Electron-photon-correlation study of the $3\,^1D$ state of helium excited by electrons 3.5 eV above threshold

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A complete electron-photon polarization-correlation study of the $3\,^1D$ state of helium excited by 26.5-eV incident electrons is carried out for electron scattering angles in the range 40°–120°. The data are analyzed in terms of the shape and dynamics of the excited-state charge cloud. When the results are considered in the natural coordinate frame, the excitation is dominated by the $M=+2$ excitation amplitude. The lack of theoretical calculations for S-D excitation processes is this energy regime is highlighted.

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I. INTRODUCTION

Relatively little is understood about electron-impact excitation processes where the angular momentum of the initial state is increased by two atomic units. Experimentally, this can be attributed to the close proximity of other $n \geq 3$ states, making resolution of the D states impossible with the best available electron energy resolution. Excitation of the $3\,^1D$ state of helium provides a simple example of such a process. In this case, the $3\,^1D$ and $3\,^3D$ states are separated by less than 1 meV and the $3\,^1P$ state is only separated from them by ~ 13 meV. This has confined studies of the individual states to those in which the decay radiation is isolated using optical spectrometers or narrow-band interference filters. Until recently, this has restricted studies to measurements of excitation functions [1] and polarization fractions [2]. Interpretation of both measurements are complicated by unknown cascade contributions from higher excited states.

The application of coincidence techniques in atomic collision physics has enabled detailed information on D-state excitation to be obtained. Kleinpoppen and McGregor [3] reported an angular differential cross section for the $3\,^3D$ state of helium at 39.7 eV using a scattered electron-photon coincidence method. The electron energy resolution was only 120 meV but observation of the photon in coincidence with the electron enabled total isolation of the $3\,^3D$ state. Batelaan *et al.* [4] have also extracted relative differential cross sections from electron-photon coincidence data for the $3\,^1D$, $3\,^3D$, and $3\,^3P$ states of helium at 40, 60, and 80 eV.

The major breakthrough in experimental studies of the $3\,^1D$ and $3\,^3D$ states of helium came with the application of electron-photon correlation techniques to these states. van Linden van den Heuvell *et al.* [5,6] used an angular-correlation method in which the angular distribution of the $(2\,^1P-1\,^1S)$ cascade photons was measured in coincidence with electrons scattered through some angle. Polarization-correlation methods in which the polarization of the $(3\,^1D-2\,^1P)$ photons is analyzed for scattering through a specific angle have provided a more comprehensive set of data. Beijers *et al.* [7,8], Batelaan, van Eck, and Heideman [9,10] and Wedding, Mikoza, and Williams [11] have studied excitation of the $3\,^1D$ state at incident electron energies in the range 40–60 eV and for electron-scattering angles in the range 10°–60°. Results have been reported from this laboratory [12–14] at 40 and 29.6 eV and for scattering angles in the range 40°–120°. An electron-photon angular-correlation study involving the $(3\,^1D-2\,^1P)$ radiation has been carried out for scattering angles $\leq 40°$ at 40 and 60 eV by Perera and Burns [15].

Some of the more recent experiments [10,13,14] in which the polarization of the radiation is determined in orthogonal directions provide an almost complete description of the excitation process. Strictly, full description of a D state [16] requires determination of state multipoles $\langle T(L)_{KQ}\rangle$ of rank K up to $K=4$. The emitted dipole radiation carries only information on multipoles of rank $K \leq 2$. Very recently, Mikoza *et al.* [17] have determined a normalized rank-four state multipole from photon-photon polarization-correlation measurements. The relative partial cross sections for excitation of the different magnetic sublevels is also extracted from these data.

Theoretically, excitation of the ^1D states in helium from the ^1S ground state contrasts sharply with the excitation of dipole allowed transitions such as the ^1P states. For example, in the low- to intermediate-energy regime, the first Born approximation overestimates the total excitation cross section for dipole allowed transitions whereas it underestimates it for ^1D excitation. An extensive discussion of both theoretical and experimental studies of $3\,^1D$ excitation in helium has been given by Mansky and Flannery [18–20]. Their calculations use a multichannel eikonal theory (DMET with a ten-channel basis set), which includes the addition of a dipole correction at large impact parameter to the original MET results. Although it does not account for electron exchange, it allows interesting conclusions to be reached both for the total excitation cross section and for small-angle scattering. Specifically, it highlights the importance of indirect excitation mechanisms involving the $2\,^1P$ and $3\,^1P$ states and shows interesting comparisons with first-order many-body theory.

