Calculation of electron-photon coincidence parameters for singlet-triplet mixed $4F$ states of helium

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(Received 8 September 1998)

We present the theoretical formalism required to interpret the electron-photon coincidence measurements of the helium $4F$ state by Cvejanović and Crowe [Phys. Rev. Lett. **80**, 3033 (1998)]. The results of the convergent close-coupling theory are compared with experiment and found to be in good qualitative agreement. We demonstrate that the singlet-triplet mixing in the $4F$ states does not affect the calculated results significantly. [S1050-2947(99)06702-5]

PACS number(s): 34.80.Bm, 34.80.Dp

I. INTRODUCTION

Electron-helium scattering is a favorite system for the experimental study of electron-atom scattering. A comprehensive set of accurate measurements has been compiled over the years which enables for thorough tests of corresponding theoretical methods. Electron-photon coincidence studies have played an important role in such testing. Observation of the scattered electron and the decay photon in coincidence allows for complete description of the scattering process, i.e., determination of absolute values and relative phases of all scattering amplitudes. These ''complete'' experiments, first suggested by Bederson [1,2], have been performed for helium P-state excitations (see Slevin and Chwirot [3] for a review and references). More recently, electron-photon coincidence measurements have been performed for helium $3\,^{1,3}D$ states (see Refs. [4,5] and references therein). Although a triple coincidence experiment [6] is required to achieve complete experimental description of the S-D transition, the missing information in the standard double coincidence experiment can be recovered with some insight from a reliable theoretical calculation [4,7].

Cvejanović and Crowe [8] have reported recently an electron-photon coincidence study of the helium $4F$ state excited from the ground state by 29.6 eV electrons. Polarization of the decay photon (Stokes parameters) for the cascade populated $3\,^1D-2\,^1P$ transition has been measured in coincidence with the scattered electron. Theoretical interpretation of such measurements is complicated due to the relativistic effects requiring the helium $4F$ states to be described as a mixture of the pure singlet and triplet states. The general formalism for such analysis was presented by Blum [9] and has been applied by Wang *et al.* [10] to the $4F$ state.

The purpose of the present paper is to present a theoretical interpretation of the Cvejanović and Crowe [8] measurements in a transparent and clear form. In Sec. II we recall the standard formalism for calculating Stokes parameters and then in Sec. III we describe how this formalism is applied to account for observation of the cascade photon. This is followed by treatment of the singlet-triplet mixing in the $4F$ state in Sec. IV. Finally, in Sec. V we compare results of a convergent close-coupling (CCC) calculation with the measurements [8], followed by our conclusions given in Sec. VI.

II. STOKES PARAMETERS

We consider electron impact excitation of a helium state with angular momentum J. It is assumed that this state is described in the nonrelativistic $LSJM_J$ coupling scheme. The dipole deexcitation radiation from the excited state is measured in coincidence with the scattered electron. In the experiment of Cvejanović and Crowe [8] the degrees of linear and circular polarization (Stokes parameters) of the radiation have been measured. The Stokes parameters P_1 and P_2 are the degrees of linear polarization and P_3 is the degree of circular polarization of the radiation propagating perpendicularly to the scattering plane. The Stokes parameter P_4 is the degree of linear polarization of the radiation propagating in the scattering plane. We refer to the review article of Andersen, Gallagher, and Hertel [11] for details of the calculation of the Stokes parameters and their relations to the shape and orientation of the excited state atomic charge cloud.

