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## Effect of higher multipole transitions in the two-photon decay spectrum of metastable hydrogen

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It is pointed out that inclusion of the effects of higher multipole transitions leads to an asymmetry in the angular distribution of the two-photon decay spectrum of metastable hydrogen.

In a recent experiment to observe the two-photon decay of metastable atomic hydrogen, O'Connell, Kollath, Duncan, and Kleinpoppen were able to measure the spectral distribution as well as the angular distribution.<sup>1</sup> The measured angular distribution basically agrees with the  $1 + \cos^2\theta$  distribution as predicted by the theory of two-electric-dipole transitions.<sup>2-7</sup>

In this note I wish to point out that if all multipole interactions are included, the angular distribution deviates from the  $1 + \cos^2\theta$  form. In particular, there are  $\cos\theta$ - and  $\cos^3\theta$ -dependent terms in the angular distribution, which result from the interference of the multipole transitions. The dominant interference comes from that between the dipole and the quadrupole. Hence, with improved precisions in angular distribution measurements, it would be interesting to look for this asymmetry. It is hoped that this paper will stimulate future experimental efforts in this regard.

The transition amplitude for the two-photon decay of metastable hydrogen in the nonrelativistic theory where spin is neglected is given by<sup>8</sup>  $\alpha^2 a M$ where  $\alpha = 1/137$ , *a* is the Bohr radius and

$$M = \vec{\epsilon}_1^{\lambda_1} \cdot \vec{\epsilon}_2^{\lambda_2} R - \sum_{i,j} \vec{\epsilon}_{ij}^{\lambda_1} \vec{\epsilon}_{2j}^{\lambda_2} \left[ T_{ij}(\Omega_1) + T_{ij}(\Omega_2) \right], \quad (1)$$

where  $\lambda_1$  and  $\lambda_2$  are the polarization indices,  $\bar{\boldsymbol{\xi}}_1$ and  $\bar{\boldsymbol{\xi}}_2$  are the polarization vectors of the emitted photons.

$$R = \langle 1S | e^{-i(\vec{k}_1 + \vec{k}_2) \cdot \vec{r}} | 2S \rangle , \qquad (2)$$
$$T_{ij}(\Omega) \equiv T_{ij}(\vec{k}_1, \vec{k}_2, \omega)$$
$$= (1/m) \langle 1S | e^{-i\vec{k}_2 \cdot \vec{r}} p_j G(\Omega) p_i e^{i\vec{k}_1 \cdot \vec{r}} | 2S \rangle , \qquad (3)$$

 $G(\Omega)$  is the Coulomb Green's function,

$$\Omega_{1,2} = |E_{2S}| - \omega_{1,2}, \qquad (4)$$

$$\omega_{1,2} = \left| \vec{k}_{1,2} \right| \,, \tag{5}$$

and  $\vec{k}_1$  and  $\vec{k}_2$  are the momenta of the emitted photons.  $\vec{p}$  is the momentum operator. *R* corresponds to the so-called "seagull" term. Its evaluation is straightforward and is easily found to be

$$R = 4\sqrt{2} \lambda^4 q^2 / (\frac{9}{4}\lambda^2 + q^2)^3 , \qquad (6)$$

where  $\lambda = \alpha Zm$ ,  $\mathbf{\bar{q}} = \mathbf{k}_1 + \mathbf{k}_2$ , and Z is the charge of the hydrogenic ion.  $T_{ij}$  can be evaluated in closed form by use of the integral representation of the Coulomb Green's function.<sup>9</sup> By taking into account of the transversality of the photon, it can be written in terms of Apelle's hypergeometric function<sup>10</sup> as

$$\begin{split} \epsilon_{1i}\epsilon_{2j}T_{ij}(\Omega) = & \frac{32\sqrt{2}\lambda^5 X^3 \vec{\epsilon}_1 \cdot \vec{\epsilon}_2}{[(X+\beta)^2 + k_1^2]! (X+\lambda)^2 + k_2^2]^2} \left\{ \frac{[X^2 - \beta^2 + k_1^2]}{[(X+\beta)^2 + k_1^2]} \frac{F_1(2 - \tau; 2, 2; 3 - \tau; x_1, x_2)}{2 - \tau} \\ & - \beta y_1 \frac{F_1(3 - \tau; 3, 3; 4 - \tau; x_1, x_2)}{3 - \tau} - \beta y_2 \frac{F_1(4 - \tau; 3, 3; 5 - \tau; x_1, x_2)}{4 - \tau} \right\} \\ & - \frac{512\sqrt{2}\lambda^5 X^5 (\vec{\epsilon}_1 \cdot \vec{k}_2) (\vec{\epsilon}_2 \cdot \vec{k}_1)}{[(X+\beta)^2 + k_1^2]! (X+\lambda)^2 + k_2^2]^3} \left\{ \frac{[X^2 - X\beta - 2\beta^2 + k_1^2]}{[(X+\beta)^2 + k_1^2]} \frac{F_1(3 - \tau; 3, 3; 4 - \tau; x_1, x_2)}{3 - \tau} + \frac{3}{2}\beta y_1 \frac{F_1(4 - \tau; 4, 4; 5 - \tau; x_1, x_2)}{4 - \tau} - \frac{3}{2}\beta y_2 \frac{F_1(5 - \tau; 4, 4; 6 - \tau; x_1, x_2)}{5 - \tau} \right\}, \end{split}$$

