Toward deformation densities for intramolecular interactions without radical reference states using the fragment, atom, localized, delocalized, and interatomic (FALDI) charge density decomposition scheme

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Supplementary Information

Section S1. Nuclear geometries for all structures

Table S1. Nuclear geometries for *fin* conformer of Hen^+

Atom	x Å	y Å	z Å
C1	-0.76910	0.66695	0.24891
H2	-1.32091	1.51662	-0.16170
H3	-0.79776	0.75597	1.33786
C4	0.68763	0.73855	-0.24081
H5	0.73613	0.82712	-1.32678
H6	1.25840	1.54733	0.21205
N7	-1.30243	-0.64948	-0.13129
N8	1.31637	-0.59366	0.11045
H9	-2.01358	-0.97754	0.51279
H10	-1.71457	-0.64609	-1.05976
H11	0.50392	-1.25830	-0.00434
H12	1.63757	-0.63175	1.08032
H13	2.09440	-0.85524	-0.49650

Table S2. Nuclear geometries for *ref* conformer of Hen^+

Atom	x Å	y Å	z Å
C1	-0.76910	0.66695	0.24891
H2	-1.32091	1.51662	-0.16170
H3	-0.79776	0.75597	1.33786
C4	0.68763	0.73855	-0.24081
H5	1.31340	0.00598	0.27034
H6	0.79038	0.61117	-1.31701
N7	-1.30243	-0.64948	-0.13129
N8	1.20275	2.10311	0.16676
H9	-2.01358	-0.97754	0.51279
H10	-1.71457	-0.64609	-1.05976
H11	0.69424	2.26808	1.07735
H12	0.93751	2.83578	-0.49514
H13	2.21443	2.13951	0.29839

Table S3. Nuclear geometries for promolecular fragments of $2F_1$ scheme of Hen^+

Atom	x Å	y Å	z Å
Fragment 1			
C1	-0.76910	0.66695	0.24891
H2	-1.32091	1.51662	-0.16170
H3	-0.79776	0.75597	1.33786
N7	-1.30243	-0.64948	-0.13129
H9	-2.01358	-0.97754	0.51279
H10	-1.71457	-0.64609	-1.05976
Fragment 2			
C4	0.68763	0.73855	-0.24081
H5	0.73613	0.82712	-1.32678
H6	1.25840	1.54733	0.21205
N8	1.31637	-0.59366	0.11045
H11	0.50392	-1.25830	-0.00434
H12	1.63757	-0.63175	1.08032
H13	2.09440	-0.85524	-0.49650

Table S4. Nuclear geometries for promolecular fragments of $2F_2$ scheme of Hen^+

Atom	x Å	y Å	z Å
Fragment 1			
N7	-1.30243	-0.64948	-0.13129
H9	-2.01358	-0.97754	0.51279
H10	-1.71457	-0.64609	-1.05976
Fragment 2			
C1	-0.76910	0.66695	0.24891
H2	-1.32091	1.51662	-0.16170
H3	-0.79776	0.75597	1.33786
C4	0.68763	0.73855	-0.24081
H5	0.73613	0.82712	-1.32678
H6	1.25840	1.54733	0.21205
N8	1.31637	-0.59366	0.11045
H11	0.50392	-1.25830	-0.00434
H12	1.63757	-0.63175	1.08032
H13	2.09440	-0.85524	-0.49650

