Generalized oscillator strength for the 3s-3p and 2p-3s transitions in the sodium atom

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The generalized oscillator strengths for the 3s-3p and 2p-3s excitation processes have been obtained from the related differential cross sections, measured at 1-keV impact energy and normalized to the first-Born-approximation (FBA) result for the 3s-3p process at the scattering angle of 2°. Theoretical values for the 3s-3p excitation process were also determined using the Glauber approximation, both considering the interaction of the incident electron with the atomic electrons not directly involved in the excitation process and $(N-1)$ nuclear charges and by neglecting this interaction. FBA results were determined for both the 3s-3p and 2p-3s processes. Theoretical values for the elastic differential cross section were also determined.

I. INTRODUCTION

The electron-impact excitation of the sodium atom has been the object of numerous experimental and theoretical investigations at the low- and intermediate-impact-energy ranges. Sodium is a particularly interesting system because its electronic structure can be treated approximately like a neon atom core with a 3s external electron. The applicability of different theoretical methods to the study of electron-atom collisions can then be adequately investigated and compared to the experimental results.

The differential cross section for the 3s-3p excitation, for example, has been previously determined both experimentally at impact energies lower than 250 eV (Refs. 1–4) and theoretically within the formalism of close-coupling,[5–7] coupled-channel,[8] distorted-wave,[9] and Glauber approximations.[10,11]

For impact energies higher than 250 eV, on the other hand, there are no experimental results for the differential cross section (DCS) for any of the known excitations in this atom, and the available theoretical results are restricted to the first Born approximation (FBA).

The generalized oscillator strength (GOS) obtained through the FBA is expected to match the high-energy experimental results. It has nevertheless been compared to the experimental results only at impact energies lower than 250 eV for the 3s-3p excitation process.[1,2] The data of Buckman and Teubner,[2] for instance, seem to approach the FBA results as the impact energy is raised from 54.4 to 217 eV but show no minimum in the GOS. The results of Shuttleworth, Newell, and Smith[1] at 250 eV do present a minimum in the GOS at the squared momentum-transfer value of $K^2=0.4$ a.u.

Recently[12] the GOS for the 3s-3p transition has been calculated using the close-coupling method; the main intention of the author consisted in discussing the Lassetre limiting theorem for the GOS.[13] The GOS results, obtained at impact energies lower than 150 eV, show the energy-dependence behavior expected in this energy range. At higher impact energies one should not expect such energy dependence for the GOS, at least for low values of the momentum transfer K.

In the present work we investigate both theoretically and experimentally the problem of GOS determination for the sodium atom, using high-impact energies. The electronic excitation from the ground state ($^2S_{1/2}$) to the $1s^2 2s^2 2p^6 3p^1(^2P_{1/2,3/2})$ and $1s^2 2s^2 2p^5 3s^2(^2P_{1/2,3/2})$ states, which we shall call, respectively, the 3s-3p and 2p-3s excitations, is studied. The differential cross section and the generalized oscillator strength for these transitions have been obtained both theoretically and experimentally using 1-keV electrons. The elastic differential cross section (EDCS) has also been determined.

II. APPARATUS AND PROCEDURE

The apparatus used for the present measurements has been described before.[14] It consists basically of a rotatable electron gun, a neutral gas (or vapor) beam, and a Möllensted velocity analyzer fixed on the vacuum chamber wall. A crossed-beam geometry is used. The electron beam, produced by a triode-type electron gun, has a typical beam current of 10 μA and a beam diameter, measured at the scattering region, of approximately 0.5 mm.

The sodium beam was produced using a cylindrical oven of 12-mm internal diameter with an exit channel of 8-mm length and 1-mm diameter. The oven was dc heated and the temperature was typically 673 K. The resistance wire was doubly wound in order to cancel the resulting magnetic field.

The scattered electrons are velocity analyzed by a Möllensted analyzer and detected by an electron multiplier (Spiraltron, Galileo Electro Optics). The energy resolution was set to 0.6 eV, as determined by the full width at half maximum (FWHM) of the elastic peak.

The analyzer viewing cone is determined by two parallel circular apertures of 200- and 50-μm diameter, located, respectively, at 120 and 150 mm from the scattering center. The apertures have been selected so that the acceptance cone includes as little excess of the scattering volume as possible. Corrections to the angular behavior of the cross sections, due to the finite size of the scattering volume, are expected to be negligible in the present work.

