

Calculation of the initial density matrix for He I in beam-tilted-foil experiments

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We investigate the validity of the surface electric-field interaction model in beam-foil experiments where the polarization of the emitted light is measured. After summarizing the theory for expanding the density operator into state multipoles and linking these to the Stokes parameters of the light, we show also how to use symmetries to reduce the size of the calculations. A very simple model is chosen for the electric field: it is perpendicular to the foil and constant over a finite distance, then zero. This model is used to find the initial density matrix for the $n=3$ singlet states of He I and the electric-field parameters from tilted-foil experiments. Reasonable results are obtained given the unrealistic electric-field model used.

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I. INTRODUCTION

The first experimental evidence of a tilt angle dependence of the beam-foil interaction was given by Berry *et al.* [1]. Further experiments have confirmed their conclusion that foil surface effects are important [2]. A number of models have been presented [3–8], but none of them gives results that agree totally with experiment [9]. In this paper, we will examine more closely a model in which we assume that the initial state is a coherent superposition of states with different L and that the interaction is due to an electric field that is normal to the foil. In the original proposal by Band [10] and the generalization by Dehaes, Schenkel, and Devooght [11], a big problem was the limited validity of the second-order expansion that they used. Numerical calculations showed discrepancies for tilt angles of more than 10° [11].

To evaluate the validity of this model, we use it in this paper to compute the initial density matrix (at the exit of the foil) for the $n=3$ singlet states of He I (neutral helium), starting from the measured relative Stokes parameters for $2s\ ^1S-3p\ ^1P$ and $2p\ ^1P-3d\ ^1D$ transitions. At the same time, we will determine the parameters of the surface electric field.

In the calculation, we use the expansion of the density operator into a sum of unit spherical tensors (irreducible tensor operators). We present a quick summary of the theory to show how the symmetries are reflected in the density operator and to allow us to reduce the number of independent variables. We also show the relation between the Stokes parameters of the emitted light and the density operator. Then we find the relation between the initial density matrix and the Stokes parameters. This allows us finally to find the initial matrix density and the surface electric field in the case of the $n=3$ singlet states of He I.

II. SPHERICAL TENSORS AND THE DENSITY OPERATOR

Let us begin by the definition of the unit spherical tensors (also called state multipoles) [12].

$$T_q^k(J_1 L_1, J_2 L_2) = \sum_{M_1, M_2} (-1)^{J_1 - M_1} \sqrt{2k+1} \times \begin{pmatrix} J_1 & J_2 & k \\ M_1 & -M_2 & -q \end{pmatrix} |J_1 L_1 M_1\rangle \langle J_2 L_2 M_2|, \quad (1)$$

where the $|JLM\rangle$ are the eigenvectors of angular momentum (the other quantum numbers will not be written, they will always be understood to be the same). These are irreducible tensor operators, and a definition equivalent to Eq. (1) can be given using the Wigner-Eckart theorem (see Appendix A). The important point is that they satisfy the following orthogonality relation:

$$\text{Tr}[T_{q_1}^{k_1}(J_1 L_1, J_2 L_2) T_{q_2}^{k_2 \dagger}(J_3 L_3, J_4 L_4)] = \delta_{k_1 k_2} \delta_{q_1 q_2} \delta_{J_1 J_3} \delta_{L_1 L_3} \delta_{J_2 J_4} \delta_{L_2 L_4}, \quad (2)$$

where the dagger denotes the adjoint.

We can then expand the density operator ρ as a linear combination of these operators:

$$\rho = \sum_{J_1, L_1} \sum_{J_2, L_2} \sum_{k, q} \rho_q^k(J_1 L_1, J_2 L_2) T_q^{k \dagger}(J_1 L_1, J_2 L_2), \quad (3)$$

where

$$\begin{aligned}
\rho_q^k(J_1 L_1, J_2 L_2) &= \text{Tr}[\rho T_q^k(J_1 L_1, J_2 L_2)] \\
&= \sum_{M_1, M_2} (-1)^{J_1 - M_1} \sqrt{2k+1} \begin{Bmatrix} J_1 & J_2 & k \\ M_1 & -M_2 & -q \end{Bmatrix} \\
&\quad \times \langle J_2 L_2 M_2 | \rho | J_1 L_1 M_1 \rangle . \quad (4)
\end{aligned}$$

With the orthogonality of the $3j$, we can invert this relation:

$$\begin{aligned}
\langle J_2 L_2 M_2 | \rho | J_1 L_1 M_1 \rangle &= \sum_{k, q} (-1)^{J_1 - M_1} \sqrt{2k+1} \begin{Bmatrix} J_1 & J_2 & k \\ M_1 & -M_2 & -q \end{Bmatrix} \\
&\quad \times \rho_q^k(J_1 L_1, J_2 L_2) . \quad (5)
\end{aligned}$$

This shows that there is total equivalence between these two representations of the density operator ρ .

The time evolution of the density operator ρ is given by

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} (H\rho - \rho H^\dagger) , \quad (6)$$

$$\frac{d}{dt} \rho_q^k(J_1 L_1, J_2 L_2) = \frac{i}{\hbar} (E_{J_1 L_1}^* - E_{J_2 L_2}^*) \rho_q^k(J_1 L_1, J_2 L_2)$$

$$\begin{aligned}
&+ \frac{i}{\hbar} \sum_{k_1, q_1} \sum_{k_2, q_2} \sum_{J_3, L_3} \sqrt{(2k+1)(2k_1+1)(2k_2+1)} (-1)^{J_1+J_2+q} \begin{Bmatrix} k_1 & k_2 & k \\ -q_1 & -q_2 & q \end{Bmatrix} \\
&\quad \times \left[\begin{Bmatrix} k_2 & k_1 & k \\ J_1 & J_2 & J_3 \end{Bmatrix} V_{q_1}^{k_1}(J_1 L_1, J_3 L_3) \rho_{q_2}^{k_2}(J_3 L_3, J_2 L_2) \right. \\
&\quad \left. - (-1)^{k+k_1+k_2} \begin{Bmatrix} k_1 & k_2 & k \\ J_1 & J_2 & J_3 \end{Bmatrix} V_{q_1}^{k_1}(J_3 L_3, J_2 L_2) \rho_{q_2}^{k_2}(J_1 L_1, J_3 L_3) \right] , \quad (9)
\end{aligned}$$

where the tensorial components of the potential $V(t)$ are defined in the same way as for the density operator:

$$V_q^k(J_1 L_1, J_2 L_2) = \text{Tr}[V(t) T_q^k(J_1 L_1, J_2 L_2)] . \quad (10)$$

In Appendix B, we give the general method for obtaining the system of equations for the evolution of the coefficients of the expansion of ρ over any basis of operators.

