

# Supplemental Material: Efficient multiphoton sampling of molecular vibronic spectra on a superconducting bosonic processor

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## I. NUMERICAL FRANCK-CONDON DATA

Additional experimental data is provided in this section. TABLE I provides an overview of the different molecular processes that are simulated and corresponding information regarding post-selection, systematic offsets, and distance metrics.

The data for each molecular process in the following tables is calculated as follows. For the “single-bit extraction” scheme, the probability and standard error for a given joint photon number of interest is:

$$q_{n',m'}^{\text{meas}} = \frac{n_{n',m'}^{\text{ee}}}{N_{n',m'}^{\text{runs}}} \quad (\text{S33})$$

$$\sigma_{n',m'} = \sqrt{\frac{q_{n',m'}^{\text{meas}}(1 - q_{n',m'}^{\text{meas}})}{N_{n',m'}^{\text{runs}}}} \quad (\text{S34})$$

where  $n_{n',m'}^{\text{ee}}$  is the number of counts where both ancillas are measured in their excited state, indicating a measure for population in  $|n', m'\rangle$  (see Eq. 4 in the main text), and  $N_{n',m'}^{\text{runs}}$  is the total number of runs of the experiment for probing  $|n', m'\rangle$ . The number of runs varies slightly among different final states due to varying post-selection probabilities. The correction protocol outlined in Appendix E of the main text is then applied to  $q_{n',m'}^{\text{meas}}$  to retrieve a new probability distribution  $p_{n',m'}^{\text{meas}}$ . The standard error  $\sigma_{n',m'}$  is truncated to one significant digit and  $p_{n',m'}^{\text{meas}}$  is then rounded to the precision set by  $\sigma_{n',m'}$ . The data reported is  $p_{n',m'}^{\text{meas}} \pm \sigma_{n',m'}$  only for probabilities with significant support relative to the precision of the experiment ( $p_{n',m'}^{\text{ideal}} \gtrsim 10^{-4}$ ).

The same method (sans the correction protocol) is applied to the data for the “sampling” scheme, except there the probabilities and standard error are given by:

$$q_{n',m'}^{\text{meas}} = \frac{n_{n',m'}}{N_{\text{runs}}} \quad (\text{S35})$$

$$\sigma_{n',m'} = \sqrt{\frac{q_{n',m'}^{\text{meas}}(1 - q_{n',m'}^{\text{meas}})}{N_{\text{runs}}}} \quad (\text{S36})$$

where  $n_{n',m'}$  is the number of times the joint photon number  $|n', m'\rangle$  is sampled from the total number of runs of the experiment  $N_{\text{runs}}$ .

Sometimes, there will be no counts reported for a given  $|n', m'\rangle$  (i.e.,  $n_{n',m'}^{\text{ee}}$  or  $n_{n',m'} = 0$ ). In this case, we simply report a probability of zero. Furthermore, sometimes the correction protocol will return negative elements in the probability distribution due to statistical noise; these unphysical cases are also nulled and a zero is reported. All distances  $D = \frac{1}{2} \sum_{i=0}^{n_{\text{max}}} \sum_{j=0}^{n_{\text{max}}} |p_{ij}^{\text{meas}} - p_{ij}^{\text{ideal}}|$  are calculated after this correction process, with the corresponding values for  $n_{\text{max}}$  specified in TABLE I.

Full time-domain master equation simulations are performed using QuTiP and consider only the Hilbert space of the two cavities with  $n_{\text{max}} = 30$ . Each Gaussian operation is simulated by evolving the associated Hamiltonian term, while also including the corresponding self-Kerr terms and photon loss for each operation. While squeezing cavity A, for instance, the native self-Kerr and photon loss rates for cavity B are used, assuming that the pumped process for squeezing cavity A does not change the participation of cavity B in any nonlinear lossy modes (and vice-versa). The simulation also takes into account photon loss during the verification measurement, which takes  $2.5 \mu\text{s}$ . The simulation does not consider imperfect state preparation and systematic errors in calibrations, which we believe to account for the remaining difference between the measured distances for the “single-bit extraction” scheme and predicted distances from the master equation simulations.

| Molecular transition   | Initial state<br>( $n, m$ ) | Percentage of data kept | $n_{\max}$ | $t_A, t_B$<br>$f_A, f_B$     | Distance to ideal distribution |          |                 | Figure  |
|--|-----------------------------|-------------------------|------------|------------------------------|--------------------------------|----------|-----------------|---------|
|  |                             |                         |            |                              | Single-bit extraction          | Sampling | Master Equation |         |
| $\text{H}_2\text{O} \xrightarrow{h\nu} \text{H}_2\text{O}^+(\tilde{B}^2\text{B}_2) + \text{e}^-$ | (0, 0)                      | ~ 95%                   | 16         | 0.937, 0.946<br>0.005, 0.002 | 0.049(1)                       | 0.151(9) | 0.0123          | FIG. 7  |
| $\text{O}_3^- \xrightarrow{h\nu} \text{O}_3 + \text{e}^-$  | (0, 0)                      | ~ 96%                   | 12         | 0.937, 0.948<br>0.005, 0.002 | 0.039(9)                       | 0.075(2) | 0.0052          | FIG. 8  |
|  | (1, 0)                      | ~ 95%                   | 10         | 0.937, 0.950<br>0.004, 0.002 | 0.057(5)                       | 0.085(5) | 0.0131          | FIG. 9  |
|  | (1, 2)                      | ~ 93%                   | 12         | 0.938, 0.950<br>0.004, 0.001 | 0.105(3)                       | 0.148(4) | 0.0217          | FIG. 10 |
| $\text{NO}_2^- \xrightarrow{h\nu} \text{NO}_2 + \text{e}^-$                                      | (0, 0)                      | ~ 94%                   | 12         | 0.935, 0.943<br>0.005, 0.003 | 0.034(0)                       | 0.110(9) | 0.0331          | FIG. 11 |
|  | (1, 0)                      | ~ 92%                   | 14         | 0.934, 0.951<br>0.004, 0.002 | 0.202(2)                       | 0.209(7) | 0.1269          | FIG. 12 |
| $\text{SO}_2 \xrightarrow{h\nu} \text{SO}_2^+ + \text{e}^-$                                      | (0, 0)                      | ~ 96%                   | 12         | 0.938, 0.950<br>0.004, 0.002 | 0.019(6)                       | 0.095(3) | 0.0065          | FIG. 13 |
|  | (0, 1)                      | ~ 94%                   | 12         | 0.931, 0.951<br>0.004, 0.001 | 0.063(7)                       | 0.136(6) | 0.0213          | FIG. 14 |

TABLE I. **Summary of experimental data.** List of molecular processes simulated and corresponding post-selection probabilities, maximum photon number probed with the “single-bit extraction” measurement scheme, and distances. Transmon offsets are independently measured after each dataset is taken.

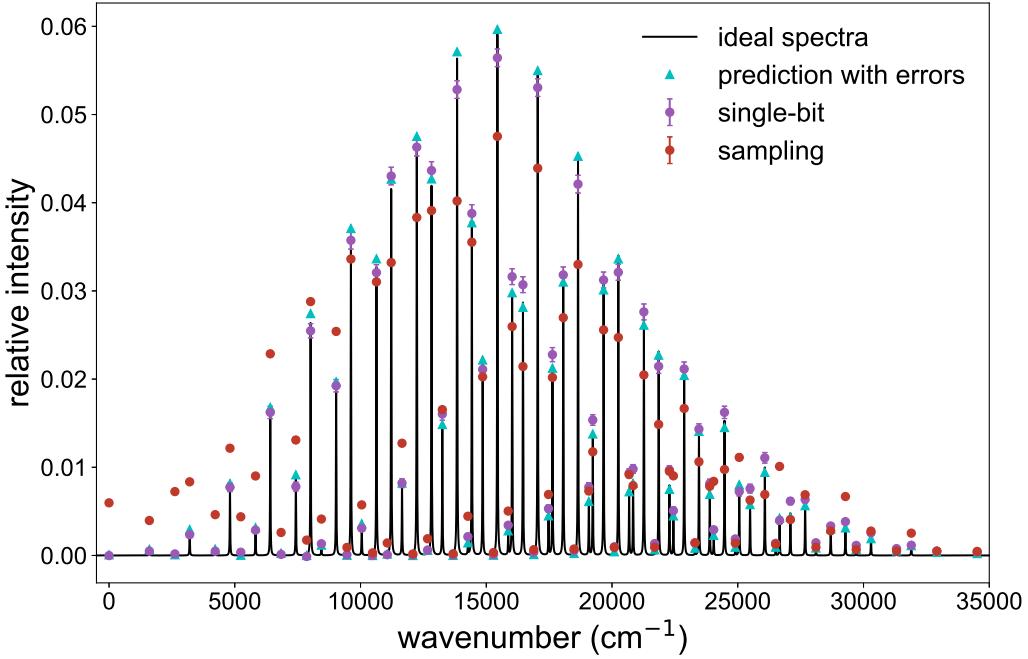


FIG. 1. Photoionization of neutral water to the ( $\tilde{B}^2\text{B}_2$ ) excited state of the cation starting in the vibrationless state  $n = 0$ ,  $m = 0$ .

