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RADIATIVE DECAY OF METASTABLE $^3P_0$ ATOMIC STATES*

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Abstract:

The radiative decay of atoms in the metastable nsnp $^3P_0$ state is found to occur only by odd parity multiphoton modes like E1M1 and 3E1. A detailed calculation of the E1M1 rate in berylliumlike argon (Ar XV) yields a lifetime of about 18 days. Estimates indicate the 3E1 rate to be several orders of magnitude smaller than the E1M1 rate.

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In atoms with two valence electrons such as the Be isoelectronic series, the lowest excited state is nsnp $^3P_0$, which is energetically allowed to decay only to the $ns^2\ ^1S_0$ ground state. In odd isotopes of such atoms, hyperfine structure mixes the $^3P_0$ and $^3P_1$ states, while spin-orbit and spin-spin interactions mix the $^3P_1$ with $^1P_1$ states, thus enabling the $^3P_0 - {}^1S_0$ transition to occur in the electric dipole (E1) mode. Garstang[1] has calculated rates for Mg I, Zn I, Cd I, and Hg I.

In the even isotopes this mode is not possible. In fact, all single photon decay modes are forbidden by the $0 \not\to 0$ selection rule, which remains valid relativistically. Furthermore, the $^3P_0 - {}^1S_0$ transition involves a parity change, which rules out all even parity multi-photon modes such as the usual 2E1 which accounts for the decay of the 2S states of hydrogenlike and heliumlike atoms.[2] We thus conclude that only odd parity multi-photon modes can contribute to the $^3P_0 - {}^1S_0$ transition. The leading modes are E1M1, E1E2, and 3E1, but for a $0 \to 0$ transition, the E1E2 is also forbidden by the E2 $0 \not\to 1$ selection rule on the intermediate states.

The main interest in these processes is probably astrophysical, since the lifetimes are expected to be large. In supernovae rich in heavy elements the energy balance could be affected by loading a sink of long-lived metastable atoms which return the energy some months or years later. Also, the metastable component could serve as a probe of ion abundances in regions of much smaller density than stellar atmospheres. In both cases, knowledge of the lifetimes would be needed. We examine the rate for the E1M1 decay mode.

The probability per second that an atom makes the transition $|i\rangle \to |f\rangle$ by emitting two photons, one of frequency ν_1 within the interval $d\nu_1$, the other of frequency ν_2 determined by $\nu_1 + \nu_2 = (E_i - E_f)/h \equiv \nu_0$, and where one photon has E1 character, the other M1 character, is

$$A(\nu_1)d\nu_1 = \frac{2^6\pi^4 e^4}{m^2 c^8}\,\nu_1^3\nu_2^3\,d\nu_1\,|M|^2_{AVG} \tag{1}$$

where

$$M = \sum_n \left[\frac{\langle f|\hat{\varepsilon}_2\cdot\vec{R}|n\rangle\langle n|\hat{k}_1 \times \hat{\varepsilon}_1\cdot(\vec{L}+2\vec{S})|i\rangle}{\nu_{ni}+\nu_1} + \frac{\langle f|\hat{\varepsilon}_1\cdot\vec{R}|n\rangle\langle n|\hat{k}_2 \times \hat{\varepsilon}_2\cdot(\vec{L}+2\vec{S})|i\rangle}{\nu_{ni}+\nu_2} \right.$$
$$\left. + \frac{\langle f|\hat{k}_2 \times \hat{\varepsilon}_2\cdot(\vec{L}+2\vec{S})|n\rangle\langle n|\hat{\varepsilon}_1\cdot\vec{R}|i\rangle}{\nu_{ni}+\nu_1} + \frac{\langle f|\hat{k}_1 \times \hat{\varepsilon}_1\cdot(\vec{L}+2\vec{S})|n\rangle\langle n|\hat{\varepsilon}_2\cdot\vec{R}|i\rangle}{\nu_{ni}+\nu_2} \right] \tag{2}$$

In these formulas, $\hat{\varepsilon}_1$, $\hat{\varepsilon}_2$ are parallel to the electric field vector of the photons, which have propagation vectors $\vec{k}_1$, $\vec{k}_2$; $\nu_{ni} = (E_n - E_i)/h$: AVG means the average over all polarization and propagation directions, as well as the average over initial and sum over final magnetic substates; and Σ_n runs over all possible states $|n\rangle$ that can be coupled to $|i\rangle$ and $|f\rangle$.

