Coherent Excitation of the 4^1F State of Helium by Electron Impact

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(Received 9 December 1997)

The Stokes parameters P_1, P_2, P_3, and P_4 have been measured in a scattered electron-polarized cascade photon correlation study of the 4^1F state of helium excited by 29.6 eV electron impact. This is the first detailed experimental study of electron impact excitation leading to the transfer of three units of angular momentum. The data are expected to be influenced by the breakdown of LS coupling for $L \geq 3$ states, even in a simple system like helium, providing a further challenge for theory. [S0031-9007(98)05736-6]

PACS numbers: 34.80.Dp

Excitation studies of low-lying states of the helium atom by electron impact have played a key role in the development of both experimental and theoretical techniques for the study of electron-atom scattering. Experimentally, helium provides a simple, stable beam of atoms. Theoretically, it is the simplest atom on which to carry out "perfect scattering experiments" in the sense originally discussed by Bederson [1,2]. In these perfect scattering experiments in which the kinematics of the collision are defined, a complete quantum mechanical description of the process is possible in terms of the complex scattering amplitudes or the shape and dynamics of the excited state. In a light two-electron target this is possible without the need for spin-polarized beams. The lowest angular momentum states can be considered to be pure Russell-Saunders coupled states, while a further simplification comes from the lack of hyperfine structure in helium.

For P states in helium, complete experiments have been available since the pioneering scattered electron photon angular correlation measurements of Eminyan *et al.* [3] and the scattered electron-polarized photon correlation measurements of Standage and Kleinpoppen [4]. However, only a new generation of theoretical methods, convergent close coupling (CCC) [5] and R matrix with pseudostates (RMPS) [6], give an accurate description of the excitation over a wide range of kinematic conditions. For S-D excitation, complete information cannot in principle be obtained from observation of a single electric dipole decay. However, a recent analysis by Andersen and Bartschat [7,8] has demonstrated that the missing information can either be extracted from a triple coincidence experiment involving the cascade P-S photons [9] or, more simply, by taking advantage of the quality of the CCC calculations [10,11].

By contrast to these "perfect experiments" completed for $2, 3^1P$, 3^3P, and $3^{1,3}D$ states of helium, no correlation measurements have been made for electron impact excitation of F, or higher angular momentum, states. We present here the first results of a scattered electron–polarized photon coincidence experiment.

There are new and intriguing aspects of F state excitation which have provoked us to carry out this study. First, given that prior to the introduction of the CCC method [5], theories which were in reasonable accord with P state excitation data could not reproduce corresponding D state results, a study involving the transfer of three units of angular momentum may provide an even greater challenge for theoretical models. Second, when $L \geq 3$, LS coupling breaks down and states arising from the $1s4f$ configuration must be described as mixed character states rather than pure singlet and triplet states. For example, using the notation of van Raan and Heideman [12],

$$|n^1F_3\rangle = \frac{|n^1F_3^0\rangle + \omega|n^3F_3^0\rangle}{(1 + \omega^2)}, \qquad (1)$$

where F^0 is a pure LS coupled state. For the $(1s4f)$ configuration, the mixing is large ($\omega = 0.43$, [13]). This mixing provides a further theoretical challenge in describing the excitation. For simplicity, the excited state is referred to as 4^1F throughout this paper.

A theoretical analysis of a coherence experiment of the type reported here, together with the predictions of a Distorted wave Born approximation, have been given by Wang *et al.* [14]. Andersen and Bartschat [8] have also performed a preliminary analysis of the electron excitation of F states relevant to coherence measurements.

Experimental studies of electron impact excitation of the $1s4f$ states of helium are complicated by their low cross sections and by their close proximity to the other $n = 4$ states which cannot be resolved within the energy resolution of electron impact spectrometers. The radiation emitted in the direct deexcitation process, 4^1F-3^1D, 4^3F-3^3D, have long wavelengths, $\lambda = 1870$ and 1869 nm, respectively, and cannot be detected as single particles. Hence information on decay of the F states can only be obtained from the $3D$-$2P$ or $2P$-$1S$ cascade radiation. A further complication affecting the quality of coincidence data comes from the relatively long lifetime of the 4^1F state, 67 ± 10 ns [15]. These difficulties explain the absence of correlation data for the $1s4f$ states so far. The only previous experimental

0031-9007/98/80(14)/3033(4)$15.00

investigation of electron impact excitation of F states is a determination of apparent cross sections from observed cascade contributions of these states to excitation of the $3^{3,1}D$ states in time resolved experiments [16,17].

