APPENDIX B

EFFECT OF THE ELECTRIC QUADRUPOLE AMPLITUDE

This appendix gives a detailed treatment of the effects of the electric dipole operator ($E2$) discussed by Bouchiat and Gúena in Ref. [27]. The angular momentum text by Zare [79] is referred to extensively in the discussion, and the notation "Zare (2.45)" means "equation 45 in Chapter 2 from Ref. [79]."

In Ref. [27] Bouchiat and Guéna introduce the phenomenological transition operator

$$\begin{aligned} T(nS{-}n'S) &= -\alpha\vec{E}\cdot\vec{\epsilon} - 2i\beta\vec{S}\times\vec{E}\cdot\vec{\epsilon} + a_1\vec{S}\cdot\vec{\epsilon}\times\vec{k} \\ &+ ia_2(\vec{S}\times\vec{I})\cdot(\vec{\epsilon}\times\vec{k}) + ia_3[(\vec{S}\cdot\vec{\epsilon})(\vec{I}\cdot\vec{k}) + (\vec{S}\cdot\vec{k})(\vec{I}\cdot\vec{\epsilon})], \end{aligned} \tag{B.1}$$

where $a_1 = -2M$, $a_2 = -M_{\rm hf}/2$, and $a_3 = -E2/2$. The first two terms are the Stark-induced amplitudes from Eq. (2.13), the terms proportional to a_1 and a_2 are the magnetic dipole amplitudes from Eq. (2.21), and the term proportional to a_3 is a new amplitude—the electric quadrupole amplitude—caused by the off-diagonal hyperfine mixing of $|nD\rangle$ states into the $|nP\rangle$ states.

In Ref [27] Bouchiat and Guéna re-analyze several different experiments that determine the value of $M_{\rm hf}/M$ and show that the contribution from the $E2$ amplitude is non-negligible in some cases. Specifically, they examine the result of Ref. [28]. In the geometry of that experiment the relevant transition rates $R(F'm_F, Fm_F)$

are given by

$$R(4 \pm 4, 3 \pm 3) = \beta E(a_1 + 4a_2)\left(1 - 3\frac{a_3}{a_1 + 4a_2}\right) \tag{B.2}$$

and

$$R(3 \pm 3, 4 \pm 4) = \beta E(a_1 - 4a_2)\left(1 + 3\frac{a_3}{a_1 - 4a_2}\right) \tag{B.3}$$

Taking the ratio of these two transition rates and inserting the values of the a_i's gives

$$R = \frac{M + M_{\rm hf} - 3E2/4}{M - M_{\rm hf} + 3E2/4}. \tag{B.4}$$

Then

$$\frac{R-1}{R+1} = \frac{M_{\rm hf}}{M}\left(1 - \frac{3}{4}\frac{E2}{M_{\rm hf}}\right). \tag{B.5}$$

Clearly, if $E2/M_{\rm hf}$ is large enough the result is not $M_{\rm hf}/M$. If the contribution from $E2$ to the present measurement of $M_{\rm hf}/\beta$ is similarly non-negligible, then we must make a similar correction.

We now consider $E2$ with the geometry in the present experiment, which is the same as in Ref. [28]: $\vec{E} = E\hat{x}$, $\vec{B} = B\hat{z}$, and $\vec{\epsilon} = \hat{z}$.

The transition operator of concern is

$$\begin{aligned} T_{E2} &= ia_3[(\vec{S}\cdot\vec{\epsilon})(\vec{I}\cdot\vec{k}) + (\vec{S}\cdot\vec{k})(\vec{I}\cdot\vec{\epsilon})] \\ &= ia_3(S_z I_y + S_y I_z). \end{aligned} \tag{B.6}$$

An angular momentum spherical tensor operator of rank one is defined as

$$J_{1,\pm1} = \mp\frac{1}{\sqrt{2}}(J_x \pm iJ_y) \quad \text{and} \quad J_{1,0} = J_z. \tag{B.7}$$

Equation B.6 can then be written as

$$T_{E2} = -\frac{a_3}{\sqrt{2}}[S_0(I_{+1} + I_{-1}) + (S_{+1} + S_{-1})I_0]. \tag{B.8}$$

The product of two spherical tensor operators is another spherical tensor operator as defined in Zare (5.36):

$$X(k,q) = \sum_{q_1 q_2} \langle k_1 q_1 k_2 q_2 \mid kq \rangle A(k_1, q_1) B(k_2, q_2). \tag{B.9}$$

We can then write down the following components of $X(k,q)$.

