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Anomalous behavior in differential cross sections for the 2 ^3S excitation in helium at minute scattering angles for fast electron collisions

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Differential cross sections (DCS) for the 1 ^1S-2 ^3S excitation in helium have been measured with a high angular resolution of about 1° as a function of the scattering angle θ from 12° to 0° for the incident energies of 200 and 500 eV. For the incident energy of 500 eV, the DCS increases drastically as the angle decreases in the angular range smaller than 4° and reaches a value 0.075 a_0^2/sr at $\theta=0°$, the increase being nearly 3 orders of magnitude. The DCS at zero scattering angle for impact energies from 200 to 800 eV have been measured. The DCS's are found to be about 1 order of magnitude greater than those of published measurements by Skerbele, Harshbarger, and Lassettre [J. Chem. Phys. **58**, 4285 (1973)] and Klump and Lassettre [J. Chem. Phys. **62**, 1838 (1975)].

It is known that differential cross sections (DCS) for excitation of the 1 ^1S-2 ^3S transition in helium show a remarkable forward peaking behavior and have unusually large values at the scattering angle $\theta=0°$ even at high incident energies.[1-4] It is recognized that this behavior is characteristic of the singlet to triplet transition where the orbital term symbols are unchanged in initial and final states as the 1 ^1S-2 ^3S transition. It is in marked contrast to the fact that the DCS's for the 1 ^1S-2 ^3P transition exhibit their greatest values at large scattering angles and low incident energies.[5]

Skerbele, Harshbarger, and Lassettre[2] measured the ratio of the DCS for the transition 1 ^1S-2 ^3S relative to that for the 1 ^1S-2 ^1S at $\theta=0°$ over the incident energy range 300–500 eV. They obtained the ratio 0.0096 $\pm$0.0024 for the incident energy 500 eV, which exceeded all theoretical evaluations then available by at least 2 orders of magnitude. Klump and Lassettre[3] extended similar measurements to the incident energy range 100–300 eV. They deduced the DCS's for the 2 ^3S excitation at $\theta=0°$ from the measured cross-section ratios using the calculated cross sections for the 2 ^1S excitation. The DCS's at $\theta=0°$ over the incident energy range 100–500 eV were compared with results of a calculation by Huo.[6] The calculation was performed applying a high-energy approximation for the second-order terms in the exchange T matrix, which was the only theoretical treatment that gave a reasonable agreement with the experimental data.

In this paper, we present results of the DCS's for the excitation of the 1 ^1S-2 ^3S transition for the scattering angle θ from 12° to 0° at the incident electron energies 500 and 200 eV. We also present the DCS's for the 1 ^1S-2 ^3S excitation for $\theta=0°$ in the incident energy range from 800 to 200 eV. The angular resolution of the electron-energy-loss spectrometer used is about 1°, which is several factors better than that in the previous experiments.[1-4] The calibration of the real 0° is the most essential procedure in this measurement, for which no description has been given in the previous papers.

A schematic diagram of an electron spectrometer used is shown in Fig. 1. It consists of an electron gun, an energy selector, a collision region, a tandem-type energy analyzer, and a channel-multiplier counter. Five sets of the electron lens system are arranged to connect the components of the instrument. A set of apertures of 0.5-mm diameter is installed in the lens system before the analyzer to obtain a desired angular resolution. Simulated hemispherical analyzers, first designed by Jost,[7] of 52-mm mean radius are employed for the energy selector and each component of the tandem analyzer. The conventional constant-resolution mode is used to measure the electron-energy-loss spectra, and the typical energy resolution is 80–100 meV in full width at half maximum (FWHM). The tandem analyzer is useful, especially to minimize the background noises in the spectrum at $\theta=0°$. The energy analyzer is rotatable around the collision center to cover the angular range from 100° to $-30°$. A

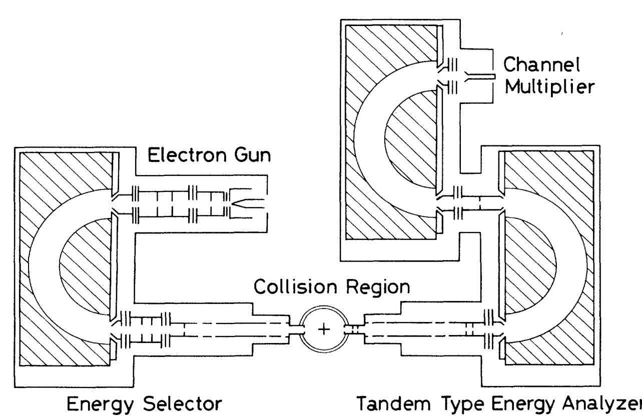

FIG. 1. Schematic diagram of the electron-energy-loss spectrometer with a tandem-simulated-hemispherical analyzer.