Here a complete determination of the polarization of the dipole radiation in coincidence with the scattered electron is carried out for an incident electron energy of 26.5 eV and scattering angles in the range 40°–120°. No other experimental data are available at an energy below 40 eV with the exception of recent data from this laboratory [14] at 29.6 eV, and no theoretical predictions are available for comparison. At 29.6-eV and 40-eV existing calculations, the DMET calculations of Mansky and Flannery, the distorted-wave Born approximation of Bartschat and Madison [21] and the first-order many-body theory [22,23] fail to reproduce the experimental data. At energies close to threshold, close-coupling methods are likely to be more appropriate and it is hoped that the present work will stimulate the application of new R-matrix calculations which include continuum states [24] to D excitation in this energy range.

II. THEORETICAL BACKGROUND

The theory of electron-photon correlation experiments relating specifically to D states has been discussed by a number of authors [8,25,26]. Only the basic principles, together with parametrization of the data and the relationships between the measured quantities and those parameters are summarized here.

In excitation studies in which a single outgoing particle is observed, the only symmetry in the system is the axial symmetry about the electron-beam axis. In correlation experiments such as those reported here, the system has symmetry about a plane defined by the momenta of the incoming and scattered electrons. Throughout this discussion the natural coordinate frame [27], in which the quantization axis z is perpendicular to the (x,y) scattering plane, will be used. The excitation of an initially pure state using identical kinematic conditions leads to the production of a pure excited state (coherent superposition of its magnetic substates). For a light atom such as helium it can be assumed that the positive reflection symmetry of the initial $^1S_0$ state is conserved in the collision. In the natural coordinate frame, only the $M=0, \pm 2$ states have positive reflection symmetry. Hence the $3\,^1D$ excited state can be described as

$$|^1D\rangle = a_{-2}(E,\theta)|2-2\rangle + a_0(E,\theta)|20\rangle + a_{+2}(E,\theta)|2+2\rangle \ , \qquad (1)$$

where the $a(E,\theta)$ are the amplitudes for excitation of the states $|LM\rangle$. They are functions of the electron energy E and scattering angle θ. Taking a_0 to be real and positive, the excitation process can then be described by the magnitudes of the amplitudes α_0, $\alpha_{\pm 2}$ and the relative phases between a_0 and $a_{\pm 2}, \beta_{\pm 2}$, i.e.,

$$a_{-2} = \alpha_{-2} e^{i\beta_{-2}} \ ,$$
$$a_0 = \alpha_0 \ , \qquad (2)$$
$$a_{+2} = \alpha_{+2} e^{i\beta_{+2}} \ .$$

If the wave function (1) is normalized to unity this gives the additional condition

$$\alpha_{+2}{}^2 + \alpha_0{}^2 + \alpha_{-2}{}^2 = 1 \ . \qquad (3)$$

The maximum information available from the dipole radiation is obtained from measurements of three Stokes parameters P_1, P_2, and P_3 of the radiation emitted in a direction perpendicular to the scattering plane and P_4 in the scattering plane and perpendicular to the incident-beam (y) direction. The Stokes parameters are readily defined both in terms of observed intensities and the scattering amplitudes [26],

$$\begin{aligned} I_z P_1 &= I(0°) - I(90°) \\ &= -(\tfrac{2}{3})^{1/2}\alpha_0(\alpha_2\cos\beta_2 + \alpha_{-2}\cos\beta_{-2}) \ , \\ I_z P_2 &= I(45°) - I(135°) \\ &= -(\tfrac{2}{3})^{1/2}\alpha_0(-\alpha_2\sin\beta_2 + \alpha_{-2}\sin\beta_{-2}) \ , \\ I_z P_3 &= I(RHC) - I(LHC) = -(\alpha_2^2 - \alpha_{-2}^2) \ , \end{aligned} \qquad (4)$$

and

$$\begin{aligned} I_y P_4 &= I(0°) - I(90°) \\ &= \tfrac{1}{2}[1 - 2\alpha_0^2 - (\tfrac{2}{3})^{1/2}\alpha_0(\alpha_2\cos\beta_2 + \alpha_{-2}\cos\beta_{-2})] \ . \end{aligned}$$

