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Lifetime of the metastable $^3P_2$ and $^3P_0$ states of rare-gas atoms*

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The lifetimes of the $^3P_2$ states of Ne, Ar, Kr, and Xe are calculated by considering the spin-dependent magnetic-quadrupole ($SM2$) radiative transition to the $^1S_0$ state as the decay mechanism. By expressing the $SM2$ operator as a contraction of the spin magnetic dipole and the electric dipole, the lifetime of the $^3P_2$ state can be expressed very simply in terms of that of the $^1P_1$ state. With the known lifetimes of the $^1P_1$ state, the calculated lifetimes of the $^3P_2$ states are 24.4, 55.9, 85.1, and 149.5 sec, respectively, for Ne, Ar, Kr, and Xe. The lifetime of the $^3P_0$ state is calculated by considering both the magnetic-dipole ($M1$) transition to $^3P_1$ and the electric-quadrupole ($E2$) transition to $^3P_2$ as the decay mechanism. The calculated lifetimes are 430, 44.9, 0.488, and 0.078 sec, respectively, for Ne, Ar, Kr, and Xe. These calculated lifetimes are above the lower limits of the experimental lifetimes measured by Van Dyck, Johnson, and Shugart.

I. INTRODUCTION

The $^1S_0$ ground state of rare-gas atoms—Ne, Ar, Kr, and Xe—has a closed-shell configuration $(np)^6$. When one of the (np) electrons is excited into $(n's)$ we have both the ^3P and the ^1P states. This excited-state configuration $(np)^5(n's)$ is similar to the configuration $(np)(n's)$ of two electrons. Here we have a (np) hole instead of a (np) electron, and the three ^3P_J states are inverted with $^3P_0$ lying on the top and $^3P_2$ at the bottom (see Fig. 1). Among the triplet states, $^3P_1$ mixes with $^1P_1$ state through spin-orbit coupling and hence decays to the $^1S_0$ ground state via electric-dipole ($E1$) transition. However, both $^3P_2$ and $^3P_0$ cannot similarly decay to the lower states via allowed electric-dipole transition, and they are metastable states. The $^3P_0$ state can decay either to the $^3P_1$ state through magnetic-dipole ($M1$) radiative transition or to the $^2P_2$ state through electric-quadrupole ($E2$) radiative transition. The $^3P_2$ state can decay only by spin-dependent magnetic-quadrupole ($SM2$) radiation to the $^1S_0$ ground state. This $SM2$ radiative interaction is a spin-dependent tensor operator of the second rank which connects states of opposite parity and differing by $\Delta S=1$ and $\Delta J=2$. Mizushima[1] has considered $SM2$ mechanism for the $^3\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ transition of H_2. $SM2$ has also been considered by Mizushima[2] and Garstang[3] as the decay mechanism in calculating the lifetimes of the $^3P_2$ states of heliumlike ions which are of astrophysical significance. Lifetimes of these ions are of the order of 10^{-9} sec, which can be measured by the time-of-flight technique.[4] However, the lifetimes of the $^3P_0$ and $^3P_2$ metastable states of Ne, Ar, Kr, and Xe are of the order of seconds or longer, and they cannot be easily measured by the time-of-flight technique. Recent lifetime measurements by Van Dyck, Johnson, and Shugart[5] can only set a lower limit for the composite lifetimes of the $^3P_2$ and $^3P_0$ states of Ne, Ar, and Kr. It is therefore very useful and interesting to calculate these lifetimes and to compare them with the experimental lower limits.

In this work we calculate the lifetimes of the $^3P_2$ states of Ne, Ar, Kr, and Xe by considering the spin-dependent magnetic quadrupole ($SM2$) radia-