III. EFFECTS OF SYMMETRIES

First, the fact that ρ is Hermitian give us

$$\rho_q^k(J_1 L_1, J_2 L_2) = (-1)^{J_1 - J_2 + q} \rho_{-q}^{k*}(J_2 L_2, J_1 L_1) , \quad (11)$$

which means that not all the tensorial components are independent variables. If there are some other symmetries, the number of independent variables will be reduced.

In Appendix C, we show how to find the relations between the coefficients in an expansion of ρ due to symmetries of the problem. Here these coefficients are the tensorial components (or state multipoles) and we will look at two symmetries that will be useful in the case of beam-foil interactions: cylindrical symmetry and

in the general case of a non-self-adjoint Hamiltonian H . If H is self-adjoint, we get the usual Fano-Liouville equation. If we want to take into account the population decay through transitions to other levels, we have to use a non-self-adjoint Hamiltonian. We will assume a Hamiltonian of the following form:

$$H = H_0 - i\frac{\Gamma}{2} + V(t) , \quad (7)$$

where H_0 is the Hamiltonian of the free atom, Γ accounts for the finite lifetime, and $V(t)$ is a time-dependent perturbation. If we take H_0 and Γ to be diagonal in the $|JLM\rangle$ basis, i.e.,

$$\begin{aligned}
\left[H_0 - i\frac{\Gamma}{2} \right] |JLM\rangle &= \left[E_{JL} - i\frac{\Gamma_{JL}}{2} \right] |JLM\rangle \\
&= E'_{JL} |JLM\rangle , \quad (8)
\end{aligned}$$

then, using Eqs. (2), (3), (7), and (8) in Eq. (6), we can write the equation for the time evolution of the components of the expansion of the density operator into unit spherical tensors as

reflection symmetry.

The first case is cylindrical symmetry. Indeed, even if the foil is tilted, the beam coming out of the foil (before it is modified by the surface interaction) should be invariant for any rotation around its axis (which we take to be the z axis). Thus the symmetry operations are rotations of axis z with an arbitrary angle α :

$$S_\alpha \equiv R_z(\alpha) = e^{-i\alpha J_z} , \quad (12)$$

where J_z is the z component of the angular momentum operator. We find then that Eq. (C3) gives us

$$g_{J_1 L_1 J_2 L_2 k q, J_3, L_3, J_4 L_4, l r}^\alpha = e^{-i\alpha q} \delta_{J_1 J_3} \delta_{L_1 L_3} \delta_{J_2 J_4} \delta_{L_2 L_4} \delta_{kl} \delta_{qr} . \quad (13)$$

And by using this in Eq. (C5), we see that cylindrical symmetry implies

$$\rho_q^k(J_1 L_1, J_2 L_2) = \delta_{q0} \rho_0^k(J_1 L_1, J_2 L_2) . \quad (14)$$

Thus all tensorial components with $q \neq 0$ are zero.

The other case is reflection symmetry with respect to the x - z plane. The symmetry group has just two ele-

ments: the reflection operator and the identity. The reflection operator can be defined as

$$S_y = S_0 R_y(\pi) . \quad (15)$$

where S_0 is the space inversion operator (parity operator) and $R_y(\pi)$ is the rotation of an angle π around the y axis. To find the g coefficient, we need the following results [13].

$$S_0 |JLM\rangle = (-1)^L |JLM\rangle , \quad (16)$$

$$\begin{aligned} \langle J_1 L_1 M_1 | R_y(\pi) | J_2 L_2 M_2 \rangle \\ = (-1)^{M_1 - M_2} \langle J_1 L_1 M_1 | R_y(-\pi) | J_2 L_2 M_2 \rangle \\ = (-1)^{J_1 + M_1} \delta_{J_1 J_2} \delta_{L_1 L_2} \delta_{M_1, -M_2} . \end{aligned}$$

They allow us to find

$$\begin{aligned} g_{J_1 L_1 J_2 L_2}^{J_3 L_3 J_4 L_4} \\ = (-1)^{L_1 + L_2 + l + r} \delta_{J_1 J_3} \delta_{L_1 L_3} \delta_{J_2 J_4} \delta_{L_2 L_4} \delta_{kl} \delta_{q, -r} , \end{aligned} \quad (17)$$

and we get for the tensorial components

$$\rho_q^k(J_1 L_1, J_2 L_2) = (-1)^{L_1 + L_2 + k + q} \rho_{-q}^k(J_1 L_1, J_2 L_2) . \quad (18)$$

Thus it gives a relation between the tensorial components with $q < 0$ and the ones with $q > 0$. It also means that some components with $q = 0$ are zero (depending on L_1 , L_2 , and k).

IV. CONSTRAINTS ON THE DENSITY MATRIX

If we want to have a well-defined ρ , we need to choose a normalization. We will take $\text{Tr}(\rho) = 1$, which gives us

$$\begin{aligned} & \left| \sum_{k,q} (-1)^{J_2 - M_2} \sqrt{2k+1} \begin{bmatrix} J_2 & J_1 & k \\ M_2 & -M_1 & -q \end{bmatrix} \rho_q^k(J_2 L_2, J_1 L_1) \right|^2 \\ & \leq \left[\sum_k (-1)^{J_1 - M_1} \sqrt{2k+1} \begin{bmatrix} J_1 & J_1 & k \\ M_1 & -M_1 & 0 \end{bmatrix} \rho_0^k(J_1 L_1, J_1 L_1) \right] \\ & \quad \times \left[\sum_k (-1)^{J_2 - M_2} \sqrt{2k+1} \begin{bmatrix} J_2 & J_2 & k \\ M_2 & -M_2 & 0 \end{bmatrix} \rho_0^k(J_2 L_2, J_2 L_2) \right] . \end{aligned} \quad (24)$$

This is a nonlinear constraint on the tensorial components of the density operator.

V. DENSITY MATRIX AND THE STOKES PARAMETERS

In the experiments, we never have a direct access to the density matrix. The only information we have is the emitted light and its polarization. This emitted light is totally described by its Stokes parameters [12]. Thus, we

(by using the formulas of Appendix A)

$$\sum_{JL} \sqrt{2J+1} \rho_0^0(JL, JL) = 1 . \quad (19)$$

With this normalization, all the diagonal elements of the density matrix are probabilities (i.e., all positive) and their sum is equal to 1. Since Eq. (4) gives us

$$\rho_0^0(JL, JL) = \sum_M \frac{1}{\sqrt{2J+1}} \langle JLM | \rho | JLM \rangle , \quad (20)$$

and we know that the sum over M will be smaller than 1, we get

$$0 \leq \sqrt{2J+1} \rho_0^0(JL, JL) \leq 1 . \quad (21)$$

And since the diagonal elements are probabilities, they are non-negative and smaller than 1, which gives, with Eq. (5).

$$0 \leq \sum_k (-1)^{J-M} \sqrt{2k+1} \begin{bmatrix} J & J & k \\ M & -M & 0 \end{bmatrix} \rho_0^k(JL, JL) \leq 1 . \quad (22)$$

We have thus one equality constraint given by Eq. (19) and four linear inequalities given by (21) and (22).