I_z and I_y are the total intensities of the radiation emitted in the z and y directions respectively and $I(\alpha)$ the intensity transmitted by a linear polarizer with its transmission axis at an angle α to the electron-beam direction. It will be seen later that it is useful to extract the individual excitation amplitudes from the measured Stokes parameters.

It is now more fashionable to analyze the results of these experiments in terms of the shape and dynamics of the excited-state charge cloud [26]. The shape is given by the alignment angle γ of the charge cloud, where

$$\tan 2\gamma = \frac{P_2}{P_1} = \frac{-\alpha_2\sin\beta_2 + \alpha_{-2}\sin\beta_{-2}}{\alpha_2\cos\beta_2 + \alpha_{-2}\cos\beta_{-2}} \ , \qquad (5)$$

and the linear polarization P_l, where

$$\begin{aligned} P_l &= (P_1^2 + P_2^2)^{1/2} \\ &= \frac{(\frac{2}{3})^{1/2}\alpha_0}{1 - \frac{2}{3}\alpha_0^2}[\alpha_2^2 + \alpha_{-2}^2 + 2\alpha_2\alpha_{-2}\cos(\beta_2 + \beta_{-2})]^{1/2} \ . \end{aligned} \qquad (6)$$

Its relative height in the z direction is given by the density-matrix element ρ_{00}:

$$\rho_{00} = \frac{\frac{3}{2}(1+P_1)(1-P_4)}{4-(1-P_1)(1-P_4)} = \alpha_0^2 \ . \qquad (7)$$

P_3 (circular polarization) is essentially a direct measure of the expectation value of the angular momentum of the excited state $L_\perp$, so called because angular momentum can only be transferred perpendicular to the scattering plane:

$$L_\perp = -2I_z P_3 = 2(\alpha_2^2 - \alpha_{-2}^2) = \frac{4P_3(1+P_4)}{(1-P_1)(1-P_4)-4} \ . \qquad (8)$$

The present data will be presented both in terms of the charge-cloud parameters γ, P_l, $L_\perp$, ρ_{00} and the square of the excitation amplitudes $\alpha_0, \alpha_{\pm 2}$.

III. EXPERIMENT

Different aspects of the apparatus have been described previously [14,28,29]. Briefly, a well-defined beam of electrons ($\sim 10^{-7}-10^{-6}$ A) crosses a beam of helium atoms. The scattered electrons are focused onto the entrance aperture of a 180° hemispherical electron energy analyzer and detected using a channel electron multiplier. The overall electron resolution of the system is always better than 500 meV so that the $n=3$ manifold of states is isolated from the $n \geq 4$ states. Both the scattered electron analyzer and a Faraday cup that continuously collects the incident electron-beam rotate about the collision center.

One photon analysis/detection system collects photons emitted from the interaction region into a small solid angle about a direction perpendicular to the scattering plane using a 50-mm focal length lens with its focus at the interaction region. Photons then fall normally on the vacuum window, the polarization analysis components, and a narrow-band interference filter isolating the 667.8-nm ($3\,^1D-2\,^1P$) radiation before being focused onto the photo cathode of a photomultiplier tube. For P_1 and P_2, the intensities transmitted by a linear polarizer with $\alpha=0°, \ldots, 315°$ in 45° steps are measured. For P_3 measurements, an aligned quarter-wave plate is inserted before the linear polarizer and $I(\alpha)$ determined for $\alpha=45°$, 135°, 225° and 315°.