| $\text{H}_2\text{O} \xrightarrow{h\nu} \text{H}_2\text{O}^+(\tilde{B}^2\text{B}_2) + \text{e}^-$ starting in $(n = 0, m = 0)$ |                        |                            |                       |                  |
|---|------------------------|----------------------------|-----------------------|------------------|
| $(n', m')$  | Classically calculated | Master equation simulation | Single-bit extraction | Sampling         |
| (0,0)   | 7.92E-05               | 8.26E-05                   | 0                     | 0.006 ± 0.0001   |
| (0,1)   | 6.67E-04               | 7.01E-04                   | 0.0005 ± 0.0002       | 0.00396 ± 0.0001 |
| (0,2)   | 2.80E-03               | 2.94E-03                   | 0.0024 ± 0.0003       | 0.0084 ± 0.0001  |
| (0,3)   | 7.76E-03               | 8.15E-03                   | 0.0077 ± 0.0005       | 0.0122 ± 0.0002  |
| (0,4)   | 1.60E-02               | 1.68E-02                   | 0.0162 ± 0.0007       | 0.0229 ± 0.0002  |
| (0,5)   | 2.64E-02               | 2.74E-02                   | 0.0255 ± 0.0008       | 0.0288 ± 0.0003  |
| (0,6)   | 3.59E-02               | 3.70E-02                   | 0.0357 ± 0.001        | 0.0336 ± 0.0003  |
| (0,7)   | 4.16E-02               | 4.26E-02                   | 0.043 ± 0.001         | 0.0332 ± 0.0003  |
| (0,8)   | 4.19E-02               | 4.27E-02                   | 0.044 ± 0.001         | 0.0391 ± 0.0003  |
| (0,9)   | 3.73E-02               | 3.77E-02                   | 0.039 ± 0.001         | 0.0355 ± 0.0003  |
| (0,10)  | 2.96E-02               | 2.98E-02                   | 0.0316 ± 0.0009       | 0.026 ± 0.0003   |
| (0,11)  | 2.13E-02               | 2.12E-02                   | 0.0228 ± 0.0008       | 0.0202 ± 0.0002  |
| (0,12)  | 1.39E-02               | 1.38E-02                   | 0.0154 ± 0.0006       | 0.0118 ± 0.0002  |
| (0,13)  | 8.33E-03               | 8.20E-03                   | 0.0098 ± 0.0005       | 0.0079 ± 0.0001  |
| (0,14)  | 4.60E-03               | 4.50E-03                   | 0.0051 ± 0.0004       | 0.009 ± 0.0001   |
| (0,15)  | 2.36E-03               | 2.29E-03                   | 0.0029 ± 0.0003       | 0.0084 ± 0.0001  |
| (1,0)   | 7.82E-05               | 8.11E-05                   | 0.0002 ± 0.0002       | 0.0072 ± 0.0001  |
| (1,1)   | 6.91E-04               | 7.19E-04                   | 0.0005 ± 0.0002       | 0.0046 ± 0.0001  |
| (1,2)   | 3.03E-03               | 3.16E-03                   | 0.0029 ± 0.0003       | 0.009 ± 0.0001   |
| (1,3)   | 8.81E-03               | 9.14E-03                   | 0.0078 ± 0.0005       | 0.0131 ± 0.0002  |
| (1,4)   | 1.91E-02               | 1.97E-02                   | 0.0192 ± 0.0007       | 0.0254 ± 0.0002  |
| (1,5)   | 3.28E-02               | 3.36E-02                   | 0.0321 ± 0.0009       | 0.031 ± 0.0003   |
| (1,6)   | 4.66E-02               | 4.75E-02                   | 0.046 ± 0.001         | 0.0383 ± 0.0003  |
| (1,7)   | 5.63E-02               | 5.71E-02                   | 0.053 ± 0.001         | 0.0402 ± 0.0003  |
| (1,8)   | 5.92E-02               | 5.97E-02                   | 0.056 ± 0.001         | 0.0475 ± 0.0003  |
| (1,9)   | 5.49E-02               | 5.50E-02                   | 0.053 ± 0.001         | 0.0439 ± 0.0003  |

|        |          |          |                 |                 |
|--------|----------|----------|-----------------|-----------------|
| (1,10) | 4.55E-02 | 4.53E-02 | 0.042 ± 0.001   | 0.033 ± 0.0003  |
| (1,11) | 3.40E-02 | 3.37E-02 | 0.0321 ± 0.0009 | 0.0247 ± 0.0002 |
| (1,12) | 2.32E-02 | 2.28E-02 | 0.0214 ± 0.0008 | 0.0149 ± 0.0002 |
| (1,13) | 1.44E-02 | 1.41E-02 | 0.0143 ± 0.0006 | 0.0106 ± 0.0002 |
| (1,14) | 8.30E-03 | 8.04E-03 | 0.0073 ± 0.0005 | 0.0111 ± 0.0002 |
| (1,15) | 4.42E-03 | 4.25E-03 | 0.004 ± 0.0004  | 0.0101 ± 0.0002 |
| (2,0)  | 2.61E-05 | 2.68E-05 | 0.0004 ± 0.0001 | 0.0044 ± 0.0001 |
| (2,1)  | 2.47E-04 | 2.52E-04 | 0.0001 ± 0.0001 | 0.00261 ± 8E-05 |
| (2,2)  | 1.15E-03 | 1.17E-03 | 0.0013 ± 0.0002 | 0.0041 ± 0.0001 |
| (2,3)  | 3.57E-03 | 3.60E-03 | 0.0031 ± 0.0003 | 0.0057 ± 0.0001 |
| (2,4)  | 8.19E-03 | 8.21E-03 | 0.0082 ± 0.0005 | 0.0127 ± 0.0002 |
| (2,5)  | 1.49E-02 | 1.48E-02 | 0.016 ± 0.0007  | 0.0165 ± 0.0002 |
| (2,6)  | 2.25E-02 | 2.22E-02 | 0.0211 ± 0.0008 | 0.0203 ± 0.0002 |
| (2,7)  | 2.87E-02 | 2.81E-02 | 0.0307 ± 0.0009 | 0.0214 ± 0.0002 |
| (2,8)  | 3.19E-02 | 3.10E-02 | 0.0318 ± 0.0009 | 0.027 ± 0.0003  |
| (2,9)  | 3.12E-02 | 3.01E-02 | 0.0312 ± 0.0009 | 0.0256 ± 0.0002 |
| (2,10) | 2.72E-02 | 2.61E-02 | 0.0276 ± 0.0009 | 0.0205 ± 0.0002 |
| (2,11) | 2.14E-02 | 2.04E-02 | 0.0211 ± 0.0008 | 0.0167 ± 0.0002 |
| (2,12) | 1.53E-02 | 1.45E-02 | 0.0162 ± 0.0007 | 0.0097 ± 0.0002 |
| (2,13) | 1.00E-02 | 9.46E-03 | 0.0111 ± 0.0006 | 0.0069 ± 0.0001 |
| (2,14) | 6.02E-03 | 5.67E-03 | 0.0064 ± 0.0004 | 0.0069 ± 0.0001 |
| (2,15) | 3.36E-03 | 3.14E-03 | 0.0038 ± 0.0003 | 0.0067 ± 0.0001 |
| (3,0)  | 2.78E-06 | 3.35E-06 | 0               | 0.00174 ± 7E-05 |
| (3,1)  | 2.97E-05 | 3.43E-05 | 8E-05 ± 8E-05   | 0.00091 ± 5E-05 |
| (3,2)  | 1.56E-04 | 1.74E-04 | 9E-05 ± 8E-05   | 0.00142 ± 6E-05 |
| (3,3)  | 5.37E-04 | 5.79E-04 | 0.0006 ± 0.0002 | 0.00189 ± 7E-05 |
| (3,4)  | 1.36E-03 | 1.43E-03 | 0.0021 ± 0.0003 | 0.0044 ± 0.0001 |
| (3,5)  | 2.73E-03 | 2.79E-03 | 0.0034 ± 0.0003 | 0.005 ± 0.0001  |
| (3,6)  | 4.50E-03 | 4.49E-03 | 0.0053 ± 0.0004 | 0.0069 ± 0.0001 |
| (3,7)  | 6.26E-03 | 6.12E-03 | 0.0078 ± 0.0005 | 0.0073 ± 0.0001 |
| (3,8)  | 7.52E-03 | 7.22E-03 | 0.0093 ± 0.0005 | 0.0092 ± 0.0002 |
| (3,9)  | 7.94E-03 | 7.50E-03 | 0.0097 ± 0.0005 | 0.0095 ± 0.0002 |
| (3,10) | 7.46E-03 | 6.94E-03 | 0.0082 ± 0.0005 | 0.0078 ± 0.0001 |
| (3,11) | 6.29E-03 | 5.78E-03 | 0.0076 ± 0.0005 | 0.0063 ± 0.0001 |
| (3,12) | 4.81E-03 | 4.37E-03 | 0.0062 ± 0.0004 | 0.0041 ± 0.0001 |
| (3,13) | 3.36E-03 | 3.01E-03 | 0.0033 ± 0.0003 | 0.00279 ± 8E-05 |
| (3,14) | 2.16E-03 | 1.91E-03 | 0.0026 ± 0.0003 | 0.00277 ± 8E-05 |
| (3,15) | 1.28E-03 | 1.12E-03 | 0.0012 ± 0.0002 | 0.00253 ± 8E-05 |
| (4,0)  | 2.66E-09 | 1.53E-07 | 0               | 0.00029 ± 3E-05 |
| (4,1)  | 1.45E-07 | 1.74E-06 | 6E-05 ± 5E-05   | 0.00017 ± 2E-05 |
| (4,2)  | 1.73E-06 | 9.99E-06 | 6E-05 ± 6E-05   | 0.0002 ± 2E-05  |
| (4,3)  | 1.02E-05 | 3.79E-05 | 0.0001 ± 8E-05  | 0.00033 ± 3E-05 |
| (4,4)  | 3.86E-05 | 1.06E-04 | 0.0004 ± 0.0001 | 0.00065 ± 4E-05 |
| (4,5)  | 1.06E-04 | 2.33E-04 | 0.0005 ± 0.0002 | 0.00073 ± 4E-05 |
| (4,6)  | 2.25E-04 | 4.19E-04 | 0.001 ± 0.0002  | 0.0009 ± 5E-05  |
| (4,7)  | 3.88E-04 | 6.34E-04 | 0.0014 ± 0.0002 | 0.00092 ± 5E-05 |
| (4,8)  | 5.63E-04 | 8.24E-04 | 0.0014 ± 0.0002 | 0.00144 ± 6E-05 |
| (4,9)  | 7.01E-04 | 9.36E-04 | 0.0019 ± 0.0003 | 0.00139 ± 6E-05 |
| (4,10) | 7.63E-04 | 9.43E-04 | 0.0014 ± 0.0002 | 0.00127 ± 6E-05 |
| (4,11) | 7.36E-04 | 8.50E-04 | 0.0014 ± 0.0002 | 0.00094 ± 5E-05 |
| (4,12) | 6.36E-04 | 6.92E-04 | 0.0011 ± 0.0002 | 0.00067 ± 4E-05 |
| (4,13) | 4.97E-04 | 5.13E-04 | 0.0008 ± 0.0002 | 0.00051 ± 4E-05 |
| (4,14) | 3.54E-04 | 3.49E-04 | 0.0005 ± 0.0001 | 0.00049 ± 3E-05 |
| (4,15) | 2.31E-04 | 2.18E-04 | 0.0004 ± 0.0001 | 0.00046 ± 3E-05 |

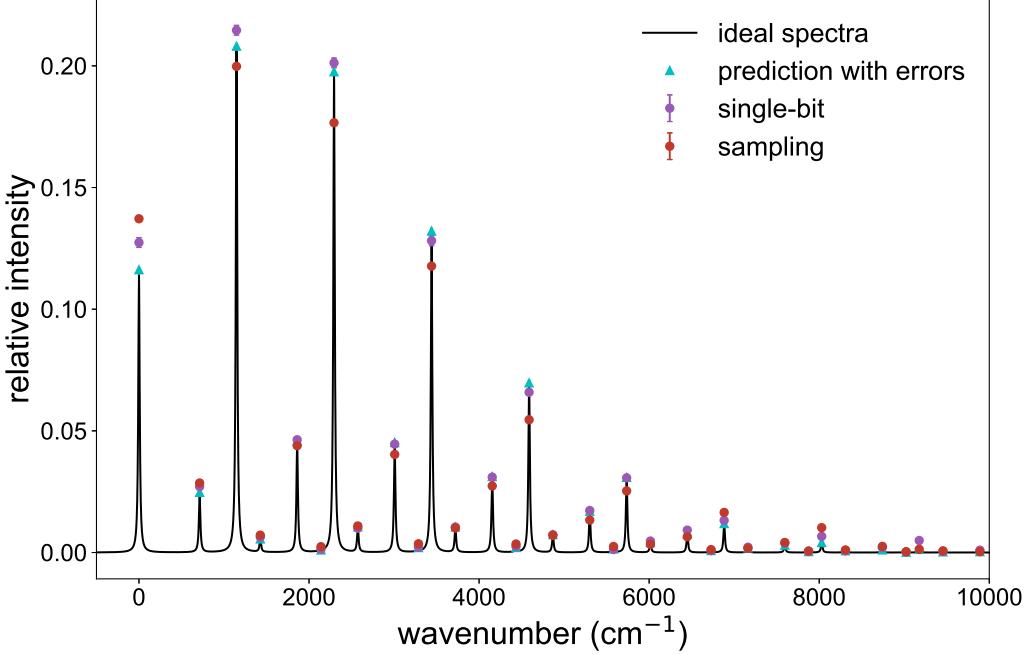


FIG. 2. Photoionization of the ozone anion to neutral ozone starting in the vibrationless state  $n = 0, m = 0$ .