There is one more constraint on the elements of the density matrix, due to the fact that ρ is Hermitian and positive definite. Indeed, we can write $\rho = \rho^{1/2} \rho^{1/2}$ and the Schwarz inequality gives us

$$\begin{aligned} & |\langle J_1 L_1 M_1 | \rho | J_2 L_2 M_2 \rangle|^2 \\ & \leq \langle J_1 L_1 M_1 | \rho | J_1 L_1 M_1 \rangle \langle J_2 L_2 M_2 | \rho | J_2 L_2 M_2 \rangle , \end{aligned} \quad (23)$$

and by using Eq. (5), we get

have to establish the relation between the density-matrix elements and the Stokes parameters.

The intensity of the light emitted in a given direction with a polarization vector \mathbf{e}_p is given by

$$I(\mathbf{e}_p, t) = I_0 \text{Tr}[\rho(t) D(\mathbf{e}_p)] , \quad (25)$$

where I_0 is a constant depending on the measurement system and on the number of ions going through the foil

(i.e., on the beam current),

$$D(\mathbf{e}_p) = \sum_f (\mathbf{e}_p \cdot \mathbf{d}) |f\rangle \langle f| (\mathbf{e}_p \cdot \mathbf{d})^\dagger, \quad (26)$$

$$I = I_0 \sum_{J_0, J_1, J_2} \sum_{L_0, L_1, L_2} \sum_{k, q} \sum_{q_1, q_2} (-1)^{J_0 + J_1 + q_2} \sqrt{2k+1} e_{q_1} e_{q_2}^* \begin{Bmatrix} 1 & 1 & k \\ q_2 & -q_1 & -q \end{Bmatrix} \begin{Bmatrix} 1 & 1 & k \\ J_1 & J_2 & J_0 \end{Bmatrix} \\ \times \langle nJ_1L_1 || d || n_0J_0L_0 \rangle \langle nJ_2L_2 || d || n_0J_0L_0 \rangle^* \rho_q^k(J_1L_1, J_2L_2), \quad (27)$$

where the e_q are the tensorial components of the polarization vector \mathbf{e}_p , with the basis defined as follows:

$$|0\rangle = |z\rangle, \\ |\pm 1\rangle = \mp \frac{1}{\sqrt{2}} (|x\rangle \pm i|y\rangle). \quad (28)$$

Now, when working in the LS scheme, if an operator A_q^k does not act on the spin, we can write [13]

$$\langle J_1L_1 || A^k || J_2L_2 \rangle \\ = (-1)^{J_2 + L_1 + S + k} \sqrt{(2J_1 + 1)(2J_2 + 1)} \\ \times \begin{Bmatrix} L_1 & k & L_2 \\ J_2 & S & J_1 \end{Bmatrix} \langle L_1 || A^k || L_2 \rangle. \quad (29)$$

In Eq. (27), the electric dipole operator \mathbf{d} does not depend on the spin and we can thus use Eq. (29). This allows us to write

$$I(\mathbf{e}_p, t) = I_0 \sum_{k, q} l_q^k(\mathbf{e}_p) B_q^k(t), \quad (30)$$

where

$$B_q^k = \sum_{J_1L_1} \sum_{J_2L_2} A^{(k)}(J_1L_1, J_2L_2) C^{(k)}(L_1, L_2) \\ \times \rho_q^k(J_1L_1, J_2L_2). \quad (31)$$

In these equations, the l_q^k are the components of an irreducible tensor that accounts for the geometry of the observation:

$$l_q^k = \sum_{q_1, q_2 = -1}^{+1} (-1)^{q_2 + 1} e_{q_1} e_{q_2}^* \begin{Bmatrix} 1 & 1 & k \\ q_1 & -q_2 & q \end{Bmatrix} \\ \times \sqrt{2k+1}, \quad (32)$$

and the $3j$ coefficient implies that $k \leq 2$ for a nonzero result. The $A^{(k)}$ account for the spin effects:

$$A^{(k)}(J_1L_1, J_2L_2) \\ = (-1)^{J_2 + L_1 + S + k} \sqrt{(2J_1 + 1)(2J_2 + 1)} \begin{Bmatrix} L_1 & k & L_2 \\ J_2 & S & J_1 \end{Bmatrix}, \quad (33)$$

and the $C^{(k)}$ account for the quantum characteristics of the system under observation:

\mathbf{d} is the electrical dipole operator, and the sum is over all possible final states $|f\rangle$ for the observed transition. Now, if we introduce the definition (3) for ρ in Eq. (25) and if we use the Wigner-Eckart theorem, we obtain

$$C^{(k)}(L_1, L_2) = \sum_{L_0} (-1)^{L_0 + L_1 + 1} \begin{Bmatrix} 1 & 1 & k \\ L_1 & L_2 & L_0 \end{Bmatrix} \\ \times \langle nL_1 || d || n_0L_0 \rangle \langle nL_2 || d || n_0L_0 \rangle^*. \quad (34)$$

Knowing that the electric dipole operator (for one electron) is given by

$$\mathbf{d} = -|e|\mathbf{r}, \quad (35)$$

we can then compute the reduced matrix elements either from theory (for hydrogenlike wave functions) or from the experimentally observed transition rates R_{fi} , since [14]

$$R_{fi} = \frac{4}{9} \frac{e^2}{4\pi\epsilon_0} \frac{\omega^3}{\hbar c^3} |\langle n_iL_i || r || n_fL_f \rangle|^2. \quad (36)$$

Now, Fig. 1 shows the two coordinate systems that we will use in this paper. The beam coordinate system has its z axis along the beam, its origin at the point where the center of the beam exits the foil, and the y axis is in the foil along the rotation axis for tilting the foil. The x axis is chosen to have a right-handed system of coordinates. The foil coordinate system has the same origin and its y' axis is the same as the y axis, but the z' axis is in the direction normal to the foil. We can thus go from one coordinate system to the other by a rotation by an angle β (the tilt angle of the foil) around the y axis. We will al-

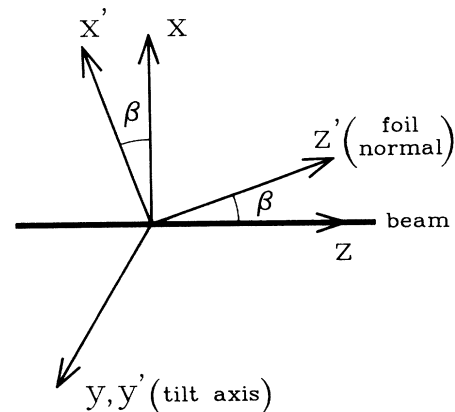


FIG. 1. Definition of the coordinate systems. The surface of the foil is in the x' - y' plane.

ways use the beam coordinate system for the observations.

