**Supporting Information**

# Machine Learning Analysis of Direct Dynamics Trajectory Outcomes for Thermal Deazetization of 2,3-Diazabicyclo[2.2.1]hept-2-ene

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## Table of Contents

***Jupyter notebooks*** S2
***Additional comments on transition-state vibrational mode excitation*** S2

***Jupyter notebooks***

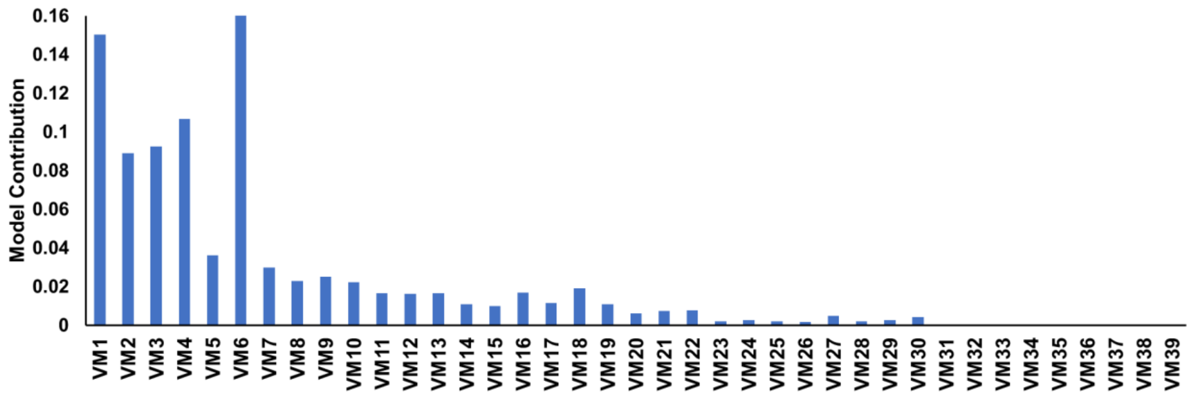
In separate Supporting Information folders there are Jupyter notebooks that contain examples of machine learning program scripts that showcase classification and feature analysis.

***Additional comments on transition-state vibrational mode excitation***

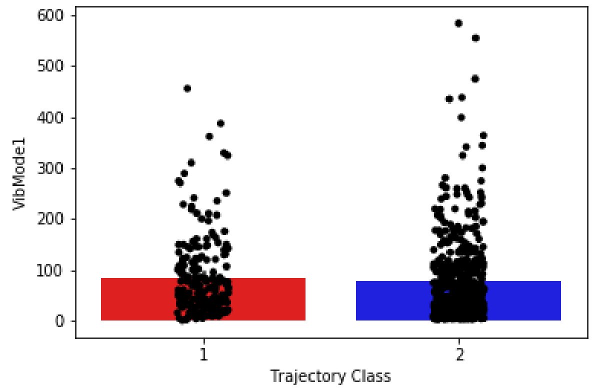
In the main manuscript we described that for all machine learning algorithms vibrational quanta alone provided poor classification. Using the 39 vibrational mode quanta values, machine learning models gave the following classification accuracy:

Random Forest = 57%
MultiLayer Perceptron = 57%
Stochastic Gradient Descent = 55%
Logistic Regression Classifier = 58%

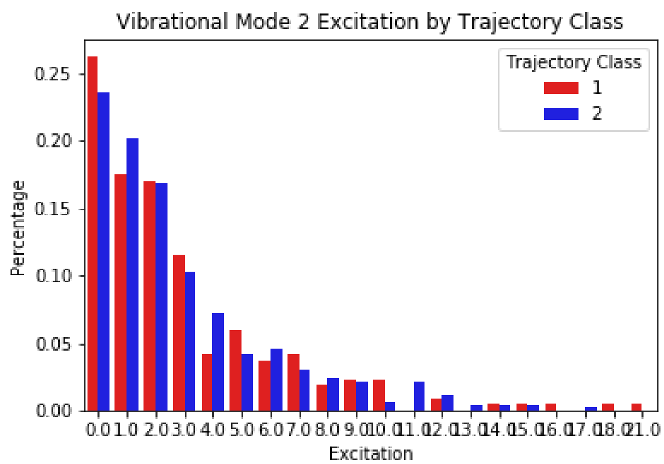
While 8% accuracy above the baseline 50% does not provide practical predictability, we did analyze the relative feature importance of each vibrational mode. Below is a plot of the model contribution by each vibrational mode (VM). Despite vibrational mode data being highly overlapping at the transition state, this does suggest that mode 1 and 6 are important for the 8% increase in accuracy above baseline.



