

Photodissociation of ultracold diatomic strontium molecules with quantum state control

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1 Supplementary Methods

1.1 Parametrizing angular distributions

1.1.1 Anisotropy (or beta) parameters

We can represent the angular distribution of any physical intensity (or differential cross section) with the expansion

$$I(\theta, \phi) \propto 1 + \sum_{l=1}^{\infty} \sum_{m=0}^l P_l^m(\cos \theta) \left[\beta_{lm} \cos(m\phi) + \gamma_{lm} \sin(m\phi) \right] \quad (7)$$

in terms of real-valued “anisotropy” coefficients β_{lm} and γ_{lm} , where $\gamma_{l0} \equiv 0$. If there is no dependence on ϕ the associated Legendre polynomials reduce to Legendre polynomials, $P_l^0(\cos \theta) = P_l(\cos \theta)$, and the remaining coefficients β_{l0} are conventionally denoted β_l .

If both atomic fragments of a dissociated homonuclear diatomic molecule are detected equally, then conservation of momentum requires the inversion symmetry $I(\pi - \theta, \phi + \pi) = I(\theta, \phi)$. The expansion (7) has this symmetry if the coefficients with odd l are zero.

If the only nonzero coefficients are those with even $l + m$, then the expansion (7) will additionally be symmetric under reflection across the equator, $I(\pi - \theta, \phi) = I(\theta, \phi)$. For homonuclear diatomic molecules, this symmetry requires the coefficients with odd m to be zero. Intensities without this symmetry display a “skewness,” such as the asymmetry in Fig. 1(b) and several other figure insets that are likely due to imperfect laser polarization or incomplete saturation correction during absorption imaging.

1.1.2 Partial scattering amplitudes

For our experiments, the measured intensity (or differential cross section) can be written as a sum of the squared absolute values of complex scattering amplitudes for separate electronic channels,

$$I(\theta, \phi) = \sum_{\Omega} \left| f^{\{\Omega\}}(\theta, \phi) \right|^2, \quad (8)$$

here indexed by the quantum number Ω for the internuclear projection of angular momentum.

To calculate the intensity, we compute the partial scattering amplitudes $f_{JM}^{\{\Omega\}}$ of a partial-wave expansion of the scattering amplitude in terms of angular basis functions,

$$f^{\{\Omega\}}(\theta, \phi) = \sum_{JM} f_{JM}^{\{\Omega\}} \psi_{JM}^{\{\Omega\}}(\theta, \phi), \quad (9)$$

as described in Sec. 1.2.1. In terms of Wigner D-functions, we chose the angular basis functions to be

$$\psi_{JM}^{\{\Omega\}}(\theta, \phi) = \sqrt{\frac{2J+1}{4\pi}} D_{M\Omega}^{J*}(\phi, \theta, 0), \quad (10)$$

so that for $\Omega = 0$ they are equivalent to spherical harmonics, $\psi_{JM}^{\{0\}}(\theta, \phi) = Y_{JM}(\theta, \phi)$.

The expansion (7) is equivalent to Eq. (8) if we write the anisotropy coefficients as the real and imaginary parts of a weighted sum over products of pairs of partial scattering amplitudes,

$$\beta_{lm} + i\gamma_{lm} = \sum_{\Omega, J, J', M} W_{lm}(\Omega, J, J', M) f_{JM}^{\{\Omega\}*} f_{J', M-m}^{\{\Omega\}} / \sum_{JM\Omega} \left| f_{JM}^{\{\Omega\}} \right|^2. \quad (11)$$

The real-valued weights may be written in terms of Wigner 3j symbols as

$$W_{lm}(\Omega, J, J', M) = (-1)^{M-\Omega} \frac{2[l]}{1 + \delta_{m0}} \sqrt{\frac{[J][J'](l-m)!}{(l+m)!}} \begin{pmatrix} J' & l & J \\ m-M & -m & M \end{pmatrix} \begin{pmatrix} J' & l & J \\ -\Omega & 0 & \Omega \end{pmatrix}, \quad (12)$$

where the shorthand $[J] \equiv 2J + 1$ and δ_{ij} is a Kronecker delta. As an aside, note that the quantities $\rho_{M, M-m}^{\{\Omega J J'\}} = f_{JM}^{\{\Omega\}*} f_{J', M-m}^{\{\Omega\}} / \sum_{JM\Omega} \left| f_{JM}^{\{\Omega\}} \right|^2$ in Eq. (11) have properties similar to density matrix elements.