The stray magnetic fields in the plane of measurements have been reduced to less than 10 mG in all directions by three pairs of square, orthogonal Helmholtz coils.

The scattering zero angle was checked by measuring the elastic cross section over a 15° range on both the right-hand and left-hand sides.

A very precise positioning mechanism allows the determination of the scattered angle with an accuracy of 0.02°. Nevertheless, the angular resolution, defined by the set of apertures in front of the Möllensted analyzer, is 0.2° in our experiment. The observed count rate was never allowed to exceed 20 kHz in order to eliminate the need for dead-time correction.

The experimental data-acquisition procedure can be described in the following way. First a set of energy-loss spectra was measured, including the elastic peak, for scattering angles from 1° to 30°, with the necessary precautions to assure the stability of the experimental conditions. For each scattering angle the relative values of the area of the elastic peak and of the $3s$-$3p$ peak were determined through a Gaussian fitting procedure. Absolute values for the $3s$-$3p$ DCS were obtained by normalization of the experimental results to the calculated cross section (FBA) at the scattering angle of 2°. From the experimental ratio between the elastic and inelastic peaks at the scattering angle of 2°, absolute values for the elastic cross section were determined.

In order to determine the GOS for the $2p$-$3s$ excitation, a new set of spectra was measured at several scattering angles, spanning an appropriate energy-loss range (0–35 eV) in order to encompass the $3s$-$3p$ and $2p$-$3p$ peaks with good statistics. In order to subtract the contribution of the $3s$ continuum to the $2p$-$3s$ area, a polynominal fitting was employed.

The absolute values for the cross section of the $2p$-$3s$ excitation process were determined from the ratios between the areas of the associated peak and of the elastic peak. The uncertainties are estimated in the following manner. The maximum statistical uncertainty was 3%, as at least 1000 counts were accumulated at the maximum of the least intense inelastic profile, related to the $2p$-$3s$ excitation process, for each scattering angle. Fluctuation in the primary beam current was of the order of 1%. An important source of error in the determination of the scattering intensity is the limited angular resolution (0.2°), which contributes with an uncertainty of about 10% to the elastic and $3s$-$3p$ cross sections and 20% to the $2p$-$3s$ cross section, reflecting the different angular dependence associated with each cross section. The area of the elastic, $3s$-$3p$, and $2p$-$3s$ processes, determined by a Gaussian fitting, is subject to an uncertainty which we estimate to be lower than 10%.

The overall uncertainty δ, defined as $\delta=(\sum_i \delta_i^2)^{1/2}$, is equal to approximately 10% (elastic differential cross section and $2p$-$3s$ GOS) and 20% in the $2p$-$3s$ GOS determination.

III. CALCULATIONS

The calculations in the Glauber approximation[11,15,16] were performed using Hartree-Fock atomic wave functions, assuming the frozen-core approximation, and expanding the monoelectronic orbitals as Slater-type functions.

The interaction between the incident electron with the core electrons and $(N-1)$ nuclear charges was approximated by a Yukawa-type potential, given by

$$V_c(r_1,\ldots,r_{N-1},r_{N+1})=A\exp(-\lambda r_{N+1})/r_{N+1}\ . \quad (1)$$

The parameters A and λ were determined by fitting V_c to a numerical potential determined by a Hartree-Fock-$X\alpha$ method. In order to generate this numerical potential, initially an atomic calculation of the ground state of the sodium atom was performed. The numerical potential was then obtained from the orbitals associated with the $1s^2 2s^2 2p^6$ electronic configuration and ten nuclear charges.

Szasz and McGinn[17] $3s$ and $3p$ Slater-type orbitals were used in the calculations. These were performed in two levels in regard to the interaction between the incident electron with the core electrons and $(N-1)$ nuclear charges. In one case this interaction was considered as described above, and we shall call it the Yukawa frozen-core (YFC) approximation. Calculations were also performed where this interaction was not considered [inert frozen-core (IFC) approximation].

The GOS was determined from the calculated values of the DCS, by using the well-known expression,

$$f_{0,n}(K)=(2k_0/k_f)K^2\Delta E\left[\frac{d\sigma}{d\Omega}\right], \quad (2)$$

where k_0 and k_f are, respectively, the momentum of the incident and scattered electrons, K the transferred momentum, ΔE the excitation energy, and $d\sigma/d\Omega$ the differential cross section.