For a given electron scattering angle the Stokes parameters P_i may be written in terms of the state multipoles T_{kq} [9,11],

$$P_1(J)=\frac{\alpha_2(J)\left[T_{22}(J)-\sqrt{\frac{3}{2}}T_{20}(J)\right]/2}{T_{00}(J)-\frac{\alpha_2(J)}{2}[T_{22}(J)+T_{20}(J)/\sqrt{6}]}, \quad (1)$$

$$P_2(J)=\frac{\alpha_2(J)T_{21}(J)}{T_{00}(J)-\frac{\alpha_2(J)}{2}[T_{22}(J)+T_{20}(J)/\sqrt{6}]}, \quad (2)$$

$$P_3(J)=-\frac{i\alpha_1(J)T_{11}(J)}{T_{00}(J)-\frac{\alpha_2(J)}{2}[T_{22}(J)+T_{20}(J)/\sqrt{6}]}, \quad (3)$$

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$$P_4(J)=-\frac{\alpha_2(J)\left[T_{22}(J)+\sqrt{\frac{3}{2}}T_{20}(J)\right]/2}{T_{00}(J)+\frac{\alpha_2(J)}{2}[T_{22}(J)-T_{20}(J)/\sqrt{6}]}, \quad (4)$$

where the coefficients α_k are

$$\alpha_k(J)=3\sqrt{2J+1}(-1)^{J+J'+k+1}\begin{Bmatrix}1 & J & J'\\ J & 1 & k\end{Bmatrix}. \quad (5)$$

Here J is the angular momentum of the atomic target state and $J'=J\pm1$ for dipole photon deexcitation. Two cases are of interest for the present study. The first is an F state, where $J=3,J'=2$ and $\alpha_1=\sqrt{2},\alpha_2=3\sqrt{2}/5$. The second is a D state, where $J=2,J'=1$ and $\alpha_1=3/2,\alpha_2=\sqrt{21/20}$.

The state multipoles $T_{kq}(J)$ are given by

$$T_{kq}(J)=\sum_{M,M'}(-1)^{J-M'}C^{kq}_{JMJ-M'}\ \rho_{MM'}(J), \quad (6)$$

where $C^{kq}_{JMJ-M'}$ are Clebsch-Gordan coefficients and the density matrix is

$$\rho_{MM'}(J)=\frac{1}{2(2J_i+1)}\times\sum_{M_i,m,m_i}f_{JM,J_iM_i}(m,m_i)f^*_{JM',J_iM_i}(m,m_i). \quad (7)$$

The state multipoles are normalized to have $T_{00}=\mathrm{tr}\,\rho/\sqrt{2J+1}$. Summation in Eq. (7) over projectile electron initial and final spin projection m_i and m indicates that no electron spin analysis is performed. The collision frame scattering amplitudes $f_{JM,J_iM_i}(m,m_i)$ for the transition from initial state with angular momentum J_i and magnetic sublevel M_i to final state with angular momentum J and magnetic sublevel M have been chosen in such a way that the trace of ρ gives the differential cross section

$$\frac{d\sigma}{d\Omega}=\mathrm{tr}\,\rho=\frac{1}{2(2J_i+1)}\sum_{M,M_i,m,m_i}|f_{JM,J_iM_i}(m,m_i)|^2. \quad (8)$$

III. ACCOUNT OF UNOBSERVED CASCADE RADIATION

Equations (1)–(4) allow for the calculation of the Stokes parameters $P_i(3)$ for excitation of the $4\,^1F$ state. However, the $4\,^1F-3\,^1D$ radiation has a long wavelength and cannot be measured as a single particle. Instead, the $3\,^1D-2\,^1P$ cascade radiation has been measured [8]. We can obtain Stokes parameters $P_i(2)$ for the latter case from the same equations if the state multipoles $T_{kq}(2)$ of the cascade populated $3\,^1D$ state are known. These state multipoles can be calculated from the known state multipoles $T_{kq}(3)$ of the $4\,^1F$ state using the following general expression [12]:

$$T_{kq}(J')=\beta_k(J\to J')\,T_{kq}(J), \quad (9)$$

where

$$\beta_k(J\to J')=(-1)^{J+J'+1+k}(2J+1)\begin{Bmatrix}1 & J & J'\\ k & J' & J\end{Bmatrix}. \quad (10)$$