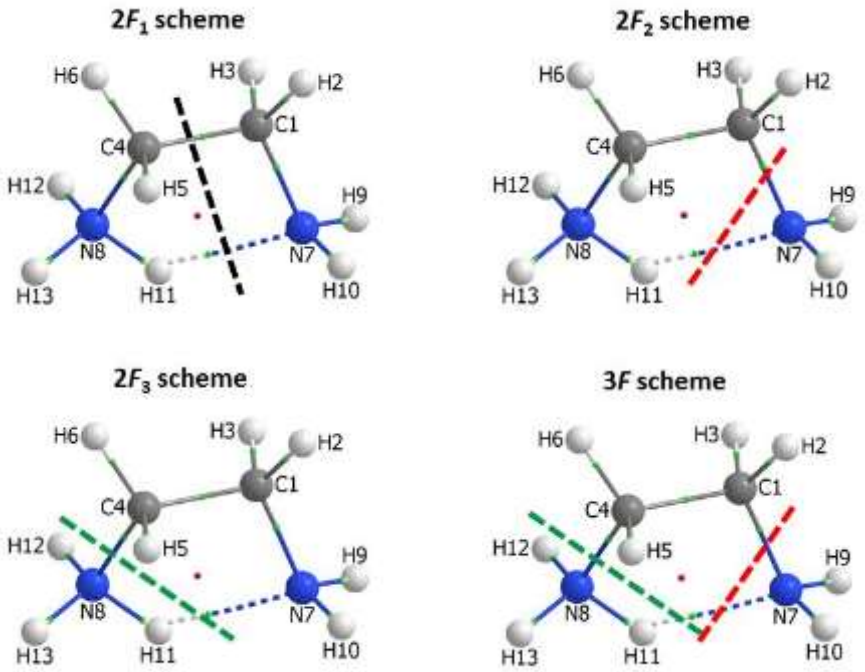
Table S5. Nuclear geometries for promolecular fragments of $2F_3$ scheme of Hen^+

Atom	x Å	y Å	z Å
Fragment 1			
N8	1.31637	-0.59366	0.11045
H11	0.50392	-1.25830	-0.00434
H12	1.63757	-0.63175	1.08032
H13	2.09440	-0.85524	-0.49650
Fragment 2			
C1	-0.76910	0.66695	0.24891
H2	-1.32091	1.51662	-0.16170
H3	-0.79776	0.75597	1.33786
C4	0.68763	0.73855	-0.24081
H5	0.73613	0.82712	-1.32678
H6	1.25840	1.54733	0.21205
N7	-1.30243	-0.64948	-0.13129
H9	-2.01358	-0.97754	0.51279
H10	-1.71457	-0.64609	-1.05976

Table S6. Nuclear geometries for promolecular fragments of 3*F* scheme of Hen^+

Atom	**x** Å	**y** Å	**z** Å
Fragment 1			
N7	-1.30243	-0.64948	-0.13129
H9	-2.01358	-0.97754	0.51279
H10	-1.71457	-0.64609	-1.05976
Fragment 2			
N8	1.31637	-0.59366	0.11045
H11	0.50392	-1.25830	-0.00434
H12	1.63757	-0.63175	1.08032
H13	2.09440	-0.85524	-0.49650
Fragment 3			
C1	-0.76910	0.66695	0.24891
H2	-1.32091	1.51662	-0.16170
H3	-0.79776	0.75597	1.33786
C4	0.68763	0.73855	-0.24081
H5	0.73613	0.82712	-1.32678
H6	1.25840	1.54733	0.21205

End of Section S1



Scheme S1. Fragmentation schemes used in this work for the orthodox deformation density calculations. Dashed lines indicate how the final molecule is fragmented into promolecules.

