Supplementary Information

Additional analysis were conducted to evaluate the error introduced by locking $\beta_0^2(02)$ to different values. We locked $\beta_0^2(02)$ to -0.4, -0.24 and 0. For each locked $\beta_0^2(02)$ value, we analyzed the angular distribution of a set of 3 images (j = 23, 33, 39). The 3 images were chosen to represent data from the low, medium, and high j regions of CO product distribution. In addition, we also conducted a separate set of analysis by locking $\beta_0^0(22)$ to -0.5, -0.24, and -0.1 and repeated the same analysis.

Regarding the choices for the value of $\beta_0^2(02)$, for a dissociation via the excitation to 2^1A' (A state) only, the theoretical value of $\beta_0^2(02)$ is - 0.5, while for a dissociation via the excitation to pure 1^1A" (B state) only, the theoretical value is 1. In this work, $\beta_0^2(20)$ data (half of the spatial anisotropy value) are extracted by locking $\beta_0^2(02)$ at -0.24, which indicates an 83% contribution of the 2^1A' (A state). Indicatively, locking $\beta_0^2(02)$ at -0.4 is equivalent to a percentage of 93 % for the 2^1A' (A state) and locking $\beta_0^2(02)$ at 0, will give a 67% percentage for the 2^1A' (A state). As the theoretical works [1–4] do not support a significant contribution of dissociation via 1^1A" (B state), we think $\beta_0^2(02)$ value should not be locked above 0.

Regarding the choices for the value of $\beta_0^0(22)$,, for a triatomic molecule dissociation, conservation of angular momentum requires that the recoil velocity vector $\vec{v}$ be perpendicular to the rotational angular momentum vector $\vec{J}$, which implies negative $\beta_0^0(22)$ values. For these reasons, we chose to evaluate the range from -0.4 to 0 for $\beta_0^2(02)$, and from -0.5 to -0.1 for $\beta_0^0(22)$. The results are shown in Figure S1 and Figure S2.

