Determination of Rank 4 Multipoles and of the Partial Cross Sections for He($3\,^1D$) Excitation by Electron Impact

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We report the first measurements of a rank 4 state multipole for He($3\,^1D$) excitation by electron impact with a polarized photon-photon coincidence technique. Our results allow, for the first time, the extraction of partial cross sections σ_m for He($3\,^1D_m$) excitation ($m=0, \pm 1, \pm 2$) which are not otherwise available. The present results put stringent tests on existing theories and, for example, point in the theoretical calculations to an overestimation of the partial cross section σ_2.

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The excitation of helium by electron impact to an $n\,^1P$ or $n\,^1D$ state is a prototype example of a coherent collision process. Most of the previous investigations have concentrated on excitation of He($n\,^1P$) states, however, and comparatively little work has been devoted so far to excitation of other states [1]. One of the reasons lies in the complexity of, for example, the He($3\,^1D$) state where a complete description of the excited state in terms of statistical tensors or state multipoles $\langle T(L)_{KQ}\rangle$ requires tensors of rank K up to $K=4$ [2-4]. All information on the collision process is then contained in these state multipoles. To determine experimentally the state multipoles one usually measured the polarization and/or angular correlation of photons emitted during the decay of the excited atom to a lower state. The information which can be extracted from such an experiment, however, is limited by the emitted photon state which can carry only the information corresponding to state multipoles of rank up to $K=2$. As a direct consequence, a complete determination of a collisionally excited D state can be carried out only under certain assumptions, for example, conservation of reflection symmetry with respect to a scattering plane and full coherence during the collision, as is often the case in electron-photon coincidence experiments [1]. Even then, there remains a phase ambiguity which leads to two possible solutions for the collisionally excited state which cannot be resolved without an external field [5].

An alternative technique to extract the maximum possible information may utilize the angular or polarization properties of the light emitted in the two-photon decay of He($3\,^1D$)$\rightarrow$He($2\,^1P$)$\rightarrow$He($1\,^1S$). As a first step we have performed a polarized first photon (γ_2)-second photon (γ_2) coincidence experiment in which the scattered electron was not detected (see Fig. 1). In this case the collision system possesses rotational symmetry about the incident electron beam axis and only the state multipoles with $Q=0$ will be observed, i.e., the excited state density matrix is incoherent. This (γ_1,γ_2) coincidence experiment determines the state multipoles $\langle T(2)_{K0}\rangle$ with $K=0$, 2, and 4, which in turn are related to the partial cross sections σ_m for population of the magnetic substates $|D_m\rangle$ ($m=0, \pm 1, \pm 2$) of the excited helium state.

While two-photon coincidence studies have been quite successful in providing specific information on the fundamental laws of physics such as parity conservation and during the search for so-called hidden variables [6,7], their application to atomic scattering processes has been quite restricted and only recently the potential and the feasibility of such two-photon coincidence studies have been explored by Williams, Kumar, and Stelbovics [8] for H($n=3$) excitation and at an incident electron energy of 290 eV. For this particular collision system, the rank $K=4$ state multipole as extracted from an angular correlation measurement was rather small and the statistical significance of the data was such that stringent tests of existing theoretical models were not permitted. The results presented here utilize a novel technique by which the polarization state of the first photon (γ_1) is measured; it yields a much higher degree of statistical significance compared to the previous angular correlation work. The selection of helium compared to collisions with hydrogen

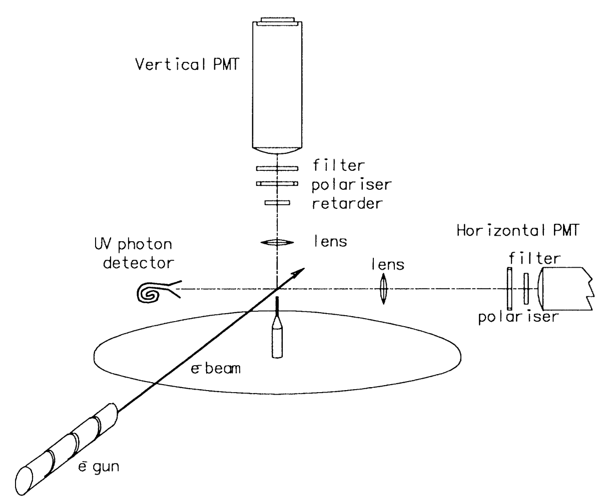

FIG. 1. Schematic diagram of photon-photon coincidence apparatus.

atoms offers several advantages, too. First, in helium the first photon from the $He(3\,^1D) \rightarrow He(2\,^1P)$ transition is uniquely selected by a wavelength filter which was not feasible in the hydrogen work because of the $H(n=3)$ degeneracy. Second, since the total electronic as well as nuclear spins are zero in helium, the present investigation does not suffer from depolarization effects due to fine and hyperfine interactions during the decay of the excited atoms.