Equation (9) relates the state multipoles $T_{kq}(J)$ of the initial state and the state multipole $T_{kq}(J')$ of the cascade populated final state (with angular momentum $J'=J\pm1$) provided that polarization and angular distribution of the dipole radiation are not registered. When applied to the $4\,^1F-3\,^1D$ transition, Eq. (9) gives

$$T_{00}(2)=\sqrt{\frac{7}{5}}T_{00}(3),\quad T_{1q}(2)=\frac{2}{3}\sqrt{\frac{14}{5}}T_{1q}(3),$$

$$T_{2q}(2)=\frac{2}{5}\sqrt{6}\,T_{2q}(3). \quad (11)$$

The Stokes parameters for the $3\,^1D-2\,^1P$ transition can be found by substituting the above $3\,^1D$ state multipoles $T_{kq}(2)$ in Eqs. (1)–(4) and yield the same Stokes parameters as for $P_i(3)$ of the $4\,^1F$ state. This is a consequence of the general conservation property of the angular distribution of the radiation for the minimum multipole cascade transitions with consequent decreasing of the atom angular momentum. The cascade $4\,^1F-3\,^1D-2\,^1P-1\,^1S$ is an example of such transitions, see Korenman [12] for detailed discussion. Thus, measurements of the Stokes parameters for any of the transition in $4\,^1F\to1\,^1S$ cascade would yield the same result,

$$P_i(3)=P_i(2)=P_i(1),\quad i=1,2,3,4. \quad (12)$$

IV. ACCOUNT OF RELATIVISTIC EFFECTS

For the helium $4F$ states the nonrelativistic Russel-Saunders coupling scheme (LS scheme) breaks down. The relativistic effects become important and total orbital angular momentum L and total spin S are not good quantum numbers any more. However, the total atom angular momentum J is still a good quantum number and is used to label atom states. These states can be described, to a good approximation, as a mixture of the singlet ($S=0$) and triplet ($S=1$) nonrelativistic Russel-Saunders wave functions,

$$\Psi^\alpha_J=\sum_{S=0,1}\omega^\alpha_S\Psi(^{2S+1}L_J),\quad \alpha=a,b, \quad (13)$$

where $\omega^\alpha_S,S=0,1$ are expansion coefficients, and

$$\Psi(^{2S+1}L_{JM})=\sum_{m_Lm_S}C^{JM}_{Lm_LSm_S}\Psi_{m_Lm_S}(^{2S+1}L). \quad (14)$$

A convenient choice for the functions $\Psi_{m_Lm_S}(^{2S+1}L)$ is to take (approximate) eigenfunctions of the nonrelativistic Hamiltonian.

The expansion coefficients satisfy the following relations:

$$(\omega^\alpha_0)^2+(\omega^\alpha_1)^2=1,\quad \alpha=a,b, \quad (15)$$

$$(\omega^a_S)^2+(\omega^b_S)^2=1,\quad S=0,1, \quad (16)$$

$$\omega^a_0=\omega^b_1=1/\sqrt{1+\omega^2}, \quad (17)$$

$$\omega_1^a = -\omega_0^b = \omega/\sqrt{1+\omega^2}, \tag{18}$$

where we have introduced a mixing coefficient $\omega = \omega_1^a/\omega_0^a = -\omega_0^b/\omega_1^b$, which specifies the degree of mixing betweens-inglet and triplet levels. If $\omega = 1$, the singlet-triplet mixing is largest and states Ψ_J^α contain an equal mixture of the singlet and triplet wave functions. If $\omega < 1$, then, in the present notation, the state labeled a is predominantly singlet and the state labeled b is predominatly triplet.

The singlet-triplet mixing coefficient ω for the $4F$ states has been calculated by Parish and Mires [13] and van den Eynde, Wiebes, and Niemeyer [14] using the Breit-Pauli Hamiltonian. Their choice of the nonrelativistic wave functions for the $4F$ state is sufficiently close to the frozen-core model we use [15]. We therefore may use their values of ω. There is, however, a discrepancy between the former and latter calculations with Ref. [13] giving the value $\omega = 0.4335$, while Ref. [14] gives $\omega = 0.59$. Nevertheless, we will demonstrate that either choice of ω leads to essentually the same results when used to calculate the Stokes parameters.

