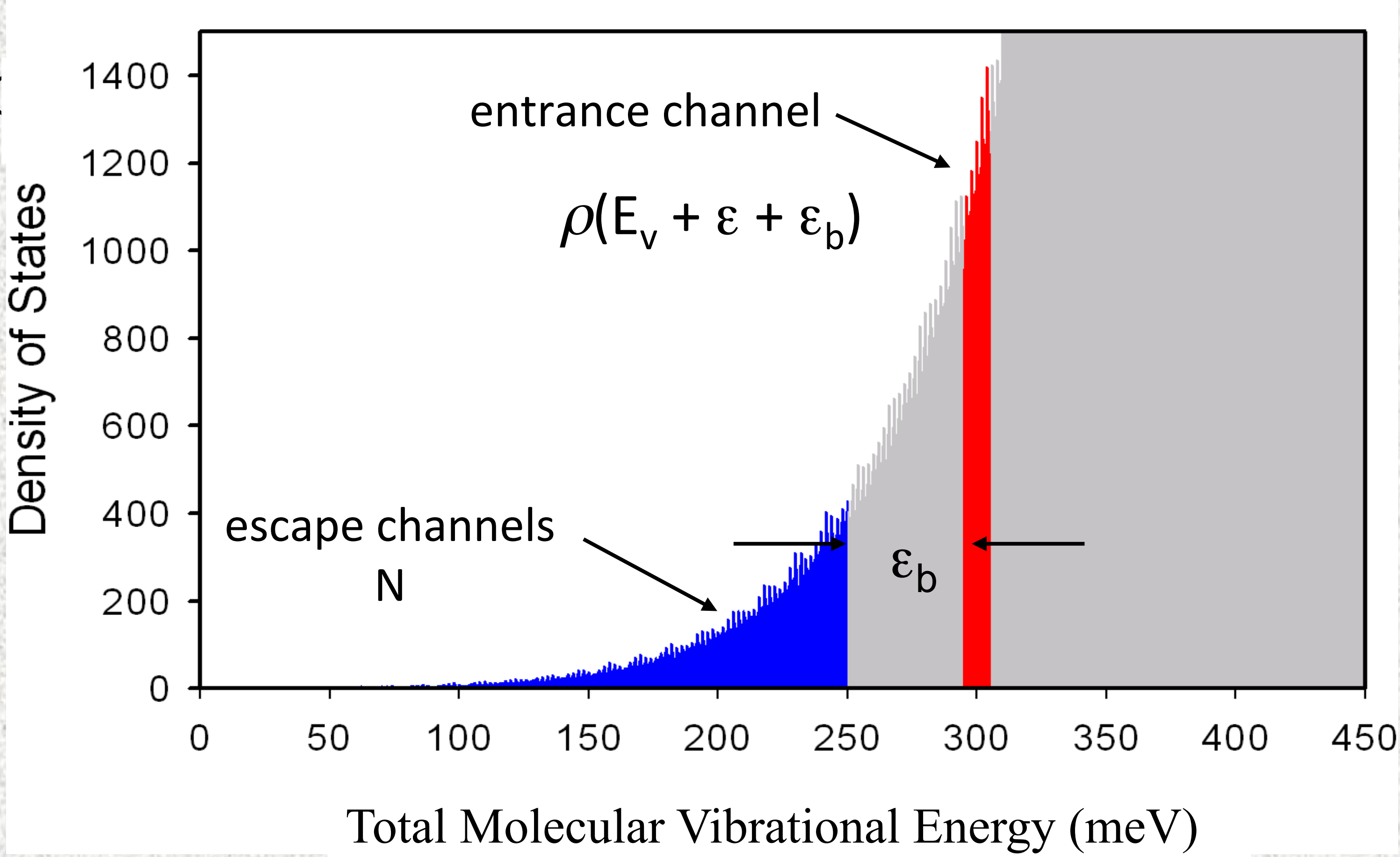
# of final states

(E_v is the molecule's thermal energy, ρ is the density of doorway states, and N is the number of vibrational final states)