The experimental method involves electron impact excitation of the target helium atoms and the time coincidence detection of two photons following the decay of $He(3\,^1D) \rightarrow He(2\,^1P) \rightarrow He(1\,^1S)$. The basic apparatus used in this work has been described recently by Mikosza *et al.* [9]. The main modification involves the simultaneous detection of two photons. The first (visible) photon (γ_1) resulting from the $He(3\,^1D) \rightarrow He(2\,^1P)$ transition at 667.8 nm passed through an optical lens (entrance solid angle 0.144 sr) to form a parallel beam of light which was subsequently directed onto the appropriate retarder and linear polarizer combination to permit a full polarization analysis of the emitted light. The polarizer was followed by an interference filter and a photomultiplier tube (EMI 9883) operated in the pulse counting mode. The second [vacuum ultraviolet (VUV)] photon from the subsequent $He(2\,^1P) \rightarrow He(1\,^1S)$ decay at 58.4 nm passed through an entrance solid angle of 0.03 sr in front of a Mullard B418BL channel electron multiplier; no polarization analysis was carried out for this second photon. Both photons were detected at 90° with respect to the incident electron beam. Measurements have been carried out with the relative (azimuthal) angle ϕ between the two detected photons chosen as $\phi = 90°$ (perpendicular) and $\phi = 180°$ (opposite). Standard coincidence electronics have been used throughout. The time coincidence spectra were recorded with a Canberra S100 pulse height analyzer controlled by an AT-386 personal computer which permitted on-line data storage and analysis.

Our results for the coincident two-photon intensity $I(\alpha)$ at $\phi = 90°$ and incident electron energy $E = 81.6$ eV as a function of polarizer angle α with respect to the z axis (chosen as the direction of the incident electron) are shown in Fig. 2. The data display a pronounced polarization correlation of the first photon (γ_1) which is symmetric about the incident electron axis. From a least-squares fit to these data using

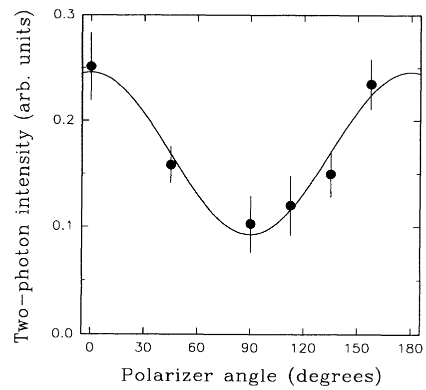

FIG. 2. The coincident two-photon intensity (●) vs polarizer angle α. The solid line is a least-squares fit [Eq. (1)] to the data points (see text).

$$I(\alpha) = \tfrac{1}{2} I_0 (1 + P_1 \cos 2\alpha + P_2 \sin 2\alpha) , \tag{1}$$

where I_0 is the total two-photon intensity, we obtained $P_1 = 0.458 \pm 0.057$ and $P_2 = -0.005 \pm 0.058$. Here the linear polarizations or Stokes parameters P_1 and P_2 are defined in the usual way as

$$P_1 = \frac{I(0°) - I(90°)}{I(0°) + I(90°)}, \quad P_2 = \frac{I(45°) - I(135°)}{I(45°) + I(135°)} . \tag{2}$$

For completeness we also measured the circular polarization P_3,

$$P_3 = \frac{I(-) - I(+)}{I(-) + I(+)} , \tag{3}$$

where the $I(\pm)$ relate to the intensities of circularly polarized light with positive and negative helicity, respectively. For the circular polarization which was measured separately we obtained $P_3 = -0.022 \pm 0.097$.