| $O_3^- \xrightarrow{h\nu} O_3 + e^-$ starting in $(n = 0, m = 0)$ |                        |                            |                       |                 |
|---|------------------------|----------------------------|-----------------------|-----------------|
| $(n', m')$  | Classically calculated | Master equation simulation | Single-bit extraction | Sampling        |
| (0,0)   | 1.14E-01               | 1.16E-01                   | 0.127 ± 0.002         | 0.1372 ± 0.0005 |
| (0,1)   | 2.42E-02               | 2.47E-02                   | 0.0271 ± 0.0008       | 0.0286 ± 0.0003 |
| (0,2)   | 5.57E-03               | 5.60E-03                   | 0.0063 ± 0.0004       | 0.0071 ± 0.0001 |
| (0,3)   | 1.06E-03               | 1.06E-03                   | 0.0013 ± 0.0002       | 0.0024 ± 8E-05  |
| (1,0)   | 2.06E-01               | 2.08E-01                   | 0.215 ± 0.002         | 0.1998 ± 0.0006 |
| (1,1)   | 4.55E-02               | 4.60E-02                   | 0.046 ± 0.001         | 0.0439 ± 0.0003 |
| (1,2)   | 1.06E-02               | 1.06E-02                   | 0.01 ± 0.0005         | 0.0109 ± 0.0002 |
| (1,3)   | 2.06E-03               | 2.04E-03                   | 0.0022 ± 0.0003       | 0.0036 ± 0.0001 |
| (2,0)   | 1.98E-01               | 1.98E-01                   | 0.201 ± 0.002         | 0.1766 ± 0.0006 |
| (2,1)   | 4.52E-02               | 4.52E-02                   | 0.045 ± 0.001         | 0.0403 ± 0.0003 |
| (2,2)   | 1.07E-02               | 1.05E-02                   | 0.0104 ± 0.0005       | 0.0101 ± 0.0002 |
| (2,3)   | 2.11E-03               | 2.06E-03                   | 0.0025 ± 0.0003       | 0.00349 ± 9E-05 |
| (3,0)   | 1.33E-01               | 1.32E-01                   | 0.128 ± 0.002         | 0.1177 ± 0.0005 |
| (3,1)   | 3.15E-02               | 3.12E-02                   | 0.031 ± 0.0009        | 0.0274 ± 0.0003 |
| (3,2)   | 7.54E-03               | 7.37E-03                   | 0.0073 ± 0.0005       | 0.0071 ± 0.0001 |
| (3,3)   | 1.51E-03               | 1.47E-03                   | 0.0012 ± 0.0002       | 0.00248 ± 8E-05 |
| (4,0)   | 7.08E-02               | 6.97E-02                   | 0.066 ± 0.001         | 0.0546 ± 0.0004 |
| (4,1)   | 1.73E-02               | 1.70E-02                   | 0.0173 ± 0.0007       | 0.0133 ± 0.0002 |
| (4,2)   | 4.19E-03               | 4.06E-03                   | 0.0047 ± 0.0004       | 0.00342 ± 9E-05 |
| (4,3)   | 8.51E-04               | 8.19E-04                   | 0.0008 ± 0.0002       | 0.0012 ± 6E-05  |
| (5,0)   | 3.15E-02               | 3.09E-02                   | 0.0307 ± 0.0009       | 0.0253 ± 0.0003 |
| (5,1)   | 7.91E-03               | 7.74E-03                   | 0.0092 ± 0.0005       | 0.0065 ± 0.0001 |
| (5,2)   | 1.94E-03               | 1.87E-03                   | 0.0022 ± 0.0003       | 0.00173 ± 7E-05 |
| (5,3)   | 4.01E-04               | 3.83E-04                   | 0.0004 ± 0.0001       | 0.00064 ± 4E-05 |
| (6,0)   | 1.22E-02               | 1.19E-02                   | 0.0132 ± 0.0007       | 0.0165 ± 0.0002 |
| (6,1)   | 3.15E-03               | 3.07E-03                   | 0.0039 ± 0.0004       | 0.0041 ± 0.0001 |
| (6,2)   | 7.83E-04               | 7.52E-04                   | 0.0007 ± 0.0002       | 0.00105 ± 5E-05 |

|       |          |          |                 |                 |
|-------|----------|----------|-----------------|-----------------|
| (6,3) | 1.64E-04 | 1.56E-04 | 0.00024 ± 9E-05 | 0.00036 ± 3E-05 |
| (7,0) | 4.23E-03 | 4.10E-03 | 0.0066 ± 0.0005 | 0.0102 ± 0.0002 |
| (7,1) | 1.12E-03 | 1.09E-03 | 0.0021 ± 0.0003 | 0.00252 ± 8E-05 |
| (7,2) | 2.81E-04 | 2.69E-04 | 0.0005 ± 0.0001 | 0.00068 ± 4E-05 |
| (7,3) | 5.96E-05 | 5.64E-05 | 0.00019 ± 8E-05 | 0.00017 ± 2E-05 |
| (8,0) | 1.33E-03 | 1.28E-03 | 0.0049 ± 0.0005 | 0.00132 ± 6E-05 |
| (8,1) | 3.61E-04 | 3.48E-04 | 0.001 ± 0.0002  | 0.00035 ± 3E-05 |
| (8,2) | 9.18E-05 | 8.73E-05 | 0.0002 ± 0.0001 | 9E-05 ± 2E-05   |
| (8,3) | 1.97E-05 | 1.85E-05 | 0.00013 ± 7E-05 | 2.8E-05 ± 8E-06 |
| (9,0) | 3.86E-04 | 3.68E-04 | 0.0047 ± 0.0005 | 0.00156 ± 6E-05 |
| (9,1) | 1.07E-04 | 1.03E-04 | 0.0012 ± 0.0002 | 0.00038 ± 3E-05 |
| (9,2) | 2.76E-05 | 2.60E-05 | 0.0004 ± 0.0001 | 8E-05 ± 1E-05   |
| (9,3) | 5.98E-06 | 5.59E-06 | 0.00013 ± 7E-05 | 3.6E-05 ± 1E-05 |

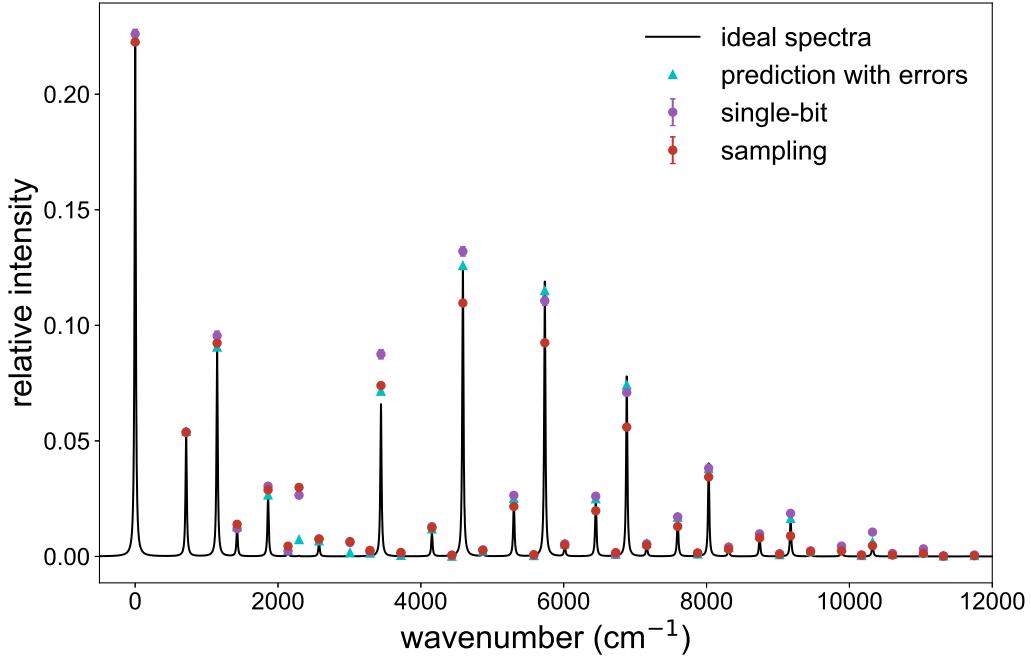


FIG. 3. Photoionization of the ozone anion to neutral ozone starting with one quanta in the symmetric-stretching mode and zero in the bending mode  $n = 1$ ,  $m = 0$ .