In the present study we used a polarization correlation method based on the coincident detection of scattered electrons with an energy loss corresponding to excitation of the $n = 4$ group of states and the 667.8 nm, 3^1D-2^1P, cascade photons whose polarization is readily analyzed. States and transitions, as well as lifetimes and branching ratios of interest, are shown in Fig. 1.

A schematic diagram of the correlation experiment is shown in Fig. 2. The momenta of the incident, $\mathbf{k}_{\text{in}}$, and scattered, $\mathbf{k}_{\text{out}}$, electrons define a scattering plane with respect to which the process conserves reflection symmetry. Therefore, within the natural coordinate frame with quantization axis perpendicular to this plane, for S-F excitations only the $M_L = -3, -1, +1$, and $+3$ substates will have nonzero excitation amplitudes. The complete set of quantities one should aim to determine will therefore consist of three relative amplitude sizes and three relative phases in addition to the differential cross section. A complete determination of these amplitudes, involving multicoincidence experiments with the scattered electron and the direct and cascade photons, is unrealistic at present.

Details of our polarization correlation experiment, as used in the study of helium 3D states, have been given by Donnelly *et al.* [18] and by Fursa *et al.* [10]. Briefly, electrons from an oxide cathode are focused into a narrow parallel beam by a set of electrostatic lenses. In the interaction region the electron beam is crossed by the atomic beam at right angles. Electrons scattered through a variable angle θ are energy analyzed and detected by a channel electron multiplier. Photons emitted perpendicular to the scattering plane are analyzed for their state of polarization, wavelength selected by an interference filter, and then detected by a photomultiplier. Pulses from the electron detector start the ramp of a time-to-amplitude converter while those from the photomultiplier stop it. The polarization of the radiation is determined from measurements of the Stokes parameters P_1,P_2,P_3. The linear polarizations are defined by

$$I_z P_1 = I_z(0°) - I_z(90°), \tag{2}$$

$$I_z P_2 = I_z(45°) - I_z(135°), \tag{3}$$

and the circular polarization P_3 by

$$I_z P_3 = I_z(\text{RHC}) - I_z(\text{LHC}), \tag{4}$$

where $I_z(\alpha°)$ is the intensity transmitted by a linear polarizer with transmission axis at an angle α to the incident electron beam direction and I_z is the total photon intensity in the z direction. RHC (LHC) refer to the handedness (right or left) of the circular polarization. Rotation of the polarization components, data accumulation, and determination of Stokes parameters are automated using a personal computer.

In general, the charge cloud would be expected to have a nonzero height along the z direction. To provide information on this component a linear polarization measurement of P_4 defined by

$$I_y P_4 = I_y(0°) - I_y(90°) \tag{5}$$

has been measured in the scattering plane using a similar photon analysis system to that in the z direction.

Considerable care was taken to optimize the coincidence spectrometer performance for these new and demanding F states studies. The long data accumulation times necessary due to the low true coincidence rates and unfavorable true to random ratios were possible only as a result of the stable operation of the spectrometer under software control.

It is also necessary to demonstrate that the observed signal does indeed arise from excitation of the 4^1F state without any significant contribution from other $n \geq 4$ states unresolved by the electron spectrometer. The energy resolution of our system ($\leq$400 meV) was sufficient to exclude signal from direct electron excitation

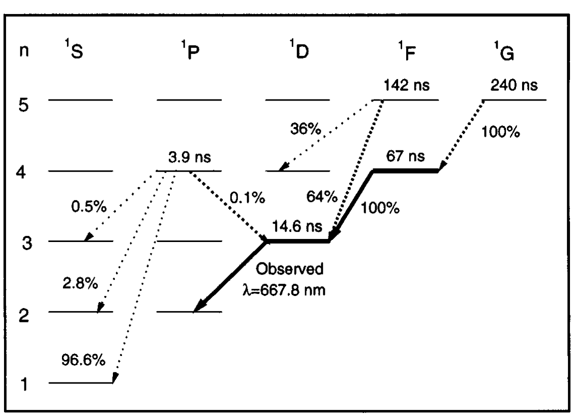

FIG. 1. Energy levels and transitions relevant for present study of excitation of the $1s4f(^1F)$ state. Branching ratios are indicated as percentages and lifetimes in ns.