After performing the indicated averages, we find, for the $^3P_0 - {}^1S_0$ transition

$$|M|^2_{AVG} = \frac{1}{27}\left|\sum_n \frac{A_n}{\nu_{ni} + \nu_1}\right|^2 + \frac{1}{27}\left|\sum_n \frac{A_n}{\nu_{ni} + \nu_2}\right|^2 \tag{3}$$

where

$$A_n = ({}^1S_0\|R\|\varphi_n)(\varphi_n\|L + 2S\|{}^3P_0) \tag{4}$$

are reduced matrix elements[3] using Edmond's convention.[4] Note that the last two terms of Eq. (2) are zero. Since neither R nor L + 2S alone can mix singlets and triplets, the decay is possible only because spin-orbit and spin-spin effects produce mixing. Thus,

$$|\varphi_n) = \begin{cases} |n^1P_1) + \sum_{n'} a_{nn'}\, |n'^3P_1) \\ |n^3P_1) + \sum_{n'} b_{nn'}\, |n'^1P_1) \end{cases} \tag{5}$$

where the coefficients $a_{nn'}$, $b_{nn'}$ must be calculated or inferred from spectroscopic measurements. We immediately note that the operator L + 2S has only one nonzero off diagonal element, namely $({}^3P_1\|L + 2S\|{}^3P_0) = -\sqrt{2}$. This means only one term $a_{nn'} = -b_{nn'} \equiv c_n$ will contribute from Eq. (5), with the result

$$|W|^2_{AVG} = \frac{2}{27}\left(|S(\nu_1)|^2 + |S(\nu_2)|^2\right) \tag{6}$$

where

$$S(\nu) = \sum_n c_n({}^1S_0\|R\|n\,{}^1P_1)\left[\frac{1}{\nu(n\,{}^1P_1 - {}^3P_0) + \nu} - \frac{1}{\nu({}^3P_1 - {}^3P_0) + \nu}\right] \tag{7}$$

In the heavier ions, the nsnp ${}^1P_1$ state is appreciably lower than any other ${}^1P_1$ state, hence one value c_n will dominate. That is, nsnp ${}^3P_1$ mixes predominantly with nsnp ${}^1P_1$ and very little with any other states. Under this condition, we can use

$$(gf)_3 = \frac{4\pi m}{3\hbar}\,\nu({}^3P_1 - {}^1S_0)\,|({}^1S_0\|R\|{}^3P_1)|^2 \tag{8}$$

with $({}^1S_0\|R\|{}^3P_1) \cong c_n({}^1S_0\|R\|{}^1P_1)$ to rewrite Eq. (7) in terms of the $(gf)_3$ values, which have been calculated by Garstang.[5] The result is

$$A(\nu_1)d\nu_1 = \frac{2^5\pi^3}{9}\;\frac{\alpha^5 a_0^3}{c^3}\,(gf)_3\,\frac{[\nu({}^1P_1 - {}^3P_1)]^2}{\nu({}^3P_1 - {}^1S_0)}\,\nu_0^3\,f(y)\,dy \tag{9}$$

where

$$f(y) \equiv y^3(1 - y)^3\left\{\left(\frac{1}{(\beta + y)(\eta + y)}\right)^2 + \left(\frac{1}{(\beta + 1 - y)(\eta + 1 - y)}\right)^2\right\} \tag{10}$$

and $\beta \equiv \nu({}^1P_1 - {}^3P_0)/\nu_0$, $\eta \equiv \nu({}^3P_1 - {}^3P_0)/\nu_0$, with $y = \nu_1/\nu_0$.