| $\text{O}_3^- \xrightarrow{h\nu} \text{O}_3 + \text{e}^-$ starting in $(n = 1, m = 0)$ |                        |                            |                       |                     |
|--|------------------------|----------------------------|-----------------------|---------------------|
| $(n', m')$   | Classically calculated | Master equation simulation | Single-bit extraction | Sampling            |
| (0,0)  | 2.24E-01               | 2.23E-01                   | 0.226 $\pm$ 0.002     | 0.2226 $\pm$ 0.0007 |
| (0,1)  | 5.42E-02               | 5.40E-02                   | 0.054 $\pm$ 0.001     | 0.0537 $\pm$ 0.0004 |
| (0,2)  | 1.31E-02               | 1.28E-02                   | 0.0121 $\pm$ 0.0006   | 0.0139 $\pm$ 0.0002 |
| (0,3)  | 2.64E-03               | 2.56E-03                   | 0.0023 $\pm$ 0.0003   | 0.0044 $\pm$ 0.0001 |
| (1,0)  | 8.96E-02               | 9.09E-02                   | 0.095 $\pm$ 0.002     | 0.0923 $\pm$ 0.0005 |
| (1,1)  | 2.67E-02               | 2.69E-02                   | 0.0304 $\pm$ 0.0009   | 0.0288 $\pm$ 0.0003 |
| (1,2)  | 6.90E-03               | 6.83E-03                   | 0.0075 $\pm$ 0.0005   | 0.0075 $\pm$ 0.0001 |
| (1,3)  | 1.51E-03               | 1.48E-03                   | 0.002 $\pm$ 0.0003    | 0.00263 $\pm$ 9E-05 |
| (2,0)  | 3.14E-04               | 5.76E-03                   | 0.0265 $\pm$ 0.0009   | 0.0299 $\pm$ 0.0003 |
| (2,1)  | 2.69E-04               | 1.39E-03                   | 0.006 $\pm$ 0.0004    | 0.0064 $\pm$ 0.0001 |
| (2,2)  | 1.59E-04               | 4.07E-04                   | 0.0013 $\pm$ 0.0002   | 0.00174 $\pm$ 7E-05 |
| (2,3)  | 6.61E-05               | 1.11E-04                   | 0.0004 $\pm$ 0.0001   | 0.00059 $\pm$ 4E-05 |
| (3,0)  | 6.59E-02               | 6.96E-02                   | 0.088 $\pm$ 0.002     | 0.074 $\pm$ 0.0004  |
| (3,1)  | 1.05E-02               | 1.14E-02                   | 0.0129 $\pm$ 0.0006   | 0.0124 $\pm$ 0.0002 |
| (3,2)  | 2.11E-03               | 2.27E-03                   | 0.0028 $\pm$ 0.0003   | 0.00267 $\pm$ 9E-05 |
| (3,3)  | 3.34E-04               | 3.67E-04                   | 0.0005 $\pm$ 0.0001   | 0.00078 $\pm$ 5E-05 |
| (4,0)  | 1.26E-01               | 1.25E-01                   | 0.132 $\pm$ 0.002     | 0.1097 $\pm$ 0.0005 |
| (4,1)  | 2.51E-02               | 2.49E-02                   | 0.0264 $\pm$ 0.0009   | 0.0216 $\pm$ 0.0002 |
| (4,2)  | 5.55E-03               | 5.42E-03                   | 0.0054 $\pm$ 0.0004   | 0.0049 $\pm$ 0.0001 |
| (4,3)  | 1.01E-03               | 9.81E-04                   | 0.0009 $\pm$ 0.0002   | 0.00165 $\pm$ 7E-05 |
| (5,0)  | 1.19E-01               | 1.16E-01                   | 0.111 $\pm$ 0.002     | 0.0925 $\pm$ 0.0005 |
| (5,1)  | 2.58E-02               | 2.51E-02                   | 0.0261 $\pm$ 0.0009   | 0.0198 $\pm$ 0.0002 |
| (5,2)  | 5.94E-03               | 5.68E-03                   | 0.0054 $\pm$ 0.0004   | 0.0048 $\pm$ 0.0001 |
| (5,3)  | 1.14E-03               | 1.08E-03                   | 0.0014 $\pm$ 0.0002   | 0.00153 $\pm$ 7E-05 |
| (6,0)  | 7.79E-02               | 7.54E-02                   | 0.071 $\pm$ 0.001     | 0.056 $\pm$ 0.0004  |
| (6,1)  | 1.79E-02               | 1.73E-02                   | 0.0171 $\pm$ 0.0007   | 0.013 $\pm$ 0.0002  |

|       |          |          |                     |                     |
|-------|----------|----------|---------------------|---------------------|
| (6,2) | 4.23E-03 | 4.01E-03 | 0.0041 $\pm$ 0.0004 | 0.00313 $\pm$ 9E-05 |
| (6,3) | 8.33E-04 | 7.82E-04 | 0.0012 $\pm$ 0.0002 | 0.00091 $\pm$ 5E-05 |
| (7,0) | 4.03E-02 | 3.89E-02 | 0.038 $\pm$ 0.001   | 0.0344 $\pm$ 0.0003 |
| (7,1) | 9.69E-03 | 9.34E-03 | 0.0098 $\pm$ 0.0005 | 0.008 $\pm$ 0.0001  |
| (7,2) | 2.33E-03 | 2.21E-03 | 0.0024 $\pm$ 0.0003 | 0.00207 $\pm$ 8E-05 |
| (7,3) | 4.70E-04 | 4.40E-04 | 0.0004 $\pm$ 0.0001 | 0.00065 $\pm$ 4E-05 |
| (8,0) | 1.75E-02 | 1.69E-02 | 0.0186 $\pm$ 0.0008 | 0.0089 $\pm$ 0.0002 |
| (8,1) | 4.38E-03 | 4.22E-03 | 0.0045 $\pm$ 0.0004 | 0.00239 $\pm$ 8E-05 |
| (8,2) | 1.07E-03 | 1.02E-03 | 0.0013 $\pm$ 0.0002 | 0.00059 $\pm$ 4E-05 |
| (8,3) | 2.20E-04 | 2.06E-04 | 0.00016 $\pm$ 8E-05 | 0.0002 $\pm$ 2E-05  |
| (9,0) | 6.66E-03 | 6.43E-03 | 0.0105 $\pm$ 0.0006 | 0.0046 $\pm$ 0.0001 |
| (9,1) | 1.72E-03 | 1.66E-03 | 0.0033 $\pm$ 0.0003 | 0.00132 $\pm$ 6E-05 |
| (9,2) | 4.28E-04 | 4.05E-04 | 0.0006 $\pm$ 0.0002 | 0.00034 $\pm$ 3E-05 |
| (9,3) | 8.93E-05 | 8.37E-05 | 3E-05 $\pm$ 6E-05   | 0.00013 $\pm$ 2E-05 |

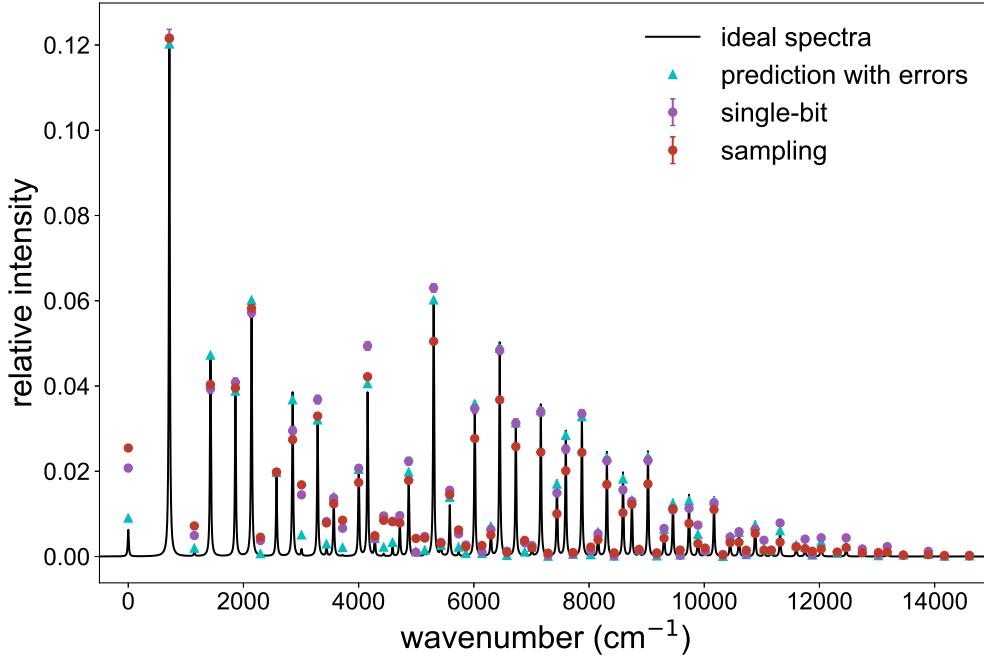


FIG. 4. Photoionization of the ozone anion to neutral ozone starting with one quanta in the symmetric-stretching mode and two in the bending mode  $n = 1, m = 2$ .

| $(n', m')$ | $O_3^- \xrightarrow{h\nu} O_3 + e^-$ starting in $(n = 1, m = 2)$ |                            |                       |                      |
|------------|---|----------------------------|-----------------------|----------------------|
|            | Classically calculated  | Master equation simulation | Single-bit extraction | Sampling             |
| (0,0)      | 6.24E-03  | 9.10E-03                   | 0.0208 $\pm$ 0.0008   | 0.0255 $\pm$ 0.0003  |
| (0,1)      | 1.21E-01  | 1.20E-01                   | 0.122 $\pm$ 0.002     | 0.1215 $\pm$ 0.0006  |
| (0,2)      | 4.73E-02  | 4.75E-02                   | 0.039 $\pm$ 0.001     | 0.0403 $\pm$ 0.0004  |
| (0,3)      | 6.01E-02  | 6.02E-02                   | 0.057 $\pm$ 0.001     | 0.0583 $\pm$ 0.0004  |
| (0,4)      | 3.85E-02  | 3.65E-02                   | 0.0295 $\pm$ 0.001    | 0.0274 $\pm$ 0.0003  |
| (0,5)      | 1.46E-02  | 1.39E-02                   | 0.0137 $\pm$ 0.0007   | 0.0124 $\pm$ 0.0002  |
| (0,6)      | 4.57E-03  | 4.18E-03                   | 0.0042 $\pm$ 0.0004   | 0.0049 $\pm$ 0.0001  |
| (0,7)      | 1.21E-03  | 1.10E-03                   | 0.001 $\pm$ 0.0002    | 0.0042 $\pm$ 0.0001  |
| (1,0)      | 9.99E-04  | 1.99E-03                   | 0.0049 $\pm$ 0.0004   | 0.0072 $\pm$ 0.0002  |
| (1,1)      | 3.79E-02  | 3.89E-02                   | 0.041 $\pm$ 0.001     | 0.0395 $\pm$ 0.0004  |
| (1,2)      | 1.90E-02  | 1.99E-02                   | 0.0198 $\pm$ 0.0008   | 0.0198 $\pm$ 0.0003  |
| (1,3)      | 3.22E-02  | 3.22E-02                   | 0.037 $\pm$ 0.001     | 0.0329 $\pm$ 0.0003  |
| (1,4)      | 2.14E-02  | 2.04E-02                   | 0.0207 $\pm$ 0.0008   | 0.0174 $\pm$ 0.0002  |
| (1,5)      | 8.78E-03  | 8.31E-03                   | 0.0096 $\pm$ 0.0006   | 0.0079 $\pm$ 0.0002  |
| (1,6)      | 2.87E-03  | 2.63E-03                   | 0.0033 $\pm$ 0.0003   | 0.0031 $\pm$ 0.0001  |
| (1,7)      | 7.99E-04  | 7.23E-04                   | 0.0011 $\pm$ 0.0002   | 0.00258 $\pm$ 0.0001 |
| (2,0)      | 4.14E-04  | 6.10E-04                   | 0.0037 $\pm$ 0.0004   | 0.0045 $\pm$ 0.0001  |
| (2,1)      | 1.56E-03  | 4.26E-03                   | 0.0145 $\pm$ 0.0007   | 0.0168 $\pm$ 0.0002  |
| (2,2)      | 2.44E-04  | 1.72E-03                   | 0.0066 $\pm$ 0.0005   | 0.0085 $\pm$ 0.0002  |
| (2,3)      | 5.64E-04  | 1.90E-03                   | 0.0095 $\pm$ 0.0006   | 0.0085 $\pm$ 0.0002  |
| (2,4)      | 6.09E-04  | 1.33E-03                   | 0.0047 $\pm$ 0.0004   | 0.0043 $\pm$ 0.0001  |
| (2,5)      | 4.53E-04  | 6.66E-04                   | 0.0027 $\pm$ 0.0003   | 0.00229 $\pm$ 9E-05  |
| (2,6)      | 1.95E-04  | 2.51E-04                   | 0.0012 $\pm$ 0.0002   | 0.00098 $\pm$ 6E-05  |
| (2,7)      | 6.97E-05  | 7.85E-05                   | 0.0003 $\pm$ 0.0001   | 0.0008 $\pm$ 5E-05   |
| (3,0)      | 2.02E-03  | 2.95E-03                   | 0.0082 $\pm$ 0.0005   | 0.0079 $\pm$ 0.0002  |
| (3,1)      | 3.84E-02  | 3.97E-02                   | 0.049 $\pm$ 0.001     | 0.0422 $\pm$ 0.0004  |