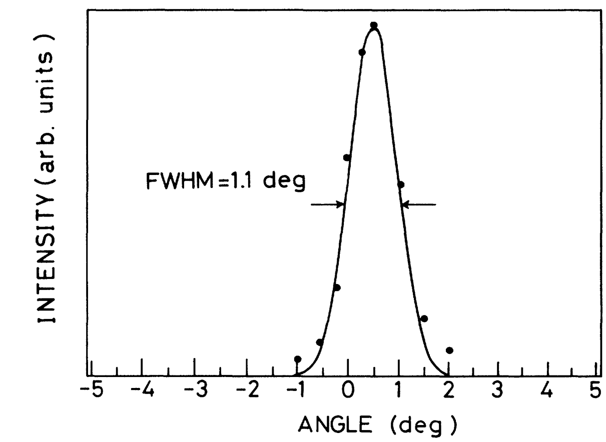

FIG. 2. Typical angular resolution of the apparatus.

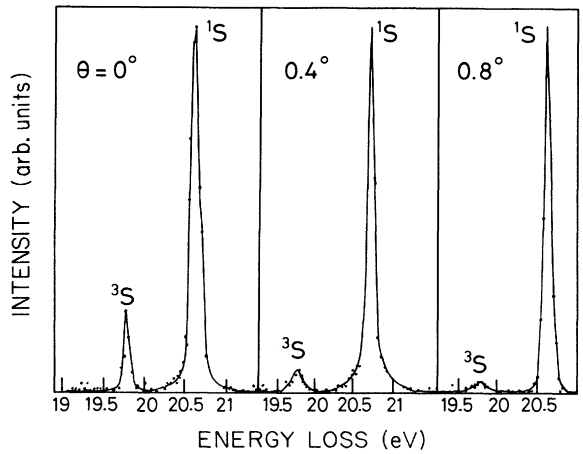

FIG. 4. Typical energy-loss spectra for the $2\,^3S$ and the $2\,^1S$ excitation in helium at scattering angles of 0°, 0.4°, and 0.8° for 500-eV incident electrons. Intensity scale is normalized to the peak for the $2\,^1S$ excitation.

detailed description of the apparatus will be reported in a succeeding paper, together with details in the results of the DCS's for the $2\,^3S$ and the $2\,^1S$ excitations. In addition to this apparatus, another electron spectrometer described in a paper by Li *et al.*[8] was used to measure a part of the DCS's in the angular region larger than several degrees.

The angular resolution of the spectrometer has been examined by measuring the angular distribution of the electron beam of 500-eV incidence directly into the analyzer system. As shown in Fig. 2, the resolution is known to be 1.1° (FWHM). Calibration of the zero scattering angle is performed utilizing the symmetry nature of the scattering intensity ratios for the $2\,^1S$ and the $2\,^3S$ excitations to the intensity of the $2\,^1P$ excitation. Figure 3 shows the intensity ratios $I_{2\,^1S}/I_{2\,^1P}$ and $I_{2\,^3S}/I_{2\,^1P}$ as a function of the geometric angle of the apparatus, telling that the true zero angle is situated at $+0.5°$ on the geometric scale.

To illustrate how drastically the $2\,^3S$ excitation peak decreases as the angle increases compared to the $2\,^1S$ peak, typical energy-loss spectra for 500-eV incident electrons are displayed in Fig. 4 at scattering angles of 0°, 0.4°, and 0.8°. Intensity scale is normalized to the peak for the $2\,^1S$ excitation.