The P_4 analyzer and photon detector, mounted in the scattering plane at right angles to the incident electron beam, are similar in principle and operation to that described for the P_1 and P_2 measurements. $I(\alpha)$ for $\alpha=0°$, 90°, 180°, and 270° is determined in this case. Both photon analysis and detection systems are automated and controlled using computer-based systems which also handle the data. Pulses from the electron detector start the ramp of two time-to-amplitude converters (TAC), one ramp being stopped by photons from one detector and the other by photons from the other detector. In this way P_4 and P_1, P_2, or P_3 data can be accumulated simultaneously. Time spectra are accumulated at a specific $I(\alpha)$ for a period of one hour and stored in a memory array. Each completed cycle of measurements is repeated many times until data of acceptable statistical accuracy and satisfying various consistency checks are obtained.

Considerable care was taken to ensure that the data were free from instrumental effects. Some of the procedures used have already been outlined [14] and detailed results of checks carried out will be provided in a forthcoming publication. The polarization components were aligned with respect to the incident electron beam using a technique previously described [29]. The alignments of the linear polarizers are readily checked during the experiment using the polarized 667.8-nm photons from the $3\,^1D$ state [2]. The P_4 analyzer is further checked using the fact that $P_4 \equiv 1$ for the $3\,^1P$ state of helium. It was also demonstrated that the photon detector efficiencies were polarization independent. Both the magnitude of retardation and handedness of the quarter-wave plate were determined in preliminary experiments. The retarder used was left-handed with a retardation consistent with 90° (92.8°+4.0°) [14].

It was established both experimentally and theoretically that it was unnecessary to correct the data to account for the finite solid angles α of the photon detectors. Experimentally, measurements of P_1 and P_2 at 40 eV showed no dependence on the solid angle. Theoretically, it has been shown [10,30] that the true Stokes parameters P_i ($i=1-4$) are related to those measured, P_i^m, with solid angle of detection α by:

$$P_i = P_i^{\,m}\left[1+\tfrac{1}{4}\alpha^2\frac{(1+P_1)(1-P_4)}{(1+P_4)}\right] \quad \text{for } i=1,2,3 ,$$

and (9)

$$P_4 = P_4^{\,m}\left[1+\tfrac{1}{4}\alpha^2\frac{(1-P_1)(1+P_4)}{(1+P_1)}\right] .$$

In the present study, α had values of 0.42 sr for the P_1, P_2 measurements, 0.25 sr for P_3, and 0.13 for P_4. For the present data, the maximum difference between $P_i^{\,m}$ and P_i is less than 1.5%, confirming the experimental observation.

Variations in electron-beam current and gas-beam density, together with electron detector efficiency, were accounted for by normalizing the true coincidence signals to the number of electrons starting the TAC ramps. Energy drifts which were not taken account of in this normalization were not normally a problem and were readily identified by continuous monitoring of the voltages defining the incident electron energy and the analyzing energy of the scattered electrons, together with frequent measurements of the electron energy-loss spectrum. The photomultipliers were found to be extremely stable over long time periods.

TABLE I. Measured values of the Stokes parameters P_1, P_2, P_3, and P_4 and the charge-cloud parameters γ, P_l, $L_\perp$, and ρ_{00}, together with the total polarization P, for the $3\,^1D$ state of helium excited by 26.5-eV electrons.

θ (deg)	P_1	P_2	P_3	P_4	γ (deg)	P_l	ρ_{00}	$L_\perp$	P
40	0.45±0.03	−0.10±0.03	−0.50±0.02	0.63±0.06	−5.9±1.9	0.46±0.03	0.21±0.04	0.86±0.05	0.68±0.03
60	0.42±0.05	−0.02±0.05	−0.63±0.04	0.66±0.10	−1.3±3.7	0.42±0.05	0.19±0.06	1.09±0.09	0.76±0.04
80	0.10±0.05	−0.04±0.05	−0.98±0.11	0.86±0.18	−11.0±12.4	0.11±0.05	0.06±0.08	1.89±0.23	0.99±0.11
100	0.12±0.05	0.06±0.06	−0.85±0.08	0.81±0.17	12.7±12.9	0.13±0.05	0.08±0.08	1.61±0.18	0.86±0.08
120	0.12±0.06	−0.08±0.07	−0.53±0.07	0.86±0.11	−16.3±13.7	0.14±0.07	0.06±0.05	1.03±0.13	0.55±0.07