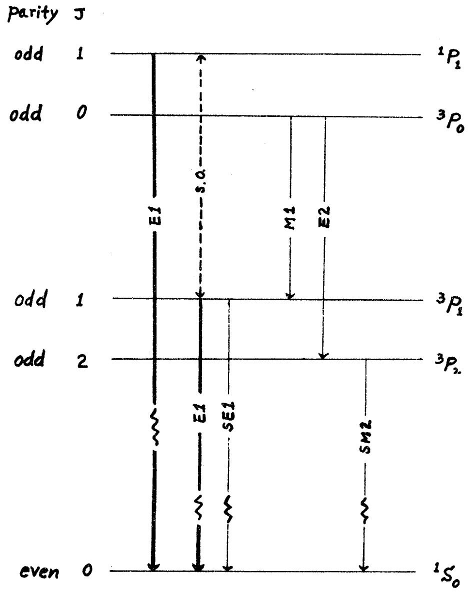

FIG. 1. Schematic energy-level diagram for the rare-gas atoms, Ne, Ar, Kr, and Xe. The $^3P_0$ state can decay to either the $^3P_1$ state by magnetic-dipole ($M1$) radiative transition or to the $^3P_2$ state by electric-quadrupole ($E2$) radiative transition. The $^3P_2$ state can decay only by spin-dependent magnetic-quadrupole ($SM2$) radiation to the $^1S_0$ ground state. The $^3P_1$ state can either (by mixing with the $^1P_1$ state) decay to the $^1S_0$ ground state via electric-dipole transition or reach the $^1S_0$ state through a much slower spin-dependent electric-dipole ($SE1$) transition.

tive transition as the decay mechanism. The $SM2$ operator is expressed as a contraction of two first-rank tensor operators, i.e., electric-dipole and spin-magnetic dipole. After evaluating the spin-dependent part, the total transition probability from ${}^3P_2$ to ${}^1S_0$ is expressed in terms of electric-dipole transition moments. To evaluate these transition dipole moments, one needs the accurate orbital wave functions both for the ${}^3P$ and the ${}^1S$ states. However, if one assumes that the ${}^3P$ and the ${}^1P$ states are constructed out of the same single-electron orbitals, u_{np} and $v_{n's}$ (but of opposite permutational symmetry with respect to interchanging two electrons), the transition dipole moment for ${}^3P \rightarrow {}^1S$ transition can be expressed in terms of the same single-electron transition dipole moment as that for ${}^1P \rightarrow {}^1S$ transition. Consequently, the lifetime of the ${}^3P_2$ state can be expressed in terms of the lifetime of the ${}^1P_1$ state which can be measured experimentally. The calculations and the numerical values of the ${}^3P_2$ state lifetimes will be presented in Sec. III. Stedman and Setser[6] have also calculated the lifetime of the ${}^3P_2$ state of rare-gas atoms by following Garstang's formulation[3b] on $SM2$ transitions. However their values are smaller and differ[7] from the present work by almost an order of magnitude. Finally in Sec. IV, the lifetimes of the ${}^3P_0$ state will be calculated by considering both the magnetic-dipole radiation and the electric-quadrupole radiation as the decay mechanism. However the magnetic-dipole radiative transition will be the dominant process for the decay of ${}^3P_0$.

II. MULTIPOLE OPERATORS FOR RADIATIVE TRANSITIONS

In this section we will express all the multipole operators for different radiative transitions as a tensor operator. The matrix elements of these operators can therefore be easily evaluated by using Racah's algebra. The total transition probability per unit time from an initial state Ψ_i which is excited to all the final states Ψ_f via radiative transitions (by emitting a photon λ of energy $\hbar c \kappa_\lambda$) is given by[8-10]

$$W = \sum_q \sum_f \frac{\kappa_\lambda}{2\pi\hbar} \int d\Omega_\lambda \, |(\Psi_f | H(r,\lambda) | \Psi_i)|^2 \qquad (2.1)$$

where $q = \pm 1$ are the transverse polarizations. The radiative interaction Hamiltonian $H(r,\lambda)$ above operates on the atomic system *only*, it is a sum of $H_r(r,\lambda)$ (which is the spin-independent interaction) and $H_s(r,\lambda)$ (which is the spin-dependent interaction) and they are given by

$$H_r(r,\lambda) = -\frac{e}{2mc} \sum_j [\vec{A}_\lambda^*(r_j) \cdot \vec{P}_j + \vec{P}_j \cdot \vec{A}_\lambda^*(r_j)] \qquad (2.2)$$

and

$$H_s(r,\lambda) = -\frac{e}{2mc} \sum_j 2\hbar \vec{S}_j \cdot \nabla \times \vec{A}_\lambda^*(r_j) \qquad (2.3)$$

where e and m are respectively the electronic charge and mass, c is the velocity of light, $\vec{P}_j$ and $\vec{S}_j$ are the linear momentum and the spin of electron j, respectively. By Taylor's expansion of the vector potential,