In order to determine the DCS's for the $2\,^3S$ excitation, we measure the intensity ratio $I_{2\,^3S}/I_{2\,^1P}$ on the energy-loss spectrum and normalize it to the absolute DCS for the $2\,^1P$ excitation, which we deduced from the generalized oscillator strengths calculated by Kim and Inokuti.[9]

The measured DCS's for the $2\,^3S$ excitation at the incident energies of 200 and 500 eV are shown in Fig. 5 as functions of the scattering angle. The experimental errors in the DCS's are estimated to be within 15% for 200 eV

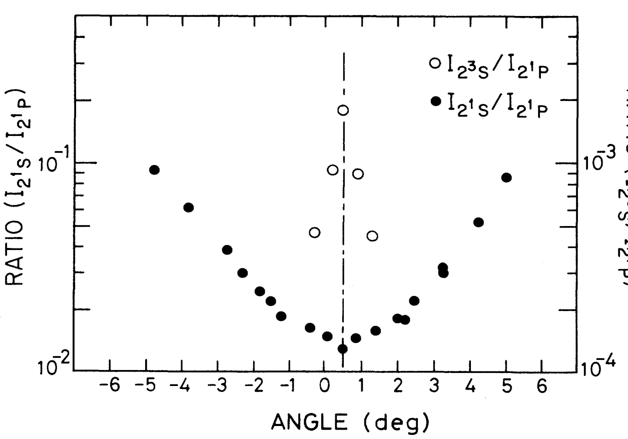

FIG. 3. Calibration of the zero angle position by means of the scattering symmetry of the DCS's for the $2\,^1S$ and the $2\,^3S$ excitation relative to the $2\,^1P$ excitation. The incident electron energy is 500 eV.

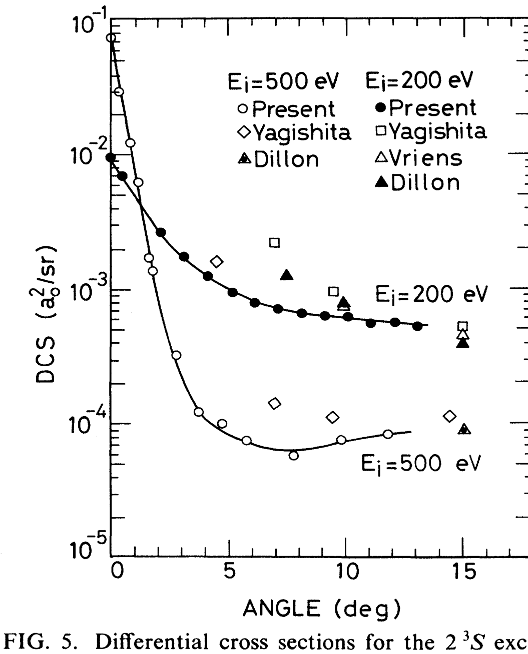

FIG. 5. Differential cross sections for the $2\,^3S$ excitation in helium as a function of the scattering angle for the incident electron energies of 200 and 500 eV.

TABLE I. Intensity ratios and DCS's for the $2\,^3S$ excitation in helium at $\theta=0°$. Corrections due to the limited angular resolution are not applied.

Impact energy (eV)	$I_{2\,^3S}/I_{2\,^1P}$ ($\times 10^{-3}$)	$I_{2\,^3S}/I_{2\,^1S}$ Present	$I_{2\,^3S}/I_{2\,^1S}$ Skerbele	DCS ($10^{-2}\,a_0^2$/sr) for the $2\,^3S$ excitation Present	DCS ($10^{-2}\,a_0^2$/sr) for the $2\,^3S$ excitation Skerbele
200	0.699	0.023	0.0071	0.996	0.18
300	2.10	0.116	0.0164	4.79	0.41
400	2.10	0.129	0.0128	6.57	0.32
500	1.88	0.149	0.0096	7.48	0.23
600	1.42			6.87	
700	1.11			6.31	
800	0.997			6.52	

and 20% for 500 eV, in the angular region larger than 2°, including the normalization error for the absolute DCS determination. Besides the present data, experimental DCS's by Yagishita, Takayanagi, and Suzuki,[10] Vriens, Simpson, and Mielczarek,[1] and Dillon[4] are also plotted in Fig. 5. For the incident energy of 500 eV, the DCS's are very small ($<1\times10^{-4}\,a_0^2$/sr) at scattering angles larger than about 4°; however, they increase drastically as the angle decreases from 4° to 0°; the increase reaches nearly 3 orders of magnitude. The DCS's for the 200-eV incident energy are naturally larger than those for 500 eV in almost the whole range of the scattering angle larger than about 1°. They also increase as the angle decreases, the increase being about 1 order of magnitude between angles of 5° and 0°. It is worth noting, however, that the DCS for the 500-eV incident energy exceeds that for 200 eV at about a 1° angle and reaches a value 0.075 a_0^2/sr at $\theta=0°$, in contrast to a value of 0.0099 a_0^2/sr for the 200-eV energy.

