Supporting Information

Bringing Molecular Dynamics and Ion-Mobility Spectrometry Closer Together: Shape Correlations, Structure-Based Predictors and Dissociation

Alexander Kulesza[a,b*], Erik Marklund[d], Luke MacAleese[a,b], Fabien Chirot[c], Philippe Dugourd[a,b]

a. Université de Lyon, F-69622, Lyon, France.
b. CNRS et Université Lyon 1, UMR5306, Institut Lumière Matière
c. Univ Lyon, Université Claude Bernard Lyon 1, Ens de Lyon, CNRS, Institut des Sciences Analytiques UMR 5280, F-69100, Villeurbanne, France
d. Department of Chemistry – BMC, Uppsala University, Box 577, SE-751 23, Uppsala, Sweden
*E-mail: mail@alexanderkulesza.de

Table S1: *Fitted parameters (eq. 1) for the CCS of all systems described in this paper (see Fig. 1 and 2 a-f) as a function of unfolding. The standard deviation σ^2 comparing so-predicted dataset with the one of the explicitly calculated CCS_{EHS} (see main text) are also given. Fitting parameters have been determined for r_{Gyr} in Å and CCS in Å^2.*

System / Conditions	a_0	a_1	a_2	σ^2
CAAKAAC 220K	246.942	325.154	4.131	14.5
CAAKAAC 850K	290.981	321.518	4.862	12.4
TrpCage 220K	395.938	143.176	5.960	9.6
TrpCage 850K	302.594	280.999	4.670	15.2
Abeta dimer 220K	958.746	126.447	9.301	25.9
Abeta dimer 850K	960.845	170.043	9.295	87.0
Ubiquitin 220K	837.639	538.287	9.220	25.1
Ubiquitin 850K	442.112	962.518	7.198	59.9
AntifreezeRD3 220K	1120.645	737.678	10.751	34.5
AntifreezeRD3 850K	973.747	1044.422	10.465	64.1
HdeA dimer 220K	1120.645	737.678	10.751	34.5
HdeA dimer 850K	1782.895	652.480	13.712	128.1

Table S2: *Fitted parameters (eq. 8) for the CCS of Ubiquitin as a function of unfolding. The standard deviation σ^2 comparing so-predicted dataset with the one of the explicitly calculated CCS_{EHS} (see main text) are also given. Fitting parameters have been determined for r_{Gyr} in Å and CCS in Å^2.*

System / Conditions	Ω_{ref}	C	r_{ref}	σ^2
Ubiquitin 220K	1828.665	10124.917	137.876	31.7

Table S3: *Fitted parameters for the dissociation-covering approach (eq. 14-16) to describe the CCS unfolding (utilizing eq. 1 for full system and fragments). Data is given for of the Aβ dimer, the HdeA dimer and the antifreeze protein RD3 (cf Fig. 1 and 2 c,f,e). The standard deviation comparing so-predicted dataset with the one of the explicitly calculated CCS_{EHS} (see main text) are also given. Fitting parameters have been determined for r_{Gyr} in Å and IA as well as CCS in Å^2.*

	a_0	a_1	a_2	a_{0f}	a_{1f}	a_{2f}	b	I_0	σ^2
Aβ dimer 850K	732.733	435.200	8.414	96.820	573.841	2.975	0.399	3.924	23.3
HdeA dimer 850K	1465.848	724.455	12.918	978.388	686.444	10.844	0.066	10.585	43.3
Antifreeze RD3 220K	1120.645	737.678	10.751	640.442	130.286	9.145	0.497	-52.420	34.5