$$\vec{A}_j^*(r_j) = \hat{e}_q e^{-i\vec{\kappa}_\lambda \cdot \vec{r}_j}, \qquad (2.4)$$

one obtains the multipole expansion of $H(r,\lambda)$.[11-14] The leading terms of the spin-independent interaction $H_r(r,\lambda)$ are $H_q(E1)$ (the electric-dipole interaction), $H_q(E2)$ (the electric-quadrupole interaction), and $H_q(M1)$ (the magnetic-dipole interaction), and they are expressed as the tensor operators in the following:

$$H_q(E1) = -i\kappa_\lambda e \sum_j \sum_m D^1_{mq}(\hat{R}) r_j C^1_m(\theta_j, \phi_j), \qquad (2.5)$$

$$H_q(E2) = \frac{e\kappa_\lambda^{\,2}}{2\sqrt{3}} \sum_j \sum_m D^2_{mq}(\hat{R}) r_j^2 C^2_m(\theta_j, \phi_j), \qquad (2.6)$$

$$H_q(M1) = -\kappa_\lambda q \mu_B \sum_j \sum_m D^1_{mq}(\hat{R}) L_m(j). \qquad (2.7)$$

In the above equations, $\mu_B = e\hbar/2mc$ is the Bohr magneton, $C^l_m(\theta_j, \phi_j)$ is the normalized spherical harmonics of electron j referring to the space-fixed coordinates, and $D^l_{mq}(\hat{R})$ is the rotation matrix element where $\hat{R}$ ($\equiv \alpha\beta\gamma$) are the Euler angles of rotation taking space-fixed z axis (quantization axis) into the direction of $\vec{\kappa}_\lambda$. Similarly, the spin-dependent Hamiltonian $H_s(r,\lambda)$ is a sum of

$$H_q(SE1) = \sqrt{2} i \kappa_\lambda^2 \mu_B \sum_j \sum_m D^1_{mq}(\hat{R}) T^1_m(j), \qquad (2.8)$$

which is the spin-dependent electric-dipole interaction, and which can cause radiative transition between the pure L-S coupled state ${}^3P_1$ and state ${}^1S_0$, and

$$H_q(SM2) = -\sqrt{2} i q \kappa_\lambda^2 \mu_B \sum_j \sum_m D^2_{mq}(\hat{R}) T^2_m(j) \qquad (2.9)$$

is the spin-dependent magnetic-quadrupole interaction which induces radiative transitions between ${}^3P_2$ and ${}^1S_0$. The spin-dependent tensor $T^l_m(j)$, appearing both in (2.8) and (2.9), is shown in the following as a contraction of two first-rank tensors, namely electric-dipole and spin-magnetic dipole,

$$T^l_m(j) = \sum_\nu C(11l; m-\nu, \nu, m) r_j C^1_{m-\nu}(\theta_j, \phi_j) S_\nu(j). \qquad (2.10)$$

After evaluating the spin-dependent part, the matrix element of the operator $T^l_m(j)$ over the initial and the final states will be reduced into the matrix elements of the electric-dipole operator. This will be illustrated in the next section where we calculate the lifetime of the $^3P_2$ state.

III. LIFETIME OF THE $^3P_2$ STATE

The $^3P_2$ state can decay only via spin-dependent magnetic-quadrupole radiative transition to the $^1S_0$ ground state. Based on this decay mechanism, we substitute (2.9) into (2.1) and perform the integration over the solid angle $d\Omega_\lambda$ by applying the following orthonormal relation[15]:

$$\int D^{l\,*}_{mq}(\alpha\beta\gamma)D^{l'}_{m'q'}(\alpha\beta\gamma)\,d\Omega_\lambda=\frac{4\pi}{2l+1}\,\delta_{ll'}\delta_{mm'}\delta_{qq'}\,. \tag{3.1}$$

