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THEORETICAL ESTIMATE OF DOUBLE QUANTUM PHOTODETACHMENT OF IODINE IONS

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The experimental value recently obtained by Hall, Robinson, and Branscomb¹ for the two-photon ionization probability of the iodine ion I^- is in disagreement with a theoretical estimate by Geltman.² It has been suggested¹ that this may be due to the inadequacy of the plane-wave approximation for the virtual states made by Geltman in evaluating the second-order contribution from the part of the electromagnetic interaction which is linear in the electron charge, $H^{(1)} = (e/mc)\vec{A}\cdot\vec{p}$.

The above discrepancy can rather be interpreted as arising from the interaction term $H^{(2)} = (e^2/2mc^2)\vec{A}^2$ having previously been disregarded.² This term, which has nonvanishing matrix elements between two-photon states and the photon vacuum, is in fact responsible for the greater part of the two-photon ionization probability in this case.

This has been ascertained by the following calculations:

(i) Using for the bound electron in I^- the approximate eigenfunction $\psi(r) = (\alpha/2\pi)^{1/2} e^{-\alpha r}/r$ [$\alpha = (2mE/\hbar^2)^{1/2}$, $E = 3.076$ eV being the electron affinity], plane waves for the positive-energy electron eigenfunctions and the incident electromagnetic wave

$$\vec{A} = \left(\frac{2}{\pi}\right)^{1/2} A \vec{\epsilon} \cos \left[\frac{2\pi}{\lambda} (\vec{n}\cdot\vec{r} - ct) \right] \quad (1)$$

($\vec{\epsilon}$ and \vec{n} denoting unit vectors in the directions of polarization and propagation, respectively;

$\lambda = 6943$ Å, $h\nu = 1.785$ eV), the transition amplitude due to $H^{(2)}$ is found to be

$$M_{fi}^{(2)} = \frac{e^2 \kappa^3}{mc^2} (2\pi\alpha)^{1/2} \left[\hbar^2 \alpha^2 + \left(\vec{p} - \frac{2\pi\hbar\vec{n}}{\lambda} \right)^2 \right]^{-1} \times A^2 \delta(E_f - E_i). \quad (2)$$

Here \vec{p} is the momentum of the ejected electron, $E_i = -E + 2h\nu$ the initial energy, and $E_f = \vec{p}^2/2m$ the final energy.

Disregarding the contribution from $H^{(1)}$, the square of $M_{fi}^{(2)}$, integrated over the momentum of the ejected electron and multiplied by the appropriate numerical factors, yields the ionization probability per unit time

$$W = \left(\frac{e^2}{4\pi\hbar c} \right)^2 \frac{[E(2h\nu - E)]^{1/2}}{(\hbar\nu)^4} \frac{\pi^2 \hbar^5 c^4}{(mc^2)^2} F^2 \approx 24.2 \times 10^{-51} F^2, \quad (3)$$

where $F = 2\nu A^2/\hbar c$ is the photon flux in the units used in references 1 and 2.

A similar calculation employing the transition amplitude $M_{fi}^{(1,1)} + M_{fi}^{(2)}$, where

$$M_{fi}^{(1,1)} = -(2c^2/h\nu) (\vec{p}\cdot\vec{\epsilon})^2 \times [4mc^2 + h\nu - 2c\vec{n}\cdot\vec{p}]^{-1} M_{fi}^{(2)} \quad (4)$$

is the second-order amplitude arising from $H^{(1)}$, yields

$$W \approx 19.8 \times 10^{-51} F^2.$$

This 20% reduction is due to the factor $[1 - \frac{2}{3} \times (2h\nu - E/h\nu)]$ by which Eq. (3) is multiplied when $M_{fi}^{(1,1)}$ is also taken into account. Note that an estimate of a lower bound for the ratio of $M_{fi}^{(2)}$ and $M_{fi}^{(1,1)}$ gives

$$\frac{M_{fi}^{(2)}}{M_{fi}^{(1,1)}} \gtrsim \frac{h\nu}{2h\nu - E}.$$

(ii) The probability as arising from $H^{(2)}$ alone has been expressed in terms of the cross section σ for one-photon ionization due to photons of energy $2h\nu = 3.57$ eV. Taking³ $\sigma = 2.1 \times 10^{-17}$ cm²,

$$W \approx 1.95 \times 10^{-32} \sigma F^2 = 410 \times 10^{-51} F^2$$

has been obtained. Considering the reduction

due to the contribution from $H^{(1)}$ and the fact that, as stated in reference 3, the value of σ may be in error by 50%, the above value does not seem incompatible with the experimental result.

