Images of photodissociation angular distributions using single-J approximation

Bart H. McGuyer

bartmcguyer.updog.co/notes/note-6-SingleJModelPD.pdf

(Dated: 19 April 2019.)

TL;DR: Simulated images using a trick to calculate the patterns that expanding clouds of atoms can form after light pulses explode certain simple molecules.

Ref. 1 studied the photodissociation of a particular molecule made of two identical atoms, $^{88}Sr_2$. There was enough experimental control that we were able to observe angular distributions formed by outgoing Sr atoms with properties that had been sought since the 1960's. In addition to presenting detailed theoretical calculations, that work also provided a simple approximation to quickly estimate many of the observed distributions.

This note presents images to highlight that "single-J" approximation presented in Section 1.2.4 of the Supplementary Information to Ref. 1. (Both the paper and its supplement are available online.[2]) Table I provides these images. What follows is a description of the single-J approximation that is adapted from Ref. 1. The equation numbers have been kept the same. Table II gives the parameters for the images in Table I produced by this approximation.

I. MODEL

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

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

in terms of real-valued "anisotropy" coefficients β_{lm} and γ_{lm}, where $\gamma_{l0} \equiv 0$.

The anisotropy coefficients can be calculated from the real and imaginary parts of a weighted sum over products of pairs of partial scattering amplitudes $f_{JM}^{\{\Omega\}}$,

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

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

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

where the shorthand $[J] \equiv 2J+1$ and δ_{ij} is a Kronecker delta.

A. Single J approximation for angular distributions

For electric-dipole (E1) photodissociation we could approximate the partial scattering amplitudes to be nonzero only for a single J but multiple M, such that

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

where $T^L_{M-M_i}(\mathbf{E})$ are the lab-frame spherical tensor components of the dissociating field. For a selected initial state, the calculation of the angular distribution then simplifies to evaluating geometrical factors that depend only on the allowed quantum numbers. This approximation works well for the energy-dependent data in Fig. 4 of Ref. 1 with odd J_i, where 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 $J_i = 1, 2$.

For the experimental conditions in Ref. 1, the field components were

$$T^1_0(\mathbf{E}) = E_z \tag{23}$$

$$T^1_{\pm1}(\mathbf{E}) = \frac{1}{\sqrt{2}}(\mp E_x - iE_y), \tag{24}$$

using the notation of Ref. 3. Equation (24) has been updated to explicitly include a third Cartesian component, E_x, that was zero for all experimental conditions. For vertical linear polarization denoted "$p = 0$", only $E_z \neq 0$. For horizontal linear polarization denoted "$|p| = 1$," only $E_y \neq 0$.

This approximation explains some interesting properties observed in Ref. 1. 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. 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. Finally, note that an azimuthal angular dependence only occurs if two or more sublevels of the single channel are produced, as expected.

Quantitatively, note that the β_{l0} coefficients are the same for $(|M_i| = 1, p = 0)$ and $(M_i = 0, |p| = 1)$, as expected. Also, the set of β_{l2} parameters are all identical for the same $|M_i|$, $|p|$, and J_c, up to a multiplicative factor. For J_i that differ by 2, they are identical.

REFERENCES

[1]M. McDonald,B. H. McGuyer, F. Apfelbeck, C.-H. Lee, I. Majewska, R. Moszynski, and T. Zelevinsky, "Photodissociation of ultracold diatomic strontium molecules with quantum state control," Nat. Phys. **535**, 122 (2016).

[2]`http://bartmcguyer.updog.co/publications.html`

[3]J. R. Brown and A. Carrington, *Rotational Spectroscopty of Diatomic Molecules*, Cambridge (2003).

TABLE I. Simulated axial images using the parameters in Table II. The coloring follows Ref. 1.