|       |          |          |                     |                     |
|-------|----------|----------|---------------------|---------------------|
| (3,2) | 1.87E-02 | 1.94E-02 | 0.0224 $\pm$ 0.0009 | 0.0179 $\pm$ 0.0003 |
| (3,3) | 1.20E-02 | 1.34E-02 | 0.0155 $\pm$ 0.0007 | 0.0145 $\pm$ 0.0002 |
| (3,4) | 6.36E-03 | 6.72E-03 | 0.0064 $\pm$ 0.0005 | 0.0051 $\pm$ 0.0001 |
| (3,5) | 1.83E-03 | 2.05E-03 | 0.0026 $\pm$ 0.0003 | 0.00206 $\pm$ 9E-05 |
| (3,6) | 4.70E-04 | 5.06E-04 | 0.0006 $\pm$ 0.0002 | 0.00093 $\pm$ 6E-05 |
| (3,7) | 9.93E-05 | 1.14E-04 | 0.0002 $\pm$ 0.0001 | 0.00086 $\pm$ 6E-05 |
| (4,0) | 2.05E-03 | 3.41E-03 | 0.0082 $\pm$ 0.0005 | 0.0082 $\pm$ 0.0002 |
| (4,1) | 6.06E-02 | 6.01E-02 | 0.063 $\pm$ 0.001   | 0.0505 $\pm$ 0.0004 |
| (4,2) | 3.64E-02 | 3.58E-02 | 0.035 $\pm$ 0.001   | 0.0277 $\pm$ 0.0003 |
| (4,3) | 3.11E-02 | 3.13E-02 | 0.031 $\pm$ 0.001   | 0.0258 $\pm$ 0.0003 |
| (4,4) | 1.77E-02 | 1.68E-02 | 0.0149 $\pm$ 0.0007 | 0.01 $\pm$ 0.0002   |
| (4,5) | 5.91E-03 | 5.69E-03 | 0.0054 $\pm$ 0.0004 | 0.0039 $\pm$ 0.0001 |
| (4,6) | 1.69E-03 | 1.55E-03 | 0.0014 $\pm$ 0.0002 | 0.00167 $\pm$ 8E-05 |
| (4,7) | 4.09E-04 | 3.80E-04 | 0.0003 $\pm$ 0.0001 | 0.00151 $\pm$ 7E-05 |
| (5,0) | 1.18E-03 | 2.29E-03 | 0.0053 $\pm$ 0.0004 | 0.0062 $\pm$ 0.0002 |
| (5,1) | 5.02E-02 | 4.93E-02 | 0.048 $\pm$ 0.001   | 0.0367 $\pm$ 0.0004 |
| (5,2) | 3.57E-02 | 3.46E-02 | 0.034 $\pm$ 0.001   | 0.0245 $\pm$ 0.0003 |
| (5,3) | 3.39E-02 | 3.31E-02 | 0.033 $\pm$ 0.001   | 0.0244 $\pm$ 0.0003 |
| (5,4) | 1.96E-02 | 1.82E-02 | 0.0156 $\pm$ 0.0007 | 0.0103 $\pm$ 0.0002 |
| (5,5) | 6.87E-03 | 6.40E-03 | 0.0066 $\pm$ 0.0005 | 0.0043 $\pm$ 0.0001 |
| (5,6) | 2.03E-03 | 1.81E-03 | 0.0012 $\pm$ 0.0002 | 0.00196 $\pm$ 8E-05 |
| (5,7) | 5.10E-04 | 4.57E-04 | 0.0005 $\pm$ 0.0002 | 0.00148 $\pm$ 7E-05 |
| (6,0) | 4.73E-04 | 1.13E-03 | 0.0032 $\pm$ 0.0003 | 0.0038 $\pm$ 0.0001 |
| (6,1) | 2.94E-02 | 2.89E-02 | 0.0252 $\pm$ 0.0009 | 0.0201 $\pm$ 0.0003 |
| (6,2) | 2.45E-02 | 2.36E-02 | 0.0225 $\pm$ 0.0009 | 0.0169 $\pm$ 0.0002 |
| (6,3) | 2.47E-02 | 2.37E-02 | 0.0226 $\pm$ 0.0009 | 0.017 $\pm$ 0.0002  |
| (6,4) | 1.44E-02 | 1.32E-02 | 0.0113 $\pm$ 0.0006 | 0.0078 $\pm$ 0.0002 |
| (6,5) | 5.18E-03 | 4.76E-03 | 0.0046 $\pm$ 0.0004 | 0.0033 $\pm$ 0.0001 |
| (6,6) | 1.56E-03 | 1.38E-03 | 0.0015 $\pm$ 0.0002 | 0.00143 $\pm$ 7E-05 |
| (6,7) | 4.00E-04 | 3.53E-04 | 0.0005 $\pm$ 0.0001 | 0.00122 $\pm$ 7E-05 |
| (7,0) | 1.44E-04 | 4.53E-04 | 0.0013 $\pm$ 0.0002 | 0.00221 $\pm$ 9E-05 |
| (7,1) | 1.37E-02 | 1.35E-02 | 0.013 $\pm$ 0.0007  | 0.0122 $\pm$ 0.0002 |
| (7,2) | 1.33E-02 | 1.28E-02 | 0.0109 $\pm$ 0.0006 | 0.0112 $\pm$ 0.0002 |
| (7,3) | 1.39E-02 | 1.33E-02 | 0.0126 $\pm$ 0.0007 | 0.011 $\pm$ 0.0002  |
| (7,4) | 8.18E-03 | 7.46E-03 | 0.0064 $\pm$ 0.0005 | 0.0054 $\pm$ 0.0001 |
| (7,5) | 3.00E-03 | 2.73E-03 | 0.0024 $\pm$ 0.0003 | 0.00221 $\pm$ 9E-05 |
| (7,6) | 9.14E-04 | 8.02E-04 | 0.001 $\pm$ 0.0002  | 0.00102 $\pm$ 6E-05 |
| (7,7) | 2.38E-04 | 2.08E-04 | 0.0004 $\pm$ 0.0001 | 0.00092 $\pm$ 6E-05 |
| (8,0) | 3.37E-05 | 1.55E-04 | 0.0005 $\pm$ 0.0002 | 0.00085 $\pm$ 6E-05 |
| (8,1) | 5.37E-03 | 5.35E-03 | 0.0074 $\pm$ 0.0005 | 0.003 $\pm$ 0.0001  |
| (8,2) | 6.07E-03 | 5.83E-03 | 0.0058 $\pm$ 0.0005 | 0.0033 $\pm$ 0.0001 |
| (8,3) | 6.54E-03 | 6.22E-03 | 0.0079 $\pm$ 0.0005 | 0.0034 $\pm$ 0.0001 |
| (8,4) | 3.86E-03 | 3.52E-03 | 0.0044 $\pm$ 0.0004 | 0.00176 $\pm$ 8E-05 |
| (8,5) | 1.44E-03 | 1.30E-03 | 0.0018 $\pm$ 0.0003 | 0.00085 $\pm$ 6E-05 |
| (8,6) | 4.42E-04 | 3.88E-04 | 0.0003 $\pm$ 0.0001 | 0.00033 $\pm$ 3E-05 |
| (8,7) | 1.17E-04 | 1.02E-04 | 0.0001 $\pm$ 7E-05  | 0.00023 $\pm$ 3E-05 |
| (9,0) | 5.90E-06 | 4.75E-05 | 0.0005 $\pm$ 0.0002 | 0.00035 $\pm$ 4E-05 |
| (9,1) | 1.84E-03 | 1.84E-03 | 0.0038 $\pm$ 0.0004 | 0.00149 $\pm$ 7E-05 |
| (9,2) | 2.42E-03 | 2.32E-03 | 0.0041 $\pm$ 0.0004 | 0.00184 $\pm$ 8E-05 |
| (9,3) | 2.67E-03 | 2.53E-03 | 0.0044 $\pm$ 0.0004 | 0.00208 $\pm$ 9E-05 |
| (9,4) | 1.58E-03 | 1.44E-03 | 0.0023 $\pm$ 0.0003 | 0.00105 $\pm$ 6E-05 |
| (9,5) | 5.95E-04 | 5.39E-04 | 0.0012 $\pm$ 0.0002 | 0.00044 $\pm$ 4E-05 |
| (9,6) | 1.85E-04 | 1.62E-04 | 0.0003 $\pm$ 0.0001 | 0.00016 $\pm$ 2E-05 |
| (9,7) | 4.94E-05 | 4.28E-05 | 0.00012 $\pm$ 7E-05 | 0.00015 $\pm$ 2E-05 |

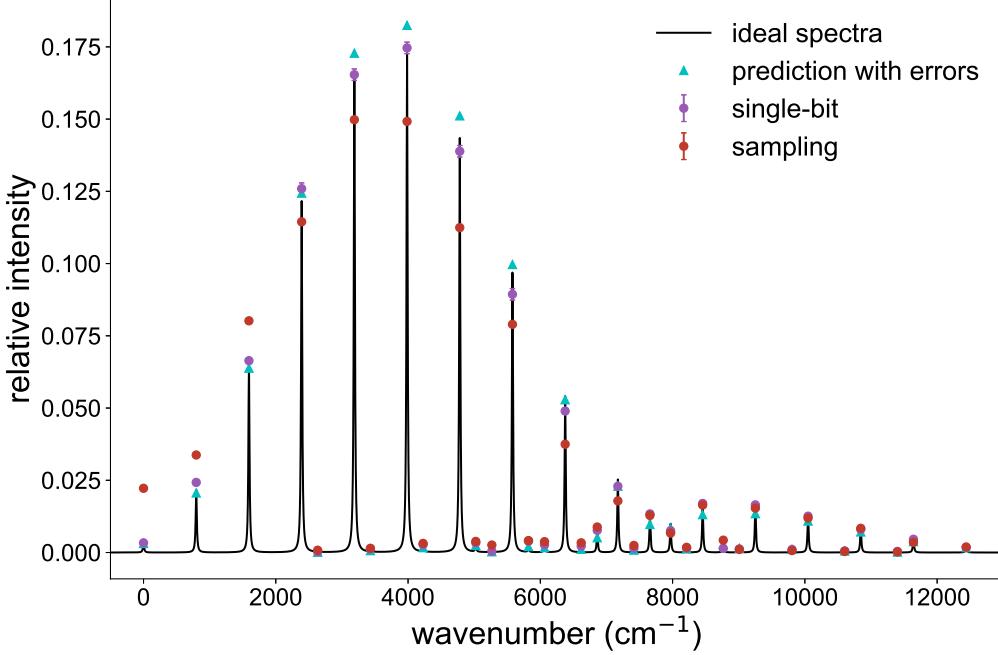


FIG. 5. Photoionization of nitrite to nitrogen dioxide starting in the vibrationless state  $n = 0$ ,  $m = 0$ .