Produces a broad spectrum

* Gribakin& Lee, EPJD 51 (2009)

Scaling of ρ/N with ε_ν's and ε_b



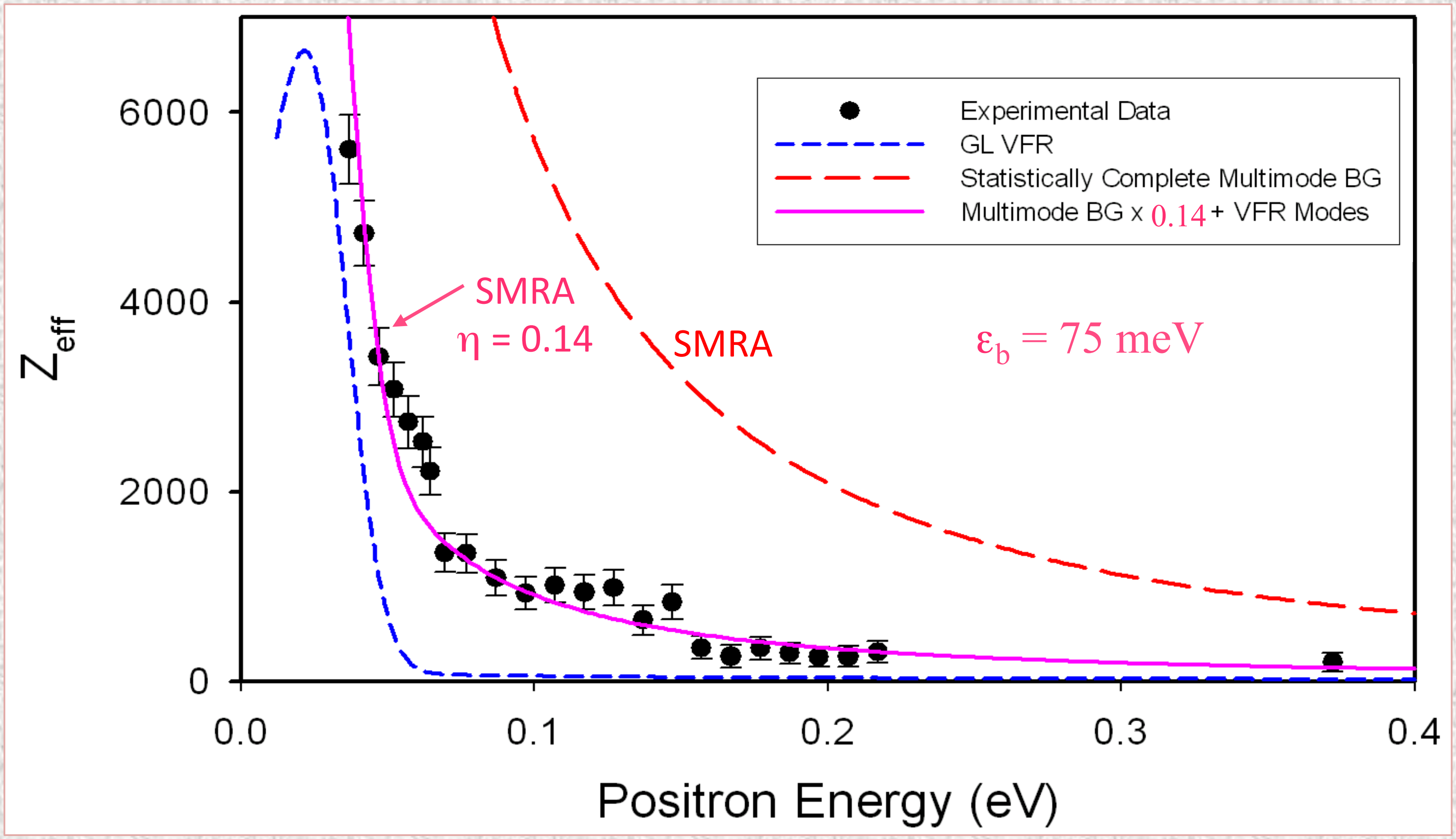
Jones et al., PRL 2012