In the beam coordinate system, if we look at the light emitted in the y direction, we obtain for the Stokes parameters (using the definitions in Ref. [12])

$$\begin{aligned} I &= \frac{2}{\sqrt{3}} B_0^0 - \frac{1}{\sqrt{6}} B_0^2 - \frac{1}{2} (B_2^2 + B_{-2}^2), \\ M &= -\frac{3}{\sqrt{6}} B_0^2 + \frac{1}{2} (B_2^2 + B_{-2}^2), \\ C &= B_1^2 - B_{-1}^2, \\ S &= -i(B_1^1 + B_{-1}^1). \end{aligned} \quad (37)$$

If we observe the light emitted in the x direction, we get instead

$$\begin{aligned} I_x &= \frac{2}{\sqrt{3}} B_0^0 - \frac{1}{\sqrt{6}} B_0^2 + \frac{1}{2} (B_2^2 + B_{-2}^2), \\ M_x &= -\frac{3}{\sqrt{6}} B_0^2 - \frac{1}{2} (B_2^2 + B_{-2}^2), \\ C_x &= -i(B_1^2 + B_{-1}^2), \\ S_x &= B_1^1 - B_{-1}^1 \end{aligned} \quad (38)$$

(but C_x and S_x will always be zero by reflection symmetry).

VI. INTERACTION POTENTIAL

In the surface electric-field model, we assume that there exists a potential $V'(d)$ that depends only on the distance d from the foil (measured along the axis normal to the foil). If \mathbf{r} is the position of the electron relative to the center of mass of the atom, it is affected by a potential

$$\begin{aligned} V(d, \mathbf{r}) &= V'(d + \mathbf{r} \cdot \mathbf{n}) - V'(d), \\ &\approx |e|(\mathbf{r} \cdot \mathbf{n})E(d), \end{aligned} \quad (39)$$

where e is the electron charge, \mathbf{n} is a unit vector along the axis normal to the foil, and $E(d)$ is the electric field normal to the foil at a distance d from the foil. We will use this first-order approximation since r is usually small compared to the distances over which there is a significant change of the electric field. In the foil coordinate system, we get [using Eq. (10)]

$$V_q^k(L_1, L_2) = \frac{(-1)^{L_1 - L_2}}{\sqrt{3}} \langle L_2 || r || L_1 \rangle \delta_{q,0} \delta_{k,1} |e| E(d), \quad (40)$$

which means that only the V_0^1 tensorial components can be nonzero. (That only, the $q=0$ components would be different from zero was to be expected because of the cylindrical symmetry of the potential.) This means that if we compute the evolution of the density operator in the foil coordinate system, we will have a decoupling in q of the system of equations (9) for the ρ_q^k .

We need again to compute the reduced matrix element of \mathbf{r} between two angular momentum eigenvectors, but this time they have the same principal quantum number n . We can write

$$\begin{aligned} \langle L_2 || r || L_1 \rangle &= \sqrt{4\pi/3} \langle L_1 || Y^1 || L_1 \rangle \\ &\times \int_0^\infty R_{n,L_2}^*(r) r R_{n,L_1}(r) r^2 dr. \end{aligned} \quad (41)$$

But the angular part is nonzero if and only if $|L_1 - L_2| = 1$, which gives us [13]

$$\begin{aligned} \langle L || r || L-1 \rangle &= -\langle L-1 || r || L \rangle \\ &= \sqrt{L} \mathcal{R}_n(L, L-1), \end{aligned} \quad (42)$$

where \mathcal{R}_n is the radial integral part of Eq. (41) (the phase convention is that of Messiah [13]). For hydrogenlike wave functions, we get [15]

$$\mathcal{R}_n(L, L-1) = \frac{3}{2} n (n^2 - L^2)^{1/2} \frac{a_0}{Z}, \quad (43)$$

where the convention is that the hydrogenic wave functions are positive as $r \rightarrow 0$. In the general case, we can use the same method as for the matrix elements for the transitions: we obtain the reduced matrix element from the transition rate with Eq. (37).

In this paper, we will use a very simple model for the surface potential. We will assume that it is linear, which has the advantage for the computer calculations of giving a constant electric field (and thus a constant V_0^1). We can write

$$E(d) = \begin{cases} E_0 & \text{if } d < d_0 \\ 0 & \text{if } d \geq d_0, \end{cases} \quad (44)$$

and $d = vt \cos\beta$, where v is the velocity of the ions and β is the tilt angle of the foil.

VII. INITIAL DENSITY-MATRIX CALCULATION

The initial density matrix (at time zero) describes the state of the beam as it exits the foil. At that point, it has cylindrical symmetry in the beam coordinate system. Then it undergoes some evolution in the surface electric field and the emitted light is observed. In the case of helium, once the atom is past the region with a strong electric field, there is no more observable evolution of the emitted light (except for an exponential decay) because the transitions are observed separately. In the case of hydrogen, the transitions are not resolved and quantum beats can be observed as we observe the light further and further from the foil.

If the foil is tilted and we want to conserve the advantages of the cylindrical symmetry of the potential, we have to compute the evolution of the density matrix in the foil coordinate system. To pass from the beam coordinate system to the foil coordinate system, we need to rotate it about the y axis by an angle β . The tensorial components of ρ transform then as follows:

$$\rho_q^k(\text{foil}) = \sum_{q_1 = -k}^k \rho_{q_1}^k(\text{beam}) r_{q_1, q}^k(\beta), \quad (45)$$

where the $r_{q_1, q}^k$ are the rotation matrix elements [13]; and to go back to the beam coordinate system to compute the Stokes parameters, we just apply a rotation of $-\beta$.

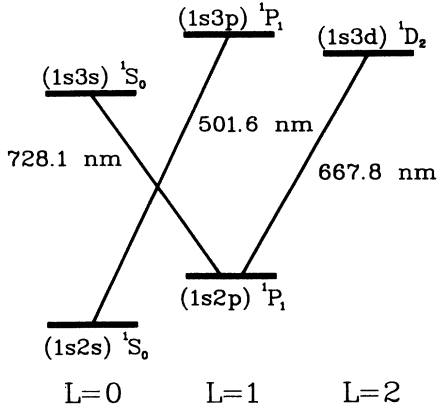


FIG. 2. Schematic representation of the energy levels of the $n=2$ and 3 singlet levels of He I. The wavelength of the transition radiation is indicated for each transition.

We can now summarize the link between the Stokes parameters and the initial density matrix $\rho(0)$ as

$$\begin{pmatrix} I \\ M \\ C \\ S \end{pmatrix} = I_0 \mathcal{O}(\lambda) R(-\beta) U(E_0, d_0) R(\beta) \rho(0), \quad (46)$$

where $R(\beta)$ is the rotation defined by Eq. (45), $I(E_0, d_0)$ is the evolution operator for the system of equations (9) with the potential defined by Eqs. (40) and (44), and $\mathcal{O}(\lambda)$ is the observation operator for the transition at a wavelength λ given by Eq. (37). As we mentioned above, the evolution operator U needs only to be computed for the region with a nonzero electric field since all the subsequent evolution is unobservable (except for the exponential decay).

