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Measurement of the spin-exchange amplitudes in the electron impact excitation of the $3^{3}P$ state of helium

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Circular and linear polarization correlation measurements have been carried out for the $3^{3}P$ state of HeI at an incident electron energy of 60 eV and for electron scattering angles from 35° to 70°. The scattering parameters λ and χ for this spin-exchange excitation process are derived from the complete set of Stokes parameters, and total polarization (|P|=1) and coherence $(|\mu|=1)$ are confirmed. Within the experimental accuracy the measured values agree with recent calculations by Cartwright and Csanak.

For more than ten years electron-photon angular and polarization correlation measurements have played a key role in the advance of our knowledge of electron-impact-excitation processes not only through the experimental results themselves but also through the stimulation of theoretical work in this area. The first angular-correlation measurements were reported by Kleinpoppen and co-workers¹⁻³ and the first polarization correlation measurements by Standage and Kleinpoppen⁴ who carried out a complete circular and linear Stokes parameter analysis of the 3¹P state of helium. The basic theory of the electron-photon coincidence method was largely developed by Macek and Jaecks, ⁵ Fano and Macek, ⁶ and Blum and Kleinpoppen. ⁷ More recent experimental and theoretical work was summarized by Blum and Kleinpoppen, ⁸ Blum, ⁹ Slevin, ¹⁰ and Andersen, Gallagher, and Hertel. ¹¹

Much of the pioneering work has concentrated on the excitation of ${}^{1}P$ states of helium, though measurements have now been extended to a large number of other excited states from H to Hg (see, e.g., Ref. 10). For light atoms, governed by LS coupling, all these measurements were carried out on excited states of the same multiplicity as the ground state. These states can be reached both by direct and exchange excitation and it is not possible to separate the direct and exchange contributions to the measured overall scattering amplitudes. Even though the direct processes are thought to be dominant in most conditions, some uncertainty remains since the measured results have to be compared with the sum (including interference effects) of the two calculated contributions.

A number of experimental reasons like long lifetimes and considerable depolarization of the emitted light through fine-structure coupling so far precluded measurements on light systems to be extended to excited states with different multiplicity from the ground state, e.g., to triplet states in systems with a singlet ground state. As long as LS coupling holds these states can only be excited by electron exchange processes so that the pure exchange amplitudes could be measured and compared with theory. First measurements on triplet states are being carried out now on the H₂ molecule^{12,13} and on the more transparent triplet system of the He atom.^{14,15} We report here details of a complete polarization correlation measurement on the $3^{3}P$ state of He over an extended range of scattering angles.

The electron impact excitation of the $3^{3}P$ state of helium is accompanied by the emission of light of the wavelength 388.9 nm,

As indicated, the process can be divided with good approximation into three independent stages: collisional excitation process ($\approx 10^{-16}$ s), fine-structure coupling into the three $3^{3}P$ states (≈ 0.1 nsec), and decay (≈ 100 nsec). The fine-structure coupling causes a reduction of the polarization of the decay light, but since the coupling is complete by the time the decay is detected, a correction can be applied to take account of the depolarization. However, the accuracy of the corrected polarization results is reduced by the depolarization, so that the measurements are extremely time consuming (nearly 1 yr for the data reported below).

In the experiment the scattered electrons and the photons are detected in coincidence in the usual way. Electrons pass a 127° monochromator and move along the z axis to cross the helium beam emanating from a long 0.5mm-diam capillary. Electrons which are scattered by an angle θ_e (measured towards the +x axis) and which have lost the energy to excite any of the n=3 states pass through a 127° analyzer and are detected by a channeltron. The photons are detected by a photomultiplier at right angles to the scattering plane in the +y direction,