The transition probability from $^3P_2$ to $^1S_0$ now becomes

$$W(^3P_2\rightarrow {}^1S_0)$$
$$=\frac{8\beta^2\kappa^5}{5\hbar}\sum_f\sum_m\left|\left(\Psi(^1S_0)\middle|\sum_j T^2_m(j)\middle|\Psi(^3P_2)\right)\right|^2, \tag{3.2}$$

where $T^2_m(j)$ is given by (2.10) and the summation over the final states f means summing over the magnetic sublevels M'_J. Since it can be easily shown that

$$\sum_{M'_J}\sum_m\left|(J'M'_J\,|\sum_j T^l_m(j)\,|JM_J)\right|^2=\frac{2J'+1}{2J+1}\left|(J'\,\|\sum_j T^l(j)\,\|J)\right|^2 \tag{3.3}$$

the transition probability $W(^3P_2\rightarrow {}^1S_0)$ is as expected independent of the magnetic quantum number M_J of the initial $^3P_2$ state. We may now evaluate (3.2) for any given value of M_J, e.g., $M_J=2$.

Representing the $^3P_2$ state by the following pure L-S coupled wave function:

$$\Psi^0_{J\,M_J}=\sum_{M_L}C(LSJ;M_L,M_J-M_L,M_J)\Phi_{L,\,M_L}\chi_{S,\,M_J-M_L} \tag{3.4}$$

(where $J=2$, $L=S=1$, and choosing $M_J=2$) and by (2.10), the summation on the right-hand side of (3.2) now becomes

$$\sum_f\sum_m\left|\left(\Psi(^1S_0)\middle|\sum_j T^2_m(j)\middle|\Psi(^3P_2)\right)\right|^2=\frac{1}{9}\left|\sum_{j=1}^{2}(^1S_0,L=0\,\|r(j)\|\,^3P,L=1)(S=0\,\|S(j)\|\,S=1)\right|^2, \tag{3.5}$$

where the reduced matrix elements for electron spin[16] can be readily evaluated as follows:

$$(S=0\,\|S(2)\|\,S=1)=-(S=0\,\|S(1)\|\,S=1)=\sqrt{3}/2\,. \tag{3.6}$$

By using the following single-configuration orbital wave functions:

$$\Phi(^3P)=(1/\sqrt{2})[u_{np}(1)v_{n's}(2)-u_{np}(2)v_{n's}(1)] \tag{3.7}$$

and

$$\Phi(^1S)=u_{np}(1)u_{np}(2)\,, \tag{3.8}$$

we therefore have

$$(^1S,L=0\,\|r(2)\|\,^3P,L=1)=-(^1S,L=0\,\|r(1)\|\,^3P,L=1)$$
$$=(3/2)^{1/2}(u_{np}\,|z|\,v_{n's})\,, \tag{3.9}$$

where $(u_{np}\,|z|\,v_{n's})$ is the single-electron transition dipole moment. Substituting (3.6) and (3.9) into (3.5), which is then substituted into (3.2), the transition probability from $^3P_2$ to $^1S_0$ is now expressed in terms of the single-electron transition dipole moment as follows:

$$W(^3P_2\rightarrow {}^1S_0)=(4/5\hbar)\kappa^5\beta^2\,|(u_{np}\,|z|\,v_{n's})|^2\,, \tag{3.10}$$

and the lifetime of the $^3P_2$ state, $\tau(^3P_2)=W^{-1}$, is just the reciprocal of the above expression.

We now express the transition probability $W(^1P_1\rightarrow {}^1S_0)$ from $^1P_1$ to $^1S_0$ also in terms of the same single-electron transition dipole matrix elements. Since $^1P_1$ can mix with $^3P_1$ through the spin-orbit interaction, $\Psi(^1P_1)$ is therefore written as a linear combination of the pure L-S coupled wave functions $\Psi^0(^1P_1)$ and $\Psi^0(^3P_1)$ as follows:

$$\Psi(^1P_1)=\cos\theta\,\Psi^0(^1P_1)+\sin\theta\,\Psi^0(^3P_1) \tag{3.11a}$$

and

$$\Psi(^3P_1)=-\sin\theta\,\Psi^0(^1P_1)+\cos\theta\,\Psi(^3P_1)\,, \tag{3.11b}$$

where the normalized coupling constants $\cos\theta$ and $\sin\theta$ will later be calculated from the spin-orbit interactions for atoms Ne, Ar, Kr, and Xe. The transition probability from $^1P_1$ to $^1S_0$ now takes the following expression:

$$W(^1P_1 \rightarrow {}^1S_0) = \sum_q \sum_f \frac{\kappa}{2\pi\hbar} \int |[\cos\theta(\Psi^0(^1S_0)|H_q(E1)|\Psi^0(^1P_1)) + \sin\theta(\Psi^0(^1S_0)|H_q(SE1)|\Psi^0(^3P_1))]|^2 , \tag{3.12}$$

where $H_q(E1)$ and $H_q(SE1)$ are given by (2.5) and (2.8), respectively. Since the transition rate for $SE1$ process is much smaller (almost a factor of 10^{-10} times smaller) than that of $E1$ process, the second term ($SE1$ term) in (3.12) can be neglected. Following the same procedure presented previously for the $^3P_2 \rightarrow {}^1S_0$ transition, and assuming[17] that the orbital wave function of the $^1P_1$ state,

$$\Phi(^1P_1) = (1/\sqrt{2})[u_{np}(1)v_{n's}(2) + u_{np}(2)v_{n's}(1)] \tag{3.13}$$

is composed of the same single-electron orbitals (u_{np} and $v_{n's}$) as that for $\Phi(^3P_1)$ given in (3.7), we have

$$W(^1P_1 \rightarrow {}^1S_0) = (8\kappa^3 e^2/3\hbar)\,|(u_{np}|z|v_{n's})|^2 \cos^2\theta . \tag{3.14}$$

The transition probability $W(^1P_1 \rightarrow {}^1S_0)$ is therefore expressed in terms of the same single-electron transition dipole matrix element as that for $W(^3P_2 \rightarrow {}^1S_0)$. Notice the κ here in (3.14) corresponds to the energy of transition between $^1P_1$ and $^1S_0$, whereas the κ appearing previously in (3.10) corresponds to the energy of the transition between $^3P_2$ and $^1S_0$. Combining (3.10) and (3.14) we obtain the following simple expression for the lifetime ratio of $\tau(^3P_2)$ to that of $\tau(^1P_1)$:

$$\frac{\tau(^3P_2)}{\tau(^1P_1)} = \frac{W(^1P_1 \rightarrow {}^1S_0)}{W(^3P_2 \rightarrow {}^1S_0)} = \frac{5(\lambda_3)^5 e^2}{6(\lambda_1)^3 \pi^2 \beta^2} \cos^2\theta \tag{3.15}$$

where $\lambda = 2\pi/\kappa$. The subscripts 1 and 3 correspond to the transitions $^1P_1 \rightarrow {}^1S_0$ and $^3P_2 \rightarrow {}^1S_0$, respectively. Knowing $\tau(^1P_1)$ and $\cos^2\theta$, the lifetime of the $^3P_2$ state, $\tau(^3P_2)$, is readily obtained.

We will now calculate the spin-orbit coupling coefficient $\cos\theta$. The spin-orbit interaction H_{so} is given as follows:

$$H_{so} = \sum_{i=1}^{2} \xi(r_i)\vec{L}_i \cdot \vec{S}_i , \tag{3.16}$$

where

$$\xi(r_i) = \frac{\hbar^2}{2m^2c^2} \frac{1}{r_i} \frac{dV(r_i)}{dr_i} \tag{3.17}$$

is expressed in terms of the central-field potential energy $V(r_i)$. Considering H_{so} (Ref. 18) and the electrostatic repulsion e^2/r_{12} as the perturbations for a two-electron system, the solution of the secular determinant gives the following energy values:

$$E(^1P_1) = F_0 + \tfrac{1}{4}\xi_p + [(G_1 - \tfrac{1}{4}\xi_p)^2 + \tfrac{1}{2}\xi_p^2]^{1/2} , \tag{3.18}$$

$$E(^3P_0) = F_0 - G_1 + \xi_p , \tag{3.19}$$

$$E(^3P_1) = F_0 + \tfrac{1}{4}\xi_p - [(G_1 - \tfrac{1}{4}\xi_p)^2 + \tfrac{1}{2}\xi_p^2]^{1/2} , \tag{3.20}$$

$$E(^3P_2) = F_0 - G_1 - \tfrac{1}{2}\xi_p , \tag{3.21}$$

where $F_0 = F_0(np, n's)$ and $G_1 = G_1(np, n's)$ are, respectively, the Coulomb and the exchange integrals of the operator e^2/r_{12}. The coupling coefficients $\cos\theta$ and $\sin\theta$ as defined in (3.11) now have the following values:

$$\cos\theta = (1+\gamma^2)^{-1/2} , \quad \sin\theta = \gamma(1+\gamma^2)^{-1/2} , \tag{3.22}$$

where

$$\gamma = \frac{\xi_p/\sqrt{2}}{(G_1 - \frac{1}{4}\xi_p) + [(G_1 - \frac{1}{4}\xi_p)^2 + \xi_p^2/2]^{1/2}} , \tag{3.23}$$