| $\text{NO}_2^- \xrightarrow{h\nu} \text{NO}_2 + e^-$ starting in $(n = 0, m = 0)$ |                        |                            |                       |                  |
|---|------------------------|----------------------------|-----------------------|------------------|
| $(n', m')$  | Classically calculated | Master equation simulation | Single-bit extraction | Sampling         |
| (0,0)   | 3.54E-03               | 3.14E-03                   | 0.0033 ± 0.0004       | 0.0222 ± 0.0002  |
| (0,1)   | 2.17E-02               | 2.07E-02                   | 0.0243 ± 0.0008       | 0.0337 ± 0.0003  |
| (0,2)   | 6.42E-02               | 6.40E-02                   | 0.066 ± 0.001         | 0.0802 ± 0.0004  |
| (0,3)   | 1.22E-01               | 1.24E-01                   | 0.126 ± 0.002         | 0.1145 ± 0.0005  |
| (0,4)   | 1.66E-01               | 1.73E-01                   | 0.165 ± 0.002         | 0.1498 ± 0.0006  |
| (0,5)   | 1.73E-01               | 1.82E-01                   | 0.175 ± 0.002         | 0.1492 ± 0.0005  |
| (0,6)   | 1.43E-01               | 1.51E-01                   | 0.139 ± 0.002         | 0.1125 ± 0.0005  |
| (0,7)   | 9.68E-02               | 9.95E-02                   | 0.089 ± 0.002         | 0.079 ± 0.0004   |
| (0,8)   | 5.41E-02               | 5.27E-02                   | 0.049 ± 0.001         | 0.0375 ± 0.0003  |
| (0,9)   | 2.53E-02               | 2.29E-02                   | 0.0229 ± 0.0008       | 0.0179 ± 0.0002  |
| (0,10)  | 9.93E-03               | 8.49E-03                   | 0.0076 ± 0.0005       | 0.0067 ± 0.0001  |
| (0,11)  | 3.29E-03               | 2.75E-03                   | 0.0014 ± 0.0003       | 0.0043 ± 0.0001  |
| (1,0)   | 2.74E-04               | 1.03E-04                   | 0.00026 ± 0.0001      | 0.0008 ± 4E-05   |
| (1,1)   | 1.18E-03               | 6.52E-04                   | 0.0012 ± 0.0002       | 0.00147 ± 6E-05  |
| (1,2)   | 2.24E-03               | 1.73E-03                   | 0.0027 ± 0.0003       | 0.00316 ± 9E-05  |
| (1,3)   | 2.32E-03               | 2.48E-03                   | 0.0033 ± 0.0003       | 0.00384 ± 0.0001 |
| (1,4)   | 1.26E-03               | 2.04E-03                   | 0.0039 ± 0.0004       | 0.00414 ± 0.0001 |
| (1,5)   | 2.02E-04               | 1.04E-03                   | 0.0022 ± 0.0003       | 0.00336 ± 9E-05  |
| (1,6)   | 6.79E-05               | 7.77E-04                   | 0.0015 ± 0.0002       | 0.00245 ± 8E-05  |
| (1,7)   | 6.34E-04               | 1.27E-03                   | 0.0015 ± 0.0002       | 0.0018 ± 7E-05   |
| (1,8)   | 1.12E-03               | 1.53E-03                   | 0.0012 ± 0.0002       | 0.00107 ± 5E-05  |
| (1,9)   | 1.13E-03               | 1.15E-03                   | 0.0011 ± 0.0002       | 0.00067 ± 4E-05  |
| (1,10)  | 8.04E-04               | 5.44E-04                   | 0.0005 ± 0.0001       | 0.00039 ± 3E-05  |
| (1,11)  | 4.38E-04               | 1.56E-04                   | 0.0004 ± 0.0001       | 0.00021 ± 2E-05  |
| (2,0)   | 4.71E-04               | 2.67E-04                   | 0.0005 ± 0.0001       | 0.0026 ± 8E-05   |
| (2,1)   | 2.62E-03               | 1.71E-03                   | 0.0026 ± 0.0003       | 0.00376 ± 9E-05  |
| (2,2)   | 7.06E-03               | 5.16E-03                   | 0.0077 ± 0.0005       | 0.0088 ± 0.0001  |

|        |          |          |                     |                     |
|--------|----------|----------|---------------------|---------------------|
| (2,3)  | 1.24E-02 | 9.77E-03 | 0.0134 $\pm$ 0.0006 | 0.0128 $\pm$ 0.0002 |
| (2,4)  | 1.58E-02 | 1.31E-02 | 0.017 $\pm$ 0.0007  | 0.0164 $\pm$ 0.0002 |
| (2,5)  | 1.58E-02 | 1.35E-02 | 0.0165 $\pm$ 0.0007 | 0.0154 $\pm$ 0.0002 |
| (2,6)  | 1.28E-02 | 1.09E-02 | 0.0126 $\pm$ 0.0006 | 0.0119 $\pm$ 0.0002 |
| (2,7)  | 8.70E-03 | 7.12E-03 | 0.008 $\pm$ 0.0005  | 0.0084 $\pm$ 0.0001 |
| (2,8)  | 5.07E-03 | 3.79E-03 | 0.0046 $\pm$ 0.0004 | 0.00354 $\pm$ 9E-05 |
| (2,9)  | 2.56E-03 | 1.67E-03 | 0.0017 $\pm$ 0.0002 | 0.00191 $\pm$ 7E-05 |
| (2,10) | 1.14E-03 | 6.20E-04 | 0.0008 $\pm$ 0.0002 | 0.00064 $\pm$ 4E-05 |
| (2,11) | 4.45E-04 | 1.96E-04 | 0.0005 $\pm$ 0.0001 | 0.00039 $\pm$ 3E-05 |

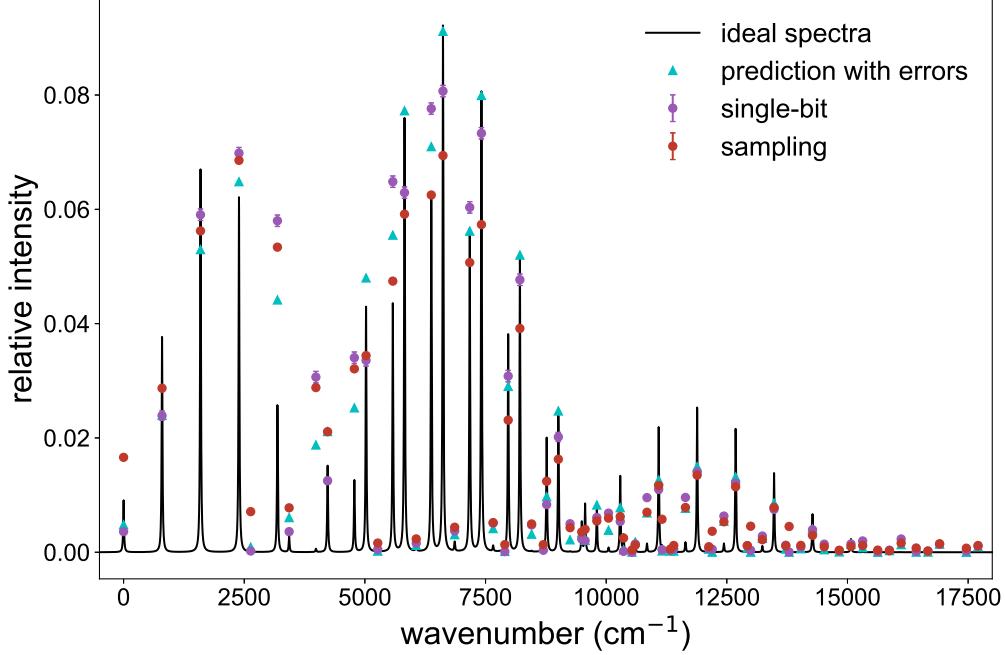


FIG. 6. Photoionization of nitrite to nitrogen dioxide starting with one quanta in the symmetric-stretching mode and zero in the bending mode  $n = 1$ ,  $m = 0$ . The more significant errors are primarily due to having a large self-Kerr on cavity A ( $\sim 30$  kHz) during the beamsplitter operation after starting in a state with higher photon number.