In Eq. (46), there is one important implicit assumption: that all the atoms contributing the observed transitions had already the same main quantum number n at the exit of the foil. This means that we neglect any contribution to the population coming from transitions from higher-energy levels with a different n .

In this paper, the goal will be to find the initial density matrix and the parameters E_0 and d_0 that define the surface potential. To do that, we are given the relative Stokes parameters M/I , C/I , and S/I for several transitions and several tilt angles β . This means that the I_0 factor, which depends on the measurement system, disap-

pears. On the other hand, the relation between the measurements and the initial density matrix is not linear, as it would be if we were given I , M , C , and S separately.

For the case of the He I $n=3$ singlet states we will observe the $2s\ ^1S-3p\ ^1P$ transition at 501.6 nm and the $2p\ ^1P-3d\ ^1D$ transition at 667.8 nm. The third transition from the singlet states $2p\ ^1P-3s\ ^1S$ at 728.1 nm, cannot give us any more information since the emitted light is not polarized (because the 1S state has spherical symmetry). Figure 2 gives a schematic representation of the energy levels and Table I gives the exact energies and lifetimes.

VIII. SOLUTION METHOD

From the results of the preceding section, we can see that for each tilt angle, we have six measurements: M/I , C/I , and S/I for each of the two observed transitions. We can put all these measurements for all the angles in a vector \mathbf{M} ; and from Eq. (46), for each of these measurements, we can write a function of $\rho(0)$, E_0 , and d_0 that should be equal to the measured value. We will call the vector of these functions \mathbf{F} . To find the initial density matrix and the electric-field parameters, we then have to solve the following constrained least-squares problem:

$$\min_{\rho(0), E_0, d_0} \sum_i \frac{1}{\sigma_i^2} [M_i - F_i(E_0, d_0, \rho(0))]^2, \quad (47)$$

with the constraints given in Sec. IV. The σ_i 's are the standard deviations on the measured values.

If the principle is simple, the number of variables involved makes it much more difficult. For example, if we take the $n=3$ singlet states of He I, that is, $S=0$ and $L=0, 1, 2$ for $\rho_q^k(L_1, L_2)$, we get k going from $|L_1 - L_2|$ to $L_1 + L_2$ for each combination of L_1 and L_2 , and each q going from $-k$ to k . This gives us a total of 81 complex variables, or 162 real variables (the real and imaginary parts each being considered as a real variable). But if we use the symmetries of Sec. III, we can substantially reduce the number of variables; for example, the Hermiticity of ρ , expressed through Eq. (11), cuts by half the number of real independent variables. Then, the reflection symmetry condition (18) links all the components with $q < 0$ to those with $q > 0$ and sets some $q=0$ components equal to zero. Together with the Hermiticity, this reduces the number of independent real variables to 45: 14 real independent variables for $q=0$, 16 for $q=1$, 10 for $q=2$, and 1 for $q=4$. And since the system of equations [Eq. (9)] is decoupled in q for the potential we have chosen, these means that we can reduce the problem of the calculation of the evolution operator to the solution of five independent systems of differential equations of sizes 14, 16, 10, 4, and 1, respectively. Now, for the initial density matrix $\rho(0)$, we are assuming cylindrical symmetry, which means by Eq. (14) that only the components with $q=0$ are nonzero. This leaves us with 14 independent real variables to totally determine $\rho(0)$. In Appendix D, we describe the method that was used to obtain systematically the reduced systems of equations.

When we say independent variables, we have to be careful, because this does not yet take into account the

TABLE I. Energy levels of He I singlet states.

n	L	Energy (m^{-1})	Lifetime \hbar/Γ (ns)
2	0	16 627 170	∞
2	1	17 112 915	0.5558
3	0	18 485 906	55.25
3	1	18 620 362	74.74
3	2	18 609 922	15.67

constraints. The normalization of the trace to 1 through Eq. (19) reduces the number of independent variables, but that relation is not easy to introduce, and we have decided not to use it to reduce the number of variables but to impose it with the other constraints of Sec. IV during the minimization procedure.

The minimization itself consisted of two nested minimization procedures. The outer one finds the minimum with respect to E_0 and d_0 by finding a zero of the gradient of the function in Eq. (47), i.e., by solving the following system of equations:

$$\begin{aligned} \sum_i \frac{1}{\sigma_i^2} (M_i - F_i) \frac{\partial F_i}{\partial E_0} &= 0, \\ \sum_i \frac{1}{\sigma_i^2} (M_i - F_i) \frac{\partial F_i}{\partial d_0} &= 0, \end{aligned} \quad (48)$$

with a method inspired by the Newton-Raphson method (we work with the tangent planes). That is, we take a first-order expansion of \mathbf{F} :

$$\mathbf{F}(E + \Delta E, d + \Delta d) = \mathbf{F}(E, d) + \Delta E \frac{\partial \mathbf{F}}{\partial E} + \Delta d \frac{\partial \mathbf{F}}{\partial d}. \quad (49)$$

where E and d are the current values of the parameters and the ΔE and Δd are what we add to them to get the next values. Introducing this into Eq. (48), we get for each step the following system of equations:

$$\begin{aligned} \left[\sum_i \frac{1}{\sigma_i^2} \frac{\partial F_i}{\partial E} \frac{\partial F_i}{\partial E} \right] \Delta E + \left[\sum_i \frac{1}{\sigma_i^2} \frac{\partial F_i}{\partial E} \frac{\partial F_i}{\partial d} \right] \Delta d \\ = \sum_i \frac{1}{\sigma_i^2} \left[(M_i - F_i) \frac{\partial F_i}{\partial E} \right], \\ \left[\sum_i \frac{1}{\sigma_i^2} \frac{\partial F_i}{\partial E} \frac{\partial F_i}{\partial d} \right] \Delta E + \left[\sum_i \frac{1}{\sigma_i^2} \frac{\partial F_i}{\partial d} \frac{\partial F_i}{\partial d} \right] \Delta d \\ = \sum_i \frac{1}{\sigma_i^2} \left[(M_i - F_i) \frac{\partial F_i}{\partial d} \right]. \end{aligned} \quad (50)$$

And at each step of this outer iteration, before solving this systems of equations, we find the minimum of the function in Eq. (47) with respect to $\rho(0)$, with the constraints on $\rho(0)$ respected. This was done with a general minimization routine from the NAG Fortran library.

The advantage of this procedure is that we need to compute the evolution operator only once each time we minimize with respect to $\rho(0)$. For each step of the outer iteration, we need to compute it twice more (when we evaluate numerically the derivatives of \mathbf{F} with respect to E and d). With the model we use in this paper (constant electric field), the equations, have constant coefficients and the evolution operator have been computed by taking the exponential of the matrix times $d_0/(v \cos \beta)$ (the interaction time). This is probably not the most efficient method, especially if the electric field is not constant.