For berylliumlike argon, Ar XV, the spectrum given by $f(y)$ is plotted in Fig. 2. Integrating this curve and using $(gf)_3 = 4.3 \times 10^{-4}$, the total transition probability

$$A = \frac{1}{2} \int_0^{\nu_0} A(\nu_1)\, d\nu_1 \tag{11}$$

is found to be

$$A(\text{Ar XV: } 2s2p\ {}^3P_0 \rightarrow 2s^2\ {}^1S_0) \cong 6.6 \times 10^{-7}\ \text{sec}^{-1} \tag{12}$$

corresponding to a mean lifetime of about 18 days.

A similar calculation for Be I yielded the value $A \sim 6 \times 10^{-15}$ sec^{-1}, or a lifetime of 50 million years, but this result is less accurate because the contributions from the higher ${}^1P_1$ states, which have been neglected here, are relatively more important.

The Z dependence of the E1M1 rate can be inferred easily from Eq. (9). The frequencies scale approximately as Z^2, while $f(y)$ is only weakly Z dependent. Hence, the dependence is nearly Z^8 times the strong Z dependence of $(gf)_3$. For $Z = 30$, if we estimate $(gf)_3 \sim 10^{-2}$, the lifetime is estimated to be $\tau \sim 30$ min.

The triple photon decay (3E1) process has several interesting properties. Since $\nu_1 + \nu_2 + \nu_3 = \nu_0$, if one photon has frequency ν_1, the other two form a continuum between zero and $\nu_0 - \nu_1$. The transition probability vanishes if any two photons have the same energy, and in the limit that one photon has zero energy. Surprisingly, the spectrum observed as single photons, unlike the symmetric two-photon spectra, is irregular and asymmetric, being peaked on the low energy side of $\nu_0/2$.

The relative importance of the 3E1 mode may be estimated by noting that adding another E1 photon introduces a factor $\sim \alpha(ka_0)^2$, while changing E1 to M1 introduces $\sim \alpha^2$. Thus, $3E1/E1M1 \sim (ka_0)^2/\alpha \sim 10^{-3}$ for ions in the Be sequence near Z = 18. Offsetting this low ratio is the fact that the rejection of 2-photon events in a triple coincidence experiment is very high. If these decays should become experimentally accessible, separation of the 3E1 mode from the E1M1 mode probably would not be difficult.

The author benefitted from fruitful discussions on this subject with Prof. R. H. Garstang.

FOOTNOTES AND REFERENCES

*Work performed under the auspices of the U. S. Atomic Energy Commission.

1. R. H. Garstang, J. Opt. Soc. Amer. 52, 845 (1962).

2. G. Breit and E. Teller, Astrophys. J. 91, 215 (1940).

3. The decaying state is $|nsnp\ ^3P_0) \equiv |^3P_0)$, the ground state is $|ns^2\ ^1S_0) \equiv |^1S_0)$, intermediate states are $|nsnp\ ^3P_1) \equiv |^3P_1)$, $|nsnp\ ^1P_1) \equiv |^1P_1)$ and others $|n'\ell'n''\ell''\ ^1P_1)$ and $|n'\ell'n''\ell''\ ^3P_1)$ are written simply as $|n\ ^1P_1)$ and $|n\ ^3P_1)$ respectively.

4. A. R. Edmonds, Angular Momentum in Quantum Mechanics, Princeton University Press, New Jersey (1957).

5. R. H. Garstang, Astrophys. J. 148, 665 (1967).

FIGURE CAPTIONS

Fig. 1. Energy levels of a berylliumlike atom. The virtual transitions enabling the E1M1 double photon emission are shown as dotted lines. H_1 represents spin-orbit and spin-spin mixing. The 3E1 mode, enabled by other virtual transitions, competes only weakly. This diagram is not to scale; the ${}^3P_1$ - ${}^3P_0$ separation is much smaller than indicated.

Fig. 2. Spectrum of the E1M1 emission from berylliumlike Ar XV. The spectra of other members of the Be I sequence are practically identical. The central dip originates from the small value of the ratio $\eta = \nu({}^3P_1 - {}^3P_0)/\nu_0$, which is present due to the additional selection rules on M1 transitions from the ${}^3P_0$ state. This dip may be slightly exaggerated due to approximations made in the calculations.