The measured polarizations are related to the same multipoles $\langle T(2)_{K0} \rangle$ with $K = 0$, 2, and 4. The theoretical description employed here rests on the work of Fano and Macek [2], Blum and Kleinpoppen [3,4], and Heck and Gauntlett [10]. Details of these calculations will be published separately [11,12]. In the frame of this theory, the $(e^-, \gamma_1, \gamma_2)$ triple coincidence rate can be calculated from the two-photon density matrix $\rho(\lambda_1, \lambda_1', \lambda_2, \lambda_2')$,

$$\begin{aligned}
\rho(\lambda_1,\lambda_1',\lambda_2,\lambda_2') = {} & C \exp[-\gamma_1(t-t_0) - \gamma_2 t] \\
& \times \sum_{kpq} (-1)^{L_1+L_2+\lambda_2} (2k+1) \begin{pmatrix} 1 & 1 & k \\ -\lambda_2' & \lambda_2 & p \end{pmatrix} \begin{Bmatrix} k & L_2 & L_2 \\ L_1 & 1 & 1 \end{Bmatrix} \\
& \times \sum_{abp'KQ} (-1)^{K+a} \sqrt{2K+1} (2b+1) \begin{pmatrix} K & b & k \\ p' & \lambda_1-\lambda_1' & a \end{pmatrix} \begin{pmatrix} b & 1 & 1 \\ \lambda_1-\lambda_1' & \lambda_1' & -\lambda_1 \end{pmatrix} \begin{Bmatrix} K & b & k \\ L_3 & 1 & L_2 \\ L_3 & 1 & L_2 \end{Bmatrix} \\
& \times \langle T(2)_{KQ} \rangle \times \mathbf{D}(0\theta\phi)^{(k)}_{pq} \times \mathbf{D}(0\alpha\beta)^{(K)}_{p'Q} \times \mathbf{D}^*(0\alpha\beta)^{(k)}_{aQ} ,
\end{aligned} \tag{4}$$

where λ_1 and λ_2 are the helicities of the first (γ_1) and second (γ_2) photon, respectively, $L_3 = 2$, $L_2 = 1$, and $L_1 = 0$ are the angular momenta of the excited He($3\,^1D$), the intermediate He($2\,^1P$), and the He($1\,^1S$) ground state, respectively, **D** is a rotation matrix as defined by Edmonds [13], and (α,β) and (θ,ϕ) are polar and azimuthal emission angles of the first and second photons, respectively. The coincident two-photon polarization correlations are derived by integrating Eq. (4) over the direction of the outgoing electron. The following explicit expressions for the geometry used in the present experiment are then obtained:

$$IP_1=\frac{3\sin^2(\beta-\phi)}{4\sqrt{5}}\langle T(2)_{00}\rangle-\frac{39-3\cos 2(\beta-\phi)}{4\sqrt{14}}\langle T(2)_{20}\rangle+\frac{54-9\cos 2(\beta-\phi)}{4\sqrt{70}}\langle T(2)_{40}\rangle \tag{5a}$$

and

$$IP_2=IP_3=0\,, \tag{5b}$$

in excellent agreement within statistical uncertainties with the above experimental result, and where the (unpolarized) intensity I is given as

$$I=\frac{81+3\cos 2(\beta-\phi)}{8\sqrt{5}}\langle T(2)_{00}\rangle-\frac{27+3\cos 2(\beta-\phi)}{4\sqrt{14}}\langle T(2)_{20}\rangle+\frac{18+9\cos 2(\beta-\phi)}{4\sqrt{70}}\langle T(2)_{40}\rangle\,. \tag{5c}$$

Using Eqs. (5a) and (5c) and performing measurements at (at least) two different azimuthal angles for $\beta-\phi$, we can extract all the state multipoles $\langle T(2)_{K0}\rangle$ and up to rank $K=4$. To determine the state multipoles $\langle T(2)_{00}\rangle$ and $\langle T(2)_{20}\rangle$ with rank $K=0$ and 2, respectively, a two-photon coincidence experiment is unnecessary, however. These two multipoles are more easily and more precisely obtained from the noncoincident intensity $I^{(\gamma_1)}$ and linear polarization $P_1^{(\gamma_1)}$ of the first photon γ_1, for which we have

$$I^{(\gamma_1)}P_1^{(\gamma_1)}=-(3\sqrt{14}/8)\langle T(2)_{20}\rangle \tag{6a}$$

and

$$I^{(\gamma_1)}=\sqrt{5}\langle T(2)_{00}\rangle-(\sqrt{14}/8)\langle T(2)_{20}\rangle\,. \tag{6b}$$

Combining Eqs. (5) and (6) we may, hence, completely determine all three state multipoles describing the excited He($3\,^1D$) state. Since the determination of $\langle T(2)_{00}\rangle$ requires the measurement of an absolute cross section for He($3\,^1D$) excitation which was not attempted here, we present the normalized state multipoles $\langle t_{K0}\rangle=\langle T(2)_{K0}\rangle/\langle T(2)_{00}\rangle$. Results for these relative multipoles are shown in Table I. Note that the range of the relative multipoles is $\pm\sqrt{\frac{10}{7}}$ for $\langle t_{20}\rangle$ and $-2\sqrt{\frac{10}{35}}$ to $+3\sqrt{\frac{10}{35}}$ for $\langle t_{40}\rangle$. The present results, for the first time, yield a $\langle T(2)_{40}\rangle$ multipole which is different from zero and positive, in the range of energies investigated here.