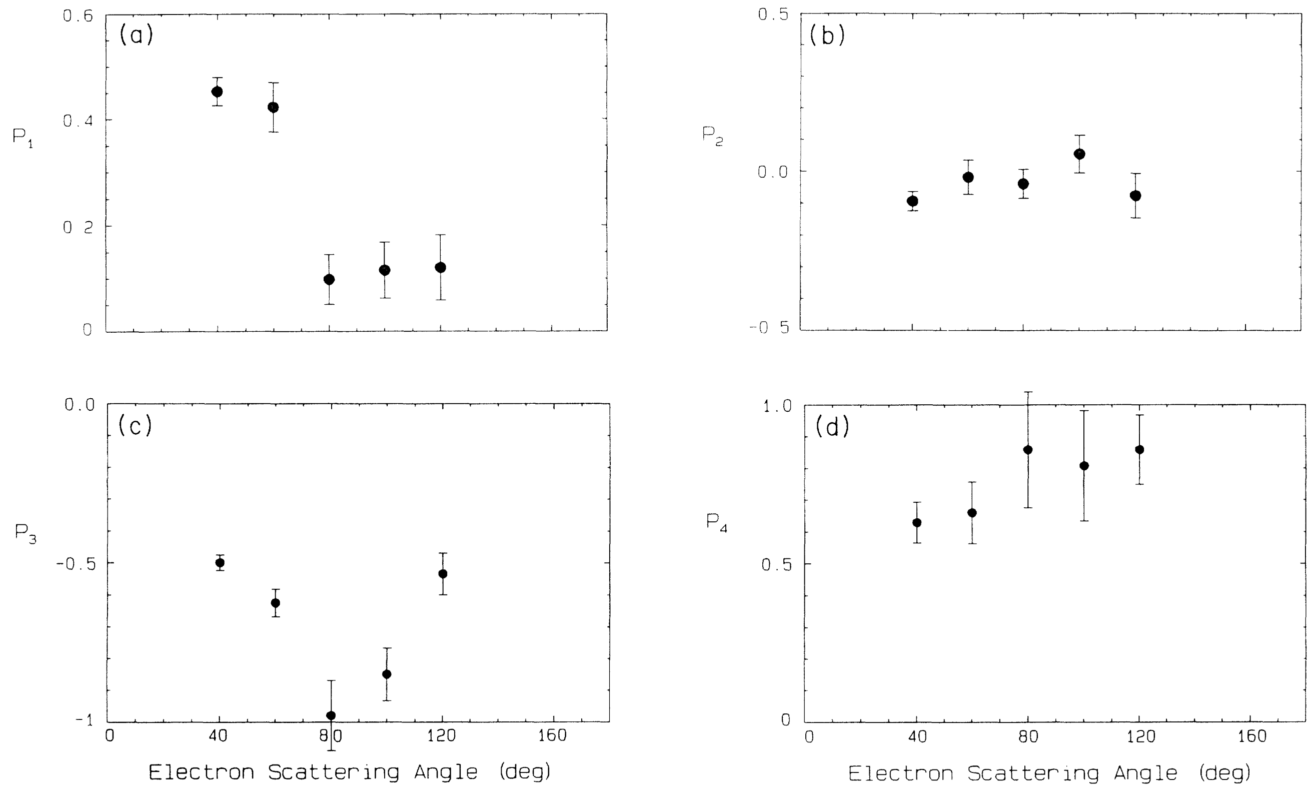

FIG. 1. Measured variation of the Stokes parameters $P_1 - P_4$ with electron-scattering angle for the $3\,^1D$ state of helium excited by 26.5-eV electrons.