From these weights W_{lm} and the properties of 3j symbols, the maximum value of l contributing in the expansion (7) is limited to twice the largest value of J for which there is a nonzero $f_{JM}^{\{\Omega\}}$. The maximum value of m is limited by the furthest off-diagonal magnetic coherence, that is, the nonzero quantity $f_{JM}^{\{\Omega\}*} f_{J', M-m}^{\{\Omega\}}$ with largest $m = M - M'$.

1.2 Calculation of angular distributions

1.2.1 Theoretical description of photodissociation

The theory of photodissociation employed here follows the seminal work of Ref. [1]. The fragmentation process is characterized by the differential cross section that is defined by Fermi's golden rule with the

electric-dipole (E1), magnetic-dipole (M1), or electric-quadrupole (E2) transition operators. Since we work with a coupled manifold of electronic states for both the ungerade bound states and ungerade continuum, we do not assume the Born-Oppenheimer approximation in contrast to Zare [1]. In this case the theory of photodissociation for diatomic molecules is very similar to the non-degenerate atom-diatom case treated in detail by Balint-Kurti and Shapiro [2, 3].

In the absence of external fields, the wave function of the initial (bound) state depends on the set of the electronic coordinates $\{\mathbf{r}\}$ and on the vector $\mathbf{R} = (R, \Theta, \Phi)$ describing the relative motion of the nuclei, and is given by

$$\Psi_{J_i M_i}^{p_i}(\{\mathbf{r}\}, \mathbf{R}) = \sum_{\Omega_i = -J_i}^{J_i} \sqrt{\frac{2J_i + 1}{16\pi^2(1 + \delta_{\Omega_i 0})}} \left(D_{M_i \Omega_i}^{J_i \star}(\Phi, \Theta, 0) \psi_{J_i \Omega_i}^{p_i}(\{\mathbf{r}\}, R) + \sigma_i D_{M_i, -\Omega_i}^{J_i \star}(\Phi, \Theta, 0) \psi_{J_i, -\Omega_i}^{p_i}(\{\mathbf{r}\}, R) \right), \quad (13)$$

where $\sigma_i = p_i(-1)^{J_i}$ is the spectroscopic parity and J_i , M_i , and p_i are the quantum numbers of the total angular momentum, its projection on the space-fixed Z axis (previously denoted z), and the parity with respect to space-fixed inversion. Note that in the above expression the quantum number π_i related to the action of the reflection in the body-fixed yz plane on the electronic coordinates does not appear. In our case it is equal to zero, and the parity of the $\Omega_i = 0$ electronic states is always “+”.

In Hund’s case (c) the internal wave function $\psi_{J_i \Omega_i}^{p_i}(\{\mathbf{r}\}, R)$ can be represented by the Born-Huang expansion [4, 5]

$$\psi_{J_i \Omega_i}^{p_i}(\{\mathbf{r}\}, R) = \sum_n \phi_{n, \Omega_i}(\{\mathbf{r}\}; R) \chi_{n, J_i \Omega_i}^{p_i}(R). \quad (14)$$

Here, the $\phi_{n, \Omega_i}(\{\mathbf{r}\}; R)$ are electronic wave functions, that is, the solutions of the electronic Schrödinger equation including spin-orbit coupling, which depend parametrically on the interatomic distance R . The $\chi_{n, J_i \Omega_i}^{p_i}(R)$ are rovibrational wave functions. Finally, the index n labels all relativistic dissociation channels. Note that for homonuclear diatomic molecules the electronic wave function has an additional gerade/ungerade (g/u) symmetry resulting from the $D_{\infty h}$ point group of the molecule. For simplicity we do not indicate the g/u symmetry in the notation $\phi_{n, \Omega_i}(\{\mathbf{r}\}; R)$. The rovibrational wave functions are solution of a system of coupled differential equations. See, for instance, Ref. [6] for the equations corresponding to the ungerade excited manifold of the electronic states.

