Unfavored angular distributions

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The recent prediction of H^- photodetachment peaking orthogonally to the light polarization, and the underlying considerations of SO(4) symmetry by Herrick are illustrated in terms of vector diagrams. We emphasize the role of the helicity of the SO(4) operators $\vec{l} \pm \vec{b}$, a phenomenon that is also apparent in the construction of the fundamental "spinor" representation of the Lorentz group and in the emerging of electron spin from the Dirac theory.

The angular distribution of fragments ejected from an atom or molecule through electric-dipole processes is described by $I(\theta) \propto 1 + \beta\frac{1}{2}(3\cos^2\theta - 1)$, where θ is the angle between the directions of emission and of incident polarization and where the coefficient β depends on the specific process. Elementary theory yields $\beta = 2$, corresponding to maximum emission in the direction of polarization, but β may actually range from 2 down to a minimum of -1 for which the emission peaks orthogonally to the polarization.

The value $\beta = -1$ has been known to occur for the entire class of "parity-unfavored" processes which result from pseudovector or pseudotensor interactions between the observed and unobserved reaction products.[1] Quite recently, the same value has also been predicted for certain parity-favored processes, where an electron is ejected from H^- (or He) leaving the target in one of its degenerate excited states.[2] This startling instance of crosswise emission has been called "dynamically unfavored." Its origin has been traced analytically to earlier results on two-electron excitations in a Coulomb field which utilize the SO(4) symmetry of hydrogenic systems[3] and the representation of the electron pair in hyperspherical coordinates.[4] This note complements the analysis of Ref. 2 mainly by articulating how SO(4) symmetry leads to two alternative couplings of orbital momenta and dipole moments, with and without a net helicity, which yield $\beta = -1$ and 2, respectively.

Briefly, photoabsorption raises H^- from its $^1S^e$ ground state to a $^1P^o$ doubly excited continuum state at an energy taken here to be slightly in excess of the threshold for dissociation into $H(n \geq 2) + e^-$. The photoelectron can escape through any superposition of $2n-1$ alternative channels characterized by orbital quantum numbers $l_1 \leq n-1$, $l_2 = l_1 \pm 1$, and $L = 1$, where the index 1 (or 2) refers to the inner (or outer) electron. However, these channels are strongly coupled, even at large r_2, by the dipole interaction $e^2\vec{r}_1 \cdot \hat{r}_2 / r_2^2$. Accordingly one considers instead the set of $2n-1$ decoupled eigenchannels which diagonalize the long-range centrifugal plus dipole potential[5]

$$(\vec{l}_2^{\,2} + 2\vec{r}_1 \cdot \hat{r}_2)/r_2^2 \,. \tag{1}$$

Different eigenvalues of this operator lead to different radial speeds in the dissociation of the negative ion, though these differences vanish as $r_2 \to \infty$. References 4 and 6 have independently shown that the dissociation to $H(n=2)$ proceeds near threshold almost entirely through a *single eigenchannel*, which for $^1P^0$ symmetry corresponds to the *second-lowest* eigenvalue of (1). This result has been verified experimentally[7] and extended to the $n=3$ threshold.[8] Herrick[3(a)] had previously shown that the lowest and second-lowest channels correspond to SO(4) quantum numbers $T=0$ and 1, respectively, for all n, to within the minor contribution of the $\vec{l}_2^{\,2}$ term of (1). The main points raised by Ref. 2 are (a) T represents the modulus of an eigenvalue of the pseudoscalar operator $\vec{L} \cdot \hat{r}_2$, where $\vec{L}$ is the orbital momentum of the $^1P^0$ state, and (b) $T=1$ (or 0) implies $\beta = -1$(or 2). The occurrence of crosswise photoemission is thus associated with the nonzero value of a pseudoscalar. This point will be amplified here by reviewing in some detail the significance of SO(4) symmetry and its implications for angular correlations.

Before going into details, we point out the blocks of our problem. The occurrence of a photoemission peak orthogonal to the incident polarization is the signature of interactions involving vector products or other screw-type mechanisms whose unraveling may hold rewards. That the channels of H^- photodetachment showing this phenomenon are most intense is made clear by the hyperspherical approach[4] and confirmed by other sources.[6,7] That these channels are labeled by nonzero values of a pseudoscalar parameter emerges from SO(4) symmetry analysis,[3,2] to be amplified below. That the +label of the hyperspherical approach and the SO(4) label $(K, T) = (n-2, 1)$ apply to the *same* channels of double excitation is borne out by em-

pirical evidence[3(b)] but the origin of this coincidence remains obscure and will not be discussed here.