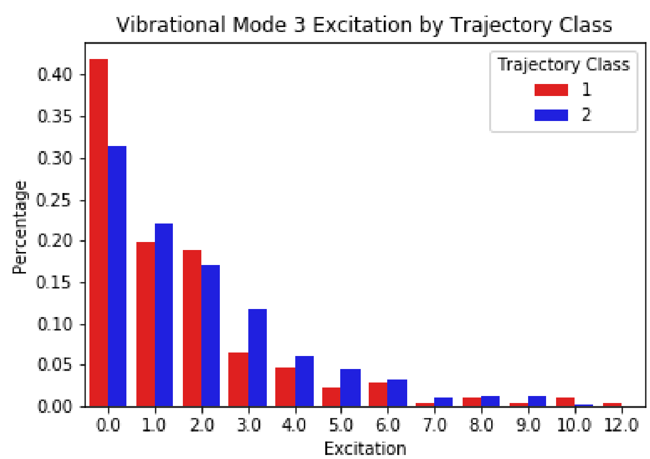

As stated in the manuscript, it was initially surprising to us that vibrational mode patterns from quanta (or total vibrational energy) did not provide high accuracy prediction of trajectory classification. However, in Figure 4 of the manuscript we demonstrated that for transition-state vibrational mode 6 the pattern of mode excitation is statistically identical between class 1 and class 2 trajectories. Below are similar plots for vibrational modes 1-5 and 7-20. Importantly, these plots indicate that a single vibrational mode excitation or just few vibrational mode patterns do not provide a direct mapping or correlation to make trajectory outcome predictions. The above machine learning results indicate that even complex vibrational mode patterns do not provide mapping or correlation to trajectory outcomes.



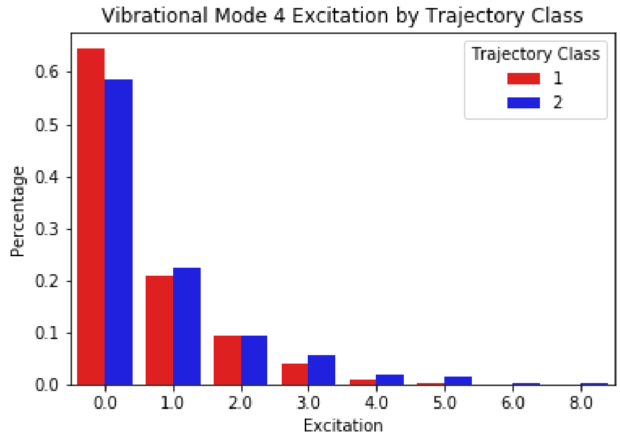
VibMode1
600
500
400
300
200
100
0
1
2
Trajectory Class

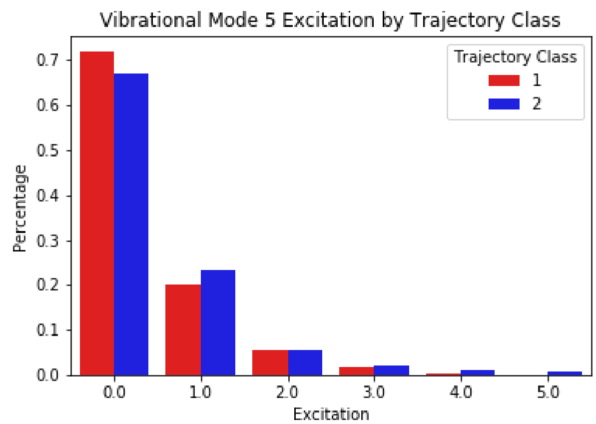



Vibrational Mode 2 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
Excitation

Vibrational Mode 3 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
Excitation


Vibrational Mode 4 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.0
1.0
2.0
3.0
4.0
5.0
6.0
8.0
Excitation