Calculations within the first Born approximation were also performed for both the $3s$-$3p$ and $2p$-$3s$ excitation processes. In this case the Hartree-Fock atomic orbitals were expanded in a Gaussian basis set [$12s$, $13p$], as determined by Meth and Nascimento.[18] This basis set produces a value of 0.986 for the optical oscillator strength of the $3s$-$3p$ excitation process in good agreement with the experimental result, 0.975.[19–21]

Calculations for the elastic differential cross section were also performed. The Yukawa-type potential for this atom determined by Cox and Bonham[22] was used in the determination of the phase shifts.

IV. RESULTS

We present in Fig. 1 energy-loss spectra for the sodium atom, measured at 2.0° scattering angle. Besides the elastic peak, the $3s$-$3p$ peak centered at 2.0 eV can be clearly

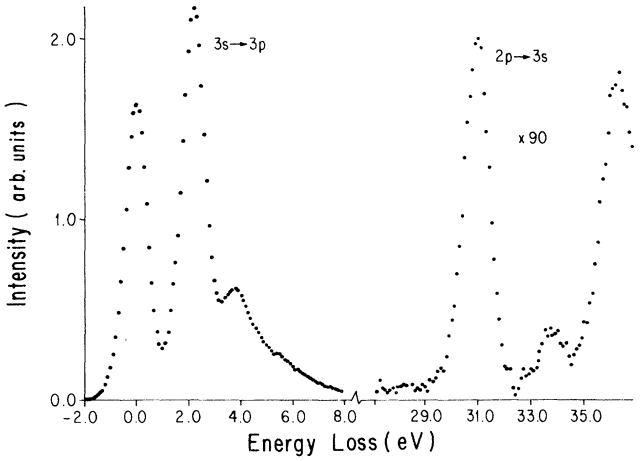

FIG. 1. Electron energy-loss spectrum for the Na atom obtained at 1 keV and 2°.

seen. Another peak, mainly related to the 3s-3d transition, can also be observed at 3.6 eV. We point out that the 3s-3p peak is well separated from the other structures and consequently the determination of its associated cross section is not hampered by the experimental energy resolution. At 31.2 eV one observes the peak related to the 2p-3s excitation process.

In order to discuss the obtained experimental and theoretical cross sections it is important to have in mind that all the experimental results were normalized to the FBA calculated theoretical value of the GOS for the 3s-3p process at the scattering angle of 2°.

As a test of the whole experimental procedure, the elastic differential cross section is compared to calculated values in Fig. 2. In these calculations, the Yukawa-type potential of Cox and Bonham[22] was used in the determination of the phase shifts. A good general agreement is observed between the two sets of results, except for small scattering angles where only a reasonable agreement is obtained. This behavior may possibly be attributed to the fact that no target polarization effects were considered in the calculations.

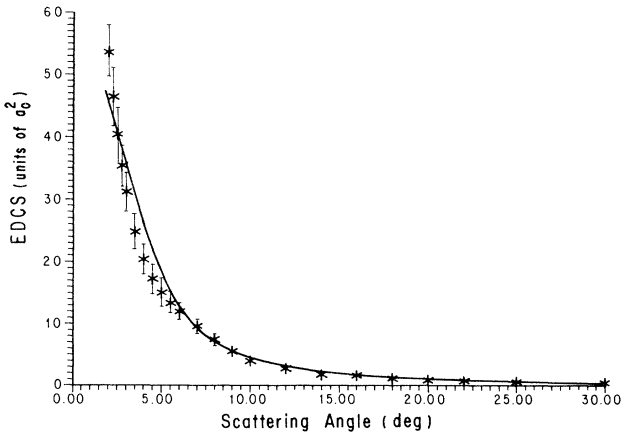

FIG. 2. Elastic differential cross section for the Na atom at 1-keV impact energy (all values from this work). *, experimental; ——, theory.

A. 3s-3p excitations

Figure 3 shows the calculated values for the GOS within the Glauber and first Born approximations, together with our experimental results. The experimental results of Buckman and Teubner[2] at the impact energy of 217 eV, and of Shuttleworth, Newell, and Smith[1] at the impact energy of 250 eV are also presented. The Glauber IFC and YFC results are seen to be practically identical at small scattering angles. This is not true for larger scattering angles, a behavior that can be seen in Fig. 4, where the DCS for this excitation process in the Glauber YFC, IFC, and Born approximation is shown.