The wave function $\Psi_{\mathbf{k}}^{p_f}(\{\mathbf{r}\}, \mathbf{R})$ of the final continuum state corresponding to the wave vector $\mathbf{k} = (k, \theta, \phi)$ can be represented by the following expansion reflecting different partial waves J of the fragmented atoms,

$$\Psi_{\mathbf{k}}^{p_f}(\{\mathbf{r}\}, \mathbf{R}) = \sum_{JM} \sum_{\Omega = -J}^J \sum_{\Omega' = -J}^J \frac{(2J+1)}{4\pi\sqrt{2\pi(1 + \delta_{\Omega' 0})}} C_{m_j \Omega}^{jp} D_{M \Omega}^J(\phi, \theta, 0) D_{m_j \Omega'}^{J \star}(\phi, \theta, 0) \times \left(D_{M \Omega'}^{J \star}(\Phi, \Theta, 0) \psi_{\Omega' \Omega}^{J p_f}(\{\mathbf{r}\}, R) + \sigma_i D_{M, -\Omega'}^{J \star}(\Phi, \Theta, 0) \psi_{-\Omega', \Omega}^{J p_f}(\{\mathbf{r}\}, R) \right), \quad (15)$$

where j denotes the total angular momentum of the photofragmented atoms, m_j is its projection in the space-fixed Z axis, and p is the product of atomic parities. The numerical coefficients $C_{m_j \Omega}^{jp}$ depend on the states of the photofragmented atoms and can be found in Ref. [7]. The internal wave function is given by the following multichannel generalization of the Born-Huang expansion,

$$\psi_{\Omega' \Omega}^{J p_f}(\{\mathbf{r}\}, R) = \sum_n \phi_{n, \Omega'}(\{\mathbf{r}\}; R) \chi_{n, \Omega' \Omega}^{J p_f}(R), \quad (16)$$

where $\chi_{n, \Omega' \Omega}^{J p_f}(R)$ is a radial channel function that satisfies the boundary condition

$$\chi_{n, \Omega' \Omega}^{J p_f}(R) \approx R \sqrt{\frac{2\mu k}{\pi}} \left(\delta_{\Omega' \Omega} j_J(kR) + S_{n, \Omega' \Omega}^{J p_f} n_J(kR) \right) \quad (\text{for } R \rightarrow \infty) \quad (17)$$

in terms of the scattering matrix $S_{n\Omega'\Omega}^{Jp_f}$. Here, μ is the reduced mass and j_J, n_J are spherical Bessel functions.

In this work we considered four different photofragmentation processes: (i) the E1 process starting from the ungerade manifold of the electronic states that correspond to the $^1S + ^3P_1$ dissociation limit and ending at the ground electronic continuum, (ii) the M1 and (iii) the E2 processes starting from the gerade manifold corresponding to the same dissociation limit and ending at the ground electronic continuum, and finally (iv) the E1 process starting from ground state molecules and ending at the ungerade manifold corresponding to the $^1S + ^3P_1$ dissociation limit.

The first three processes begin with manifolds that are described by two coupled electronic states: 0_u^+ and 1_u for the E1 process and 0_g^+ and 1_g for the M1 and E2 processes. The corresponding wave functions for the initial states are given by Eqs. (13) and (14) with the summation over Ω_i limited to $-1, 0, 1$, and with n fixed to $^1S + ^3P_1$. The wave function for the final continuum state, however, corresponds to the Born-Oppenheimer approximation and is given by Eqs. (15) and (14) with only the $\Omega = 0$ term and with n fixed to $^1S + ^1S$. Note that in the single-channel approximation $\Omega = \Omega'$, so for simplicity we denote the rovibrational wavefunction by $\chi_{n\Omega}^{Jp_f}(R)$. The electronic transition operator for the E1 process was assumed to be constant and proportional to the atomic value, while the operators for the M1 and E2 transitions followed the asymptotic form of Refs. [8, 9]. Otherwise, the remaining derivation of the expression for the differential cross section follows Ref. [1] and is not reproduced here, although the multichannel character of the initial state wave functions complicates the angular momentum algebra.