Vibrational Mode 5 Excitation by Trajectory Class
Trajectory Class
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2
Percentage
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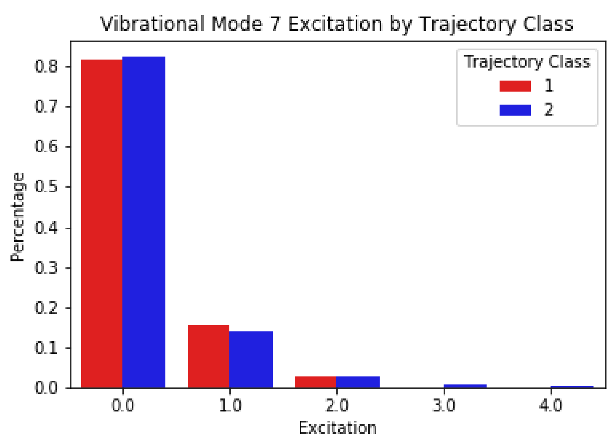
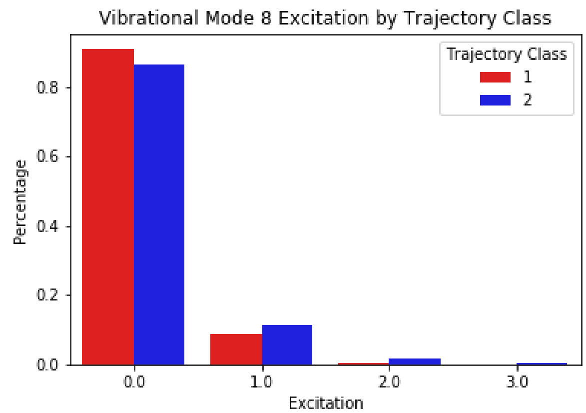
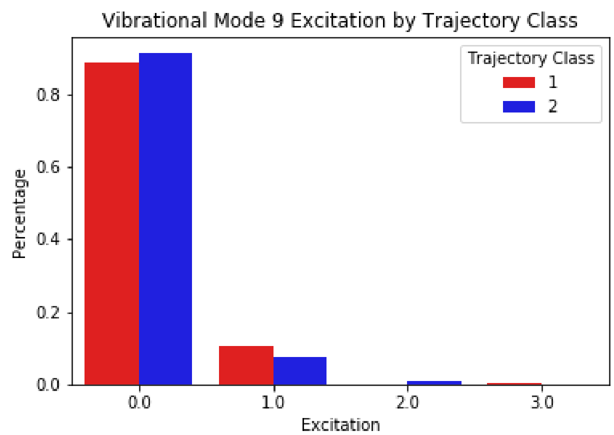
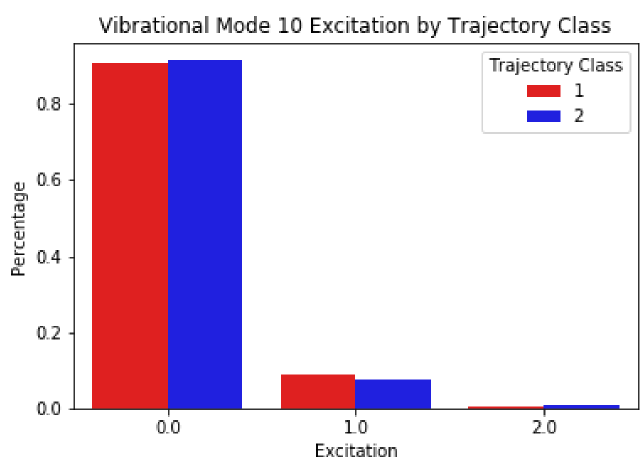
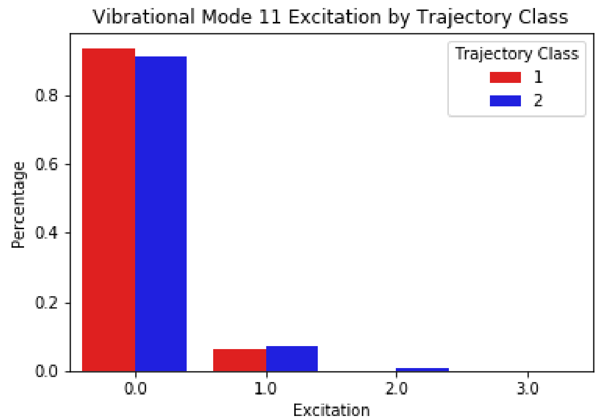
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.0
1.0
2.0
3.0
4.0
5.0
Excitation

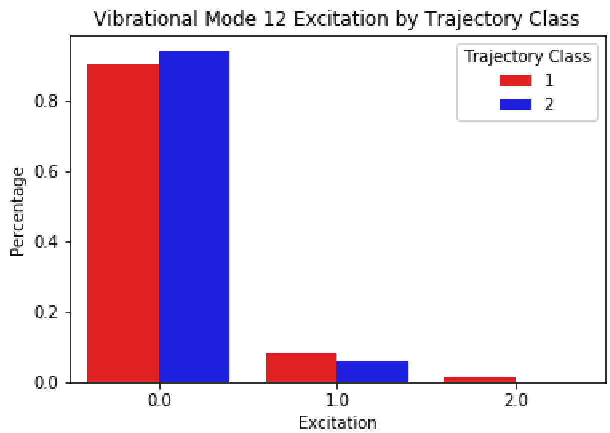

Vibrational Mode 7 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.0
1.0
2.0
3.0
4.0
Excitation