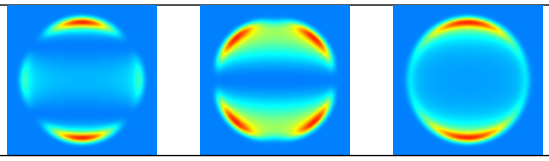
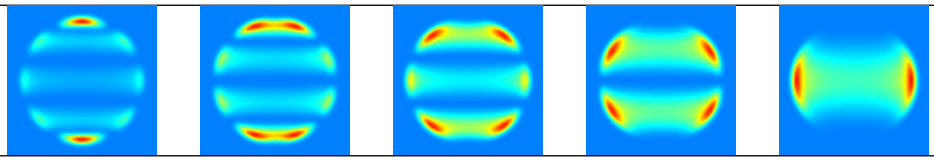
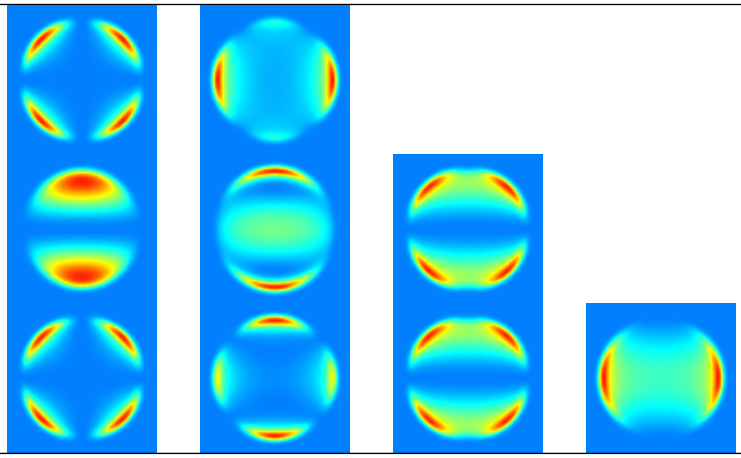
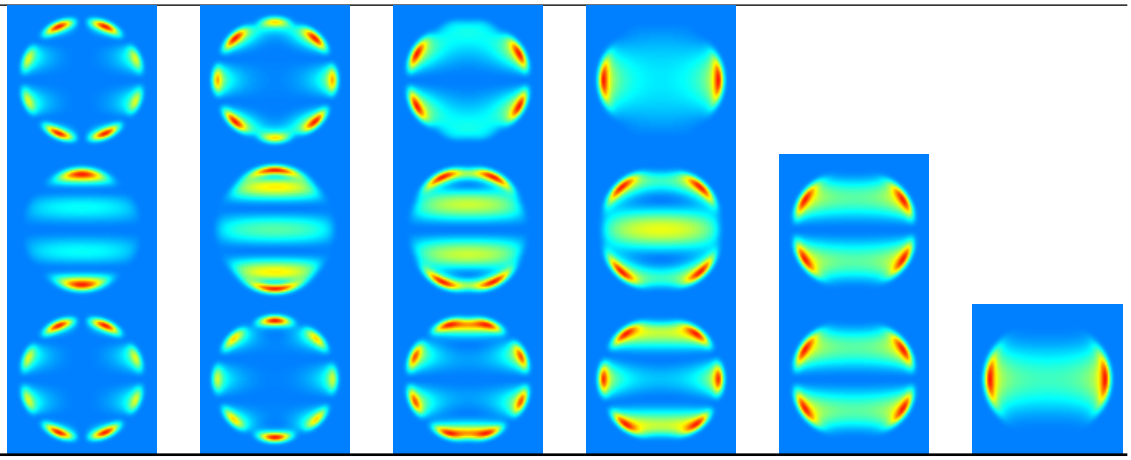
J_i	$\lvert M_i\rvert$ 0	1	2	3	4	5
$X(J_c = 2)$ channel for vertical linear polarization ($p = 0$)						
1, 2, 3						
$X(J_c = 4)$ channel for vertical linear polarization ($p = 0$)						
3, 4, 5						
$X(J_c = 2)$ channel for horizontal linear polarization ($\lvert p\rvert = 1$)						
1						
2						
3						
$X(J_c = 4)$ channel for horizontal linear polarization ($\lvert p\rvert = 1$)						
3						
4						
5						

TABLE II. Single-channel parameters calculated for photodissociation of an initial molecule with (J_i, M_i) at the ground-state $X(J_c)$ continuum. For the polarization cases of interest, the sign of M_i doesn't seem to matter. The accidentally forbidden cases when $J_i = J_c$ and $M_i = 0$ are omitted, because their experimental patterns come from admixing with $J_i \neq J_c$.

$\lvert M_i\rvert$	J_i	β_{20}	β_{40}	β_{60}	β_{80}	β_{22}	β_{42}	β_{62}	β_{82}
$X(J_c = 2)$ channel for vertical linear polarization ($p = 0$)									
0	1, 3	10/7	18/7						
1	1, 2, 3	5/7	−12/7						
2	2, 3	−10/7	3/7						
$X(J_c = 4)$ channel for vertical linear polarization ($p = 0$)									
0	3, 5	100/77	1458/1001	20/11	490/143				
1	3, 4, 5	85/77	729/1001	−1/11	−392/143				
2	3, 4, 5	40/77	−81/91	−2	196/143				
3	3, 4, 5	−5/11	−243/143	17/11	−56/143				
4	4, 5	−20/11	162/143	−4/11	7/143				
$X(J_c = 2)$ channel for horizontal linear polarization ($\lvert p\rvert = 1$)									
0	1, 3	5/7	−12/7	0	0	−5/14	−1/7		
0	2	5/7	−12/7	0	0	5/14	1/7		
1	1	−50/49	36/49	0	0	−10/49	3/49		
1	2	2/7	12/7	0	0	2/7	−3/35		
1	3	50/49	111/49	0	0	−10/49	3/49		
2	2, 3	5/7	−12/7						
3	3	−10/7	3/7						
$X(J_c = 4)$ channel for horizontal linear polarization ($\lvert p\rvert = 1$)									
0	3, 5	85/77	729/1001	−1/11	−392/143	−25/77	−81/1001	−1/22	−7/143
0	4	85/77	729/1001	−1/11	−392/143	25/77	81/1001	1/22	7/143
1	3	400/539	−1539/7007	−10/11	280/143	−150/539	−27/637	0	5/143
1	4	1360/1463	6561/19019	2/209	6664/2717	450/1463	81/1729	0	−105/2717
1	5	580/539	5508/7007	8/11	406/143	−150/539	−27/637	0	5/143
2	3	−20/77	−5589/4004	59/44	−98/143	−15/88	27/1144	3/88	−7/572
2	4	65/154	−243/728	5/8	−245/143	45/176	−81/2288	−9/176	21/1144
2	5	10/11	243/572	5/44	−350/143	−15/88	27/1144	3/88	−7/572
3	3	−3880/2233	30861/29029	−134/319	392/4147	−20/319	162/4147	−4/319	7/4147
3	4	−40/121	−243/1573	−170/121	1400/1573	20/121	−162/1573	4/121	−7/1573
3	5	140/319	−3402/4147	−620/319	5495/4147	−20/319	162/4147	−4/319	7/4147
4	4, 5	−5/11	−243/143	17/11	−56/143				
5	5	−20/11	162/143	−4/11	7/143				