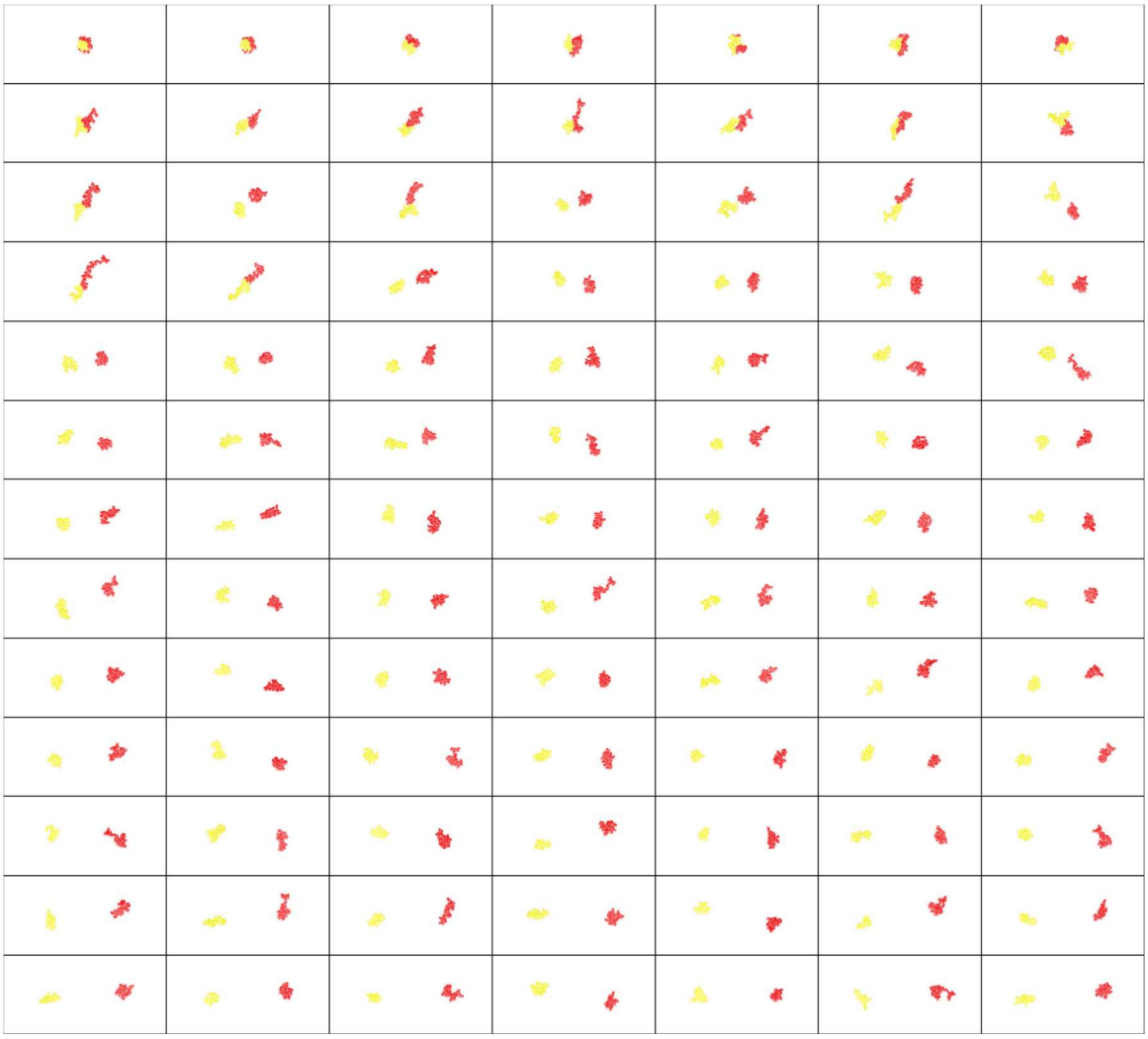

Figure S1: *Snapshots of the 220 K PT-WTMetaD trajectory for the Aβ dimer (cf Fig. 1c), sorted with increasing value of system's radius of gyration* $r_{Gyr.}$ *Red and yellow denote the different monomers of the dimeric system.*