Vibrational Mode 8 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
3.0
Excitation


Vibrational Mode 9 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
3.0
Excitation


Vibrational Mode 10 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
Excitation


Vibrational Mode 11 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
3.0
Excitation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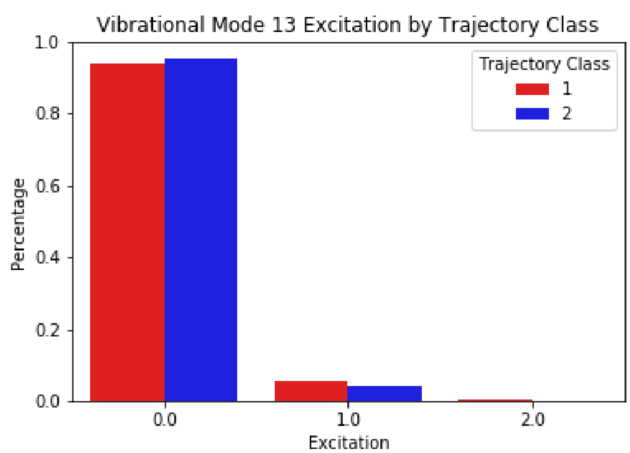
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
Excitation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Trajectory Class
1
2
Percentage
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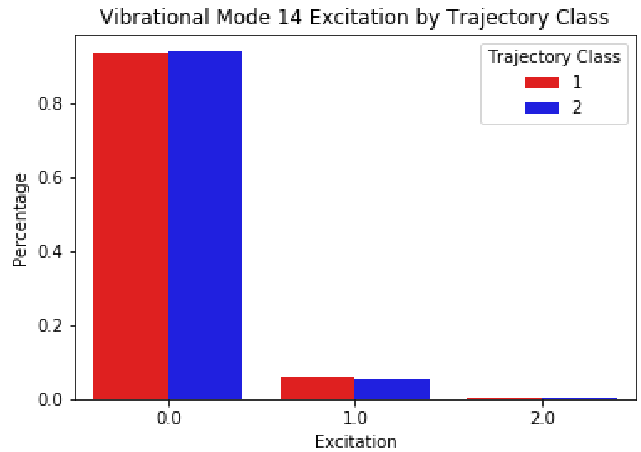
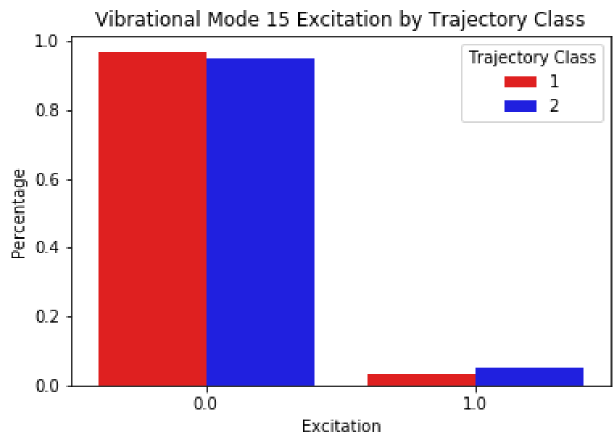
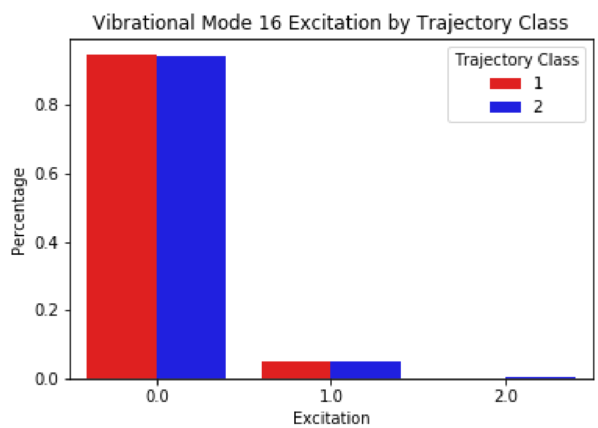
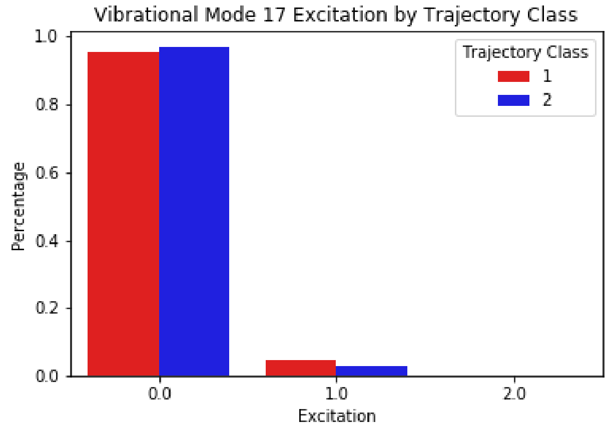
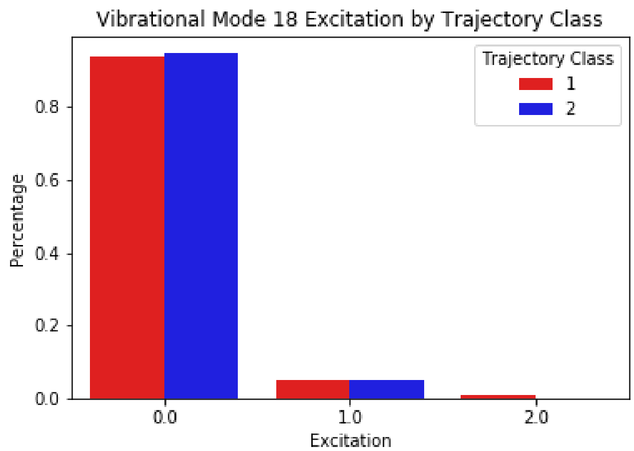
0.2
0.4
0.6
0.8
1.0
0.0
1.0
2.0
Excitation