IX. RESULTS FOR He I

As we have already mentioned above, in this paper our main interest is the singlet ($S=0$) states of the $n=3$ level of He I (neutral helium). We assume that there is no mixing with the $S=1$ triplet states. Because $S=0$, we have $J=L$ and some of the formulas obtained above can be somewhat simplified. Because the transitions are between states with fixed L , the $C^{(k)}(L_1, L_2)$ coefficient defined by Eq. (34) becomes

$$\begin{aligned} C^{(k)}(L_i, L_i) &= (-1)^{L_i + L_f + 1} \begin{Bmatrix} 1 & 1 & k \\ L_i & L_i & L_f \end{Bmatrix} \\ &\times |\langle n_i L_i || d || n_f L_f \rangle|^2, \end{aligned} \quad (51)$$

where L_i and L_f are the initial and final L 's for the observed transition, respectively. And the $A^{(k)}$ coefficient defined by Eq. (33) is then equal to 1. We can compute the reduced matrix elements using the transition rates from the tables [14,16] and Eq. (36). The results are nearly the same as for hydrogen. Moreover, since there is only one reduced matrix element coming in for each transition, it will cancel out when we compute the relative Stokes parameter ($M/I, C/I, S/I$). Thus, we do not need to compute them.

For the potential, we need the reduced matrix elements of r between the $3p$ and $3s$ states and between $3d$ and $3p$ states. From the tables [16] and Eq. (36), we get

$$\begin{aligned} \langle 3p || r || 3s \rangle &= 6.566 \times 10^{-10} \text{ m} \\ (\lambda &= 7435.1 \text{ nm}, R_{s,p} = 2.53 \times 10^5 \text{ s}^{-1}), \\ \langle 3d || r || 3p \rangle &= 7.821 \times 10^{-10} \text{ m} \\ (\lambda &= 95760 \text{ nm}, R_{p,d} = 168 \text{ s}^{-1}), \end{aligned} \quad (52)$$

which differs only by a few percent from the results obtained with Eq. (43) for hydrogen (i.e., with $Z=1$). This is understandable since the helium nucleus with an electron in a $1s$ state looks very much like a hydrogen nucleus from a certain distance.

The measurements we used are those from Brooks and Pinnington [17,18]. They used helium ions at 160 keV, through a carbon foil with $5 \mu\text{g}/\text{cm}^2$. Since this is a low energy, we can use the classical formula to compute the

TABLE II. Independent tensorial components of the initial density matrix.

k	L_1	L_2	$\rho_0^k(L_1, L_2)$
0	0	0	0.7014
1	1	0	-0.1268 + i 0.0534
0	1	1	0.1618
2	1	1	-0.0264
2	2	0	-0.042 + i 0.0274
1	2	1	-0.006 - i 0.0095
3	2	1	0.0112 - i 0.0262
0	2	2	0.0082
2	2	2	-5.6 $\times 10^{-6}$
4	2	2	-1.2 $\times 10^{-4}$

TABLE III. Initial density matrix elements (upper triangle).

L_1	M_1	L_2	M_2	$\langle L_1 M_1 \rho L_2 M_2 \rangle$
0	0	0	0	0.7014
0	0	1	0	$-0.1268 + i0.0534$
0	0	2	0	$-0.042 + i0.0274$
1	-1	1	-1	0.0826
1	-1	2	-1	$0.0047 - i0.0169$
1	0	1	0	0.115
1	0	2	0	$-0.009 + i0.0142$
1	1	1	1	0.0826
1	1	2	1	$0.0047 - i0.0169$
2	-2	2	-2	0.0036
2	-1	2	-1	0.0037
2	0	2	0	0.0036
2	1	2	1	0.0037
2	2	2	2	0.0036

speed: $E = mv^2/2$ (the energy loss in the foil can be neglected). Since the mass of helium is 4.003 amu, we get a speed of 2.777×10^6 m/s. The measurements were made for tilt angles from 0° to 70° , by steps of 10° . Thus, we have eight sets of six measurements (two transitions, three Stokes parameters for each), which gives us 48 elements for \mathbf{M} . As mentioned above, we have 14 unknowns for $\rho(0)$ and two unknown parameters for the electric field (E_0 and d_0), thus a total of 16 parameters for the least-squares adjustment. We also have one equality constraint (trace normalization). If we assume that the measurement errors are normally distributed, we see that the function in Eq. (47) should have a χ^2 distribution with $48 - 16 - 1 = 31$ degrees of freedom (if we neglect all the inequality constraints that would reach equality).

The 14 independent tensorial components of the den-

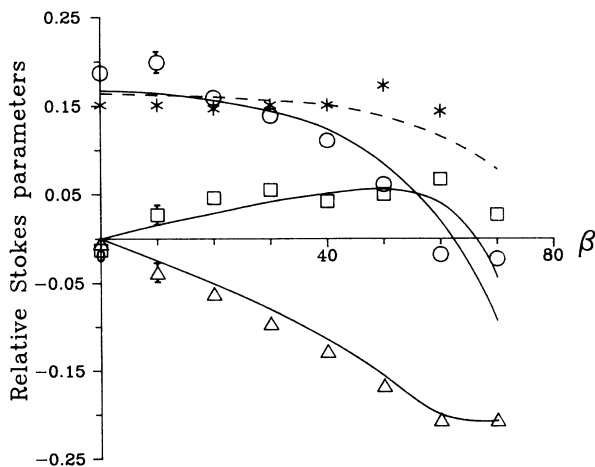


FIG. 3. Measured relative Stokes parameters M/I (\circ), C/I (\square), and S/I (\triangle) with their typical error bars [17] for the $2s\ ^1S-3p\ ^1P$ transitions. The solid curves show the same parameters calculated with the best fit of the initial density matrix and electric-field parameters. They are plotted as a function of the foil tilt angle β . The measured [19] M_x/I_x (*) for 501.6 nm (not used in the fit) are compared to the calculated one (dashed curve).

