Manifestation of Dynamically Unfavored Transitions in the Angular Distribution of Photofragments

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A class of parity-favored electric dipole transitions are classed as "dynamically unfavored" with a photoparticle angular distribution parameter $\beta \sim -1$. This deviation from the elementary-model value $\beta = 2$ results when the squared pseudoscalar operator $(\vec{j}_t \cdot \hat{r})^2$ is nonzero. Herrick's SO(4) classification singles out doubly excited ${}^1P^\circ$ continuum eigenchannels of H$^-$ and He with $(\vec{j}_t \cdot \hat{r})^2 \sim 0$ or ~ 1. Most of the photoelectrons should have $(\vec{j}_t \cdot \hat{r})^2 \sim 1$, i.e., $\beta \sim -1$.

Photoparticle angular distributions are known[1] to be very sensitive to the parity of the angular momentum transferred to the target residue,

$$\vec{j}_t = \vec{j}_\gamma - \vec{l} = \vec{l}_f - \vec{L}_i . \tag{1}$$

Here $j_\gamma = 1$ for electric dipole processes, l is the orbital momentum of the observed fragment, and L_i and l_f are the unobserved angular momenta of the target and residue, respectively. In particular a parity-unfavored transition, for which $j_t + j_\gamma + l$ is odd, automatically has an asymmetry parameter $\beta_{\text{unf}} = -1$ in the distribution

$$d\sigma/d\hat{k} \propto 1 + \beta P_2(\hat{\epsilon}\cdot\hat{k}) \xrightarrow[\beta = -1]{} \tfrac{3}{2}|\hat{\epsilon}\times\hat{k}|^2, \tag{2}$$

where $\hat{\epsilon}$ is the photon's polarization. By contrast, parity-favored transitions can have any asymmetry between the extremes -1 and 2, depending on the specific dynamics.

Reference 1 emphasized that parity-unfavored angular distributions result from pseudovector or pseudotensor interactions between the observed and unobserved reaction products. I show here how an unfavored asymmetry parameter results even for a parity-favored reaction, in the presence of *pseudoscalar* dynamical elements.

Consider an even-parity spinless atom with $L_i = 0$ absorbing a linearly polarized photon. Photoionization leaves the residual ion in a state with orbital momentum $l_f = j_t$. The transition is thus parity favored. In the elementary case where $\beta = 2$ we have $d\sigma/d\hat{k} \propto |\hat{\epsilon}\cdot\hat{k}|^2$, i.e., the electron escapes preferentially along the polarization axis $\hat{\epsilon}$, in contrast to the orthogonal escape given by Eq. (2). The mean value of the squared pseudoscalar, $(\vec{L}\cdot\hat{r})^2 = [(\vec{l}_f + \vec{l})\cdot\hat{r}]^2$, then vanishes at large photoelectron distances r where $\hat{r} = \hat{k}$, because $\vec{L} = \vec{j}_\gamma$ and $\vec{j}_\gamma \cdot \hat{\epsilon} = 0$. On the other hand, for dynamically unfavored transitions with $\beta \sim -1$, the electron is most likely to escape along $\hat{r} = \pm\hat{L}$ and the mean value of $(\vec{L}\cdot\hat{r})^2$ is of order unity. [Note that the identity $\vec{l}\cdot\hat{r} = 0$ implies that $(\vec{L}\cdot\hat{r})^2 = (\vec{l}_f\cdot\hat{r})^2 = (\vec{j}_t\cdot\hat{r})^2$, and also note that β may differ from 2 for other trivial reasons, e.g., when the photoelectron escapes with $l = 0$ only.]

The relationship between β and the dynamical unfavoredness $(\vec{j}_t\cdot\hat{r})^2$ will be derived here for a specific problem, namely, H$^-$ photodetachment processes which leave the atom in some excited state with principal quantum number $n \geqslant 1$. The "accidental" degeneracy of hydrogen implies that the atom and the photoelectron need not separate in eigenstates of $\vec{l}_f^{\,2}$ and $\vec{l}^{\,2}$, respectively. Rather, as shown by Seaton[2] and by Gailitis and Damburg,[3] the representation appropriate to this separation diagonalizes the operator

$$(\vec{l}^{\,2} + 2\vec{r}_f\cdot\hat{r})/2r^2, \tag{3}$$

which combines the centrifugal field and the dipole interaction. This operator has $2n-1$ eigenvectors ${}^1P^\circ$, and as many for ${}^3P^\circ$, related to the Stark eigenstates of the residual H as shown below.