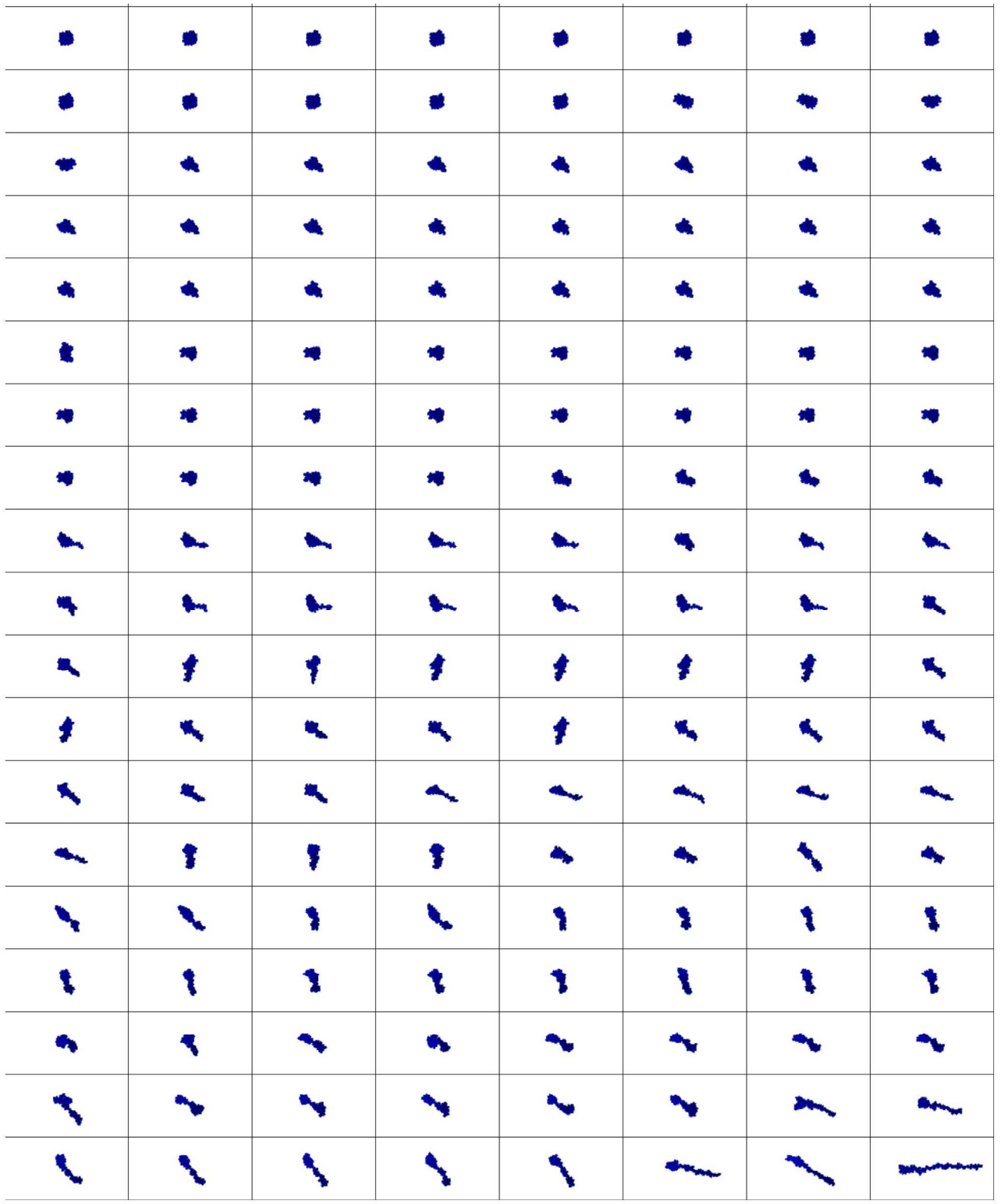

Figure S2: Snapshots of the 220 K PT-WTMetaD trajectory for Ubiquitin (cf Fig. 1d), sorted with increasing value of system's radius of gyration r_{Gyr}.