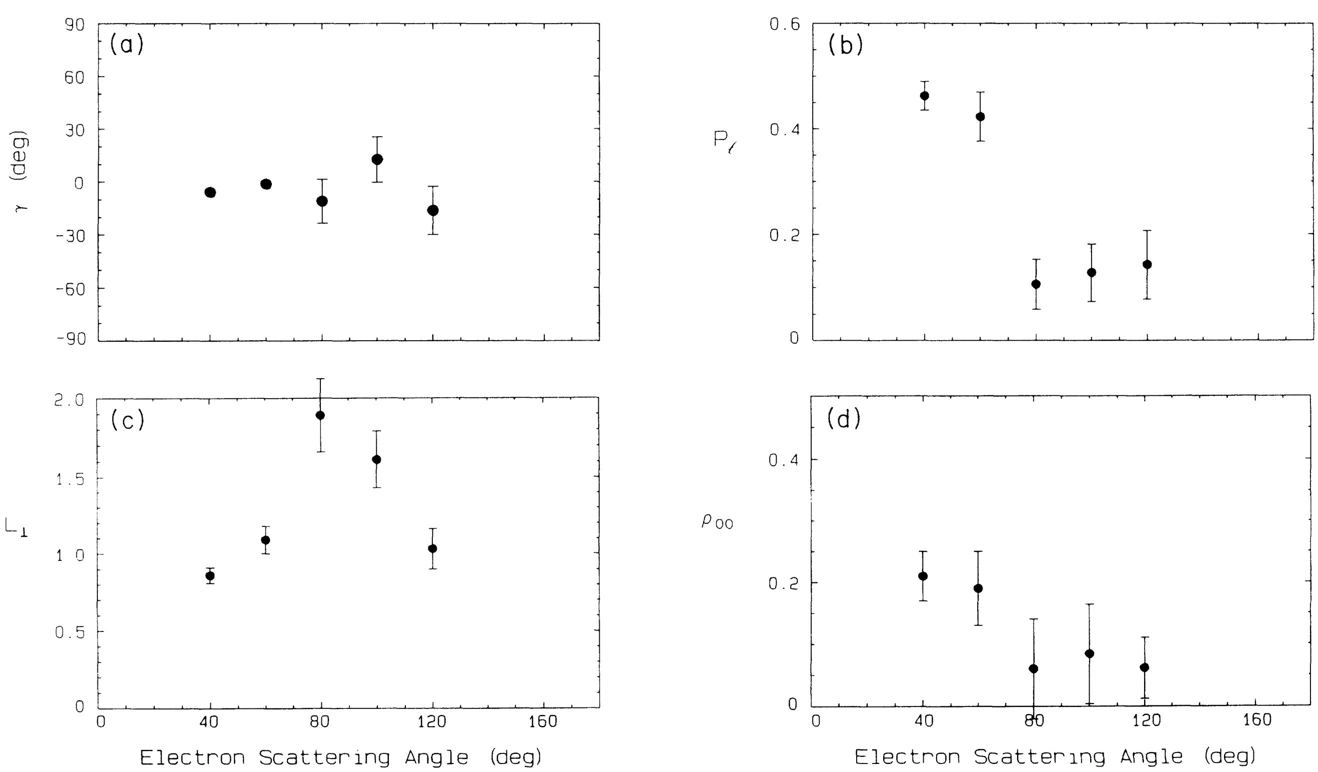

FIG. 2. Variation of the excited-state charge-cloud parameters for the $3\,^1D$ state of helium at 26.5 eV.

IV. RESULTS AND DISCUSSION

Table I shows the values of the Stokes parameters measured at an incident energy of 26.5 eV together with the charge-cloud parameters γ, P_l, $L_\perp$, and ρ_{00}. Also shown is the total polarization P defined by

$$P=(P_l{}^2+P_3{}^2)^{1/2} . \qquad (10)$$

The variation of the Stokes parameters with scattering angle is shown in Fig. 1. The sharp drop in P_1 from a value in the range 0.4–0.5 at 40° appears to be characteristic of the $3\,^1D$-state excitation for energies measured (up to 40 eV). In this case, the drop occurs between 60° and 80° compared with 40°–60° at 29.6 eV [14] and 40 eV [12]. Only at 26.5 eV are consistently positive values observed at higher angles. With the exception of 40°, the P_2 values are essentially consistent with zero. This is not unlike the situation at 29.6 eV, but a more dramatic variation was observed at 40 eV. P_3 is clearly large and negative at all scattering angles. Again, as for P_1, there is evidence that the dominant feature, in this case the maximum in $|P_3|$, moves towards larger angles as the incident electron energy decreases. Similarly, the maximum in P_4 observed at 60° at 29.6 eV has moved to a larger angle with all values for $\theta \geq 80°$ being nearly consistent with unity.