Herrick[4] has analyzed this problem in terms of the Runge-Lenz vectors $\vec{b}_f$ and $\vec{b}$, which are separate constants of the motion for the H atom and for the photoelectron as $r \to \infty$. For each of these separate systems the operators $\{\vec{b}_f, \vec{l}_f\}$ or $\{\vec{b}, \vec{l}\}$ are the generators of an SO(4) symmetry group. The dipole interaction term $A_0 = 2\vec{r}_f\cdot\hat{r}$ in (3) is an invariant of the product group SO(4) = SO(4)$_f \otimes$ SO(4) and is diagonalized automatically by casting the product representation in irreducible form.

The key point is the following. The degenerate states of the H atom and of the ejected electron do not separately have a definite parity. The SO(4) generators $\vec{b}$ which intermix them are odd-parity vectors, while the $\vec{l}$ are pseudovectors. Moreover, the reduction of the product group involves operators of opposite helicity, $\vec{l} \pm \vec{b}$. Pseudoscalar elements thus enter the dynamics of our problem even though the Hamiltonian (3) is a scalar. (It may also be noted that while the operator

$A_0 = 2\vec{r}_f \cdot \hat{r}$ is constructed with vectors, it is to be diagonalized within a Hilbert space whose dimensions are limited by eigenvalues of the pseudovectors $\vec{l}_f$ and $\vec{L}$.) Thus it is not surprising that the correlations between the residual H and the photoelectron are represented in part by nonzero values of the pseudoscalar operator $\vec{l}_f \cdot \hat{r}$. However, no complete analysis is available of the role of parity in the reduction of the SO(4) group.

In any event, it is apparent from Ref. 4 that the reduction of the product representation splits the matrix of $A_0 = 2\vec{r}_f \cdot \hat{r}$, firstly, into two diagonal blocks labeled by eigenvalues 0 and 1 of the squared pseudoscalar operator $T^2 = (\vec{L} \cdot \hat{r})^2 = (\vec{l}_f \cdot \hat{r})^2$. (Following Herrick, I use T as the nonnegative square root of T^2.) Within each block the eigenvectors of the irreducible representation are classified by a further quantum number

$$K = n-1-T,\, n-3-T, \ldots, -(n-1-T), \tag{4}$$

where n is the principal quantum number of the H residue. Each of these eigenvectors is also an eigenvector of A_0 with the eigenvalue

$$A_0 = -3nK/Z, \tag{5}$$

where $Z=1$ is the nuclear charge of H.

The dynamical unfavoredness T *alternates* along the sequence of successive eigenstates of K and T arranged in order of increasing A_0:

$$(K,T) = (n-1,0), (n-2,1), (n-3,0), \ldots. \tag{6}$$

The very same order applies also to the eigenvalues of the complete operator (3), which identify successive eigenchannels of photoelectron escape at large r. This ordering is verified by the numerical evaluation of (3) discussed in Ref. 3, and it is justified qualitatively by considering that $\langle K,T|\vec{l}_f{}^2|K,T\rangle$ increases linearly[4] with n, while A_0 if of order n^2 according to (4) and (5). Accordingly Herrick's classification of eigenchannels becomes exact in the limit $n \to \infty$. Note that this alternation of values of $T = 0,1,0,\ldots$ coincides with the alternation of ${}^1P^{\circ}$ hyperspherical quantum numbers $-$, $+$, $-$, ... in the larger-r limit, which has emerged in independent studies of doubly excited states of H$^-$ and He.[5,6]

The value of the asymmetry parameter $\beta(K,T)$ for a specific escape eigenchannel is given according to Ref. 1 by

$$\beta(K,T) = \textstyle\sum_{j_t} \sigma(j_t)\beta(K,T;j_t)/\sum_{j_t}\sigma(j_t), \tag{7}$$

where $j_t = l_f$ in this problem. Each separate $\beta(K, T; j_t)$ involves the coherent superposition of two contributions $D_\pm$ from photoelectron escape with $l = l_f \pm 1$. For eigenvectors of A_0 alone the dipole amplitudes $D_\pm$ are proportional to the Wigner coefficient $(l_f - T;\, LT|\, l0)$, implying a ratio

$$\gamma = \frac{D_-}{D_+} = \frac{(l_f - T; 1T\,|\,l_f - 1, 0)}{(l_f - T; 1T\,|\,l_f + 1, 0)} = \begin{cases} -[l_f/(l_f+1)]^{1/2}, & T=0 \\ [(l_f+1)/l_f]^{1/2}, & T=1, \end{cases} \tag{8}$$

independently of K. [This result follows from Eq. (32) of Ref. 4.] The presence of this Wigner coefficient shows how the relative orientations of $\vec{l}_f$, $\vec{L}$, $\vec{l}$, and $\hat{r}$ are fixed by the value of T; i.e., for $T=0$, $\vec{L}$ has no projection along $\hat{r}$, while for $T=1$ $\vec{L}$ is aligned along $\hat{r}$. Equation (12) of Ref. 1 gives then