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and an interference filter for $\lambda = 389$ nm restricts the coincidence signal to the $3^{3}P$ state. The linear polarization of the light is analyzed by a linear polarizer whose angle α , with respect to the z axis, is measured in the same way as θ_e . For the circular polarization analysis a $\lambda/4$ plate with its fast axis parallel to z is inserted in front of the linear polarizer. The time correlation spectrum on the multichannel analyzer shows a "coincidence" peak with a sharp onset determined by the time resolution of the apparatus of a few nanoseconds and a long tail determined by the lifetime of the $3^{3}P$ state of approximately 100 nsec. The signal shape may be used for a cascade-free measurement of the lifetime of the $3^{3}P$ state.^{14,16} A time-toamplitude conversion range of 800 nsec was used and, after subtraction of the random coincidence background, the true coincidence signal was normalized to the total number of electrons which caused valid starts in the timeto-amplitude converter. For each value of θ_e , 8-12 runs of approximately one day each were taken for different values of the linear polarizer angle α , and the normalized coincidence rates were fitted to the function

$$F(\alpha) = A(1 + B\cos 2\alpha + C\sin 2\alpha)$$

where B and C correspond to the Stokes parameters P'_1 and P'_2 , respectively. The measurements and the fit were then repeated with the $\lambda/4$ plate in place when B and C correspond to the Stokes parameters P'_1 and P'_3 , respectively (the spectroscopic definition of the handedness of circularly polarized light is used). Using relations given by Blum and Kleinpoppen⁸ the measured Stokes parameters P'_{ν} can be linked to the "original" Stokes parameters P_{ν} , corrected for the depolarization caused by the finestructure coupling, and to the scattering parameters λ and χ :

$$P_{1}' = (I_{0} - I_{90})/(I_{0} + I_{90}) = \frac{15}{41}P_{1} = \frac{15}{41}(2\lambda - 1), \quad P_{2}' = (I_{45} - I_{135})/(I_{45} + I_{135}) = \frac{15}{41}P_{2} = -\frac{30}{41}\sqrt{\lambda(1 - \lambda)}\cos\chi$$
$$P_{3}' = (I_{RHC} - I_{LHC})/(I_{RHC} + I_{LHC}) = \frac{27}{41}P_{3} = \frac{54}{41}\sqrt{\lambda(1 - \lambda)}\sin\chi$$

where I_0 , I_{45} , I_{90} , I_{135} are the normalized coincidence rates for linear polarizer angles $\alpha = 0^{\circ}$, 45°, 90°, 135° and I_{RHC} , I_{LHC} the normalized coincidence rates for rightand left-hand circularly polarized light (using the spectroscopic definition).

Only the spin-exchange amplitudes $g_{0,1}$ (for excitation of the substates with $M_L = 0, 1$) contribute to the excitation of the ³P state so that $\lambda = |g_0|^2/(|g_0|^2 + 2|g_1|^2)$ and χ is defined by $g_1 = |g_1|e^{i\chi}, g_0 = |g_0|$.

The depolarization factors show that the maximum possible linear polarization for the decay of the ${}^{3}P$ state is 0.37 and the maximum possible circular polarization 0.66 compared to 1.0 in both cases for the measurements on the ${}^{1}P$ state.

The measured Stokes parameters P'_{ν} for the electronimpact excitation of the 3 ³P state of helium at 60 eV are listed in Table I and shown in Fig. 1 for a range of scattering angles. The values shown for P'_1 are the average of the results from linear and circular polarization measure-

TABLE I. Stokes parameters for the HeI $3^{3}P-2^{3}S$ photons measured in coincidence with the scattered electrons. E = 60 eV.

θε	<i>P</i> ['] ₁	P'2	P'3	
35	-0.15 ± 0.18	-0.34 ± 0.28	-0.35 ± 0.18	
37.5	0.05 ± 0.11	-0.09 ± 0.17	-0.13 ± 0.16	
40	0.11 ± 0.14	$+0.01 \pm 0.14$	-0.40 ± 0.20	
45	0.00 ± 0.09	-0.05 ± 0.09	-0.56 ± 0.17	
50	0.10 ± 0.05	-0.18 ± 0.07	-0.43 ± 0.09	
55	0.10 ± 0.06	-0.14 ± 0.08	-0.39 ± 0.08	
57.5	0.10 ± 0.08	-0.09 ± 0.13	-0.63 ± 0.12	
60	0.17 ± 0.08	-0.09 ± 0.09	-0.52 ± 0.12	
62.5	0.08 ± 0.08	-0.07 ± 0.08	-0.37 ± 0.13	
65	0.16 ± 0.08	-0.12 ± 0.09	-0.56 ± 0.13	
70	0.18 ± 0.08	-0.14 ± 0.11	-0.51 ± 0.13	