The SO(4) symmetry holds for the motion of any body subject to a potential $\propto 1/r$. Central symmetry of this potential leads to invariance of the orbital momentum $\vec{l}$, its $1/r$ form leads to invariance of a second vector $\vec{b}$ ("Runge-Lenz" vector) parallel to the major axis of a classical orbit. Quantum mechanically $\vec{l}$ and $\vec{b}$ do not commute. In fact $\vec{b}$ transforms, e.g., a p eigenstate of an H atom into an s or d state with the same principal number n, being analogous to a dipole-moment operator in this respect. The six operators representing the components of $\vec{l}$ and $\vec{b}$ thus suffice to transform any H eigenstate with a given n into any other state of the same n manifold. In mathematical language, $\vec{l}$ and $\vec{b}$ thus constitute the "generators of an SO(4) algebra." Note particularly how the two operators l_z and b_z represent an infinitesimal rotation and a polarization, respectively, with the same z axis; accordingly they commute. By combining them into $\frac{1}{2}(l_z \pm b_z)$ one obtains screw-type operations of opposite helicity. Each set of screw operations $\frac{1}{2}(\vec{l} \pm \vec{b})$ forms a separate subgroup of SO(4), and each element of one of these sets commutes with all operators of opposite helicity. Within each of these sets the commutation rules are the same as for $\vec{l}$ alone, if $\vec{b}$ is suitably normalized, whereby $\frac{1}{4}|\vec{l} \pm \vec{b}|^2$ are two invariants with eigenvalues $j(j+1)$ and j integer or half-integer. A single electron in the nth level of H has $j = \frac{1}{2}(n-1)$ for both operators $\frac{1}{4}|\vec{l} \pm \vec{b}|^2$; this relationship requires $\vec{l} \cdot \vec{b}$ to vanish, as it does for a classical orbit.

[Note, incidentally, how screw-type operators occur also in analogous and fundamental contexts. The Lorentz group of transformations, SO(3, 1), also centers on six infinitesimal operators, three pseudovector rotations and three vector shifts to a moving frame. Screw-type combinations of these operators serve to construct a two-dimensional ("spinor") linear representation of the proper Lorentz group, shorn of the inversion of space coordinates. To include the inversion a second representation must be added, contragredient to the first one and with infinitesimal operators of opposite helicity. These circumstances presumably underlie the spontaneous emergence of the electron spin from Dirac's relativistic equation, even though the spin operator manifests itself through different steps as the vector product of two velocity operators.]

For a pair of *non interacting* electrons in a Coulomb field we set $\vec{L} = \vec{l}_1 + \vec{l}_2$ and $\vec{B} = \vec{b}_2 - \vec{b}_1$ as in Ref. 3. Here, as for a single electron, the squared operators $\frac{1}{4}|\vec{L} \pm \vec{B}|^2$ with eigenvalues $J_\pm(J_\pm + 1)$ remain invariant; the corresponding quantum numbers $J_\pm$ serve to classify levels. For two (or more) particles, however, these two quantum numbers need not be equal, which allows nonzero values of the pseudoscalar $\vec{L} \cdot \vec{B}$. Indeed the integer $T = |J_+ - J_-| = \vec{L} \cdot \hat{B}$, which measures the excess of one helicity over the opposite one, also indicates to what extent $\vec{L}$ is parallel to $\vec{B}$ rather than orthogonal to it, whereby the combinations $\vec{L} \pm \vec{B}$ represent a net helicity of the electron pair. We shall see that $T = 1$ implies $\beta = -1$ for $^1P^0$ photodetachment from H^-, while $T = 0$ leads to $\beta = 2$.

Upon introducing the electron interaction $1/r_{12}$, Ref. 3(b) has verified that its matrix elements off-diagonal in J_+ and/or J_- remain remarkably small. Reference 3(a) has traced the origin of this fact for the long-range dipole component of this interaction, represented by $\vec{r}_1 \cdot \hat{r}_2 / r_2^2$ in the operator (1), by pointing out that the matrix of $\vec{r}_1 \cdot \vec{r}_2$ is proportional to that of $\hat{b}_1 \cdot \hat{b}_2$ owing to a Wigner-Eckart theorem and hence is diagonal in J_+ and J_-; Ref. 2 rests on this argument.