Increasing ε_b increases ρ/N

Decreasing ω's also increases ρ/N

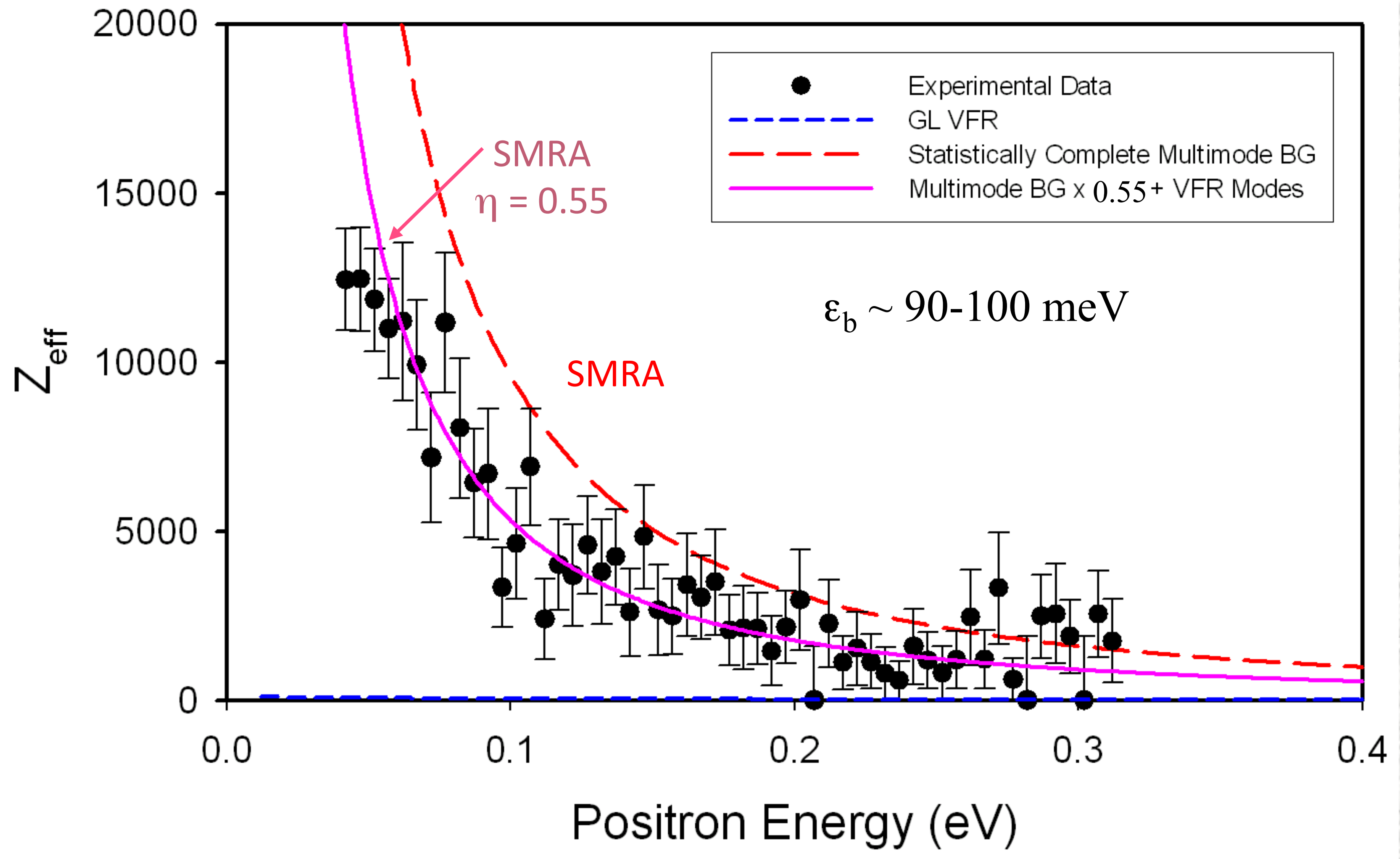
Jones et al., PRL 2012

Carbon Tetrachloride (CCl_4)