Indebtedness to G. J. Schulz for drawing attention to this problem and for many discussions is gratefully acknowledged.

¹J. L. Hall, E. J. Robinson, and L. M. Branscomb, Phys. Rev. Letters **14**, 1013 (1965).

²S. Geltman, Phys. Letters **4**, 168 (1963).

³B. Steiner, M. L. Seman, and L. M. Branscomb, J. Chem. Phys. **37**, 1200 (1962).

⁴The idea of expressing two-photon in terms of one-photon ionization amplitudes is due to Geltman (see reference 2).

CORRECTION TO THE ($\text{He}^3, 2^3S_1$) TO ($\text{He}^{3+}, 2^2S_{1/2}$) HYPERFINE-STRUCTURE RATIO*

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Tests of the theory of hfs in simple atoms which do not depend upon nuclear structure are of special interest because of the (45±17) parts per million (ppm) difference¹ between the theoretical and experimental hfs of atomic hydrogen. Various ratios of hfs measurements have this property and therefore afford unambiguous tests of the quantum-electrodynamic corrections. The theoretical ratio of the 2S state hfs to the 1S state hfs agrees^{2,3} with the measured value to a precision of several parts in 10⁸ for the H atom, and a precision of several parts in 10⁷ for D and He³⁺. There has been, however, a discrepancy in the ratio of the 2³S₁ state hfs of the He³ atom⁴ to the 2²S_{1/2} state hfs of the He³⁺ ion,⁵ the calculated^{6,7} value being too large by 10 ppm. The purpose of this note is to point out the omission of a correction of -4 ppm in this theoretical ratio.

The second-order radiative corrections to hfs have been calculated in powers of the Coulomb field, i.e., in powers of $Z\alpha$. They give a correction to the hfs of the form

$$1 + \alpha[a + b(Z\alpha) + c(Z\alpha)^2 + \dots].$$

Here⁸ $a = \frac{1}{2}\pi$ and $b = \frac{5}{2} - \log 2$; c depends upon the atomic state and contains $\log(Z\alpha)$ and $\log^2(Z\alpha)$. Thus the atom/ion ratio should have a previous-

ly omitted factor

$$F = \frac{1 + \alpha[a + b(Z\alpha)] + [\lambda c_1^a + (1-\lambda)c_2^a] \alpha (Z\alpha)^2}{1 + \alpha[a + b(Z\alpha) + c_2^i (Z\alpha)^2]} \\ = 1 + [c_1^a - c_2^i + (1-\lambda)(c_2^a - c_1^a)] \alpha (Z\alpha)^2 + \dots,$$

where λ is the fraction of the atomic hfs due to the interaction of the nucleus and the 1s electron, and $(1-\lambda)$ is the fraction due to the 2s electron; exchange effects are neglected.

The atomic hfs can be obtained⁹ to within 3.6% by assuming that all the hfs arises from a single unscreened 1s electron, or by using the simplest two-parameter variational wave function.¹⁰ In the first case $\lambda = 1$, and in the second $\lambda = 0.95$; we will use $\lambda = 1$ for simplicity. Since the radiative corrections arise at distances from the origin comparable to the electron Compton wavelength, screening is negligible, and $c_1^a \approx c_1^i$. Thus,

$$F \approx 1 + (c_1^i - c_2^i) \alpha (Z\alpha)^2 = 1 - (4.2 \pm 0.4) \times 10^{-6},$$

using Zwanziger's result,¹¹ $c_1^i - c_2^i = -2.7$ for $Z = 2$, and allowing for a 10% error due to our approximations.

The remaining discrepancy of 6 ppm may be due to inaccuracies in the relativistic corrections,⁶ which have an estimated uncertainty