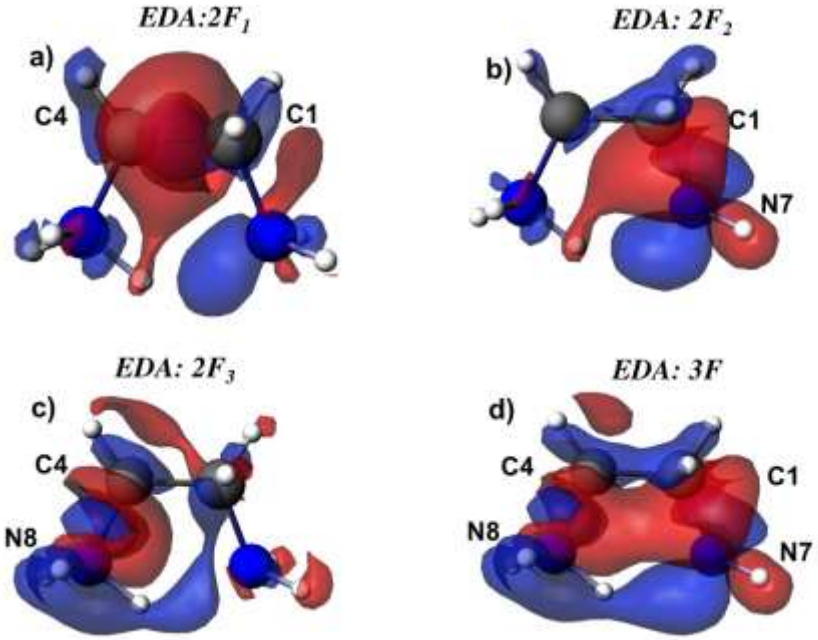


Figure S1. Total deformation densities using orthodox fragmentation methods, for schemes a) $2F_1$ ($CH_2NH_2 + \cdot CH_2NH_3^+$), b) $2F_2$ ($NH_2 + \cdot C_2H_4NH_3^+$), c) $2F_3$ ($NH_3^+ + \cdot C_2H_4NH_2$) and d) $3F$ ($NH_2 + \cdot NH_3^+ + \cdot C_2H_4$), as calculated in ADF. Red and blue regions indicate a decrease and increase in density, respectively, relative to the radical fragments. All isosurfaces are at 0.0025 au.

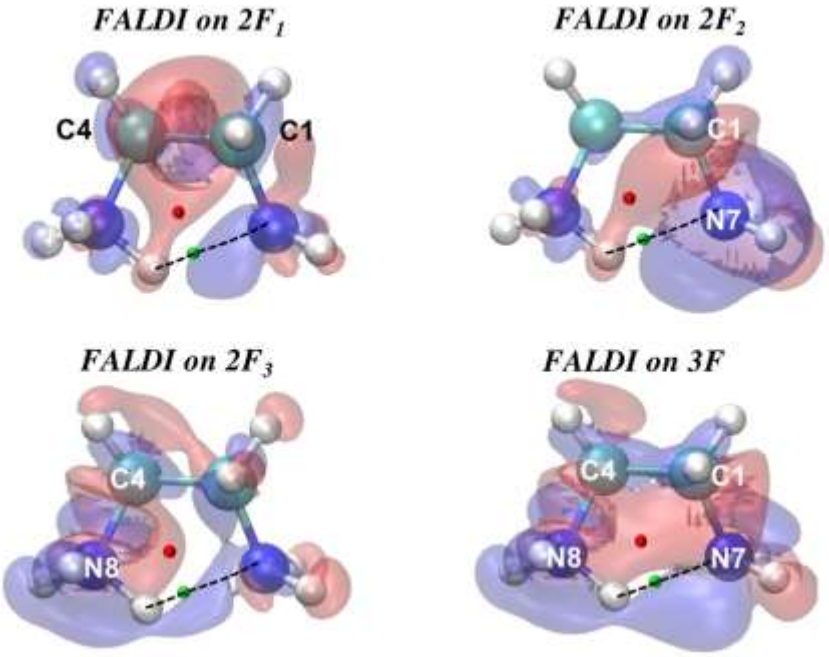


Figure S2. Total deformation densities using the FALDI-on-promolecules approach for the fragmentation schemes: $2F_1$ (CH_2NH_2 +·$CH_2NH_3^+$), $2F_2$ (NH_2 +·$C_2H_4NH_3^+$), $2F_3$ (NH_3^+ +·$C_2H_4NH_2$) and $3F$ (NH_2 + ·NH_3^+ +·C_2H_4), as calculated in Gaussian. Red and blue regions indicate a decrease and increase in density, respectively, relative to the radical fragments. All isosurfaces are at 0.0025 au.