Reduced SMRA contribution (the η factor) likely due to fact that higher-order multimode states are more weakly coupled – reduces ρ faster than N

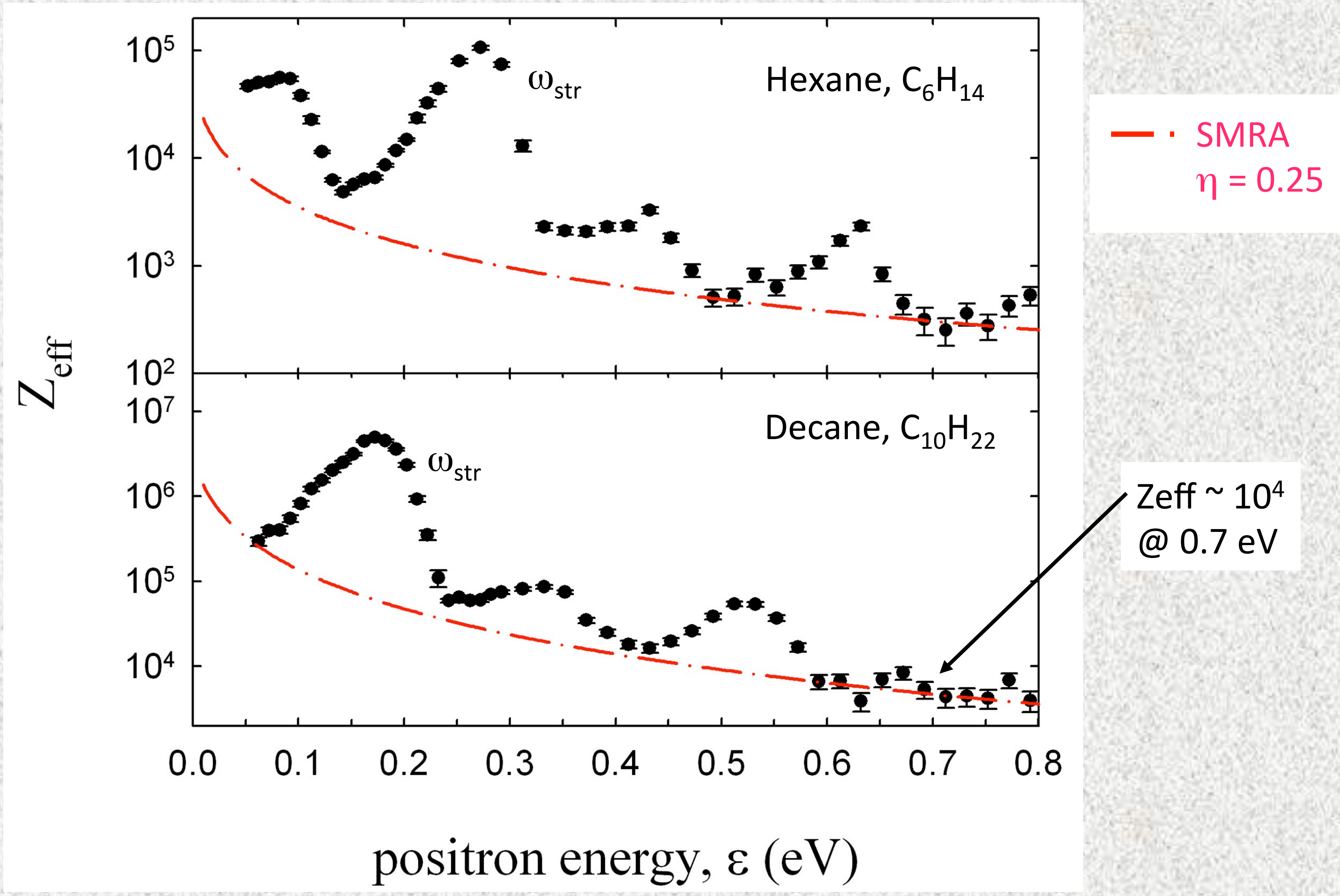
Carbon Tetrabromide (CBr_4)