The excited-state parameters γ, P_l, $L_\perp$, and ρ_{00} are shown in Fig. 2. As a consequence of the near-zero values of P_2, the values of γ lie very close to zero at all electron-scattering angles and the values of P_l show the same variation as P_1. The large positive values of $L_\perp$ are expected, given the large negatively measured P_3. The small ρ_{00} are a reflection of the large P_4.

Although it is generally accepted that the description of the excitation process in terms of the shape and dynamics of the excited-state charge cloud is most transparent physically, the variation of the square of the relative scattering amplitudes shown in Fig. 3 provides a useful complement to the (γ, P_l, $L_\perp$, ρ_{00}) presentation. The dominant feature of the excitation process is excitation of the $M=+2$ sublevel at all scattering angles, the data at 80° being consistent with this being the sole excitation mechanism. The values of $|\alpha_0|^2 \equiv \rho_{00}$ show that excitation of the $M=0$ state is only of some significance at 40° and 60°. With the exception of 120°, it is also clear that within experimental error $|\alpha_0|^2 \approx |\alpha_{-2}|^2$. Analysis of the data in this way can also give an indication of the internal consistency of the measured Stokes parameters. For example, the result at $\theta=80°$ (when $P_3 \approx -1$, implying domination of the $M=2$ state) is consistent with the observation $P_4 \approx 1$ at this angle implying $\alpha_0 \sim 0$.

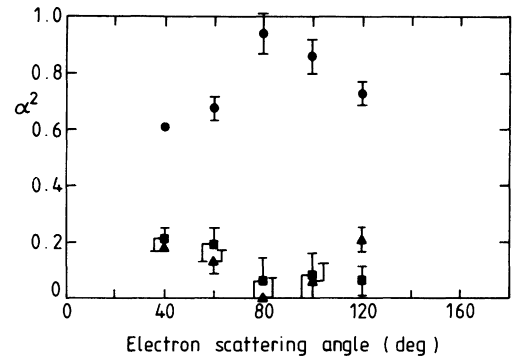

FIG. 3. α_{+2}^2 (●), α_0^2 (■), and α_{-2}^2 (▲) at 26.5 eV.

Considerable attention has been given to understanding the sign of $L_\perp$ for the excited P states; for example, by Andersen and Hertel [31]. For P states analyzed in the natural frame of reference, $L_\perp$ is determined by the relative excitation of the $M=\pm 1$ excitation processes. For the $n\,^1P$ states of helium, the $L_\perp$ behavior is generally characterized by positive values at small scattering angles, becoming negative at larger angles. However, at 26.5 eV the sign of $L_\perp$ is only observed to be negative at an angle of 120° [29] for He($3\,^1P$). No sign change is observed for the $3\,^1D$ state in these measurements; otherwise, the qualitative behavior of $L_\perp$ for the $3\,^1P$ and $3\,^1D$ states at 29.6 eV and in the angular range $40 \leq \theta \leq 120°$ is similar. This similarity needs to be treated with caution given the lack of data at both smaller and larger scattering angles. At higher energies (60 eV) Batelaan, van Eck, and Heideman [10] show $L_\perp$ distinctly negative for the $3\,^1D$ state for $\theta \leq 31.2°$, although their only marginally negative values at 45 eV may suggest that $L_\perp$ is positive for small scattering angles at lower energies. Clearly, the behavior of $L_\perp$ as a function of scattering angle and incident electron energy is more complex than for P states and is not explained qualitatively by simple semiclassical models.

In conclusion, the lack of theoretical calculations for this process at incident electron energies below 29.6 eV and the failure of existing perturbative models to reproduce experimental data at higher energies [12,14] is emphasized.

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