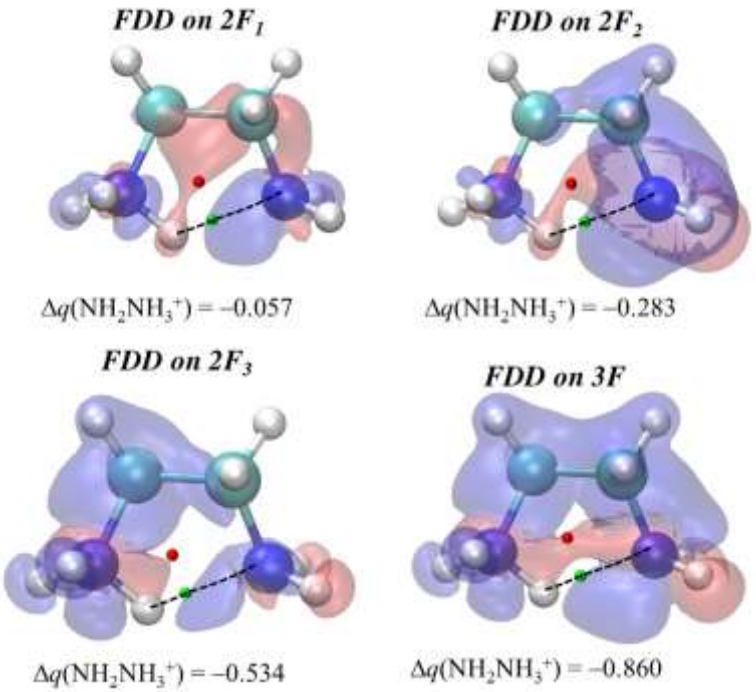


Figure S3. Fragment deformation densities (FDD) obtained from indicated partitioning schemes, using the FALDI-on-promolecules approach, for the $NH_2NH_3^+$ fragment. All isosurfaces are at 0.0025 au.

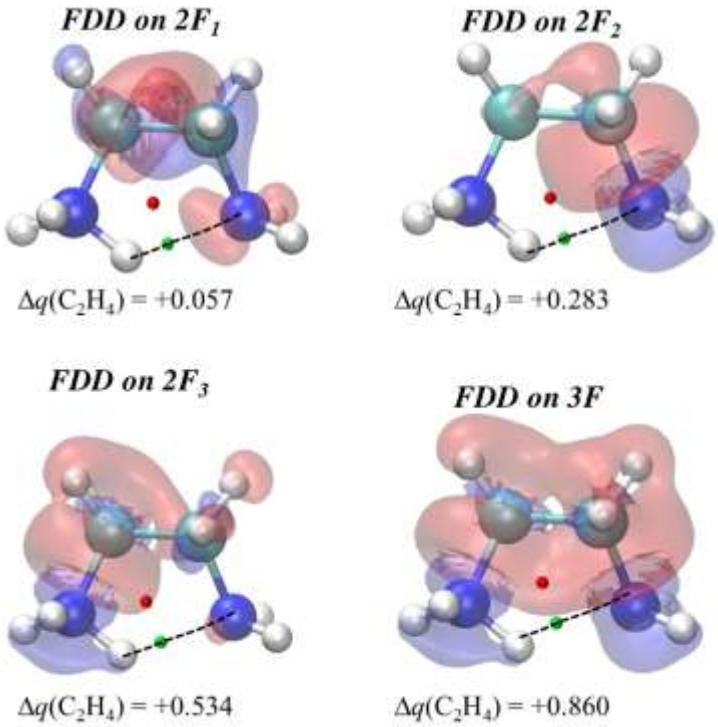


Figure S4. Fragment deformation densities (FDD) obtained from indicated partitioning schemes, using the FALDI-on-promolecules approach, for the $–CH_2CH_2–$ fragment. All isosurfaces are at 0.0025 au.