When considering electron-impact excitation of the $4F$ states, we can use Eq. (13) to calculate scattering amplitudes for the excitation of singlet-triplet mixed $4F$ states,

$$f^\alpha_{JM,J_iM_i}(m,m_i) = \sum_{S=0,1} \omega_S^\alpha f^S_{JM,J_iM_i}(m,m_i), \quad \alpha = a,b, \tag{19}$$

where on the right-hand side the singlet and triplet scattering amplitudes are obtained from a nonrelativistic calculation. In what follows we consider scattering from helium ground state ($J_i = M_i = 0$) and drop initial state indexes.

We now proceed to the calculation of the radiation from the $4F$ levels to the $3\,^1D$ state. The major difference with what was presented in Sec. II is that radiation to the $3\,^1D$ state can occur from both Ψ_J^a and Ψ_J^b levels simultaneously. Due to the energy difference between Ψ_J^a and Ψ_J^b levels, $\omega_{ab} = (E_a - E_b)/\hbar = 704$ MHz, the observed radiation can be time modulated (quantum beats). We refer to Blum [9] for a general discussion of this phenomenon and to Wang *et al.* [10] for the specific application to the $4F \rightarrow 3\,^1D$ transition. The formalism presented in Sec. II for the calculation of the Stokes parameters requires modification. We will outline below how this can be done while keeping the structure of Eqs. (1)–(4) unchanged.

Both of the Ψ_J^a and Ψ_J^b levels are excited by electron impact from the helium ground state and their density matrix can be expressed via scattering amplitudes (19),

$$\rho^{\alpha\beta}_{MM'}(J) = \frac{1}{2} \sum_{m,m_i} f^\alpha_{JM}(m,m_i) f^{\beta *}_{JM'}(m,m_i), \tag{20}$$

assuming no spin analysis is performed. The summation over projectile electron spin magnetic sublevels in Eq. (20) results in cancelation of the singlet-triplet terms,

$$\rho^{\alpha\beta}_{MM'}(J) = \frac{1}{2} \sum_S \sum_{m,m_i} \omega_S^\alpha \omega_S^\beta f^S_{JM}(m,m_i) f^{S*}_{JM'}(m,m_i)$$

$$= \sum_S \omega_S^\alpha \omega_S^\beta \rho^S_{MM'}(J). \tag{21}$$

The interesting consequence of the last relation is, for example, that the differential cross section for excitation of the levels Ψ_J^α is just a sum of the cross section for the excitation of the corresponding singlet and triplet states multiplied by their weight factors,

$$\frac{d\sigma^\alpha}{d\Omega} = \mathrm{tr}\,\rho^{\alpha\alpha} = \sum_S (\omega_S^\alpha)^2 \frac{d\sigma^S}{d\Omega}, \tag{22}$$

and does not depend on the sign of the expansion coefficients ω_S^α. Similar expressions hold for any other quantity diagonal in index α.

The state multipoles corresponding to the density matrix $\rho^{\alpha\beta}_{MM'}(J)$ are defined according to Eq. (6),

$$T^{\alpha\beta}_{kq}(J) = \sum_{M,M'} (-1)^{J-M'} C^{kq}_{JMJ-M'} \rho^{\alpha\beta}_{MM'}(J)$$

$$= \sum_S \omega_S^\alpha \omega_S^\beta T^S_{kq}(J). \tag{23}$$

They satisfy the same symmetry properties in the collision frame as the state multipoles $T^S_{kq}(J)$ [9],

$$T_{kq} = (-1)^{k+q} T_{k-q} = (-1)^k T^*_{kq}. \tag{24}$$

They are real for even k and imaginary for odd k, and T_{k0} are zero for odd k.