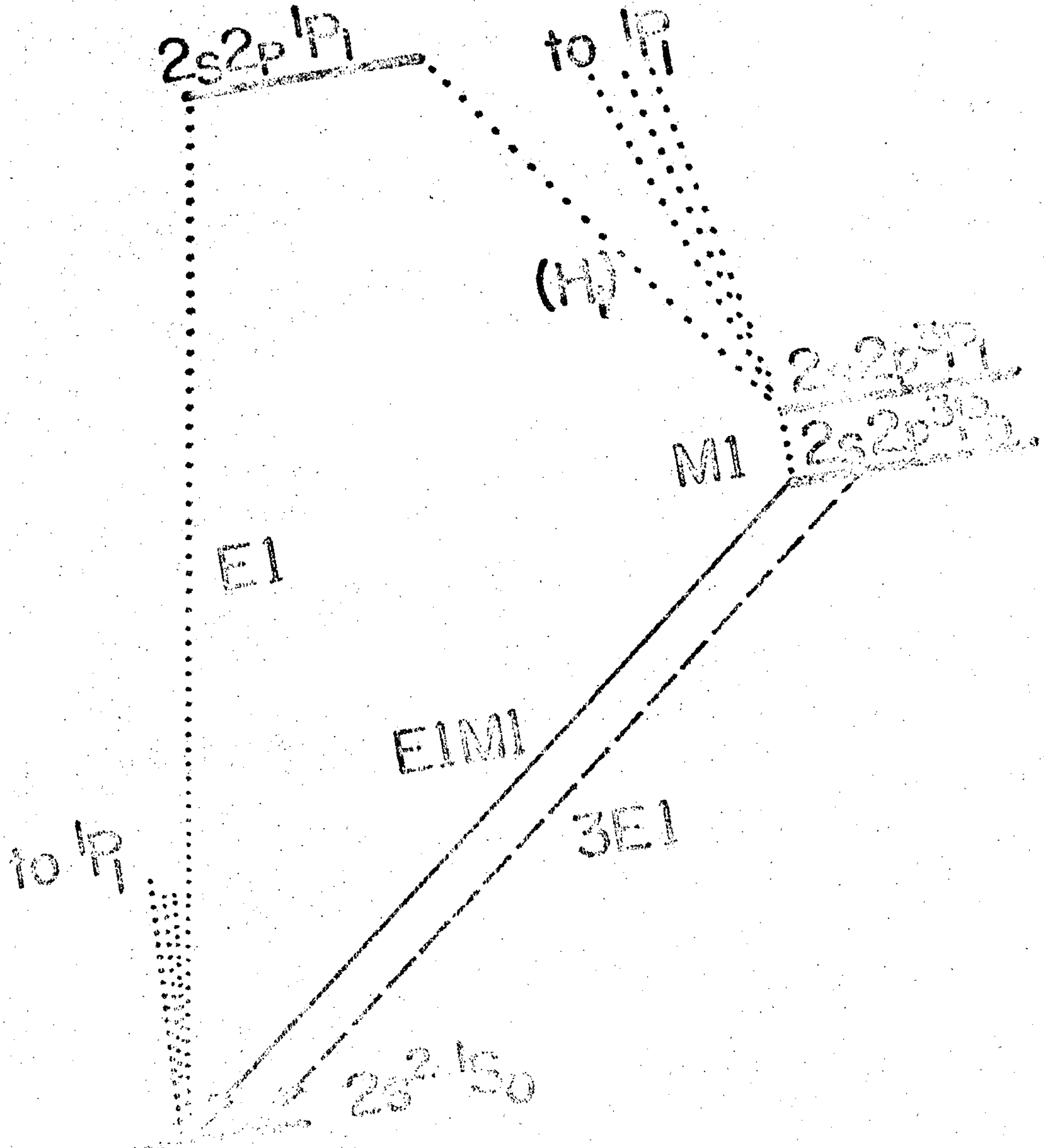

Fig. 1

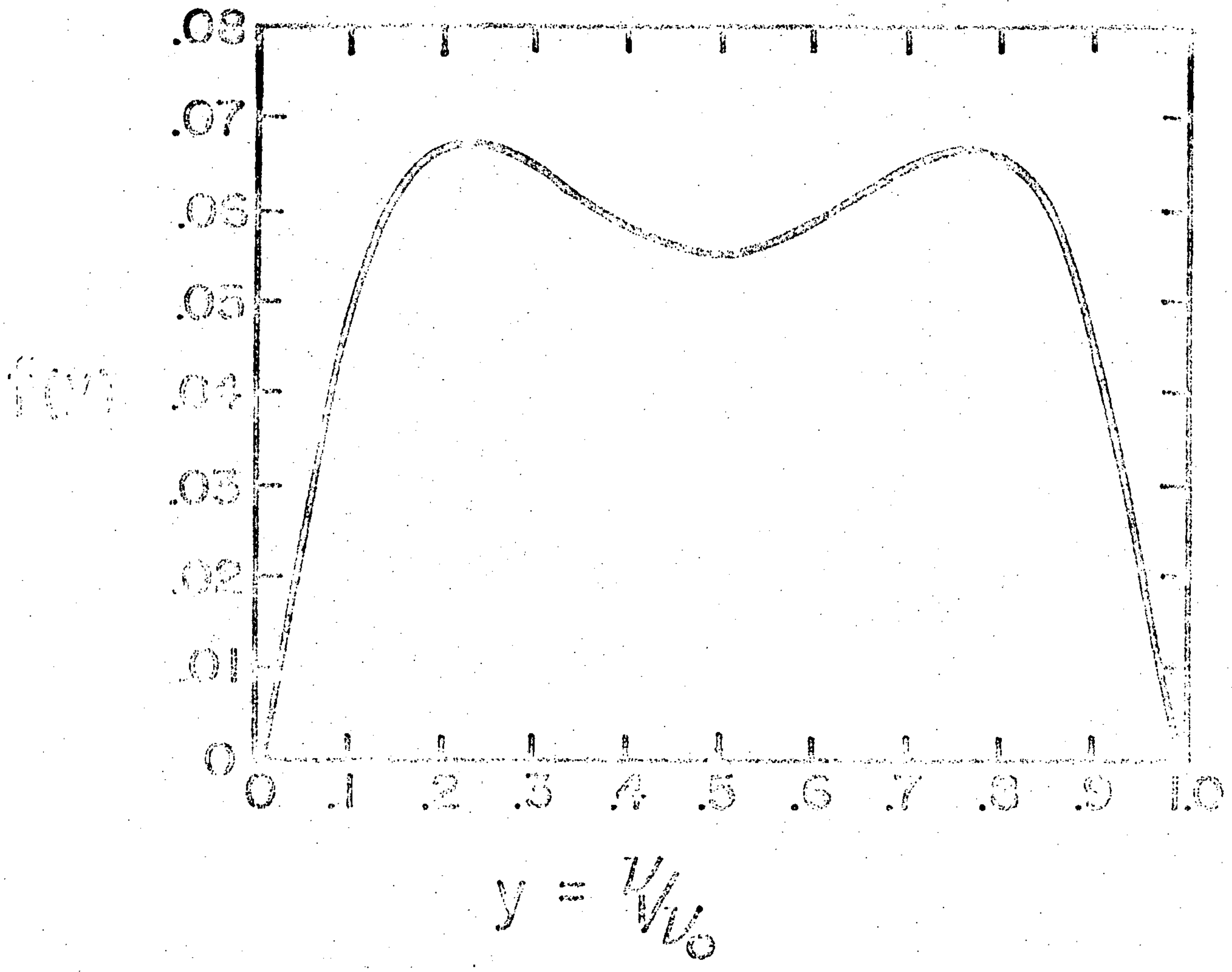

Fig. 2

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