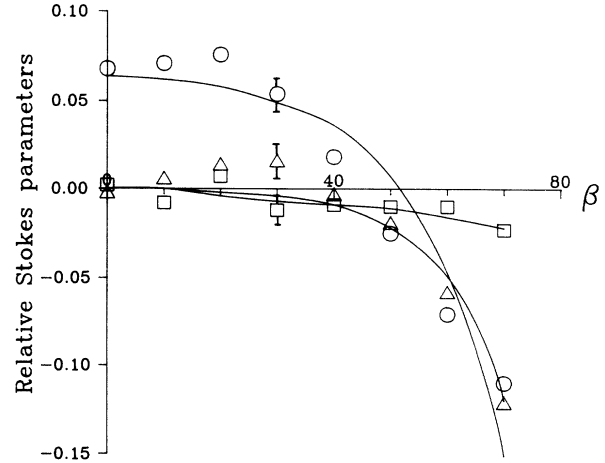


FIG. 4. Measured relative Stokes parameters M/I (\circ), C/I (\square), and S/I (\triangle) with their typical error bars [17] for the $2p\ ^1P-3d\ ^1D$ transitions and the same parameters calculated with the best fit of the initial density matrix and electric-field parameters. They are plotted as a function of the foil tilt angle β .

ty operator calculated by the program are given in Table II and the upper triangular part of the corresponding density matrix is given in Table III. In brief, it says that about 70% of the population is in the 1S state, 28% in the 1P state, and 2% in the 1D state. The $S-P$ coherence is weak and the $S-D$ and $P-D$ coherences are maximum. But given the low population of the 1D states, the measurement errors, and the simple model used, the coherences with the D states probably have no meaning. It is not a pure state, since $\text{Tr}(\rho^2) = 0.5636$. The electric-field parameters found are

$$E_0 = 6.2 \times 10^7 \text{ V/m}, \quad d_0 = 8.46 \text{ nm}, \quad (53)$$

which gives a total electrical potential of 0.525 V. The residue of Eq. (47) was 291.5, which is well beyond the value of 55 that corresponds to the 0.5% probability of rejecting a true assumption for a χ^2_{31} distribution. One should, however, notice that this was obtained with the σ_i given by Brooks and Pinnington [17], which may be underestimated, especially for the large angles. A factor of 2 on the σ_i 's would already bring the residue down to 72.8. Several trials showed that the residue is not very sensitive to a change of a few percent in either E_0 or d_0 . This result should not be taken as exact, but more as an indication of order of magnitude.

Figures 3 and 4 show the measured relative Stokes parameters, with the error bars used in the calculation and the relative Stokes parameters calculated with the electric-field parameters and the initial density matrix that we found. We see that the agreement is good. Figure 3 also shows the calculated M_x/I_x [observation in the x direction, see Eq. (38)] for the $2s\ ^1S-3p\ ^1P$ transition at 501.6 nm. We have also indicated the results of the measurements of M_x/I_x for He I at 150 keV by Dehaes and Carmeliet [19] (the energy is close enough so that there

should not be too much difference); and we see that the agreement with these measures (which were not used in the calculation) is also good.

X. CONCLUSIONS

In this paper, we have sketched the theory of state multipoles applied to density-matrix calculations and we have given a general method for finding the effects of symmetries on tensorial components. We have then applied this theory to the calculation of the evolution of the density matrix and of the polarization of the emitted light in beam-foil spectroscopy for the surface electric-field model. We chose to look at one of the simplest models of the electric field: a constant field E_0 over a distance d_0 and then nothing. We looked specifically at the $n=3$ singlet states of He I and, starting from the measured relative Stokes parameters for several tilt angles of the foil, we found an initial density matrix and electric-field parameters. The results were good, considering the simplicity and lack of realism of the chosen model for the electric field. The assumptions made on the electric field may appear unduly simplistic. This is not so, because the electric field appears linearly in Eq. (9). Therefore, referring to Eq. (D1), the main contribution to the solution, neglecting contributions from $[\Omega(t'), \Omega(t'')]$ and higher-order commutators, will involve $\int_0^t \Omega(t') dt'$, which is linear in the potential difference between the foil and the vacuum and therefore independent of the actual shape of the electric field.

This seems to indicate that the idea of a surface electric field being responsible for the observed polarization is valid. Now, the problem arises of the physical origin of this electric field and of giving a good model of that field. The hypothesis that appears to us to be the most promising is that secondary electrons created by the ion going through the foil and ejected at the same time are at least partly responsible for this field [9,20,21]. The problem is then to compute the interaction between these electrons and the atom.

The methods developed in this paper can also be applied to other ions, such as hydrogen. In the case of hydrogen, the levels are nearly degenerate in energy and quantum beats can be observed as a function of the distance from the foil. This gives us more information about the state of the beam than in the case of He I, so that it may not be necessary to tilt the foil to obtain more information. On the other hand, it can be shown that there will then be some undetermined parameters. This can be compensated for by making some assumptions, such as maximum entropy [22].

APPENDIX A: SOME STATE MULTIPOLE PROPERTIES

A definition of the state multipoles equivalent to Eq. (1) can be given using the Wigner-Eckart theorem [12]:

$$\begin{aligned} & \langle J_1 L_1 M_1 | T_q^k(J_3 L_3, J_4 L_4) | J_2 L_2 M_2 \rangle \\ &= (-1)^{J_1 - M_1} \begin{pmatrix} J_1 & k & J_2 \\ -M_1 & q & M_2 \end{pmatrix} \\ & \times \langle J_1 L_1 | T^k(J_3 L_3, J_4 L_4) | J_2 L_2 \rangle, \end{aligned} \quad (\text{A1})$$

where the reduced matrix element is

$$\begin{aligned} & \langle J_1 L_1 | T^k(J_3 L_3, J_4 L_4) | J_2 L_2 \rangle \\ &= \sqrt{2k+1} \delta_{J_1 J_3} \delta_{L_1 L_3} \delta_{J_2 J_4} \delta_{L_2 L_4}. \end{aligned} \quad (\text{A2})$$

And two other useful relations are

$$T_q^{k\dagger}(J_1 L_1, J_2 L_2) = (-1)^{J_1 - J_2 + q} T_{-q}^k(J_2 L_2, J_1 L_1), \quad (\text{A3})$$

$$\text{Tr}[T_q^k(J_1 L_1, J_2 L_2)] = \sqrt{2J_1 + 1} \delta_{J_1 J_2} \delta_{L_1 L_2} \delta_{k_0} \delta_{q_0}. \quad (\text{A4})$$

APPENDIX B: ORTHOGONAL OPERATORS EXPANSION

This appendix generalizes the work of Fano [23]. Given a set $\{U_j\}$ of orthogonal operators, i.e., such that

$$\text{Tr}(U_j U_k^\dagger) = \delta_{jk}, \quad (\text{B1})$$

which are in general non-Hermitian ($U_j \neq U_j^\dagger$), if the U_j 's form a complete basis, we can expand any operator Q on that basis:

$$Q = \sum_j Q_j U_j^\dagger = \sum_j Q_j^\dagger U_j, \quad (\text{B2})$$

where the Q_j and Q_j^\dagger are complex numbers defined by

$$\begin{aligned} Q_j &= \text{Tr}(Q U_j), \\ Q_j^\dagger &= \text{Tr}(Q U_j^\dagger). \end{aligned} \quad (\text{B3})$$

We can do this for the density operator ρ also; and since ρ is Hermitian, we find that

$$\rho_j^\dagger = \rho_j^* \quad (\text{B4})$$

(i.e., the coefficients of the two expansions are related by complex conjugation). We also have

$$\text{Tr}(\rho) = \sum_j \rho_j \text{Tr}(U_j^\dagger), \quad (\text{B5})$$

$$\text{Tr}(\rho^2) = \text{Tr}(\rho \rho^\dagger) = \sum_j |\rho_j|^2. \quad (\text{B6})$$