The explicit time dependence of the Stokes parameters for the $4F \rightarrow 3\,^1D$ transition can be obtained using the following substitution in Eqs. (1)–(4) [9,10]:

$$T_{kq}(J) = \sum_{\alpha,\beta} \omega_0^\alpha \omega_0^\beta T^{\alpha\beta}_{kq}(J) \exp[-i\omega_{\alpha\beta} t - \gamma t], \tag{25}$$

where γ is the decay constant and is related to the mean lifetime $\tau = 1/\gamma = 67 \pm 10$ ns [16] for the helium $4F$ state. An example of the $\cos(\omega_{\alpha\beta} t)$ modulation of the radiation was presented by Wang *et al.* [10]. Experimental observation of such modulation requires very high time resolution as well as much better coincidence statistical accuracy, which currently is not feasible. In the experiment of Cvejanović and Crowe [8] the observation time was much longer than the mean lifetime of the $4F$ state. In order to compare with experiment we must integrate Eq. (25) over the time. Extending the integration limit to infinity [9], we obtain

$$T_{kq}(J) = \sum_{\alpha,\beta} \omega_0^\alpha \omega_0^\beta T^{\alpha\beta}_{kq}(J) \frac{\gamma}{\gamma^2 + \omega^2_{\alpha\beta}}. \tag{26}$$

The last relation allows for a significant simplification due to the substantial difference between the values of $\gamma = 15$ MHz and $\omega_{ab} = 704$ MHz. The $\alpha \neq \beta$ terms are much smaller than $\alpha = \beta$ terms and, therefore,

$$T_{kq}(J) \approx \frac{1}{\gamma} \sum_{\alpha} (\omega_0^{\alpha})^2 T_{kq}^{\alpha\alpha}(J)$$

$$= \frac{1}{\gamma} \sum_{S} \left(\sum_{\alpha} (\omega_0^{\alpha})^2 (\omega_S^{\alpha})^2 \right) T_{kq}^{S}(J). \quad (27)$$

This relation can be interpreted as if radiation is originated not from the two Ψ_J^a and Ψ_J^b levels, but rather from a single level whose singlet-triplet mixing is given by the expansion coefficients

$$(\tilde{\omega}_S)^2 = \sum_{\alpha} (\omega_0^{\alpha})^2 (\omega_S^{\alpha})^2, \quad (28)$$

$$(\tilde{\omega}_0)^2 + (\tilde{\omega}_1)^2 = 1. \quad (29)$$

When expressed through the mixing coefficient ω, they are

$$\tilde{\omega}_0 = \frac{\sqrt{1+\omega^4}}{1+\omega^2}, \quad \tilde{\omega}_1 = \frac{\sqrt{2}\omega}{1+\omega^2}, \quad (30)$$

and the new mixing coefficient is

$$\tilde{\omega} = \tilde{\omega}_1 / \tilde{\omega}_0 = \frac{\sqrt{2}\omega}{\sqrt{1+\omega^4}}. \quad (31)$$

The value of $\tilde{\omega}$ is larger than the value of ω $(0<\omega<1)$ and is 0.603 or 0.788 depending on if $\omega=0.4335$ [13] or $\omega=0.59$ [14], respectively.

Finally, in order to compare theoretical results with the experimental data of Cvejanović and Crowe [8], the following substitution should be made in Eqs. (1)–(4):

$$T_{kq}(J) = \frac{1}{\gamma} [\tilde{\omega}_0^2 T_{kq}^0(J) + \tilde{\omega}_1^2 T_{kq}^1(J)]. \quad (32)$$

Since the Stokes parameters are relative quantities, they are a function of $\tilde{\omega}^2$, i.e., we may divide Eq. (32) by $\tilde{\omega}_0^2$ and multiply by γ. The account of the unobserved cascade radiation from the $4F$ to $3\,^1D$ state is identical with what was presented in Sec. III.

V. RESULTS AND DISCUSSION

We now compare in Fig. 1 the results of our CCC calculation for Stokes parameters with the experimental data of Cvejanović and Crowe [8]. We use the same 111-state calculation at 30 eV given by Fursa and Bray [17]. The states comprise 14 ^1S states, 13 ^3S and $^{3,1}P$ states, 12 $^{3,1}D$ states, 10 $^{3,1}F$ states, and 7 $^{3,1}G$ states.