Now we discuss the boundary condition for the final continuum rovibrational wave function $\chi_{n\Omega}^{Jp_f}(R)$. The single-channel approximation is valid for the 0_g^+ ground electronic continuum. In this case, at large internuclear distances R the partial wave expansion (15) becomes [10]

$$\chi_{n\Omega}^{Jp_f}(R) \approx i^J e^{i\delta_J} \sqrt{\frac{2\mu}{\pi\hbar^2 k}} \sin(kR + \delta_J + J\pi) \quad (\text{for } R \rightarrow \infty), \quad (18)$$

where δ_J is the phase shift for a given partial wave J . However, in practice it is more convenient to work with real functions than with complex functions that satisfy this boundary condition. Therefore, we chose to instead use the real-valued large- R boundary condition [10]

$$\chi_{n\Omega}^{Jp_f}(R) \approx \sqrt{\frac{2\mu}{\pi\hbar^2 k}} \sin(kR + \delta_J + J\pi) \quad (\text{for } R \rightarrow \infty), \quad (19)$$

and to include the phase factor $i^J e^{i\delta_J}$ in the partial scattering amplitudes.

Therefore, for the first three processes the differential cross section is given by Eq. (8) with $\Omega = 0$. The partial scattering amplitudes in the expansion (9) for this scattering amplitude are then given by

$$f_{JM}^{\{\Omega\}} = \sum_{\Omega_i=-J_i}^{J_i} 2i^J e^{i\delta_J} \sqrt{\frac{4\pi(2J_i+1)}{(1+\delta_{\Omega_i 0})(1+\delta_{\Omega 0})}} \langle \chi_{n_k J \Omega}^{p_f} | T_{\Omega-\Omega_i}^L | \chi_{n J_i \Omega_i}^{p_i} \rangle \begin{pmatrix} J & L & J_i \\ -\Omega & \Omega - \Omega_i & \Omega_i \end{pmatrix} \begin{pmatrix} J & L & J_i \\ -M & M - M_i & M_i \end{pmatrix}, \quad (20)$$

where $T_{\Omega-\Omega_i}^L$ is the electronic transition operator of rank $L = 1$ for E1 or M1 transitions and $L = 2$ for E2 transitions for the experimental conditions described in Sec. 1.2.2. The anisotropy parameters in the expansion (7) then follow from using these partial amplitudes with Eqs. (11) and (12).

The M1 and E2 processes were not observed separately because of selection rules. In this case, both processes must be included and the observed cross section may be written as the sum

$$I(\theta, \phi) = \left| f_{JM, M1}^{\{0\}} + f_{JM, E2}^{\{0\}} \right|^2, \quad (21)$$

of separate scattering amplitudes (9) using Eq. (20). This expression explicitly allows for interference between the M1 and E2 processes. Note that this interference may affect the angular distribution even if it does not affect the strength of the transition, which is proportional to the integral of the differential cross section over all angles. The strength curves in Fig. 3(b) were calculated this way, and were included in the figure

by scaling their amplitudes together to better match experiment, while keeping their relative amplitudes fixed by theory. Note that the experimental procedure used to produce the data in Fig. 3(b) is an imperfect measure of the photodissociation lineshape, so the comparison with theory is qualitative.