$$X(1,\pm 1) = \frac{1}{\sqrt{2}}(\pm S_{\pm 1} I_0 \mp S_0 I_{\pm 1}). \tag{B.10}$$

$$X(2,\pm 1) = \frac{1}{\sqrt{2}}(S_{\pm 1} I_0 + S_0 I_{\pm 1}). \tag{B.11}$$

Then, the products of two spherical tensor operators may be written as

$$S_0 I_{\pm 1} = \frac{\mp 1}{\sqrt{2}}[X(1,\pm 1) \mp X(2,\pm 1)], \text{ and} \tag{B.12}$$

$$S_{\pm 1} I_0 = \frac{\pm 1}{\sqrt{2}}[X(1,\pm 1) \pm X(2,\pm 1)]. \tag{B.13}$$

Thus, we need to find the matrix elements of $S_0 I_{\pm 1} + S_{\pm 1} I_0$:

$$\begin{aligned} &\langle \psi' \mid S_0 I_{\pm 1} + S_{\pm 1} I_0 \mid \psi \rangle \\ &= \frac{1}{\sqrt{2}} \langle \psi' \mid \mp [X(1,\pm 1) \mp X(2,\pm 1)] \pm [X(1,\pm 1) \pm X(2,\pm 1)] \mid \psi \rangle \\ &= \frac{1}{\sqrt{2}} \langle \psi' \mid \mp X(1,\pm 1) \pm X(1,\pm 1) + X(2,\pm 1) + X(2,\pm 1) \mid \psi \rangle \\ &= \sqrt{2}\, \langle \psi' \mid X(2,\pm 1) \mid \psi \rangle. \end{aligned} \tag{B.14}$$

Thus, we have

$$\langle \psi' \mid T_{E2} \mid \psi \rangle = -a_3 \langle \psi' \mid X(2,1) + X(2,-1) \mid \psi \rangle. \tag{B.15}$$

This matrix element can be evaluated using the formulas Zare(5.64):

$$\langle \gamma j_1 j_2 j m \mid X(k,q) \mid \gamma' j_1' j_2' j' m' \rangle \tag{B.16}$$
$$= (-1)^{j-m} \begin{pmatrix} j & k & j' \\ -m & q & m' \end{pmatrix} \langle \gamma j_1 j_2 j \mid\mid X^k \mid\mid \gamma' j_1' j_2' j' \rangle,$$

and Zare (5.68):

$$\langle \gamma j_1 j_2 j \mid\mid X^k \mid\mid \gamma' j_1' j_2' j' \rangle = [(2j+1)(2j'+1)(2k+1)]^{1/2} \tag{B.17}$$

$$\times \begin{Bmatrix} j_1 & j_1' & k_1 \\ j_2 & j_2' & k_2 \\ j & j' & k \end{Bmatrix} \sum_{\gamma''} \langle \gamma j_1 \mid\mid A^{k_1} \mid\mid \gamma'' j_1' \rangle \langle \gamma'' j_2 \mid\mid B^{k_2} \mid\mid \gamma' j_2' \rangle.$$

For our situation these give

$$\langle 7SIF'm_F' \mid X(k,q) \mid 6SIFm_F \rangle = (-1)^{F'-m_F'} \tag{B.18}$$

$$\times [(2F'+1)(2F+1)(2k+1)]^{1/2} \begin{pmatrix} F' & k & F \\ -m_F' & q & m_F' \end{pmatrix}$$
$$\times \begin{Bmatrix} S & S & 1 \\ I & I & 1 \\ F' & F & k \end{Bmatrix} \sum_{\gamma''} \langle \, 7sS \mid\mid S^1 \mid\mid \gamma'' S \rangle \langle \gamma'' I \mid\mid I^1 \mid\mid 6sI \rangle.$$

Note that in the 3-j symbol we must satisfy the triangle condition with $m_F' - m_F = q$ with $q = \pm 1$. That means we either need the matrix elements of $X(2,+1)$ or of $X(2,-1)$ but never both.

The reduced matrix elements in Eq. (B.18) can be evaluated using the Wigner-Eckart Theorem. The results are

$$\langle 1/2 \mid\mid S^{(1)} \mid\mid 1/2 \rangle = \sqrt{3/2} \quad \text{and} \quad \langle 7/2 \mid\mid I^{(1)} \mid\mid 7/2 \rangle = 3\sqrt{14}. \tag{B.19}$$

We can now use Eq. (B.18) to calculate the relative size of the $E2$ amplitude compared with the M and $M_{\rm hf}$ amplitudes as well as calculate the size of the $E2$-M and $E2$-$M_{\rm hf}$ interference terms. Tables B.1 and B.2 show the relevant calculations if we assume the population in the $|\,6SF\rangle$ state is uniform across the m_F sublevels. In that case, the interference terms between the $M1$ amplitudes and the $E2$ amplitude cancel and the pure $E2$ rate is tiny. Therefore, with uniform populations, the contributions from the $E2$ amplitude can be ignored.