where

$$x_1 + x_2 = \frac{2[k_1^2 + \beta^2 - X^2][k_2^2 + \lambda^2 - X^2] - 8X^2 \vec{k}_1 \cdot \vec{k}_2}{[(X + \beta)^2 + k_1^2][(X + \lambda)^2 + k_2^2]}, \quad (8)$$

$$x_1 - x_2 = \frac{\left[ (X - \beta)^2 + k_1^2 \right] \left[ (X - \lambda)^2 + k_2^2 \right]}{\left[ (X + \beta)^2 + k_1^2 \right] \left[ (X + \lambda)^2 + k_2^2 \right]},$$
(9)

$$\beta = \lambda/2 , \qquad (10)$$

$$X = [2m(\omega + |E_{2s}|)]^{1/2}, \qquad (11)$$

$$\tau = \lambda / X , \qquad (12)$$

$$y_{1} = \frac{-4X[X^{2} - \beta^{2} + k_{1}^{2}][(X - \lambda)^{2} + k_{2}^{2}]}{[(X + \beta)^{2} + k_{1}^{2}][(X + \lambda)^{2} + k_{2}^{2}]},$$
(13)

$$y_{2} = \frac{4X[k_{2}^{2} + \lambda^{2} - X^{2}][(X + \beta)^{2} - k_{1}^{2}] + 16X^{2}(X + \beta)\vec{k}_{1} \cdot \vec{k}_{2}}{[(X + \beta)^{2} + k_{1}^{2}][(X + \lambda)^{2} + k_{2}^{2}]}.$$
(14)

From Eqs. (6)-(14), we see that to order  $(Z\alpha)^2$  the transition amplitude can be written as

$$M = \vec{\epsilon}_1^{\lambda_1} \cdot \vec{\epsilon}_2^{\lambda_2} \left\{ g(\omega_1) + (Z\alpha)^2 [f(\omega_1) + \vec{f}(\omega_1) \cos\theta] \right\} + (Z\alpha)^2 h(\omega_1) \vec{\epsilon}_1^{\lambda_1} \cdot \hat{k}_2^{\lambda_2} \vec{\epsilon}_2^{\lambda_2} \cdot \hat{k}_1 , \qquad (15)$$

where g, f,  $\overline{f}$ , and h are functions of comparable order. g corresponds to the two-electric-dipole transition without retardation and agrees with Klarsfeld's result.<sup>6</sup> f is the retardation correction to g to order  $(Z\alpha)^2$ .  $\overline{f}$  and h come from the two-electric-quadrupole transition, but in addition,  $\overline{f}$  contains a contribution from the "seagull" term. The interference between these quadrupolar terms and the dominant dipole term would lead to a correction of order  $(Z\alpha)^2$  in the decay cross section. The one-photon magnetic dipole transition amplitude, though comparable to the quadrupolar corrections, cannot give rise to an interference term with the two-electric-quadrupole transition and only gives a correction of order  $Z^4 \alpha^3$  to the total decay cross section.<sup>7</sup> Furthermore, it does not affect the asymmetry of the angular distribution of the two-photon decay mode.

Upon squaring M and summing over the polarizations of the omitted photons, we obtain, after retaining only terms to order  $(Z\alpha)^2$ ,

$$\sum_{\lambda_1\lambda_2} |M|^2 = [g^2 + 2gf(Z\alpha)^2](1 + \cos^2\theta) + 2(Z\alpha)^2g[(\overline{f} - h)\cos\theta + (\overline{f} + h)\cos^3\theta].$$
(16)

We define a dimensionless energy variable  $\eta$  by

$$\omega_1 \equiv \eta (E_{2s} - E_{1s}) = \frac{3}{8} (Z^2 \alpha / a) \eta ; \qquad (17)$$

thus

$$\omega_2 = (1 - \eta)^{\frac{3}{8}} Z^2 \alpha / a \,. \tag{18}$$

Then the decay spectrum is given by

$$\frac{d^2 W_{1s+2s}}{d\eta d\cos\theta} = \frac{1}{2} \left(\frac{3}{8}\right)^3 \eta (1-\eta) \frac{Z^6}{\pi} \alpha^8 m \sum_{\lambda_1,\lambda_2} |M|^2.$$
(19)

The decay rate in sec<sup>-1</sup> can be obtained by replacing m with  $mc^2/\hbar$ .