Jones et al., PRL 2012

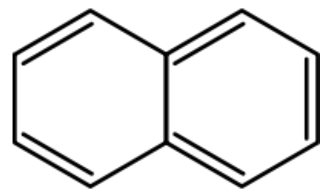
Jones et al., PRL 2012

"Ubiquitous" SMRA (e.g. alkanes)

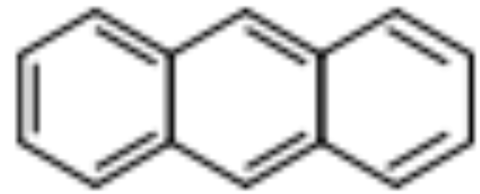


C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

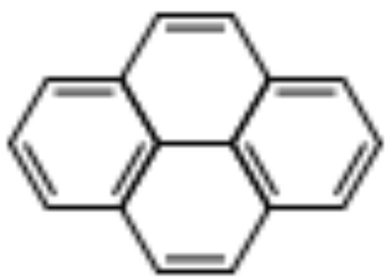
Polycyclic Aromatic Hydrocarbons (PAH molecules)



Naphthalene, $C_{10}H_8$



Anthracene, $C_{14}H_{10}$



Pyrene, $C_{16}H_{10}$

Coronene, $C_{24}H_{12}$

C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

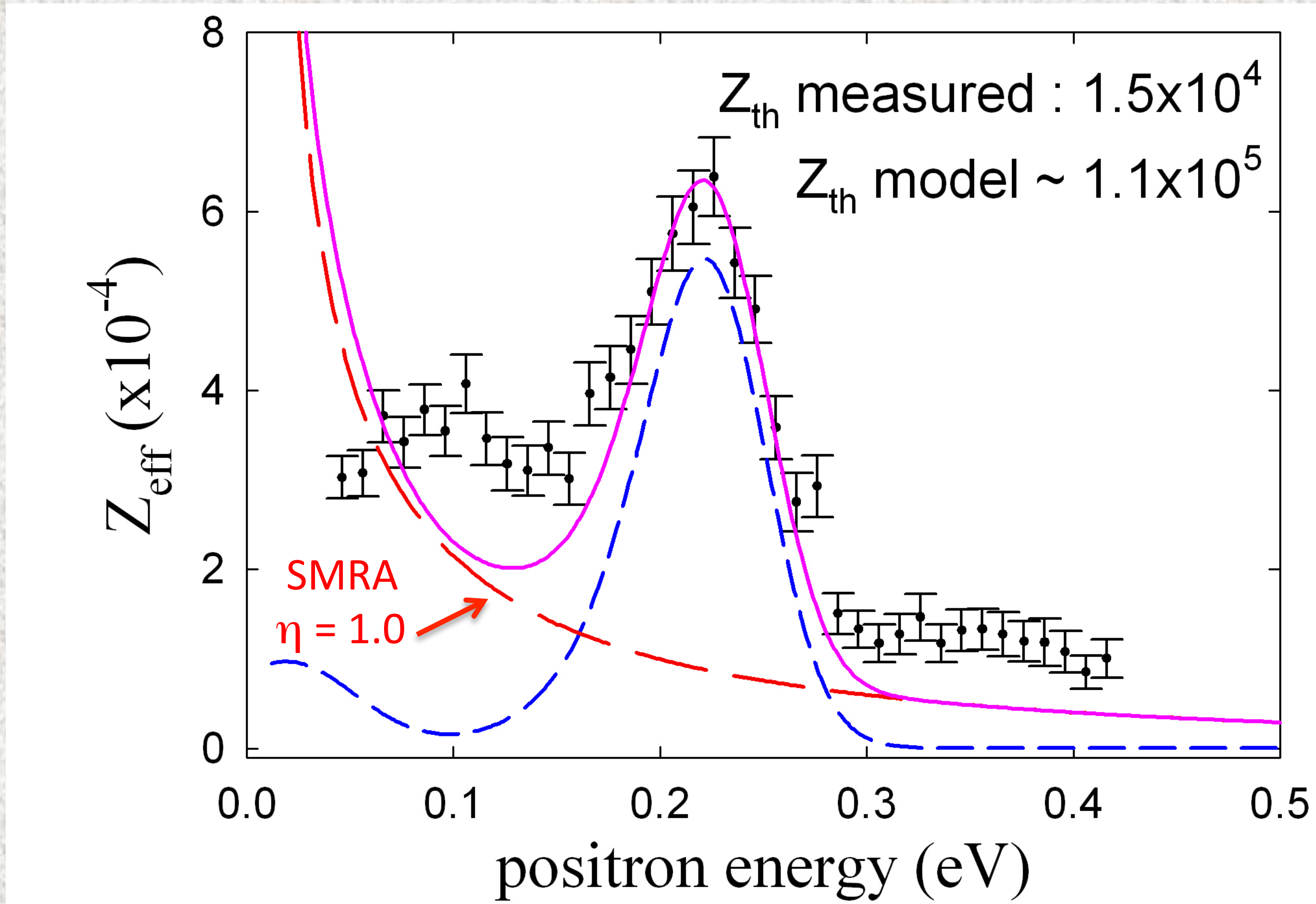
Annihilation on PAH Molecules

Molecule	Formula	I. P. (eV)	Ps Formation Threshold	Binding Energy (meV)	Primary C-H str. VFR (meV)
Benzene	C_6H_6	9.2	2.4	150	225
Naphthalene	$C_{10}H_8$	8.1	1.3	288	87
Anthracene	$C_{14}H_{10}$	7.4	0.6	421*	cut off
Pyrene	$C_{16}H_{10}$	7.4	0.6	525*	cut off
Coronene	$C_{24}H_{12}$	7.3	0.5	835*	cut off

* empirical formula (Danielson, JPB '09)

For anthracene and larger, VFR cut off – Z_{eff} dominated by SMRA, further enhanced by the large fraction on low-frequency (C-C) modes