The population across the hyperfine sublevels is not uniform, however, as we discussed in Chapter 5. Tables B.3 and B.4 show the effect of the nonuniform population distribution calculated by Peter Marte. (See Chapter 5.) In this case, the R^+ we measure is 0.084% too large, and R^- is 0.068% too small. If we are conservative and assume a 50% uncertainty in the $E2$ correction because of uncertainties in the size of $E2$ and the exact distribution of the population, then the correction makes our result for $|\,M_{\rm hf}/\beta\,|$ smaller by 0.2% and increases our uncertainty from 0.15% to 0.16%.

These results can also be confirmed by rewriting the basis states $|\,Fm_F\rangle$ in terms of the electron and nucleus spin quantum numbers and Clebsch-Gordan coefficients using

$$|\,Fm_F\rangle = \sum_{m_S m_I} |\,Sm_S I m_I\rangle\langle S m_S I m_I\,|\,F m_F\rangle. \qquad \text{(B.20)}$$

Using that formalism gives identical results.

Table B.1: Table showing coefficients for $F = 3$ to $F' = 4$ magnetic dipole and electric quadrupole amplitudes for various transitions. The transition rates in the table are the contributions to the total transition rate relative to the M amplitude assuming uniform populations in the hyperfine levels, $E2/M_{\rm hf} = 0.053$, and $M_{\rm hf}/M = -0.1906$. The sum of all the hyperfine transitions is shown at the bottom and the fractional correction needed to account for the presence of $E2$ is shown in the "Correction" row.

$F = 3$ to $F' = 4$ Uniform Population							
		Coefficients			Transition Rates ($\times 100/M^2$)		
m_F	m'_F	$C_{Fm_F}^{F'm'_F}$	$E2$	Pop	$2ME2$	$2M_{\rm hf}E2$	$(E2)^2$
3	4	0.93541	−0.70151	0.1429	−0.1486	0.0280	0.0004
3	2	0.17678	−0.39772	0.1429	0.0159	−0.0030	0.0001
2	3	0.81009	−0.20251	0.1429	−0.0371	0.0070	0.0000
2	1	0.30619	−0.53578	0.1429	0.0371	−0.0070	0.0003
1	2	0.68465	0.17115	0.1429	0.0265	−0.0050	0.0000
1	0	0.43301	−0.54122	0.1429	0.0531	−0.0100	0.0003
0	1	0.55902	0.41923	0.1429	0.0531	−0.0100	0.0002
0	−1	0.55902	−0.41923	0.1429	0.0531	−0.0100	0.0002
−1	0	0.43301	0.54122	0.1429	0.0531	−0.0100	0.0003
−1	−2	0.68465	−0.17115	0.1429	0.0265	−0.0050	0.0000
−2	−1	0.30619	0.53578	0.1429	0.0371	−0.0070	0.0003
−2	−3	0.81009	0.20251	0.1429	−0.0371	0.0070	0.0000
−3	−2	0.17678	0.39772	0.1429	0.0159	−0.0030	0.0001
−3	−4	0.93541	0.70151	0.1429	−0.1486	0.0280	0.0004
Sum($\times 100$)					0	0	0.0027
Correction to R^+					−0.002%		

Table B.2: Table showing coefficients for $F = 4$ to $F' = 3$ magnetic dipole and electric quadrupole amplitudes for various hyperfine transitions. The transition rates in the table are the contributions to the total transition rate relative to the M amplitude assuming uniform populations in the hyperfine levels, $E2/M_{\rm hf} = 0.053$, and $M_{\rm hf}/M = -0.1906$. The sum of all the hyperfine transitions is shown at the bottom and the fractional correction needed to account for the presence of $E2$ is shown in the "Correction" row.

$F = 4$ to $F' = 3$ Uniform Population							
		Coefficients			Transition Rates ($\times 100/M^2$)		
m_F	m'_F	$C_{Fm_F}^{F'm'_F}$	$E2$	Pop	$2ME2$	$2M_{\rm hf}E2$	$(E2)^2$
4	3	0.93541	0.70151	0.1111	−0.1155	−0.0218	0.0003
3	2	0.81009	0.20251	0.1111	−0.0289	−0.0054	0.0000
2	3	0.17678	0.39772	0.1111	0.0124	0.0023	0.0001
2	1	0.68465	−0.17115	0.1111	0.0206	0.0039	0.0000
1	2	0.30619	0.53578	0.1111	0.0289	0.0054	0.0002
1	0	0.55902	−0.41923	0.1111	0.0412	0.0078	0.0001
0	1	0.43301	0.54122	0.1111	0.0412	0.0078	0.0002
0	−1	0.43301	−0.54122	0.1111	0.0412	0.0078	0.0002
−1	0	0.55902	0.41923	0.1111	0.0412	0.0078	0.0001
−1	−2	0.30619	−0.53578	0.1111	0.0289	0.0054	0.0002
−2	−1	0.68465	0.17115	0.1111	0.0206	0.0039	0.0000
−2	−3	0.17678	−0.39772	0.1111	0.0124	0.0023	0.0001
−3	−2	0.81009	−0.20251	0.1111	−0.0289	−0.0054	0.0000
−4	−3	0.93541	−0.70151	0.1111	−0.1155	−0.0218	0.0003
Sum(×100)					0	0	0.0021
Correction to R^-					0.002%		