On this basis consider now the correlation of the directions of electron ejection—i.e., of $\hat{r}_2$ as $r_2 \to \infty$—and of incident light polarization. The light polarization is orthogonal to its angular momentum $\vec{J}_T$ which coincides with the orbital momentum $\vec{L}$ of the excited H^-, since H^- was initially in $^1S_0$. Insofar as the excited H^- is classified by the SO(4) invariants $\frac{1}{4}|\vec{L} \pm \vec{B}|^2$, the direction of $\vec{L}$ is identified as $\frac{1}{2}(\vec{L} + \vec{B}) + \frac{1}{2}(\vec{L} - \vec{B})$. This direction is thus related to those of $\hat{r}_2$ or of $\vec{l}_2$ indirectly through a $9j$ recoupling coefficient

$$[(2l_1+1)(2l_2+1)(2J_+ + 1)(2J_- + 1)]^{1/2} \begin{Bmatrix} \frac{1}{2}(n_1-1) & \frac{1}{2}(n_2-1) & J_+ \\ \frac{1}{2}(n_1-1) & \frac{1}{2}(n_2-1) & J_- \\ l_1 & l_2 & L \end{Bmatrix}, \tag{2}$$

represented graphically in Fig. 1. Herrick has obtained a simple analytic form of this recoupling for the limit of very high excitation of electron No. 2,[3(a)] stressing that this limit also represents the relations relevant to electron detachment into the continuum just above the threshold.

Herrick's limit procedure may be illustrated with reference to Fig. 1 by noticing that for large

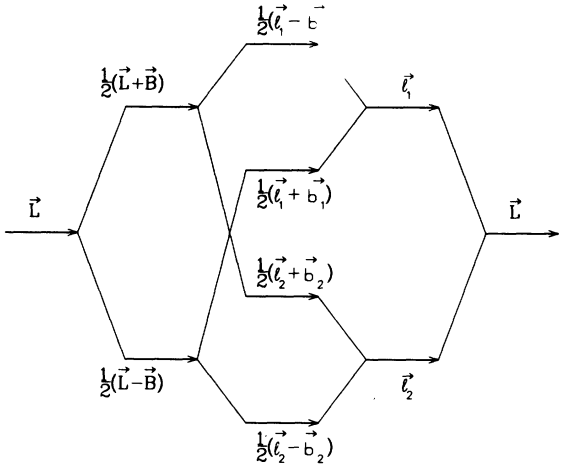

FIG. 1. Vector diagram of the recoupling transformation element Eq. (2).

values of the quantum number n_2, the Runge-Lenz vector $\vec{b}_2$ is very large indeed, equal to $n_2+O(1/n_2)$, and so are the vectors $\vec{L}\pm\vec{B}=\vec{L}\pm(\vec{b}_2-\vec{b}_1)$. Figure 2, which represents the several vectors in proportions approximating the case of large n_2, shows how the magnitude of $\vec{L}\pm\vec{B}$ departs from that of $\vec{b}_2$ primarily by the projection of other vectors onto $\vec{b}_2$. Residual departures are of the order of the obliquity of $\vec{L}\pm\vec{B}$ with respect to $\vec{b}_2$, i.e., of $O(1/n_2)$. Owing to this stretching of the vector diagram, most of the vector addition operations required to construct the $9j$ recoupling become trivial, namely, all those involving the addition of nearly parallel or antiparallel vectors. The relevant Wigner coefficients reduce then to unity, to $O(1/n_2)$, and to within normalization.

To implement these approximations we begin by evaluating the magnitude of the quantum numbers $J_\pm$ which represent the magnitudes $\frac{1}{2}|\vec{L}\pm\vec{B}|$ and serve to label the two-electron excitations. That is, we solve the equation

$$J_\pm(J_\pm+1)=\tfrac{1}{4}|\vec{L}\pm\vec{B}|^2, \tag{3}$$

discarding terms of $O(1/n_2)$. The result is

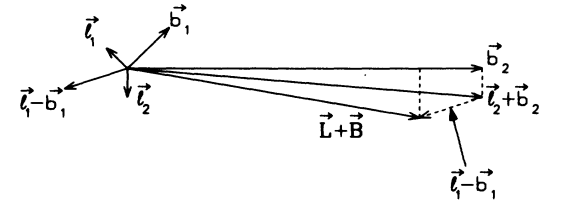

FIG. 2. Diagram showing how the component of $\vec{L}+\vec{B}$ on the axis $\hat{b}_2$ differs from b_2 by the projection of $\vec{l}_1-\vec{b}_1$ on the same axis, to within $O(1/n_2)$,