Vibrational Mode 14 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
Excitation


Vibrational Mode 15 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
1.0
0.0
1.0
Excitation


Vibrational Mode 16 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
Excitation


Vibrational Mode 17 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
1.0
0.8
0.6
0.4
0.2
0.0
0.0
1.0
2.0
Excitation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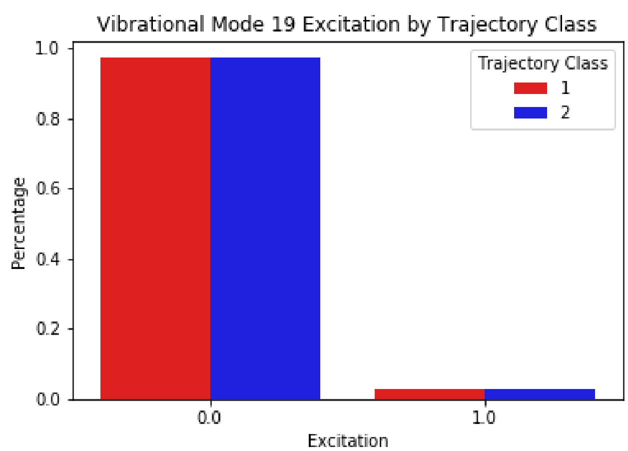
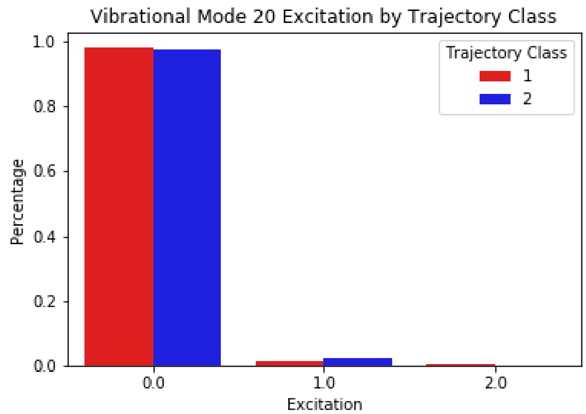
Trajectory Class
1
2
Percentage
0.8
0.6
0.4
0.2
0.0
0.0
1.0
2.0
Excitation


Vibrational Mode 19 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
1.0
0.8
0.6
0.4
0.2
0.0
0.0
1.0
Excitation


Vibrational Mode 20 Excitation by Trajectory Class
Trajectory Class
1
2
Percentage
1.0
0.8
0.6
0.4
0.2
0.0
0.0
1.0
2.0
Excitation