$\xi_p = (u_{np}|\xi(r)|u_{np})$ is the spin-orbit coupling constant.[19] By (3.18)–(3.21), ξ_p and G_1 can be evaluated from the measured energy separations.[20] With these values for ξ_p and G_1, we have calculat-

TABLE I. Lifetimes of the $^3P_2$ and the $^3P_0$ states of rare-gas atoms.

Atom	$\cos^2\theta$ [a]	$(\lambda_1)^{-1}$ (cm^{-1}) [b]	$(\lambda_3)^{-1}$ (cm^{-1}) [b]	$\tau(^3P_2)/\tau(^1P_1)$	$\tau(^1P_1)$ (nsec) [c]	$\tau(^3P_2)$ (sec)	$\kappa_{0,1}$ (cm^{-1}) [d]	$\tau(^3P_0)$ (sec)
Ne	0.930	13.589×10^4	13.404×10^4	1.221×10^{10}	2.0	24.4	2257.9	430
Ar	0.797	9.540×10^4	9.314×10^4	2.236×10^{10}	2.5	55.9	5045.8	44.9
Kr	0.486	8.585×10^4	7.997×10^4	2.128×10^{10}	4.0	85.1	26860	0.488
Xe	0.438	7.719×10^4	6.707×10^4	3.361×10^{10}	4.45	149.5	51218	0.0782

[a] $\cos\theta$ is the spin-orbit coupling coefficient defined in (3.13) and (3.25).

[b] $(\lambda_1)^{-1} = [E(^1P_1) - E(^1S_0)]/hc$, $(\lambda_3)^{-1} = [E(^3P_2) - E(^1S_0)]/hc$. The numerical values are from *Atomic Energy Levels*, Natl. Bur. Std. (U. S.) Circ. No. 467 (U. S. GPO, Washington D. C., 1949, 1952, 1958), Vols. 1–3.

[c] Semiempirical values given in Ref. 20a.

[d] $\kappa_{0,1} = [E(^3P_0) - E(^3P_1)]/\hbar c$. The numerical values are from the same reference as quoted in footnote b of this table.

ed the coupling constant $\cos\theta$ for Ne, Ar, Kr, and Xe and they are listed in Table I. Substituting these values into (3.15), we obtain the ratio of the lifetime of the $^3P_2$ state to that of the $^1P_1$ state. With the known lifetime[21] of the $^1P_1$ state, the lifetime of the $^3P_2$ state of Ne, Ar, Kr, and Xe are readily calculated and they are listed in Table I. The lifetime of the $^3P_2$ state calculated by Stedman and Setser[6] are respectively 2.0, 8.0, 13, and 33 sec for Ne, Ar, Kr, and Xe. Although they calculated these lifetimes by following Garstang's[3b] formulation on *SM*2 transitions, their values differ[7] from the present results in Table I by almost an order of magnitude. This discrepancy is too large to be accounted for even if they might have neglected the spin-orbit coupling between $^3P_1$ and $^1P_1$.

IV. LIFETIME OF THE $^3P_0$ STATE

The $^3P_0$ state can decay either to the $^3P_1$ state via magnetic dipole transition (*M*1) or to the $^3P_2$ state via electric quadrupole transition (*E*2). Substituting (2.6) and (2.7) into (2.1) and after carrying out the integration over the solid angle $d\Omega_\lambda$, we obtained the total transition probability W from the $^3P_0$ state to both the $^3P_1$ and the $^3P_2$ states as follows:

$$W=\frac{e^2(\kappa_{0,2})^5}{15\hbar}\sum_m\sum_{M_J}\left|\left(\Psi(^3P_2)\middle|\sum_j r_j^2C_m^2(\theta_j,\phi_j)\middle|\Psi(^3P_0)\right)\right|^2+\frac{4(\kappa_{0,1})^3}{3\hbar}\mu_B^2\sum_m\sum_{M_J'}\left|\left(\Psi(^3P_1)\middle|\sum_j L_m(j)\middle|\Psi(^3P_0)\right)\right|^2, \tag{4.1}$$