Calculated Stokes parameters are given for two cases. The first gives nonrelativistic results for excitation of the $4\,^1F$ state and the second gives the results that account for the singlet-triplet mixing as decribed in the preceding section. For maximal difference we take the largest value of the $4F$ mixing coefficient $\omega=0.59$ [14]. We find very close agreement between the results of the two models. The primary reason for this is that the state multipoles $T_{kq}^0(3)$ corresponding to excitation of the singlet state have larger absolute values than the state multipoles $T_{kq}^1(3)$ correponding to the triplet state. The coefficient $\tilde{\omega}^2=0.621$ reduces their contribution even more.

Comparing results of the CCC calculation with experimental values of the Stokes parameters, we find good qualitative agreement. As in the experiment, we find that values of the Stokes parameter P_2 are close to zero. Parameter P_4 does not differ substantially from its nonrelativistic value $(P_4=0.5)$ at zero scattering angle. Agreement between theory and experiment is good for Stokes parameter P_1 at 20°, 30°, and 40°, but the experimental value at 10° is substantially below the theoretical prediction. The largest discrepancy between theory and experiment is for the parameter P_3. Similar to the P_1 case, the 10° point exhibits the largest problem. Given that $P_1=0.5$ and $P_3=0$ at zero degrees, it is difficult to reconcile the discrepancy at 10°.

We have also presented in Fig. 1 the alignment angle γ of the atomic charge cloud and the angular momentum $L_\perp$ transfered to the atom perpendicular to the scattering plane,

$$\gamma = \mathrm{ATAN}\,2(P_2, P_1)/2,$$

$$L_\perp = 2\sqrt{2} i T_{11}/T_{00} = -\frac{6P_3(P_4+1)}{4-(P_1-1)(P_4-1)}. \quad (33)$$

The near zero values of alignment angle γ are due to the near zero values of the parameter P_2. The discrepancy between theoretical and experimental values of $L_\perp$ has its origin in the corresponding discrepancy for Stokes parameters P_1, P_3, and P_4.

Our results indicate that at the scattering angles 10° $-40°$, where experimental data of Cvejanović and Crowe [8] are available, the relativistic effects are negligible. Therefore, we consider it unlikely that a more consistent incorporation of the relativistic formalism in the scattering calculation would lead to a better agreement with experiment. Apart from the difficulty for theory and experiment to deal with such a complex transition, there is an uncertainty associated with the cascade contributions to the $3\,^1D$ state from levels other than $4\,^1F$. It follows from our calculation that cascades from $n=5,6$ F and G levels are comparable with the cascade from the $4\,^1F$ level. These states have longer lifetimes and their contributions have been substantially reduced in the experiment of Cvejanović and Crowe [8]. However, this may be the source of the occasional discrepancies.

VI. CONCLUSIONS

We have shown how the helium Stokes $3\,^1D-2\,^1P$ parameters measured by Cvejanović and Crowe [8] in coincidence with electron-impact 4F excitation may be calculated from the nonrelativistic scattering amplitudes for $4\,^1F$ and $4\,^3F$ excitation. The state multipoles describing the dipole radiation from the $4F$ states may be simply obtained, to a good approximation, from the nonrelativistic singlet and triplet state multipoles via Eq. (32). At the considered 30-eV energy, the CCC-calculated triplet state multipoles are considerably smaller than the corresponding singlet ones result-