$$\beta(K,T;l_f) = \frac{(l_f+2) + (l_f-1)\gamma^2 - 6[l_f(l_f+1)]^{1/2}\gamma}{(2l_f+1)(1+\gamma^2)}, \tag{9}$$

where γ is given approximately by (8); in general γ must be evaluated by direct diagonalization of (3). The approximation (8) yields simply

$$\beta(K,T;l_f) = \begin{cases} 2, & T=0 \\ -1, & T=1, \end{cases} \tag{10}$$

establishing a *one-to-one correspondence* between β and T. The values of $\beta(K,T)$ obtained from (7) and (9) with the exact γ tend to the limits (10) for large n, as shown in Fig. 1. Deviations of β from the limiting values -1 and 2 are proportional to

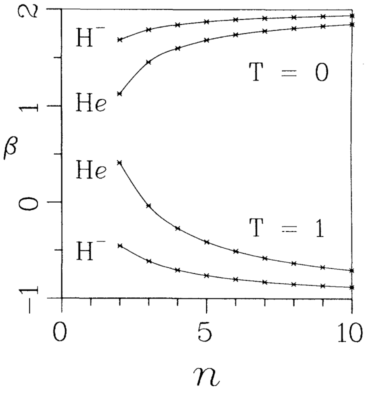

FIG. 1. Asymmetry parameter $\beta(K,T)$ characterizing the angular distribution of photoelectrons from H$^-$ or He, as a function of the excited state n of the residual H or He$^+$. Results shown are for the two most attractive channels only, for which $K = n-1-T$. Note the strong dependence of β on the dynamical unfavoredness, $T^2 = (\vec{j}_t \cdot \hat{r})^2$.

Z/n; this convergence for $n \to \infty$ is slower for He than H^- because He^+ dipole moments are smaller than those of H by roughly $\frac{1}{2}$.

Theory[5,7] has long predicted that photoemission from H^- and He would occur mostly in "+" eigenchannels. This prediction has been verified experimentally[8] for the normal $^1P^o$ emission from H^-, for which $+ \equiv (K = n-2, T = 1)$; for $^3P^o$ one has instead $+ \equiv (K = n-1, T = 0)$. Low-energy photoelectrons emitted when the $^1S^e$ ground state is photoionized should thus have the values $\beta \sim -1$ plotted in the lower two curves of Fig. 1. These values will probably hold only for photoelectron energies within a fraction of an electron volt. In sharp contrast, the asymmetry of photoelectrons from a $^3S^e$ metastable state of He should have $\beta \sim 2$, as in the upper two curves of Fig. 1. These properties should also be reflected in the alignment of highly excited residual H atoms or He^+ ions and, consequently, in the angular distribution and polarization of their fluorescence.

Note that the above results assume that the electronic spins are coupled in either a singlet for a triplet state. This should be valid for photoelectron energies much larger than the fine-structure splittings of the hydrogenic levels, for which the different fine-structure components may retain a phase coherence. The coherence aspect will be discussed separately.

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Observation of Stimulated Level Shifting in Inverted Atomic Thallium Populations

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Level shifting has been observed in a two-level, two-isotope atomic system which is undergoing stimulated emission. The optical fields which produce the level shifts are generated by the same stimulated process.

In this Letter we report the first observation of level shifting in an amplifying medium which is undergoing stimulated emission ("stimulated level shifting"). The material medium is atomic thallium which is produced with total population inversion in the Tl $7^2S_{1/2}$ state as a result of photodissociation of TlI by 193-nm radiation. The pulsed, stimulated, optical fields evolve within the inverted medium as a result of superfluorescent emission along the axis of uv dissociation pump beam. Since there are two naturally occurring isotopes of thallium, Tl^{203} (30%) and Tl^{205} (70%), each isotopic system produces an optical field, the frequency of which is nearly resonant with the unperturbed frequency connecting the atomic levels of the other isotope. As the magnitude of each field increases, the atomic levels of one isotope are shifted by the high-frequency Stark (HFS) effect due to the field of the other isotopes. In the high-field limit of our experiment, the two separate systems are coupled, i.e., the frequency shift is much greater than the zero-field isotope splitting.

Level shifts due to the interaction of spontaneously emitted light from one isotopic system with atoms of a second isotope have been important in developing an understanding of the HFS effect, and in the spectroscopy of microwave resonance studies, although typical shifts in these experiments are small, ~1 Hz.[1] Recently, Liao