Finally, for the fourth process of photofragmentation beginning with ground state molecules and ending at the $^1S + ^3P_1$ continuum, the wave function of the initial (bound) state satisfies the Born-Oppenheimer approximation. Therefore we set $\Omega_i = 0$ in Eq. (13) and limit the Born-Huang expansion (14) to a single product. However, the partial wave expansion (15) for the final continuum must explicitly account for the Coriolis coupling between the 0_u^+ and 1_u electronic states, and for the angular momentum $j = 1$ and total parity $p = -1$ of the atomic fragments. For this multichannel continuum case, we imposed the complex boundary conditions of Eq. (17). The remaining derivation of the expression for the differential cross section follows Refs. [11, 2, 3, 12]. The differential cross section $I(\theta, \phi)$ then follows from Eqs. (8) and (9) using the partial scattering amplitudes

$$f_{JM}^{\{\Omega\}} = \sum_{\Omega_i = -J_i}^{J_i} \sum_{\Omega' = -J_i}^{J_i} 2 \sqrt{\frac{4\pi(2J_i + 1)}{(1 + \delta_{\Omega_i 0})(1 + \delta_{\Omega' 0})}} \langle \chi_{n\Omega'\Omega}^{Jp_f} | T_{\Omega' - \Omega_i}^L | \chi_{nJ_i\Omega_i}^{p_i} \rangle \begin{pmatrix} J & j & J_i \\ -\Omega' & \Omega' - \Omega_i & \Omega_i \end{pmatrix} \begin{pmatrix} J & j & J_i \\ -M & M - M_i & M_i \end{pmatrix}. \quad (22)$$

1.2.2 Field components for the experimental conditions

For E1 transitions in our experiment, the lab-frame spherical tensor components of the field driving the transition are

$$T_0^1(\mathbf{E}) = E_z \quad (23)$$

$$T_{\pm 1}^1(\mathbf{E}) = -\frac{i}{\sqrt{2}} E_y, \quad (24)$$

using the notation of Ref. [13]. For linear polarization parallel to the z axis, which is labeled “ $p = 0$ ” in Fig. 4, $E_y = 0$. For linear polarization along the y axis, which is labeled “ $|p| = 1$ ” in Fig. 4, $E_z = 0$. Likewise, for M1 transitions these components are

$$T_0^1(\mathbf{B}) = B_z \quad (25)$$

$$T_{\pm 1}^1(\mathbf{B}) = -\frac{i}{\sqrt{2}} B_y. \quad (26)$$

Note that in Fig. 3 “ $p = 0$ ” now corresponds to linear polarization along the y axis, such that $B_y = 0$, and “ $|p| = 1$ ” to linear polarization parallel to the z axis, such that $B_z = 0$. For electric E2 transitions, these components are

$$T_0^2(\nabla\mathbf{E}) = 0 \quad (27)$$

$$T_{\pm 1}^2(\nabla\mathbf{E}) = \mp \frac{1}{2} (ikE_z) \quad (28)$$

$$T_{\pm 2}^2(\nabla\mathbf{E}) = \pm \frac{i}{2} (ikE_y) \quad (29)$$

for traveling-wave light propagating along the x axis with wavenumber k [14].

For θ and ϕ defined as in Fig. 1, these experimental conditions produce angular distributions that can be described only with β_{lm} coefficients in the expansion (7), leading to the simplified form of Eq. (2).

1.2.3 Energy independent angular distributions

For $^{88}\text{Sr}_2$ photodissociation, there are a few cases where the angular distributions are independent of the continuum energy. For E1 photodissociation to the ground continuum (where symmetry restricts J to even values) this occurs if J_i is even because $\Delta J = 0, \pm 1$. This also occurs with odd J_i for either $|M_i| = J_i$

with $p = 0$ or $J_i = 1$ and $M_i = 0$ with $|p| = 1$, because selection rules only allow a single value of J in the continuum. For M1/E2 photodissociation to the ground continuum from $J_i = 1$, $M_i = 0$, this occurs when the dissociation laser is linearly polarized along the z axis, because of ΔM selection rules. In these cases, the energy-dependent radial integrals in Eq. (20) are common to all partial scattering amplitudes, so the angular distributions are independent of the continuum energy. They are also relatively simple to calculate, because they reduce to evaluating geometrical factors.