| $\text{NO}_2^- \xrightarrow{\hbar\nu} \text{NO}_2 + e^-$ starting in $(n = 1, m = 0)$ |                        |                            |                       |                 |
|---|------------------------|----------------------------|-----------------------|-----------------|
| $(n', m')$  | Classically calculated | Master equation simulation | Single-bit extraction | Sampling        |
| (0,0)   | 9.08E-03               | 4.87E-03                   | 0.0036 ± 0.0004       | 0.0166 ± 0.0002 |
| (0,1)   | 3.77E-02               | 2.40E-02                   | 0.0239 ± 0.0008       | 0.0287 ± 0.0003 |
| (0,2)   | 6.70E-02               | 5.28E-02                   | 0.059 ± 0.001         | 0.0562 ± 0.0004 |
| (0,3)   | 6.21E-02               | 6.36E-02                   | 0.07 ± 0.001          | 0.0686 ± 0.0004 |
| (0,4)   | 2.57E-02               | 4.12E-02                   | 0.058 ± 0.001         | 0.0534 ± 0.0004 |
| (0,5)   | 5.78E-04               | 1.47E-02                   | 0.0306 ± 0.001        | 0.0288 ± 0.0003 |
| (0,6)   | 1.25E-02               | 2.20E-02                   | 0.034 ± 0.001         | 0.0321 ± 0.0003 |
| (0,7)   | 4.34E-02               | 5.43E-02                   | 0.065 ± 0.001         | 0.0474 ± 0.0004 |
| (0,8)   | 6.17E-02               | 7.15E-02                   | 0.078 ± 0.001         | 0.0625 ± 0.0004 |
| (0,9)   | 5.65E-02               | 5.71E-02                   | 0.06 ± 0.001          | 0.0507 ± 0.0004 |
| (0,10)  | 3.80E-02               | 2.95E-02                   | 0.0308 ± 0.001        | 0.0231 ± 0.0003 |
| (0,11)  | 2.00E-02               | 9.85E-03                   | 0.0083 ± 0.0005       | 0.0124 ± 0.0002 |
| (0,12)  | 8.41E-03               | 2.11E-03                   | 0.002 ± 0.0003        | 0.004 ± 0.0001  |
| (0,13)  | 2.87E-03               | 4.62E-04                   | 0.0002 ± 0.0002       | 0.00248 ± 9E-05 |
| (0,14)  | 8.00E-04               | 2.41E-04                   | 0.0004 ± 0.0002       | 0.0058 ± 0.0001 |
| (1,0)   | 1.46E-04               | 9.10E-04                   | 0.0002 ± 0.0002       | 0.0071 ± 0.0001 |
| (1,1)   | 2.73E-03               | 6.17E-03                   | 0.0036 ± 0.0004       | 0.0078 ± 0.0002 |
| (1,2)   | 1.51E-02               | 2.15E-02                   | 0.0125 ± 0.0006       | 0.0211 ± 0.0002 |
| (1,3)   | 4.29E-02               | 4.89E-02                   | 0.0336 ± 0.001        | 0.0344 ± 0.0003 |
| (1,4)   | 7.59E-02               | 7.90E-02                   | 0.063 ± 0.001         | 0.0592 ± 0.0004 |
| (1,5)   | 9.21E-02               | 9.34E-02                   | 0.081 ± 0.001         | 0.0694 ± 0.0004 |
| (1,6)   | 8.05E-02               | 8.20E-02                   | 0.073 ± 0.001         | 0.0573 ± 0.0004 |
| (1,7)   | 5.16E-02               | 5.33E-02                   | 0.048 ± 0.001         | 0.0392 ± 0.0003 |
| (1,8)   | 2.39E-02               | 2.52E-02                   | 0.0202 ± 0.0008       | 0.0163 ± 0.0002 |

|        |          |          |                     |                     |
|--------|----------|----------|---------------------|---------------------|
| (1,9)  | 7.45E-03 | 8.37E-03 | 0.0061 $\pm$ 0.0005 | 0.0055 $\pm$ 0.0001 |
| (1,10) | 1.24E-03 | 1.78E-03 | 0.0014 $\pm$ 0.0003 | 0.00124 $\pm$ 6E-05 |
| (1,11) | 1.95E-05 | 1.99E-04 | 0.0006 $\pm$ 0.0002 | 0.00118 $\pm$ 6E-05 |
| (1,12) | 8.94E-05 | 1.33E-05 | 0.0005 $\pm$ 0.0002 | 0.0037 $\pm$ 0.0001 |
| (1,13) | 1.56E-04 | 1.69E-05 | 0.0003 $\pm$ 0.0002 | 0.0046 $\pm$ 0.0001 |
| (1,14) | 1.08E-04 | 2.40E-05 | 0                   | 0.0045 $\pm$ 0.0001 |
| (2,0)  | 3.83E-04 | 1.99E-04 | 0.0005 $\pm$ 0.0001 | 0.00161 $\pm$ 7E-05 |
| (2,1)  | 1.23E-03 | 1.20E-03 | 0.0016 $\pm$ 0.0002 | 0.00233 $\pm$ 8E-05 |
| (2,2)  | 1.61E-03 | 3.06E-03 | 0.0037 $\pm$ 0.0004 | 0.0044 $\pm$ 0.0001 |
| (2,3)  | 9.74E-04 | 4.06E-03 | 0.0051 $\pm$ 0.0004 | 0.0052 $\pm$ 0.0001 |
| (2,4)  | 1.41E-04 | 2.92E-03 | 0.0048 $\pm$ 0.0004 | 0.005 $\pm$ 0.0001  |
| (2,5)  | 1.05E-04 | 1.84E-03 | 0.005 $\pm$ 0.0004  | 0.0043 $\pm$ 0.0001 |
| (2,6)  | 7.93E-04 | 3.63E-03 | 0.0068 $\pm$ 0.0005 | 0.006 $\pm$ 0.0001  |
| (2,7)  | 1.49E-03 | 6.90E-03 | 0.0096 $\pm$ 0.0005 | 0.007 $\pm$ 0.0001  |
| (2,8)  | 1.71E-03 | 7.86E-03 | 0.0096 $\pm$ 0.0005 | 0.0078 $\pm$ 0.0002 |
| (2,9)  | 1.50E-03 | 5.59E-03 | 0.0063 $\pm$ 0.0004 | 0.0053 $\pm$ 0.0001 |
| (2,10) | 1.08E-03 | 2.48E-03 | 0.0028 $\pm$ 0.0003 | 0.00219 $\pm$ 8E-05 |
| (2,11) | 6.70E-04 | 6.10E-04 | 0.0009 $\pm$ 0.0002 | 0.0012 $\pm$ 6E-05  |
| (2,12) | 3.63E-04 | 5.24E-05 | 0.0003 $\pm$ 0.0001 | 0.00034 $\pm$ 3E-05 |
| (2,13) | 1.72E-04 | 1.32E-05 | 0.0001 $\pm$ 8E-05  | 0.00037 $\pm$ 3E-05 |
| (2,14) | 7.12E-05 | 2.40E-05 | 0.00014 $\pm$ 9E-05 | 0.00074 $\pm$ 5E-05 |
| (3,0)  | 8.70E-05 | 1.50E-04 | 0.00012 $\pm$ 9E-05 | 0.00133 $\pm$ 6E-05 |
| (3,1)  | 1.12E-03 | 9.95E-04 | 0.0003 $\pm$ 0.0001 | 0.00132 $\pm$ 6E-05 |
| (3,2)  | 5.23E-03 | 3.46E-03 | 0.0023 $\pm$ 0.0003 | 0.0036 $\pm$ 0.0001 |
| (3,3)  | 1.33E-02 | 7.92E-03 | 0.0054 $\pm$ 0.0004 | 0.0062 $\pm$ 0.0001 |
| (3,4)  | 2.19E-02 | 1.29E-02 | 0.011 $\pm$ 0.0006  | 0.0117 $\pm$ 0.0002 |
| (3,5)  | 2.53E-02 | 1.53E-02 | 0.014 $\pm$ 0.0007  | 0.0135 $\pm$ 0.0002 |
| (3,6)  | 2.16E-02 | 1.35E-02 | 0.0121 $\pm$ 0.0006 | 0.0115 $\pm$ 0.0002 |
| (3,7)  | 1.39E-02 | 8.77E-03 | 0.0075 $\pm$ 0.0005 | 0.0078 $\pm$ 0.0002 |
| (3,8)  | 6.67E-03 | 4.15E-03 | 0.0039 $\pm$ 0.0004 | 0.00296 $\pm$ 9E-05 |
| (3,9)  | 2.31E-03 | 1.37E-03 | 0.0014 $\pm$ 0.0002 | 0.00103 $\pm$ 6E-05 |
| (3,10) | 5.13E-04 | 2.86E-04 | 0.0004 $\pm$ 0.0001 | 0.00025 $\pm$ 3E-05 |
| (3,11) | 4.40E-05 | 3.18E-05 | 0.0002 $\pm$ 0.0001 | 0.00021 $\pm$ 2E-05 |
| (3,12) | 2.11E-06 | 7.28E-06 | 0.0002 $\pm$ 0.0001 | 0.00071 $\pm$ 5E-05 |
| (3,13) | 1.79E-05 | 8.77E-06 | 0.00018 $\pm$ 9E-05 | 0.00091 $\pm$ 5E-05 |
| (3,14) | 1.77E-05 | 7.78E-06 | 0.0001 $\pm$ 8E-05  | 0.00084 $\pm$ 5E-05 |
| (4,0)  | 9.46E-06 | 3.58E-05 | 0                   | 0.0004 $\pm$ 3E-05  |
| (4,1)  | 8.74E-06 | 2.20E-04 | 0.0004 $\pm$ 0.0001 | 0.00055 $\pm$ 4E-05 |
| (4,2)  | 2.89E-07 | 5.52E-04 | 0.0006 $\pm$ 0.0002 | 0.00096 $\pm$ 5E-05 |
| (4,3)  | 3.10E-05 | 7.20E-04 | 0.0011 $\pm$ 0.0002 | 0.00115 $\pm$ 6E-05 |
| (4,4)  | 8.57E-05 | 5.26E-04 | 0.0012 $\pm$ 0.0002 | 0.00112 $\pm$ 6E-05 |
| (4,5)  | 9.82E-05 | 3.86E-04 | 0.0014 $\pm$ 0.0002 | 0.00095 $\pm$ 5E-05 |
| (4,6)  | 6.28E-05 | 7.62E-04 | 0.002 $\pm$ 0.0003  | 0.00118 $\pm$ 6E-05 |
| (4,7)  | 2.51E-05 | 1.35E-03 | 0.0023 $\pm$ 0.0003 | 0.0016 $\pm$ 7E-05  |
| (4,8)  | 7.91E-06 | 1.48E-03 | 0.0014 $\pm$ 0.0002 | 0.00149 $\pm$ 7E-05 |
| (4,9)  | 3.98E-06 | 1.03E-03 | 0.0011 $\pm$ 0.0002 | 0.00117 $\pm$ 6E-05 |
| (4,10) | 5.12E-06 | 4.58E-04 | 0.0004 $\pm$ 0.0001 | 0.00043 $\pm$ 4E-05 |
| (4,11) | 8.37E-06 | 1.20E-04 | 0.00015 $\pm$ 8E-05 | 0.00027 $\pm$ 3E-05 |
| (4,12) | 1.07E-05 | 1.67E-05 | 0                   | 0.0001 $\pm$ 2E-05  |
| (4,13) | 1.00E-05 | 5.98E-06 | 0                   | 7E-05 $\pm$ 1E-05   |
| (4,14) | 7.02E-06 | 5.32E-06 | 0.00015 $\pm$ 7E-05 | 0.00013 $\pm$ 2E-05 |