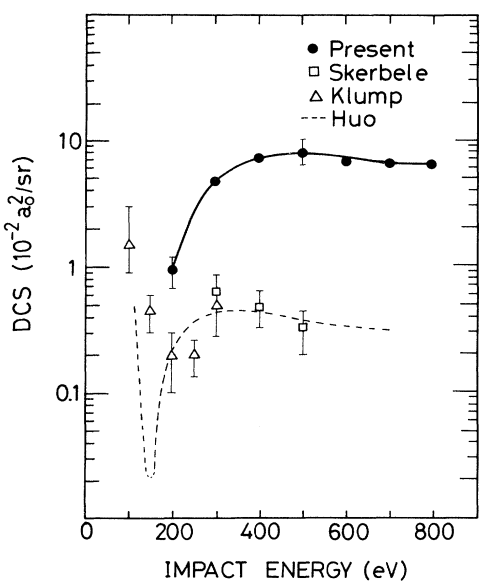

FIG. 6. Differential cross sections for the $2\,^3S$ excitation in helium at $\theta=0°$ as a function of the incident electron energy. Corrections due to the limited angular resolution are not applied.

We have measured the DCS's for the $2\,^3S$ excitation in helium at $\theta=0°$ for the incident energy region between 800 and 200 eV. Numerical data of the intensity ratios $I_{2\,^3S}/I_{2\,^1P}$ and $I_{2\,^3S}/I_{2\,^1S}$ as well as the DCS's for the $2\,^3S$ excitation at $\theta=0°$ are tabulated in Table I together with the data by Skerbele et al.[2] and Klump and Lassettre[3] for a comparison. Figure 6 shows the present DCS's at $\theta=0°$ together with the data of Skerbele et al.[2] and Klump and Lassettre.[3] The present DCS's show nearly constant values above 400-eV energy, which are about 1 order of magnitude larger than those of the previous measurements. The DCS's decrease rapidly as the energy decreases below 400 eV. The decrease in the DCS seems not to cease at 200 eV, suggesting a minimum of the DCS at the energy between 100 and 200 eV, which might be justified if we recall a value of the DCS $3.4\times10^{-2}\,a_0^2$/sr at $\theta=0°$ for 40.1 eV by Trajmar,[11] in addition to the value $1.5\times10^{-2}\,a_0^2$/sr for 100 eV by Klump and Lassettre.[3] A dashed line in Fig. 6 represents a result of a calculation by Huo.[6] Although the calculated values of the DCS do not reproduce those of the present experiment, the curve suggests the variation in the DCS as a function of the impact energy and the existence of a minimum at around the energy of 150 eV.

Accuracies of the measured DCS's are drastically affected by the limited angular resolution of the apparatus, especially when the DCS is a steep function of the scattering angle like the case of the impact energies 400–800 eV. For instance, the DCS for the $2\,^3S$ excitation varies by nearly a factor of 10 from $\theta=0°$ to 1° for 500 eV, therefore the angular resolution of 1° is still quite insufficient. A rough estimation using a procedure discussed in the appendix of the paper by Li et al.[8] gives a result that the measured intensity ratio $I_{2\,^3S}/I_{2\,^1P}$ at $\theta=0°$ should be corrected by a factor 1.67, in other words, a correction +67% should be applied to the measured ratio. On the other hand, a correction to be applied to the same ratio for the 200-eV impact energy is estimated to be +13%; it is due to the difference in the steepness of the DCS vs angle function. If we assume that the angular resolution used in the experiments by Skerbele et al.[2] and Klump and Lassettre[3] was about 3° in FWHM, which is deduced from a description in their paper that their resolution was limited by the use of slits of 3 mm length, the correction factors for the measured ratio $I_{2\,^3S}/I_{2\,^1S}$ at $\theta=0°$ are crudely estimated to be about 5 for the impact energy of 500 eV, and 1.5 for the impact energy of 200 eV. The enormous discrepancy between the present intensity ratios $I_{2\,^3S}/I_{2\,^1S}$ and the previous ones is not fully accounted for by the difference in the angular resolution between the two. We suggest that incorrect determination of the zero angle position, together with poorer signal-to-noise ratios on the earlier work, may have contributed to the discrepancy between that work and the present data.

Recently, Nakazaki *et al.*[12] performed a new calculation of the DCS for the $2\,^3S$ excitation at 100 and 200 eV using the *R*-matrix method. The results, however, do not reproduce the large DCS's at small angles, although they give a good agreement with the experiments at large angles.

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