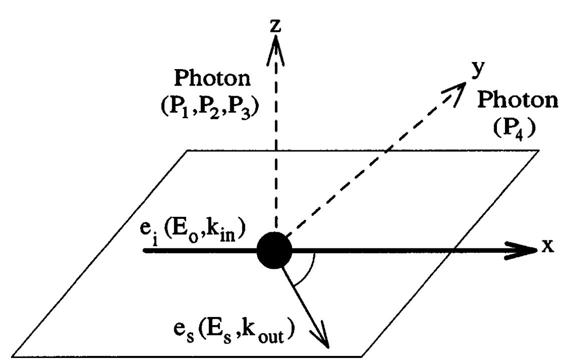

FIG. 2. Schematic illustration of polarization-correlation method.

of the 3^1D state while the $n \geq 5$ states would be detected with reduced efficiency in a wing of the spectrometer transmission function. Many of the $n \geq 4$ states will simply increase the random coincidence signal. States with greatest potential to contribute to the real coincidence signal are those which decay by dipole allowed transitions to the 3^1D state or the 4^1F state and hence contribute to the 3^1D-2^1P observed transition. Of these the 4^1P and $5F, G$ states are of greatest concern.

The contribution from the 4^1P state has been estimated from available cross section data and known branching ratios. An estimate at 30 eV, close to the present incident energy of 29.6 eV, was hampered by the lack of differential cross sections for the 4^1F state at this energy. An upper limit was obtained using differential cross sections from CCC calculations at 30 eV for the 4^1P state and at 40 eV for the 4^1F state [19]. Of utmost importance in this case is the fact that only 0.11% of atoms excited to the 4^1P state decay to the 3^1D state [20] compared with 100% of the excited 4^1F state. On this basis, a 4^1P contribution to the true coincidence signal of 1.5% at a scattering angle of 10° rising to 6% at 40° is obtained. Since the $4F$ differential cross section at 30 eV is likely to be greater than that at 40 eV, the true 4^1P contribution is likely to be even lower.

Because of the considerably shorter lifetime of the 4^1P state ($\tau = 3.9$ ns from A coefficients [20]) and the 3^1D state ($\tau = 14.61$ ns [21]) through which it cascades, compared to the lifetime of the 4^1F state (67 ± 10 ns) [15], it was possible to test the experimental data for the presence of a short lived component from 4^1P excitation. We have compared the values of Stokes parameters obtained from different time windows across the measured time spectra corresponding to varying the relative $4^1P/4^1F$ contribution to the signal. No effects dependent on the time window used were observed confirming that no significant contribution from a short lived component, such as 4^1P, was detected.

The situation is different when $5F$ and $5G$ states, the major possible contributors from the $n \geq 5$ group of states, are considered. In both cases smaller cross section values are expected compared to excitation of the $4F$ state and this is supported by energy loss spectra where a decrease of intensity is observed at an energy loss corresponding to $n = 5$ states. However in the present experiment the $n = 5$ states are observed only in a wing of the electron analyzer transmission function. An estimate based on the Gaussian shape of this function gives a scattered electron transmission efficiency for the $5F$ and $5G$ states which is 24% of that for the $4F$ state. The time distribution of true coincidencies due to the long lifetimes of the $5F$ (142 ns [15]) and $5G$ (240 ns [17]) states can be used to further decrease their possible influence by careful setting of the time window used to determine the true coincidence signal. In principle when a time spectrum is composed of contributions from states with substantially different lifetimes, as in the present case, the contribution of the longer–lived states to the time spectrum can be eliminated by a careful choice of time window, with only a relatively small signal loss from the state of interest [22]. While this technique was not completely optimized in the present study, it leads to an estimated upper limit for the contribution of the longer lived 5^1G state of 3.6%.

The $5F$ state ($\tau = 142$ ns [15]) contribution to the number of true coincidences expected inside the time window used for determination of Stokes parameters is somewhat larger, but then only 64% of all the atoms excited into this state decay to the 3^1D state [20]. Combined with the decrease in analyzer transmission efficiency for $n = 5$ states this gives an estimated maximum contribution of 4.6%. These quoted upper limits of possible contributions from both the $5F$ and $5G$ states are obtained assuming the same cross section values for all three states. As cross sections for excitation of the $5F$ and $5G$ states are expected to be smaller than that for the $4F$ state, the real contributions will be significantly smaller.

Negligible contributions arise from the $n \geq 5^1P$ and $n \geq 6^1F, {}^1G$ states, making the overall contributions from other states to the 4^1F data small in comparison to the statistical uncertainties in the data.

The Stokes parameters P_1, P_2, P_3, and P_4 measured in the present experiment are shown in Fig. 3. The scattering angular range was limited in the cases of P_3 and P_4 by low coincidence counting rates at the larger scattering angles. The alignment angle γ of the charge cloud in the scattering plane [23], defined as

$$\tan 2\gamma = \frac{P_2}{P_1} \tag{6}$$

is also shown in Fig. 3.