FIG. 1. Stokes parameters of the He I $3^{3}P-2^{3}S$ (388.9 nm) photons measured in coincidence with the scattered electrons following spin exchange excitation from the ground state of helium. E = 60 eV. Full lines are theoretical results by Cartwright and Csanak (Ref. 17).

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ments. The present values are compared with the theoretical results of Cartwright and Csanak¹⁷ (derived from their Figs. 1 and 2) and show general agreement within the error bars. The negative value of the circular polarization parameter P'_3 proves that the orientation of the atom and thus the angular momentum transfer during the collision are positive for these scattering angles as in the case of the ¹P state excitation. No change of sign of P_3 was found within the angular range used, in agreement with the calculations¹⁷ which indicate a sign change at $\theta_e = 105^\circ$. At $\theta_e \approx 60^\circ$ the measured P_3 (corrected for the depolarization) nearly reach the value -1.0. The calculations¹⁷ show a minimum of $P_3 \approx -1$ at $\theta_e = 70^\circ$.



Scattering angle (degrees)

The measured Stokes parameters in Table I were used to calculate the scattering parameters λ and χ shown in Fig. 2. Within the range 0 to -2π adopted for χ , P_2 , and P_3 each allow two possible values of χ in line with the symmetry of the sine and cosine functions. Only two of these four results are consistent with each other and the average of these is shown in Fig. 2(b). The present values of the phase angle χ for spin-exchange excitation are quite considerable and not too different from those found for $3^{1}P$ state excitation.¹⁸ Within the error bars the values for λ and χ are in agreement with the theory by Cartwright and Csanak.¹⁷

The total polarization $P = (P_1^2 + P_2^2 + P_3^2)^{1/2}$ and the coherence parameter $|\mu| = |(P_2 - iP_3)|/(1 - P_1^2)^{1/2}$ are shown in Fig. 3 and confirm complete polarization and coherence for the spin-exchange process.

Instead of λ and χ , alternative parameters have recently been used.^{11,19} These are P_3 , the degree of circular polarization P_l , the degree of linear polarization, and γ , the alignment angle of the electron cloud with respect to the zaxis. γ is the same as θ_{\min} introduced by Eminyan and co-workers.¹⁻³ If the total polarization can be taken to be 1, P_3 and γ are sufficient to describe the excitation process completely. According to the calculation of Cartwright and Csanak¹⁷ who used this two-parameter approach, γ tends to change sharply in the vicinity of scattering angles where $|P_3|$ to close to 1. As shown in Fig. 2(c) for 60 eV, γ is expected to change sign rapidly at $\theta_e = 68^\circ$. This is near the maximum θ_e used in the present experiment which was completed before the calculations became known. No obvious indication of a sign change is found,



FIG. 2. Scattering parameters λ , χ , and γ as a function of the scattering angle for $3^{3}P$ state excitation in helium. E = 60 eV. Full lines are theoretical results by Cartwright and Csanak (Ref. 17).

FIG. 3. Total polarization and coherence parameters for electron-impact excitation of the $3^{3}P$ state of helium as a function of the scattering angle. E = 60 eV.

but further measurements are required and are being carried out to establish the behavior in this range more accurately.

An earlier *R*-matrix calculation of the 3 ${}^{3}P$ state excitation parameters by Fon *et al.*²⁰ may also be considered in this context even though it does not cover the energy of 60 eV. The λ and χ parameters for 81.63 eV appear not to

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differ substantially from the values shown in Fig. 2. However, the sign change of γ occurs at a lower scattering angle ($\theta_e = 60^\circ$) and the transition is much slower than calculated by Cartwright and Csanak.¹⁷

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