Further insight in the collision process may be provided by relating the state multipoles $\langle T(2)_{K0}\rangle$ to the partial cross sections σ_m for excitation of the He($3\,^1D_m$) substates ($m=0,\pm1,\pm2$),

$$\begin{aligned}\langle T(2)_{00}\rangle&=\sqrt{\tfrac{1}{5}}(\sigma_0+2\sigma_1+2\sigma_2)\,,\\ \langle T(2)_{20}\rangle&=-\sqrt{\tfrac{2}{7}}(\sigma_0+\sigma_1-2\sigma_2)\,,\\ \langle T(2)_{40}\rangle&=\sqrt{\tfrac{2}{35}}(3\sigma_0-4\sigma_1+\sigma_2)\,,\end{aligned} \tag{7}$$

where for symmetry reasons the relation $\sigma_m=\sigma_{-m}$ holds [4]. In Fig. 3 we display the normalized (using $\sigma_0+2\sigma_1+2\sigma_2=1$) partial cross sections σ_m. As it turns out, the cross section σ_0 is by far the largest while σ_2 is smallest and even zero (within error bars) at low incident energies. This energy dependence is expected, since at threshold we expect $\sigma_0=1$ and $\sigma_1=\sigma_2=0$ (Ref. [14]). The theoretical calculations based on various models [distorted wave Born approximation with excited state distorting potentials (DWBA-EP), Ref. [15]; 22-state second-order diagonalization method, Ref. [16]; 10-state eikonal calculations, Ref. [17]; multichannel eikonal theory (DMET), Ref. [18]] are in qualitative agreement with this behavior. On a more quantitative basis, all calculations predict values for σ_2 which are too large. They overestimate the contributions from that part of the electron charge cloud which is aligned perpendicular to the direction of the incident electron and which becomes excited in those collisions that involve large momentum transfers. The present results thus put stringent test on existing theoretical calculations and are, therefore, helpful in obtaining a deeper understanding of these fundamental collision processes.

TABLE I. The coincident two-photon [P_1, Eq. (5)] and the noncoincident one-photon [$P_1^{(\gamma_1)}$, Eq. (6)] polarization correlation and the extracted normalized state multipoles $\langle t_{K0}\rangle=\langle T(2)_{K0}\rangle/\langle T(2)_{00}\rangle$ ($K=2$ and 4) for different incident energies E and azimuthal angles ϕ.

E (eV)	ϕ (deg)	P_1	$P_1^{(\gamma_1)}$	$\langle t_{20}\rangle$	$\langle t_{40}\rangle$
40	180	0.390 ± 0.044	0.491 ± 0.019	-0.936 ± 0.043	0.301 ± 0.299
60	90	0.627 ± 0.108	0.423 ± 0.008	-0.785 ± 0.017	0.575 ± 0.364
81.6	90	0.458 ± 0.058	0.336 ± 0.018	-0.603 ± 0.038	0.234 ± 0.183

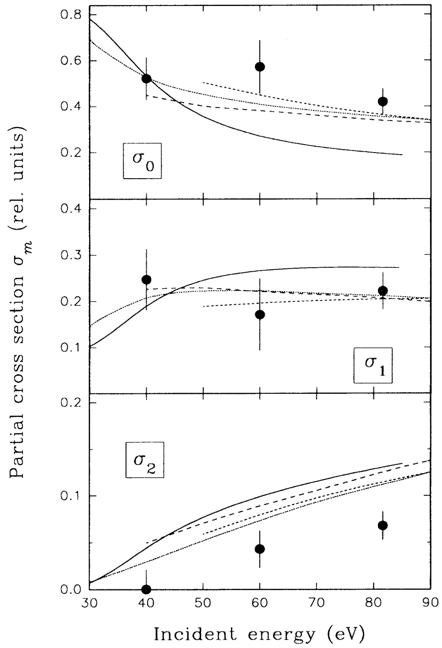

FIG. 3. The relative partial cross sections σ_m ($m = 0,1,2$) vs incident energy. The present results (●) are compared with theoretical calculations using the DWBA-EP (solid lines, Ref. [15]), 22-state second-order diagonalization (long-dashed lines, Ref. [16]), 10-state eikonal (short-dashed lines, Ref. [17]), and full DMET (dotted lines, Ref. [18]) methods.

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