Assuming that the U_j form a Lie algebra (i.e., closed under commutation), we can define

$$i[U_j, U_k^\dagger]_- = \sum_n c_{jk}^n U_n = \sum_n c_{jk}^{\dagger n} U_n^\dagger, \quad (\text{B7})$$

$$i[U_j, U_k^\dagger]_+ = \sum_n b_{jk}^n U_n = \sum_n b_{jk}^{\dagger n} U_n^\dagger, \quad (\text{B8})$$

where $[\ ,]_-$ and $[\ ,]_+$ are the commutator and the anticommutator, respectively. The coefficients of these expansion are easily found by using the orthogonality relation [Eq. (B1)]

$$\begin{aligned} c_{jk}^n &= i \text{Tr}(U_n^\dagger [U_j, U_k^\dagger]_-), & c_{jk}^{\dagger n} &= i \text{Tr}(U_n [U_j, U_k^\dagger]_-), \\ b_{jk}^n &= i \text{Tr}(U_n^\dagger [U_j, U_k^\dagger]_+), & b_{jk}^{\dagger n} &= i \text{Tr}(U_n [U_j, U_k^\dagger]_+), \end{aligned} \quad (\text{B9})$$

and we can easily find the following relations:

$$c_{jk}^{n*} = -c_{kj}^{\dagger n}, \quad (\text{B10})$$

$$b_{jk}^{n*} = -b_{kj}^{\dagger n}. \quad (\text{B11})$$

We can now find the evolution equation for the components ρ_j of the density operator, given a Hamiltonian H . Let us assume that we can write H as

$$H = H_1 - i \frac{\Gamma}{2}, \quad (\text{B12})$$

$$H^\dagger = H_1 + i \frac{\Gamma}{2}, \quad (\text{B13})$$

where H_1 and Γ are Hermitian. Then, from the Liouville equation,

$$i\hbar \frac{\partial \rho}{\partial t} = H\rho - \rho H^\dagger, \quad (\text{B14})$$

and what we wrote above, we can obtain the following system of equations for the components ρ_j :

$$\frac{d\rho_j}{dt} = \sum_j \Omega_{jk} \rho_k = \sum_j \frac{1}{\hbar} (V_{jk} + iW_{jk}) \rho_k, \quad (\text{B15})$$

where

$$V_{jk} = \sum_n c_{jk}^n \text{Tr}(H_1 U_n), \quad (\text{B16})$$

$$W_{jk} = \sum_n b_{jk}^n \text{Tr} \left[\frac{\Gamma}{2} U_n \right], \quad (\text{B17})$$

and we can show that V and W are anti-Hermitian, i.e., that

$$V_{jk} = -V_{kj}^*, \quad (\text{B18})$$

$$W_{jk} = -W_{kj}^*. \quad (\text{B19})$$

APPENDIX C: EXPRESSION OF SYMMETRIES

If we are given a group G of transformations S_i under which the system described by ρ is invariant, we must have

$$[\rho, S_i] = 0, \quad \forall S_i \in G. \quad (\text{C1})$$

Taking a complete basis of orthogonal operators $\{U_j\}$ (as in Appendix B), we have in general

$$S_i U_j^\dagger S_i^{-1} = \sum_k g_{jk}^i U_k^\dagger, \quad (\text{C2})$$

where

$$g_{jk}^i = \text{Tr}(S_i U_j^\dagger S_i^{-1} U_k). \quad (\text{C3})$$

Using the expansion of ρ

$$\rho = \sum_j \rho_j U_j^\dagger \quad (\text{C4})$$

and the orthogonality relation (B1), Eq. (C1) gives us

$$\sum_j \rho_j (\delta_{jk} - g_{jk}^i) = 0, \quad \forall i, k. \quad (\text{C5})$$

This is the relation between the expansion coefficients that is due to the symmetry group considered.

APPENDIX D: INDEPENDENT-VARIABLE EQUATIONS

We have seen in Sec. VIII that the system of equations (9) decouples in q . Thus, for each q , we get a system of equations that can be written as

$$\frac{d\rho}{dt} = \Omega \rho, \quad (\text{D1})$$

where Ω is a complex matrix (which is time dependent in the general case, but not for the model chosen here) and ρ is the vector of all the tensorial components. Separating the real and imaginary parts, we define

$$\Omega = \Omega_0 + i\Omega_1, \quad (\text{D2})$$

$$\rho = \mathbf{A} + i\mathbf{B}, \quad (\text{D3})$$

and we can then write Eq. (D1) as a real equation:

$$\frac{d}{dt} \begin{bmatrix} \mathbf{A} \\ \mathbf{B} \end{bmatrix} = \begin{bmatrix} \Omega_0 & -\Omega_1 \\ \Omega_1 & \Omega_0 \end{bmatrix} \begin{bmatrix} \mathbf{A} \\ \mathbf{B} \end{bmatrix}. \quad (\text{D4})$$

Now, if we define \mathbf{a} to be the vector of all *independent* variables in \mathbf{A} , we can write

$$\mathbf{A} = \begin{bmatrix} I \\ E \\ 0 \end{bmatrix} \mathbf{a} = L\mathbf{a}, \quad (\text{D5})$$

where I is a unit matrix of dimension equal to the number of elements of \mathbf{a} , E is a matrix with one nonzero element per line (and that element is 1 or -1 , because of the symmetry relations of Sec. III), and 0 is a null matrix (for the elements of \mathbf{A} that are zero, again with the relations of Sec. III). We can do the same thing with \mathbf{B} and define

$$\mathbf{B} = M\mathbf{b}, \quad (\text{D6})$$

where \mathbf{b} contains the independent variables and M has the same structure as L . The Moore-Penrose pseudoinverses are defined as follows [24]:

$$\begin{aligned} L^+ L &= I, & L^+ &= (L^T L)^{-1} L^T, \\ M^+ M &= I, & M^+ &= (L^T L)^{-1} M^T. \end{aligned} \quad (\text{D7})$$

The pseudoinverses of L and M exist since it is easy to show that $(L^T L)$ and $(M^T M)$ are diagonal positive-definite matrices (because of the structure of L and M). It is then easy to obtain from Eq. (D4) the following reduced system of equations:

$$\frac{d}{dt} \begin{bmatrix} \mathbf{a} \\ \mathbf{b} \end{bmatrix} = \begin{bmatrix} L^+ \Omega_0 L & -L^+ \Omega_1 M \\ M^+ \Omega_1 L & M^+ \Omega_0 M \end{bmatrix} \begin{bmatrix} \mathbf{a} \\ \mathbf{b} \end{bmatrix}, \quad (\text{D8})$$

which has only independent variables. This method gives us a systematic way to eliminate the dependent variables and to construct the reduced system of equations in a computer program.

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