1.2.4 Single- J approximation for angular distributions

In a similar fashion, Eqs. (20) and (22) can be used to explore what range of angular distributions may occur in an experiment by making simplifying assumptions about the radial integrals. For example, for E1 photodissociation to the ground continuum we could approximate the radial matrix elements in Eq. (20) to be nonzero only for a single J but multiple M , such that

$$f_{JM}^{\{\Omega\}} \propto T_{M-M_i}^L(\mathbf{E}) \begin{pmatrix} J & L & J_i \\ -M & M - M_i & M_i \end{pmatrix}, \quad (30)$$

where $T_{M-M_i}^L(\mathbf{E})$ are the lab-frame spherical tensor components of the dissociating field, as described in Sec. 1.2.2 for our experimental conditions. For a selected initial state, the calculation of the angular distribution simplifies to evaluating geometrical factors that depend only on the allowed quantum numbers.

In addition to energy-independent cases, this approximation works well for the energy-dependent data in Fig. 4 with odd J_i , because the continuum energies were chosen so that a single J was responsible for most of each angular distribution: $J = 4$ for $J_i = 3, 4$ and $J = 2$ for $J_i = 1, 2$. This approximation also explains some interesting properties that we observe. For example, the $(M_i = 0, |p| = 1)$ cases are identical except for a 90° rotation in ϕ , which corresponds to alternating the sign of the β_{l2} parameters with J_i . For $M_i \neq 0$, a qualitatively similar rotation often occurs. Finally, for $|p| = 1$ the cases of $|M_i| = J$ have the same coefficients as those of $p = 0$ for $|M_i| = J - 1$, since they produce the same single sublevels M in the continuum.

1.3 Parameters for theoretical images in figures

Supplementary Tables 1, 2, and 3 list the parameters used to generate the theoretical images shown in Fig. 4 and Extended Data Figs. 1–3.

2 Supplementary Tables

Supplementary Table 1: Parameters β_{lm} of the theoretical images in Extended Data Fig. 1 for the $p = 0$ case of M1/E2 photodissociation of $1_g(v_i = -1, J_i = 1, M_i = 0)$.

ε/h (MHz)	β_{20}	β_{22}	β_{40}	β_{42}	β_{44}
4	0.4178	−0.3599	0.1095	−0.01290	0.002280
8	0.2475	−0.3563	0.3077	−0.03626	0.006409
12	−0.06407	−0.2117	0.4362	−0.05140	0.009087

Supplementary Table 2: Parameters β_{lm} of the theoretical images in Fig. 4 and Extended Data Fig. 2 for E1 photodissociation of $1_u(v_i = -1, J_i, M_i)$. Values for odd J_i that depend on the continuum energy ε (given below in MHz) are rounded to four significant figures. However, the energies for odd J_i were chosen so that Eq. (30) with $J = J_i + 1$ is a good approximation. Fig. 4 uses the values from this approximation, which are given on a second line as fractions. Extended Data Fig. 2 uses the unapproximated values. For even J_i , the values are independent of the continuum energy. Where the transition is forbidden by symmetry ($p_i = M_i = 0$, even J_i), the experimental pattern is enabled by an applied magnetic field that admixes excited states [15] such that the parameters correspond to the approximation (30) with the substitution $J_i \rightarrow J_i \pm 1$. Omitted values are zero because of symmetry.