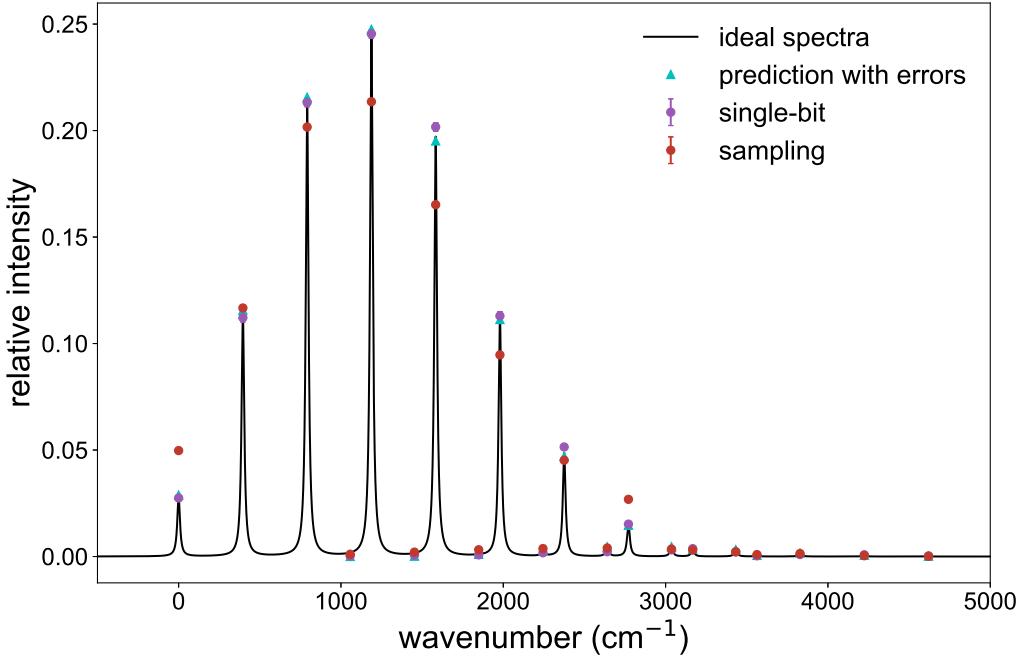


FIG. 7. Photoionization of sulfur dioxide to the cation starting in the vibrationless state  $n = 0, m = 0$ .

| $\text{SO}_2 \xrightarrow{h\nu} \text{SO}_2^+ + \text{e}^-$ starting in $(n = 0, m = 0)$ |                        |                            |                       |                      |
|--|------------------------|----------------------------|-----------------------|----------------------|
| $(n', m')$   | Classically calculated | Master equation simulation | Single-bit extraction | Sampling             |
| (0,0)  | 2.82E-02               | 2.88E-02                   | 0.0275 $\pm$ 0.0009   | 0.0497 $\pm$ 0.0003  |
| (0,1)  | 1.14E-01               | 1.15E-01                   | 0.112 $\pm$ 0.002     | 0.1167 $\pm$ 0.0005  |
| (0,2)  | 2.13E-01               | 2.15E-01                   | 0.213 $\pm$ 0.002     | 0.2017 $\pm$ 0.0006  |
| (0,3)  | 2.47E-01               | 2.47E-01                   | 0.245 $\pm$ 0.002     | 0.2136 $\pm$ 0.0006  |
| (0,4)  | 1.97E-01               | 1.95E-01                   | 0.202 $\pm$ 0.002     | 0.1652 $\pm$ 0.0006  |
| (0,5)  | 1.14E-01               | 1.11E-01                   | 0.113 $\pm$ 0.002     | 0.0947 $\pm$ 0.0005  |
| (0,6)  | 4.85E-02               | 4.72E-02                   | 0.051 $\pm$ 0.001     | 0.0452 $\pm$ 0.0003  |
| (0,7)  | 1.54E-02               | 1.48E-02                   | 0.0152 $\pm$ 0.0007   | 0.0268 $\pm$ 0.0003  |
| (0,8)  | 3.57E-03               | 3.38E-03                   | 0.0037 $\pm$ 0.0004   | 0.00302 $\pm$ 9E-05  |
| (0,9)  | 5.68E-04               | 5.28E-04                   | 0.0003 $\pm$ 0.0002   | 0.00091 $\pm$ 5E-05  |
| (1,0)  | 9.40E-06               | 2.89E-05                   | 0.0003 $\pm$ 0.0001   | 0.00108 $\pm$ 5E-05  |
| (1,1)  | 4.35E-05               | 1.53E-04                   | 0.0007 $\pm$ 0.0002   | 0.00206 $\pm$ 7E-05  |
| (1,2)  | 7.70E-04               | 9.82E-04                   | 0.0008 $\pm$ 0.0002   | 0.00318 $\pm$ 9E-05  |
| (1,3)  | 2.67E-03               | 2.84E-03                   | 0.0017 $\pm$ 0.0003   | 0.00374 $\pm$ 0.0001 |
| (1,4)  | 4.54E-03               | 4.53E-03                   | 0.0023 $\pm$ 0.0003   | 0.004 $\pm$ 0.0001   |
| (1,5)  | 4.78E-03               | 4.60E-03                   | 0.0031 $\pm$ 0.0003   | 0.0035 $\pm$ 9E-05   |
| (1,6)  | 3.44E-03               | 3.22E-03                   | 0.0022 $\pm$ 0.0003   | 0.00219 $\pm$ 7E-05  |
| (1,7)  | 1.76E-03               | 1.62E-03                   | 0.0009 $\pm$ 0.0002   | 0.0014 $\pm$ 6E-05   |
| (1,8)  | 6.53E-04               | 5.89E-04                   | 0.0008 $\pm$ 0.0002   | 0.00038 $\pm$ 3E-05  |
| (1,9)  | 1.73E-04               | 1.54E-04                   | 0.0003 $\pm$ 9E-05    | 0.00011 $\pm$ 2E-05  |

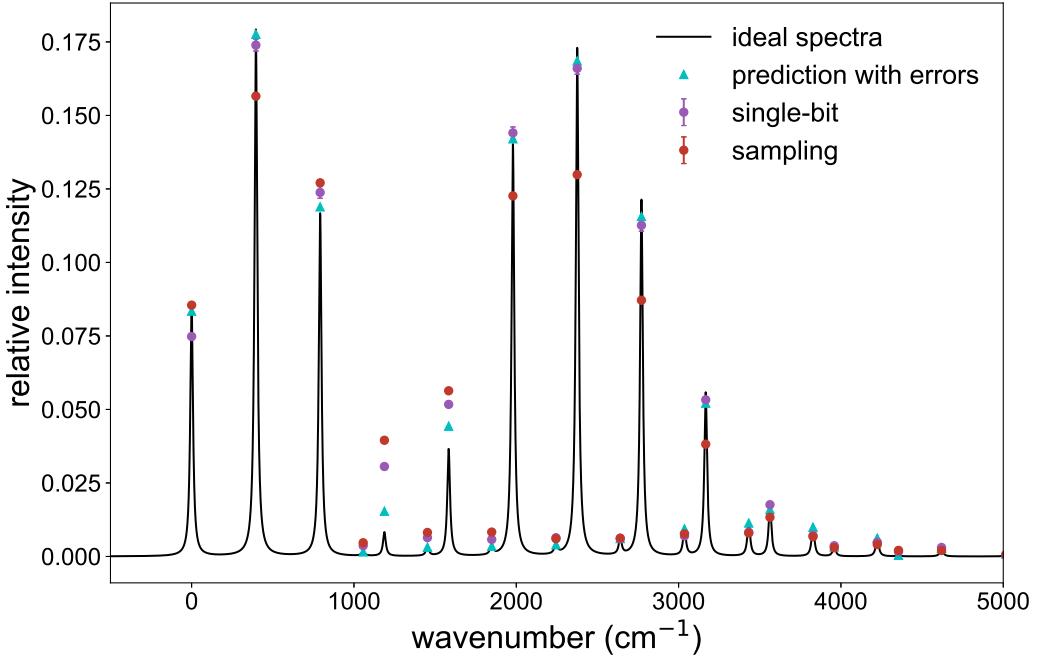


FIG. 8. Photoionization of sulfur dioxide to the cation starting with zero quanta in the symmetric-stretching mode and one quantum in the bending mode  $n = 0, m = 1$ .

| $\text{SO}_2 \xrightarrow{h\nu} \text{SO}_2^+ + e^-$ starting in $(n = 0, m = 1)$ |                        |                            |                       |                 |
|---|------------------------|----------------------------|-----------------------|-----------------|
| $(n', m')$  | Classically calculated | Master equation simulation | Single-bit extraction | Sampling        |
| (0,0)   | 8.52E-02               | 8.36E-02                   | 0.075 ± 0.001         | 0.0855 ± 0.0005 |
| (0,1)   | 1.79E-01               | 1.78E-01                   | 0.174 ± 0.002         | 0.1566 ± 0.0006 |
| (0,2)   | 1.17E-01               | 1.19E-01                   | 0.124 ± 0.002         | 0.1271 ± 0.0006 |
| (0,3)   | 8.11E-03               | 1.50E-02                   | 0.0306 ± 0.0009       | 0.0395 ± 0.0003 |
| (0,4)   | 3.64E-02               | 4.42E-02                   | 0.052 ± 0.001         | 0.0564 ± 0.0004 |
| (0,5)   | 1.40E-01               | 1.42E-01                   | 0.144 ± 0.002         | 0.1226 ± 0.0006 |
| (0,6)   | 1.73E-01               | 1.69E-01                   | 0.166 ± 0.002         | 0.1299 ± 0.0006 |
| (0,7)   | 1.21E-01               | 1.16E-01                   | 0.113 ± 0.002         | 0.0872 ± 0.0005 |
| (0,8)   | 5.56E-02               | 5.22E-02                   | 0.053 ± 0.001         | 0.0382 ± 0.0003 |
| (0,9)   | 1.74E-02               | 1.60E-02                   | 0.0176 ± 0.0007       | 0.0133 ± 0.0002 |
| (0,10)  | 3.62E-03               | 3.24E-03                   | 0.0036 ± 0.0004       | 0.00296 ± 9E-05 |
| (0,11)  | 4.51E-04               | 3.96E-04                   | 0.0016 ± 0.0003       | 0.00206 ± 8E-05 |
| (1,0)   | 1.68E-03               | 1.55E-03                   | 0.0037 ± 0.0003       | 0.0047 ± 0.0001 |
| (1,1)   | 3.27E-03               | 3.16E-03                   | 0.0064 ± 0.0005       | 0.0082 ± 0.0002 |
| (1,2)   | 3.48E-03               | 3.48E-03                   | 0.0058 ± 0.0004       | 0.0083 ± 0.0002 |
| (1,3)   | 3.89E-03               | 3.99E-03                   | 0.0064 ± 0.0004       | 0.006 ± 0.0001  |
| (1,4)   | 5.95E-03               | 6.09E-03                   | 0.006 ± 0.0004        | 0.0062 ± 0.0001 |
| (1,5)   | 9.44E-03               | 9.38E-03                   | 0.007 ± 0.0005        | 0.0077 ± 0.0001 |
| (1,6)   | 1.18E-02               | 1.14E-02                   | 0.0083 ± 0.0005       | 0.008 ± 0.0001  |
| (1,7)   | 1.07E-02               | 9.95E-03                   | 0.0072 ± 0.0005       | 0.0067 ± 0.0001 |
| (1,8)   | 6.90E-03               | 6.24E-03                   | 0.0048 ± 0.0004       | 0.0041 ± 0.0001 |
| (1,9)   | 3.18E-03               | 2.79E-03                   | 0.0031 ± 0.0003       | 0.00203 ± 8E-05 |
| (1,10)  | 1.03E-03               | 8.79E-04                   | 0.0005 ± 0.0001       | 0.00061 ± 4E-05 |
| (1,11)  | 2.26E-04               | 1.86E-04                   | 4E-05 ± 6E-05         | 0.00035 ± 3E-05 |

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