where $\kappa_{0,2}=[E(^3P_0)-E(^3P_2)]/\hbar c$ and $\kappa_{0,1}=[E(^3P_0)-E(^3P_1)]/\hbar c$. The summation indices M_J and M_J' refer to the magnetic sublevels of the final states $^3P_2$ and $^3P_1$, respectively. The lifetime of the $^3P_0$ state, $\tau(^3P_0)=W^{-1}$, is the reciprocal of the total transition probability given by (4.1).

Both $\Psi(^3P_2)$ and $\Psi(^3P_0)$ are the pure *L-S* coupled states which can be represented by (3.4). However $\Psi(^3P_1)$, as given by (3.11) is a linear combination of the pure *L-S* coupled states $\Psi^0(^3P_1)$ and $\Psi^0(^1P_1)$. Following (3.3), the summation of the first term (electric quadrupole term) in (4.1) becomes

$$\sum_m\sum_{M_J}\left|\left(\Psi(^3P_2)\middle|\sum_j r_j^2C_m^2(\theta_j,\phi_j)\middle|\Psi(^3P_0)\right)\right|^2=5\left|\left(J=2,L=S=1\middle\|\sum_j r_j^2C^2(\theta_j,\phi_j)\middle\|J=0,L=S=1\right)\right|^2. \tag{4.2}$$

After decomposing[16] the reduced matrix element which is in the coupled (J) representation into that of the uncoupled representation (LS), (4.2) now reads

$$\sum_m\sum_{M_J}|(\cdots)|^2=15W(1012;12)^2\left|\left(\Phi(^3P),L=1\middle\|\sum_j r_j^2C^2(\theta_j,\phi_j)\middle\|\Phi(^3P),L=1\right)\right|^2, \tag{4.3}$$

where $W(abcd;ef)$ is the Racah coefficient.[16] By the Wigner-Eckart relation[17] and by using the orbital wave function given in (3.7) for $\Phi(^3P)$, we have

$$\begin{aligned}\left|\left(\Phi(^3P)\middle\|\sum_{j=1}^{2} r_j^2C^2(\theta_j,\phi_j)\middle\|\Phi(^3P)\right)\right|^2&=\tfrac{5}{2}|(u_{np_z}|r^2C_0^2(\theta_j,\phi_j)|u_{np_z})|^2\\&=\frac{2}{5}\left|\int_0^\infty R_{np}(r)r^4\,dr\right|^2,\end{aligned} \tag{4.4}$$

where $R_{np}(r)$ is the radial part of the orbital wave function u_{np}, and (4.3) now reads

$$\sum_m\sum_{M_J}|(\cdots)|^2=\frac{2}{5}\left|\int_0^\infty R_{np}^2(r)r^4\,dr\right|^2. \tag{4.5}$$

Similarly, the second summation (magnetic-dipole term) in (4.1) becomes

$$\sum_m\sum_{M_J'}\left|\left(\Psi(^3P_1)\middle|\sum_j L_m(j)\middle|\Psi(^3P_0)\right)\right|^2=9W(1011;11)^2C(111;101)^{-2}|(u_{np_z}|L_z|u_{np_z})|^2\cos^2\theta=2\cos^2\theta. \tag{4.6}$$

Substituting both (4.5) and (4.6) into (4.1), the total transition probability from ${}^3P_0$ to both ${}^3P_1$ and ${}^3P_2$ is given by

$$W=\frac{2e^2(\kappa_{0,2})^5}{75\hbar}\left|\int_0^\infty R_{np}^2(r)r^4\,dr\right|^2+\frac{8\beta^2}{3\hbar}(\kappa_{0,1})^3\cos^2\theta . \tag{4.7}$$

The radial integral above can be integrated numerically by using the Hartree-Fock wave function of Knox *et al.*[20] As expected, the first term in (4.7) (which is due to electric-quadrupole transition) is found[22] to be a factor of 10^{-5} to 10^{-3} smaller than the second term (which is due to magnetic-dipole transition), we therefore calculated the lifetime of the ${}^3P_0$ state, $\tau({}^3P_0)$, by considering the magnetic-dipole transition only. The values are listed in the last column of Table I.