J_i	$ M_i $	ε/h	β_{20}	β_{22}	β_{40}	β_{42}	β_{60}	β_{62}	β_{80}	β_{82}
$ p = 1$										
4	0	76	85/77	25/77	729/1001	81/1001	-1/11	1/22	-392/143	7/143
4	1	74	1360/1463	450/1463	6561/19019	81/1729	2/209	0	6664/2717	-105/2717
4	2	71	65/154	45/176	-243/728	-81/2288	5/8	-9/176	-245/143	21/1144
4	3	68	-40/121	20/121	-243/1573	-162/1573	-170/121	4/121	1400/1573	-7/1573
4	4	—	-5/11	0	-243/143	0	17/11	0	-56/143	0
3	0	71	0.3097	-0.3909	0.3584	-0.05354	0.8934	-0.009862	-2.561	-0.04574
			85/77	-25/77	729/1001	-81/1001	-1/11	-1/22	-392/143	-7/143
3	1	71	0.1445	-0.2850	-0.1389	-0.01507	-1.653	-0.007708	1.816	0.03244
			400/539	-150/539	-1539/7007	-27/637	-10/11	0	280/143	5/143
3	2	72	-0.4966	-0.1545	-1.515	0.02140	1.633	0.03091	-0.6213	-0.01110
			-20/77	-15/88	-5589/4004	27/1144	59/44	3/88	-98/143	-7/572
3	3	72	-1.835	-0.05559	1.208	0.03464	-0.4569	-0.01112	0.08381	0.001497
			-3880/2233	-20/319	30861/29029	162/4147	-134/319	-4/319	392/4147	7/4147
2	0	52	5/7	5/14	-12/7	1/7				
2	1	48	2/7	2/7	12/7	-3/35				
2	2	44	5/7	0	-12/7	0				
1	0	32	5/7	-5/14	-12/7	-1/7				
1	1	31	-0.3445	-0.1311	0.1921	0.01601				
			-50/49	-10/49	36/49	3/49				
$p = 0$										
4	0	77	100/77		1458/1001		20/11		490/143	
4	1	78	85/77		729/1001		-1/11		-392/143	
4	2	71	40/77		-81/91		-2		196/143	
4	3	68	-5/11		-243/143		17/11		-56/143	
4	4	—	-20/11		162/143		-4/11		7/143	
2	0	56	10/7		18/7					
2	1	55	5/7		-12/7					
2	2	44	-10/7		3/7					

Supplementary Table 3: Parameters β_{lm} of the theoretical images in Extended Data Fig. 2 and 3 for E1 photodissociation of $0_u^+(v_i, J_i, M_i)$, with $v_i = -3$ for $J_i = 3$ and $v_i = -4$ for $J_i = 1$. Only odd J_i are allowed by symmetry. The values that depend on the continuum energy ε (given below in MHz) are rounded to four significant figures. However, as in Supplementary Table 2, the energies were chosen so that Eq. (30) with $J = J_i + 1$ is a good approximation. Extended Data Fig. 3 uses these approximate values, which are given on a second line as fractions below. Extended Data Fig. 2 uses the unapproximated values. Omitted values are zero because of symmetry.

J_i	$ M_i $	ε/h	β_{20}	β_{22}	β_{40}	β_{42}	β_{60}	β_{62}	β_{80}	β_{82}
$ p = 1$										
3	0	72	0.5258	-0.3728	0.4946	-0.05980	0.6317	-0.01999	-2.652	-0.04736
			85/77	-25/77	729/1001	-81/1001	-1/11	-1/22	-392/143	-7/143
3	1	71	0.3251	-0.2842	-0.2059	-0.02529	-1.437	-0.005318	1.893	0.03381
			400/539	-150/539	-1539/7007	-27/637	-10/11	0	280/143	5/143
3	2	70	-0.4253	-0.1640	-1.463	0.02271	1.548	0.03281	-0.6596	-0.01178
			-20/77	-15/88	-5589/4004	27/1144	59/44	3/88	-98/143	-7/572
3	3	69	-1.801	-0.06003	1.161	0.03740	-0.4509	-0.01201	0.09050	0.001616
			-3880/2233	-20/319	30861/29029	162/4147	-134/319	-4/319	392/4147	7/4147
1	0	33		5/7	-5/14	-12/7	-1/7			
1	1	32	-0.4751	-0.03716	0.3941	0.03284				
			-50/49	-10/49	36/49	3/49				
$p = 0$										
3	0	72	2.131		2.272		3.022		3.223	
			100/77		1458/1001		20/11		490/143	
3	1	73	1.861		0.7914		-1.079		-2.573	
			85/77		729/1001		-1/11		-392/143	
3	2	74	0.9400		-1.677		-1.561		1.298	
			40/77		-81/91		-2		196/143	
3	3	75	-5/11		-243/143		17/11		-56/143	
1	0	33	0.4265		1.035					
			10/7		18/7					
1	1	32		5/7	-12/7					

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