$$|\vec{L}+\vec{B}|\sim(\vec{L}+\vec{B})\cdot\hat{b}_2=b_2+(\vec{l}_1-\vec{b}_1)\cdot\hat{b}_2 .$$

$$J_\pm=\tfrac{1}{2}[n_2-1-\vec{b}_1\cdot\hat{b}_2\pm\vec{L}\cdot\hat{b}_2]+O(1/n_2)$$
$$=\tfrac{1}{2}(n_2-1+K\pm T)\ , \tag{4}$$

where the last expression serves to define the pair of quantum numbers (K,T) which replace $J_\pm$ in Ref. 3. Equation (4) complements Ref. 3 by interpreting K and T as $-\vec{b}_1\cdot\hat{b}_2$ and $\vec{L}\cdot\hat{b}_2$, respectively, to $O(1/n_2)$.

Returning now to the $9j$-recoupling diagram of Fig. 1, recall that it represents a set of products of six Wigner vector addition coefficients, one for each node of the diagram, to be summed over magnetic quantum numbers. Notice now that Eq. (4) permits us to interpret $\frac{1}{2}(T\mp K)$ as the magnetic quantum numbers of the diagram vectors $\frac{1}{2}(\vec{l}_1\pm\vec{b}_1)$ if the coordinate axis is laid along $\vec{b}_2$ (recalling that $\vec{l}_2\cdot\hat{b}_2=0$). Similarly $\pm\frac{1}{2}n_2$ represents the magnetic quantum number for the diagram vectors $\frac{1}{2}(\vec{l}_2\pm\vec{b}_2)$. All the magnetic quantum numbers are thus fixed for given values of n_2, K, and T, and the sum over products of Wigner coefficients reduces to a single term. Four of the six Wigner coefficients for the diagram of Fig. 1 reduce then to unity as noted above, namely, all those involving vectors of order $\vec{b}_2$. The entire $9j$ coefficient is finally represented by the residual product of two Wigner coefficients

$$(\tfrac{1}{2}(n_1-1)\tfrac{1}{2}(T-K),\tfrac{1}{2}(n_1-1)\tfrac{1}{2}(T+K)|\,l_1T)(l_1T,\,l_20|\,LT), \tag{5}$$

to within normalization and terms of $O(1/n_2)$. [The relevance of this equation is also subject to the condition that the first term of (1), $\vec{l}_2^{\,2}$, be small as compared to $\vec{r}_1\cdot\hat{r}_2$, as discussed in Ref. 2.]

The second factor of Eq. (5), pertaining to the addition $\vec{l}_1+\vec{l}_2=\vec{L}$, provides the core of the angular distribution law for electron ejection near threshold. Firstly, the zero value of the magnetic index following l_2 reminds us that $\vec{l}_2$ is orthogonal to the quantization axis $\hat{b}_2$. Recall also that $\vec{l}_2$ is orthogonal to the electron escape direction $\hat{r}_2$; $\hat{r}_2$ itself is strongly correlated to the Runge-Lenz vector $\vec{b}_2$ for a highly elongated escape orbit. The key element is the value of the magnetic index T pertaining to $\vec{L}$:

(1) For $T=0$, $\vec{L}$ is orthogonal to $\hat{b}_2$ as well as to the direction of incident polarization; hence the incident polarization, the escape $\hat{r}_2$ and the axis $\hat{b}_2$ may all be parallel and closely correlated. The same set of quantum numbers holds for the elementary photoionization of neutral H and leads to $\beta=2$.

(2) For $T=1$, $\vec{L}$ is parallel to the axis—since it has unit magnitude—and hence is orthogonal to $\vec{l}_2$. The escape direction $\hat{r}_2$ is thus correlated

with $\vec{L}$ rather than with the direction of polarization, which leads to $\beta = -1$.

Note finally that Eq. (5) includes also a connection with the alignment of the excited H residue of the photodetachment. This alignment, which manifests itself in the distribution and polarization of fluorescence, is represented by the parameter

$$3T^2 - l_1(l_1+1) \tag{6}$$

in the frame of the quantization axis. This parameter is positive for $T = 1$ and excitation to $n_1 = 2$, which restricts to $l_1 < 2$ and thus requires $\vec{l}_1$ to be parallel to $\hat{b}_2$, but higher excitations allow the direction of $\vec{l}_1$ to depart from $\hat{b}_2$ thus leading to a sign reversal of the mean alignment.

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