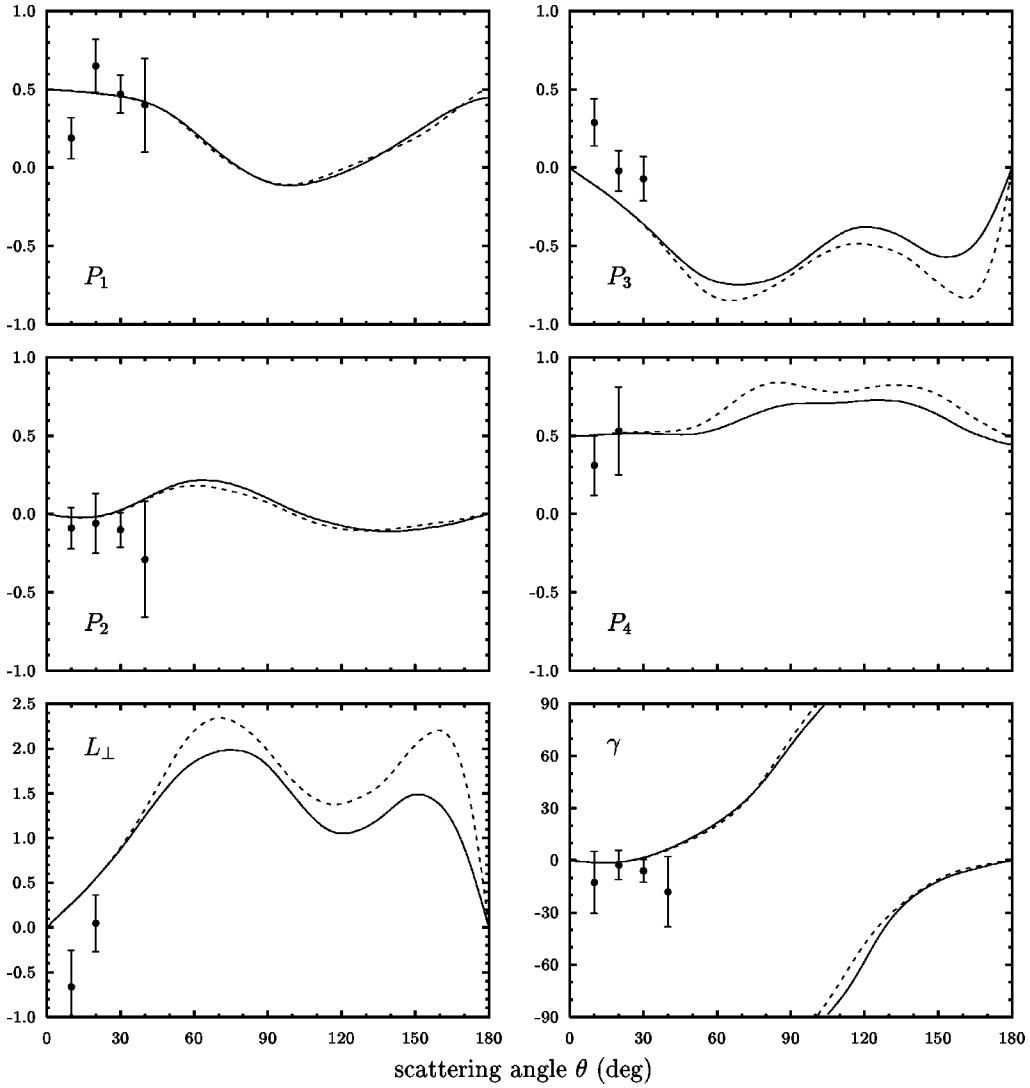

FIG. 1. Stokes parameters for electron-impact helium $4F$ state excitation. The present CCC(111) calculation is described in the text. The solid line is the result of a model which accounts for the singlet-triplet mixing in the $4F$ state. The dashed line is the result of the corresponding nonrelativistic calculation for the $4\ ^1F$ state. The experiment is due to Cvejanović and Crowe [8].

ing in the Stokes parameters being predominantly determined by the singlet-state multipoles. As a result, the $4F$ singet-triplet mixing does not affect the Stokes parameters significantly.

The agreement with experiment is qualitative rather than quantitative. We have checked for convergence in the CCC calculations and find little variation between various calculations at the small scattering angles. It would be desirable to have more angles measured to pin down the remaining small discrepancies.

ACKNOWLEDGMENTS

Support of the Australian Research Council and the Flinders University of South Australia is acknowledged. We are also indebted to the South Australian Center for High Performance Computing and Communications.

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