Table B.3: Table showing coefficients for $F = 3$ to $F' = 4$ magnetic dipole and electric quadrupole amplitudes for various transitions. The transition rates in the table are the contributions to the total transition rate relative to the M amplitude assuming nonuniform populations in the hyperfine levels, $E2/M_{\rm hf} = 0.053$, and $M_{\rm hf}/M = -0.1906$. The sum of all the hyperfine transitions is shown at the bottom and the fractional correction needed to account for the presence of $E2$ is shown in the "Correction" row.

$F = 3$ to $F' = 4$ Nonuniform Population							
		Coefficients			Transition Rates ($\times 100/M^2$)		
m_F	m'_F	$C^{F'm'_F}_{Fm_F}$	$E2$	Pop	$2ME2$	$2M_{\rm hf}E2$	$(E2)^2$
3	4	0.93541	−0.70151	0.1163	−0.1548	0.0295	0.0006
3	2	0.17678	−0.39772	0.1163	0.0166	−0.0032	0.0002
2	3	0.81009	−0.20251	0.1442	−0.0480	0.0091	0.0001
2	1	0.30619	−0.53578	0.1442	0.0480	−0.0091	0.0004
1	2	0.68465	0.17115	0.1632	0.0388	−0.0074	0.0000
1	0	0.43301	−0.54122	0.1632	0.0776	−0.0148	0.0005
0	1	0.55902	0.41923	0.1666	0.0792	−0.0151	0.0003
0	−1	0.55902	−0.41923	0.1666	0.0792	−0.0151	0.0003
−1	0	0.43301	0.54122	0.1593	0.0757	−0.0144	0.0005
−1	−2	0.68465	−0.17115	0.1593	0.0379	−0.0072	0.0000
−2	−1	0.30619	0.53578	0.1369	0.0455	−0.0087	0.0004
−2	−3	0.81009	0.20251	0.1369	−0.0455	0.0087	0.0001
−3	−2	0.17678	0.39772	0.1134	0.0162	−0.0031	0.0002
−3	−4	0.93541	0.70151	0.1134	−0.1509	0.0288	0.0006
Sum($\times 100$)					0.1150	−0.0220	0.0042
Correction to R^+					−0.094%		

Table B.4: Table showing coefficients for $F = 4$ to $F' = 3$ magnetic dipole and electric quadrupole amplitudes for various hyperfine transitions. The transition rates in the table are the contributions to the total transition rate relative to the M amplitude assuming uniform populations in the hyperfine levels, $E2/M_{\rm hf} = 0.053$, and $M_{\rm hf}/M = -0.1906$. The sum of all the hyperfine transitions is shown at the bottom and the fractional correction needed to account for the presence of $E2$ is shown in the "Correction" row.

$F = 3$ to $F' = 4$ Nonuniform Population							
		Coefficients			Transition Rates ($\times 100/M^2$)		
m_F	m'_F	$C_{Fm_F}^{F'm'_F}$	$E2$	Pop	$2ME2$	$2M_{\rm hf}E2$	$(E2)^2$
4	3	0.93541	0.70151	0.123	−0.1637	−0.0312	0.0006
3	2	0.81009	0.20251	0.1061	−0.0353	−0.0067	0.0000
2	3	0.17678	0.39772	0.1038	0.0148	0.0028	0.0002
2	1	0.68465	−0.17115	0.1038	0.0247	0.0047	0.0000
1	2	0.30619	0.53578	0.104	0.0346	0.0066	0.0003
1	0	0.55902	−0.41923	0.104	0.0494	0.0094	0.0002
0	1	0.43301	0.54122	0.1109	0.0527	0.0100	0.0003
0	−1	0.43301	−0.54122	0.1109	0.0527	0.0100	0.0003
−1	0	0.55902	0.41923	0.1028	0.0489	0.0093	0.0002
−1	−2	0.30619	−0.53578	0.1028	0.0342	0.0065	0.0003
−2	−1	0.68465	0.17115	0.1068	0.0254	0.0048	0.0000
−2	−3	0.17678	−0.39772	0.1068	0.0152	0.0029	0.0002
−3	−2	0.81009	−0.20251	0.1104	−0.0367	−0.0070	0.0000
−4	−3	0.93541	−0.70151	0.1321	−0.1758	−0.0335	0.0007
Sum($\times 100$)					−0.0589	−0.0112	0.0034
Correction to R^-					0.079%		