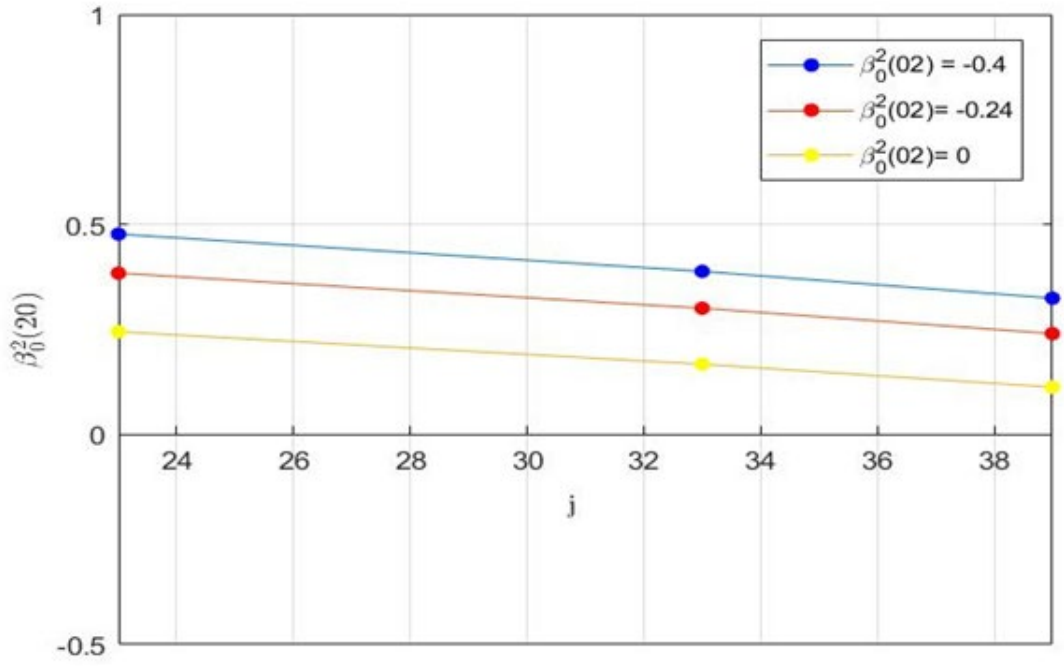

Figure S1: Trend of $\beta_0^2(20)$ with rotational quantum number j, for $\beta_0^2(02)$ locked at -0.4(blue line), -0.24 (red line) and 0 (yellow line).

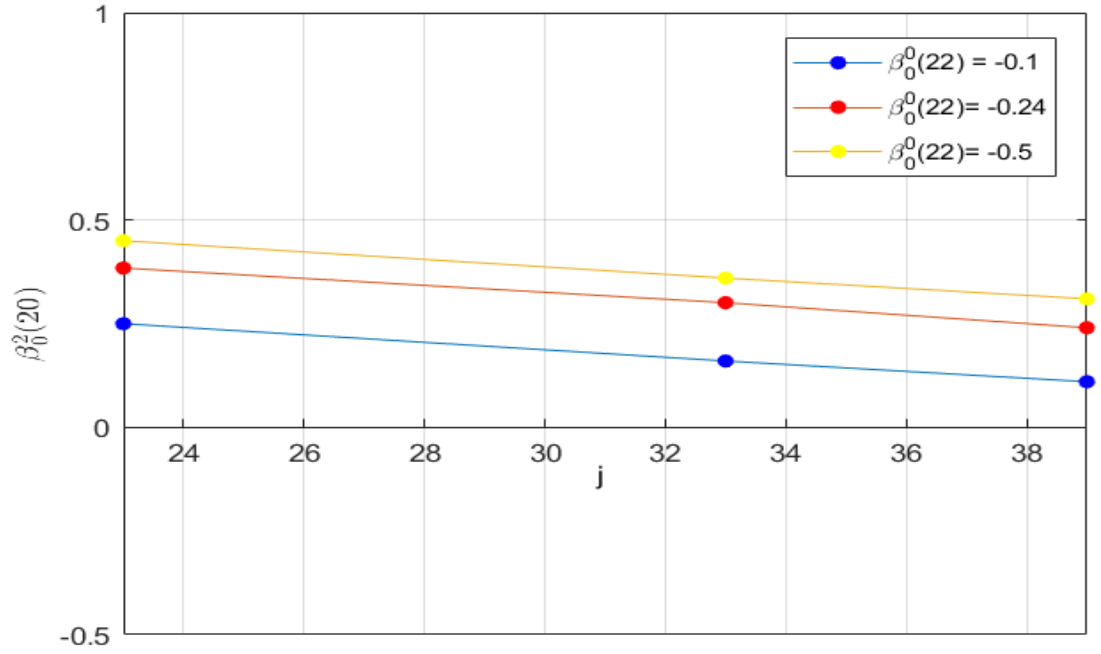

Figure S2: Trend of $\beta_0^2(20)$ with rotational quantum number j, for $\beta_0^0(22)$ locked at -0.1(blue line), -0.24 (red line) and -0.5 (yellow line).

It can be clearly seen that locking $\beta_0^2(02)$ and $\beta_0^0(22)$ at different values will generally shift the entire distribution of extracted $\beta_0^2(20)$ up or down but will not affect the general trend of $\beta_0^2(20)$ as a function of CO j states. An error range of ± 0.14 for extracted $\beta_0^2(20)$ can be estimated from this figure.

References

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2. S. Y. Grebenshchikov and R. Borrelli, *J. Phys. Chem. Lett.* 3, 3223 (2012).
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4. J. A. Schmidt, M. S. Johnson, and R. Schinke, *Proc. Natl. Acad. Sci. U. S. A.* 110, 17691 (2013).