I have calculated  $\eta(1-\eta)g^2$ ,  $\eta(1-\eta)gf$ ,  $\eta(1-\eta)g(\overline{f}-h)$ , and  $\eta(1-\eta)g(\overline{f}+h)$  for  $\eta=0$  to 0.5, since the quantities are symmetric in  $\eta$  and  $1-\eta$ . The results are shown in Table I. In the nonrelativistic dipole approximation,  $\sum_{\lambda_1\lambda_2} |M|^2$ would reduce to  $g^2(1+\cos^2\theta)$ . I have checked these results against previous work by calculating the decay rate in this approximation by a simple numerical integration and obtained a value of  $8.24Z^6$ sec<sup>-1</sup>, as compared to previously given values of  $8.2282Z^6$  sec<sup>-1</sup> (Klarsfeld<sup>6</sup>) and  $8.226Z^6$  sec<sup>-1</sup> (Shapiro and Breit<sup>5</sup>). The angular distribution in this nonrelativistic calculation to order  $(Z\alpha)^2$  is obtained as

$$\frac{dW_{2s+1s}}{d\cos\theta} = Z^{6} \{ [3.09 - 0.97(Z\alpha)^{2}](1 + \cos^{2}\theta) \\ - 0.28(Z\alpha)^{2}\cos\theta \\ - 0.21(Z\alpha)^{2}\cos^{3}\theta \} \sec^{-1}.$$
(20)

TABLE I. Two-photon decay spectrum. (-n) indicates a multiplicative factor  $10^{-n}$ . The spectrum is symmetric in  $\eta$  and  $1 - \eta$ .

η	$\eta(1-\eta)g^2$	$-\eta(1-\eta)gf$	$-\eta(1-\eta)g(\bar{f}-h)$	$-\eta(1-\eta)g(\bar{f}+h)$
0.05	0.2692 (-1)	0.1687 (-1)	0.4745 (-3)	0.1334(-3)
0.10	0.4338 (-1)	0.2334 (-1)	0.1571(-2)	0.7164(-3)
0.15	0.5423 (-1)	0.2479(-1)	0.2962(-2)	0.1697(-2)
0.20	0.6170 (-1)	0.2359(-1)	0.4441(-2)	0.2904(-2)
0.25	0.6696 (-1)	0.2165(-1)	0.5869(-2)	0.4174(-2)
0.30	0.7067(-1)	0.1922(-1)	0.7146(-2)	0.5375(-2)
0.35	0.7327(-1)	0.1697(-1)	0.8206(-2)	0.6408(-2)
0.40	0.7494(-1)	0.1519(-1)	0.8991(-2)	0.7191(-2)
0.45	0.7591(-1)	0.1405(-1)	0.9477(-2)	0.7682(-2)
0.50	0.7623 (-1)	0.1367 (-1)	0.9641 (-2)	0.7849(-2)

Johnson<sup>7</sup> has calculated the relativistic and the retardation correction to the two-electric-dipole transition. The first correction term in Eq. (20), ~0.97( $Z\alpha$ )<sup>2</sup>, represents the retardation correction, and is contained in Johnson's results. I have fitted Johnson's results for the two-electric-dipole radiation decay rate with a series in ( $Z\alpha$ )<sup>2</sup>. To order ( $Z\alpha$ )<sup>2</sup>, Johnson's relativistic corrections modify the angular distribution to

$$\frac{dW_{2s+1s}}{d\cos\theta} = Z^{6} \{ [3.09 - 2.81(Z\alpha)^{2}](1 + \cos^{2}\theta) \\ - 0.28(Z\alpha)^{2}\cos\theta \\ - 0.21(Z\alpha)^{2}\cos^{3}\theta \} \sec^{-1}.$$
(21)

Relativistic effects only yield corrections of order  $(Z\alpha)^4$  in the  $\cos\theta$  and  $\cos^3\theta$  terms. The one-photon magnetic dipole transition, whose decay rate is given by  $2.50 \times 10^{-6}Z^{10} \text{ sec}^{-1}$ , gives a further correction of order  $Z^4\alpha^3$ . I have not included the reduced-mass correction for each element, but this can be easily incorporated from Johnson's results.<sup>7</sup>

The author thanks Professor Gerald Feinberg for helpful comments.

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