In Table I, as we run from Ne to Xe, the lifetime[23] of the ${}^3P_2$ state increases from 24.4 to 149.5 sec while the lifetime of the ${}^3P_0$ state decreases from 430 to 0.078 sec. Van Dyck, Johnson, and Shugart[5] have measured the composite lifetime τ of the metastable rare-gas atoms, i.e., the lifetime of composite atoms in both ${}^3P_2$ and ${}^3P_0$ states. They obtained a lower limit for τ by beam-of-flight experiment, and their values are 0.8, 1.3, and 1.0 sec, respectively, for Ne, Ar, and Kr. In the beam-of-flight experiment, one measures the ratio of the number of atoms, $N_a(v)/N_b(v)$, passing through the two separate points a and b in the beam. This ratio is related[5] to the lifetime by

$$\frac{N_a(v)}{N_b(v)}=C\left[\tfrac{5}{6}e^{-t/\tau_2}+\tfrac{1}{6}e^{-t/\tau_0}\right]=Ce^{-t/\tau} \tag{4.8}$$

where τ_2 and τ_0 are the lifetimes of ${}^3P_2$ and ${}^3P_0$, respectively, and τ is the composite lifetime that has been measured. The numerical factors $\frac{5}{6}$ and $\frac{1}{6}$ are the statistical weighting factors for the states ${}^3P_2$ and ${}^3P_0$, respectively, and C is the proportionality constant. When τ_2 is much larger than τ_0, as in the cases for Kr and Xe, the second term (τ_0 term) can be dropped. The measured composite lifetime τ in this case becomes the lifetime of the ${}^3P_2$ state, τ_2. Although the calculated lifetime of the ${}^3P_0$ of Kr (possibly also Xe) lies below the measured lower limit of the composite lifetime τ, there is no real disagreement between the calculated and the measured values because the calculated lifetime of the ${}^3P_2$ state is well above the measured lower limit for Kr.

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[17] This is a good assumption which can also be justified by the discussions in footnote 19.

[18] Evaluation of the H_{so} over the $(np)^5(n's)$ configuration is the same as that over the $(np)(n's)$ configuration, except that a minus sign arises due to the (np) hole which results in inverted fine-structure levels.

[19] If the single-electron orbital wave function u'_{np} chosen for the ${}^1P$ state differs from u_{np} chosen for the ${}^3P$ state, the off-diagonal matrix element of spin-orbit interaction (between ${}^3P_1$ and ${}^1P_1$) will give rise to $(u'_{np}|\xi(r)|u_{np})=\lambda\xi_p$ instead of ξ_p, where λ is the so-called King–Van Vleck parameter. Dow and Knox (Ref. 18) have calculated λ for Ne, Ar, Kr, and Xe, and their values are practically equal to 1 (within 0.1%). This fact certainly justifies our approximation that u_{np} and u'_{np} are the same.

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[21] The experimental lifetimes of the ${}^1P_1$ state are not available for all the atoms considered here. The $\tau({}^1P_1)$

values used here are the semiempirical values from Ref. 20.

[22] Using the wave function in Ref. 18, we have calculated the radial integral $|\int_0^\infty R_{np}^2(r) r^4\, dr|^2$ numerically, and it is 8.50×10^{-34}, 3.91×10^{-33}, 9.95×10^{-33}, or 2.088×10^{-32} cm^4, respectively, for Ne, Ar, Kr, or Xe. The ratio of the first term (electric-quadrupole transition probability term) in (4.7) to the second term (magnetic-dipole transition term) are therefore 0.590×10^{-5}, 5.58×10^{-5}, 1.076×10^{-3}, and 5.91×10^{-3}, respectively, for Ne, Ar, Kr, and Xe.

[23] The preliminary values of $\tau(^3P_2)$ were presented at the Fourth International Conference on Atomic Physics, Heidelberg, 1974. The values quoted there [see p. 201, *Proceedings of the Fourth International Conference on Atomic Physics, Abstracts of Contributed Papers, Heidelberg, 1974,* edited by J. Kowalski and H. G. Weber (Heidelberg Univ. Press, 1974)] are in error by approximately a factor of 10.