It can be seen that nonzero values of all Stokes parameters are found with the exception of P_2. For this reason the alignment angle γ is always consistant with zero. The P_2 values are also consistent with corresponding data for the 3^1D state at 29.6 eV [24]. This is also true for P_1 and P_4 except at the smallest scattering angle of 10° where the P_1 and P_4 values are consistently lower than the corresponding ${}^1D$ results. This result may reflect the mixed nature of the $1s4f$ states. The small angle 3^1D values are consistent with their theoretical value of 0.6 at zero scattering angle, but the F state values of P_1 and P_4 at 10° scattering angle are considerably lower than the value of 0.5 expected at zero scattering angle for a pure ${}^1F$ state [26]. P_3 values are closely related to the angular momentum transfer in the excitation process, so significant differences observed in the P_3 values for 3^1P [25], 3^1D [24], and 4^1F states are to be expected.

In conclusion, we believe this new data on excitation of a simple atom involving the transfer of three units of angular momentum presents a new challenge for recently

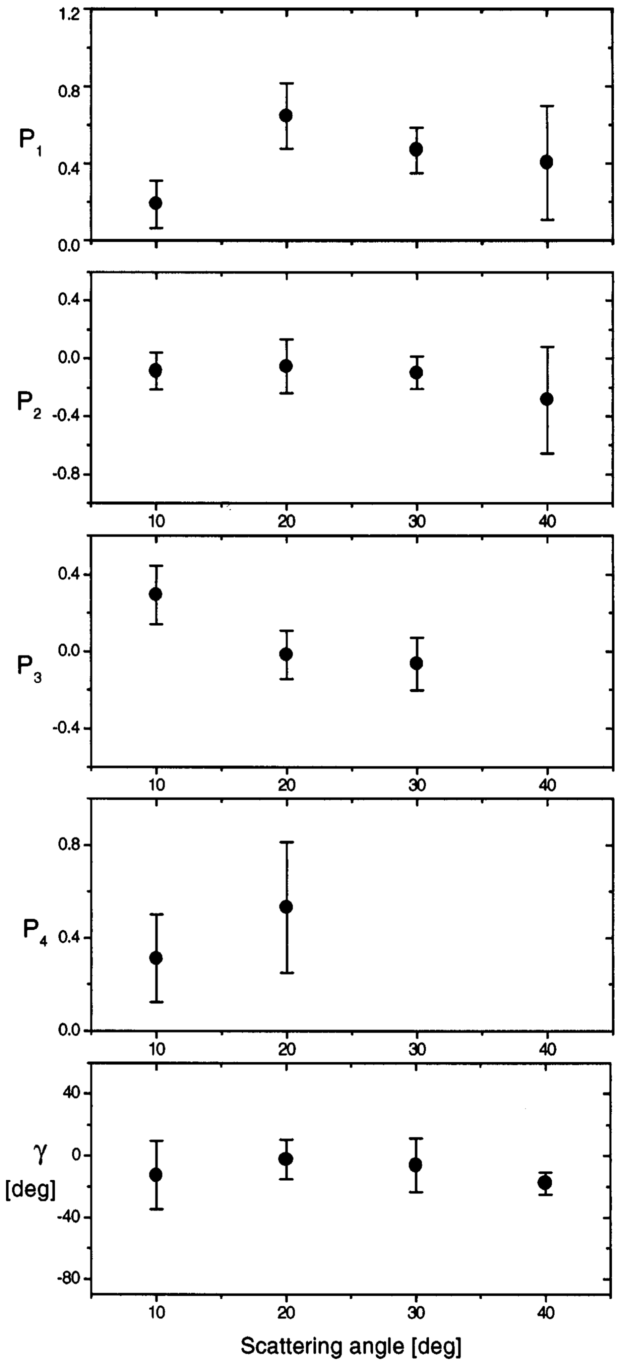

FIG. 3. Stokes parameters P_1, P_2, P_3, and P_4 and the alignment angle γ for the $1s4f(^1F)$ state excited by 29.6 eV electrons.

developed theories which have been highly successful in describing data for lower angular momentum states. In particular, it provides an unusual opportunity to consider the breakdown of *LS* coupling without the complications of a multielectron target system.

This work is supported by the United Kingdom Engineering and Physical Sciences Research Council. We thank Dr. Igor Bray for unpublished data.

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