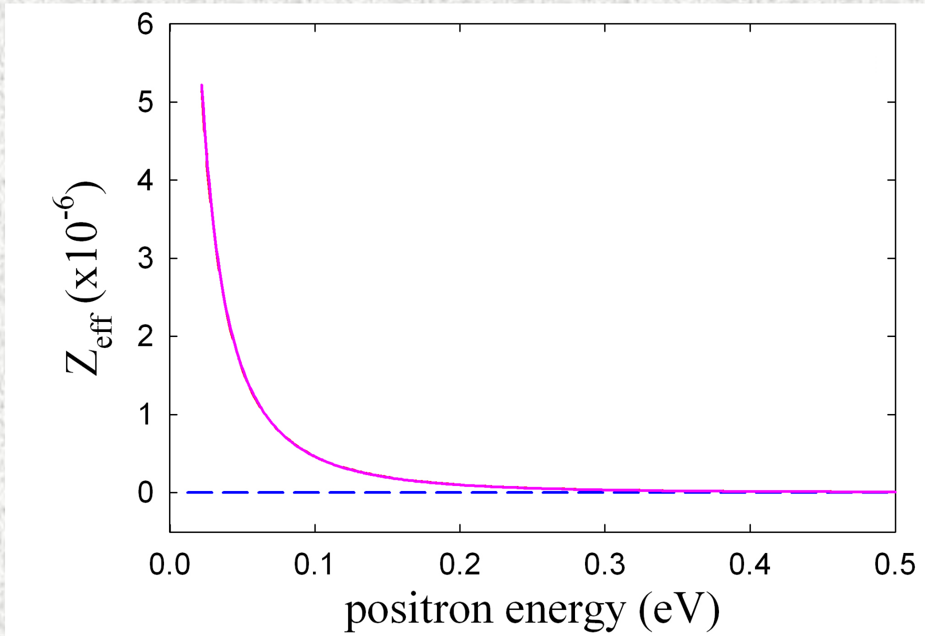
Benzene

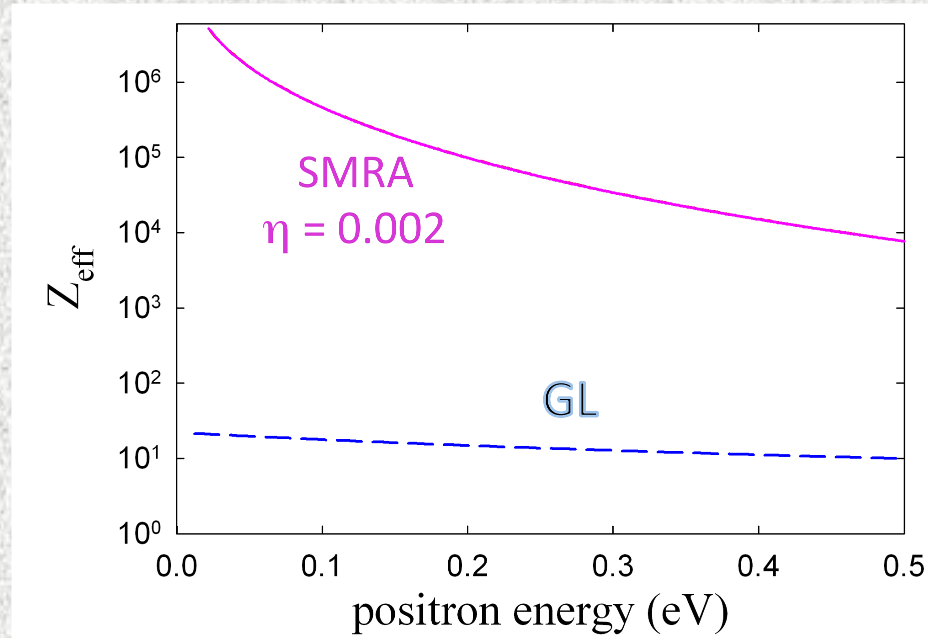


C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

C. M. Surko, Workshop on Positrons in Astrophysics, 3.12

Anthracene





Thermal Z_{eff} measured: 4.3×10^6

model: fit ($\eta = 0.002$)

Now building a "hot cell" to study this and larger PAH molecules

Resonant Annihilaiton Due to Statistical Ensembles of Multi-mode Vibrations

- Multimode states play an important (previously un-appreciated) role in annihilation on molecules
- Provide a generic new annihilation mechanism - prominent/dominant in molecules with large binding
- Directly relevant to annihilation rates for cool/cold positrons from PAH molecules (e.g., in the ISM)
- Determining η and the "tapering" of high order multimode states is "a work in progress"

"Ubiquitous nature of multi-mode vibrational resonances in positron-molecule annihilation," Jones, Danielson, Natisin, Surko, Gribakin, *Phys. Rev. Lett.* (2012).

"Positron-molecule interactions: resonant attachment, annihilation and bound states" Gribakin, Young, Sukro, *Rev. Mod. Phys.* (2010).

positrons.ucsd.edu