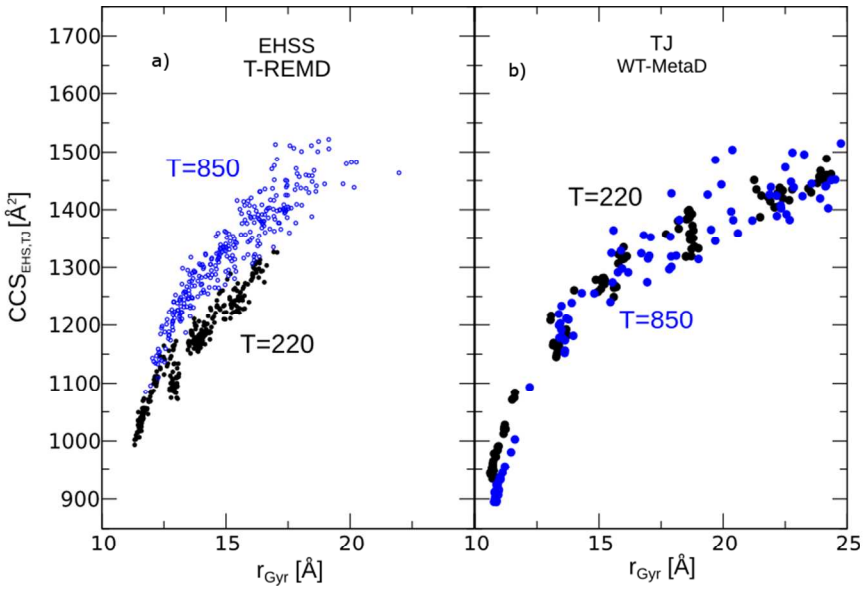

Figure S3: *Correlations between explicitly calculated collisional cross-sections (CCS) and radii of gyration (r_{Gyr}) for Ubiquitin (compare Figure 1 and 2 c). In a) the EHSS algorithm has been used for configurations sampled with T-REMD. In b the TJ method for calculating CCS has been employed for configurations obtained along PT-WTMetaD. Blue and black denote different generation conditions (thermostat temperature).*

Eq. 7 can be further simplified:

$$\Omega_{sMS} = \pi r_{HS}^2 + \pi r_{HS}^3 \left[\frac{1}{r_{HS}} - \frac{1}{r}\right] = \pi r_{HS}^2 \left(1 + r_{HS}\left[\frac{1}{r_{HS}} - \frac{1}{r}\right]\right) = \pi r_{HS}^2 \left(2 - \frac{r_{HS}}{r}\right) \quad (eq.S1)$$

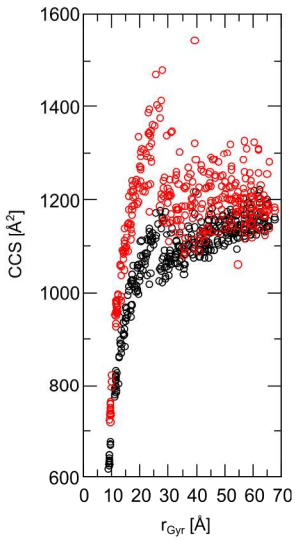

Figure S4: *Correlations between explicitly calculated collisional cross-sections (CCS_{EHS}) and radii of gyration (r_{Gyr}) for the Amyloid-β dimer (compare Figure 1 and 2 c given in red. In blue, Ω from the dissociation-capable approach of eq. 14-15 is given, utilizing the predictors of eq. 11 for the full system and its fragments.*

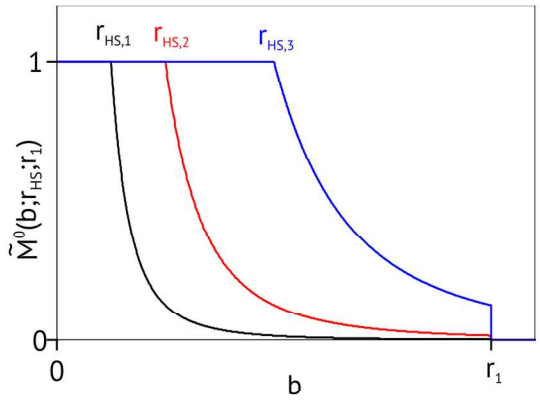

Figure S5: Plot of the approximated radius-dependent collision probability $\widetilde{M}^0(b; r_{HS}, r_1)$ along the scanned impact parameter b. The curves correspond to three different proteins/folding state combinations represented by three different radii describing the limit where the hard-spheres picture is applicable r_{HS} whereas the outer radius enclosing all atoms r_1 is the same for the three systems. Here the black line corresponds to a "small" protein that is highly unfolded, while the blue line represents a much "bigger" protein that is much less unfolded. Notice that these systems would have the same cross-section if the hard-sphere-only picture had been applied ($\Omega = \pi r_1^2$).