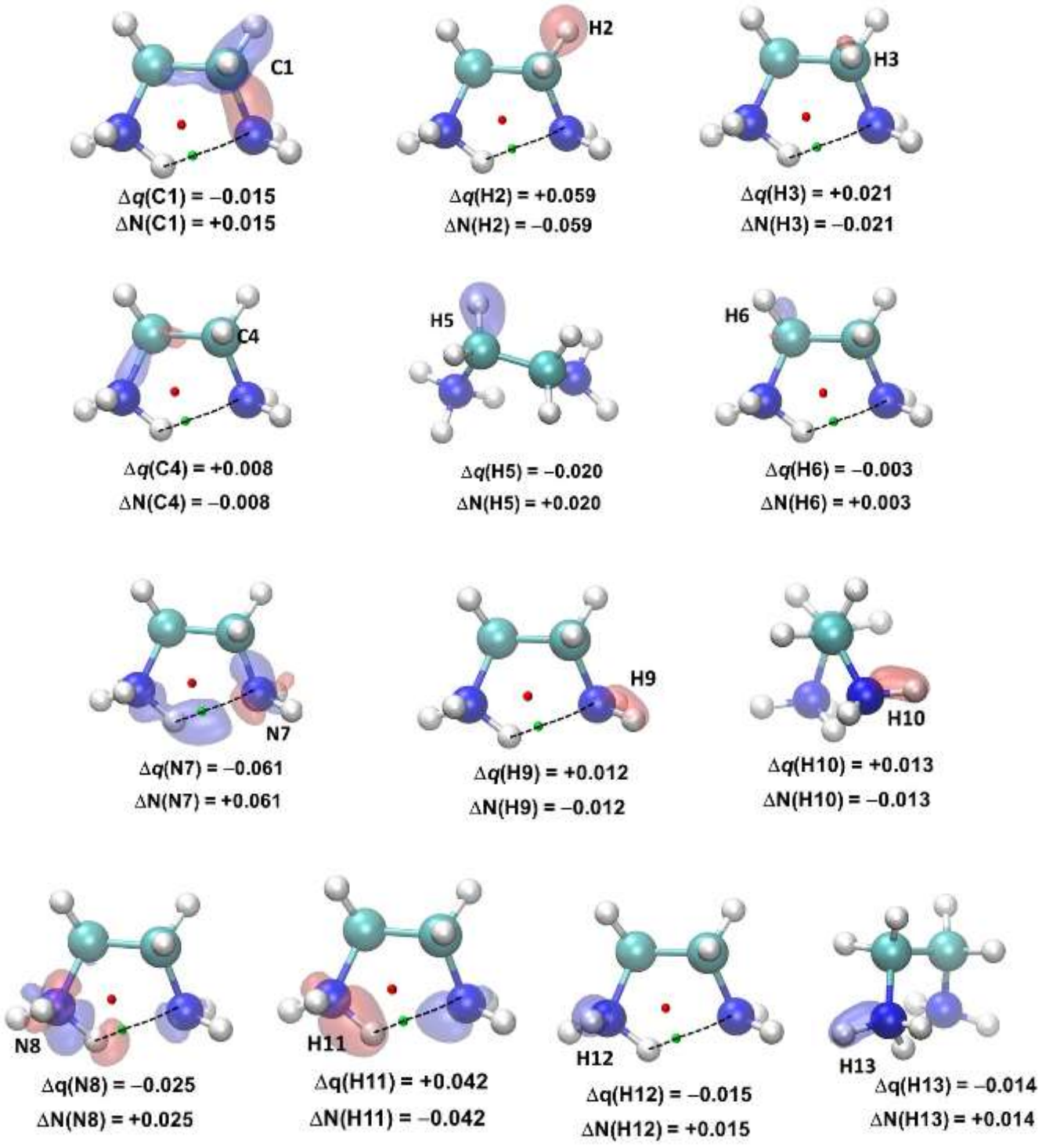


Figure S5. FALDI's atomic deformation densities (ADD), and changes in QTAIM's net charge and atomic populations (both in *e*) obtained for indicated atoms on the *ref* → *fin* structural change. Isosurfaces for C4, H6, H9, H10, H12 and H13 are at 0.001 au; all other isosurfaces are at 0.0025 au.