Figure 3 shows several interesting features. A very good general agreement is observed between our experimental and theoretical results for small (less than 0.6 a.u.) values of the square of the momentum transfer K^2. In this region the Born and Glauber results agree with each other. For K^2 values between 0.6 and 0.9 a.u. the experimental results slightly disagree with the Born results, but still show a good agreement with the Glauber results, probably because the Born approximation begins to fail in this K^2 region. Clearly no minimum in the GOS is observed in the momentum-transfer region between 0 and 0.9 a.u.

Concerning the other experimental results, Buckman and Teubner[2] present a reasonable agreement with our theoretical and experimental results, the difference being probably due to the lower impact energy (217 eV) used in their work, that is, the Born limit has not been completely attained in their case. The results of Shuttleworth, Newell, and Smith[1] also agree with the present results and the Buckman and Teubner[2] results for K^2 values lower than 0.2 a.u. For higher values of K^2, although the

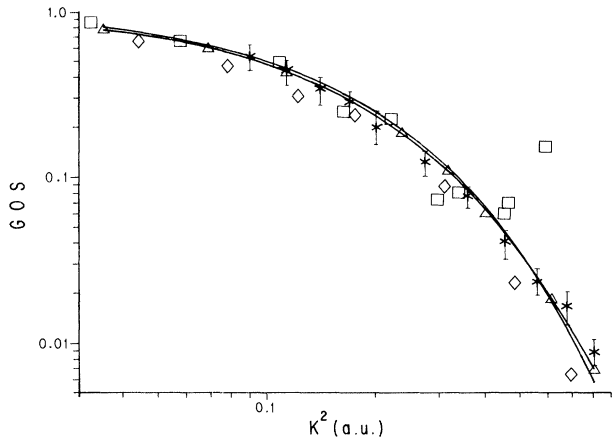

FIG. 3. Generalized oscillator strength (dimensionless units) for the 3s-3p excitation of the Na atom. *, experimental, this work; □, experimental, Shuttleworth, Newell, and Smith (Ref. 1); ◇, experimental, Buckman and Teubner (Ref. 2); ——, FBA, this work; –△–, Glauber approximation, this work.

Shuttleworth, Newell, and Smith[1] results agree within the error bars with both sets of results (except for the last point), they show a minimum in the GOS for this process at $K^2=0.4$ a.u. which is not seen either in our results or in Buckman and Teubner's[2] results.

As to the minimum in the GOS, Shimamura[23] predicted a minimum in the GOS with the FBA using Slater orbitals. He predicted the minimum position to appear between $K^2=0.72$ and 0.93 a.u., depending on the choice of the exponent on the Slater orbitals. In the present FBA calculations, the first local minimum in the GOS is obtained only at $K^2=2.0$ a.u. This corresponds, for 1-keV impact electrons, to a scattering angle larger than 10° where, as discussed below, the FBA is not expected to properly describe the collision process anymore. One should also expect our FBA results to be more precise than the previously available results[23] once we have used more precise target wave functions, that is, self-consistent-field Hartree-Fock wave functions.

In order to discuss the behavior of the theoretical results for larger scattering angles, Fig. 4 shows the values for the DCS calculated within the Glauber YFC and IFC and with FB approximations. One observes a good agreement between the Glauber and Born results for small scattering angles, but an increasing disagreement for larger scattering angles. This situation, which has also been observed before,[16,24,25] shows clearly that for 1-keV impact energy, higher-order corrections to the first Born approximation are necessary in order to properly describe the collision process for scattering angles greater than 10°.

Figure 4 also shows a negligible difference between the Glauber YFC and IFC results for small scattering angles. For larger scattering angles this difference begins to be considerable. This behavior can be explained by the following argument: the difference between the YFC and IFC results is related to the difference in accounting for the interaction between the incident electron with the core electron and $(N-1)$ nuclear charges. This effect is expected to be small for small scattering angles, a situation in which the incident electron interacts very little with the core electrons. Larger scattering angles, on the contrary, represent a situation where the incident electron penetrates more in the atom and thus interacts strongly with the core electrons.

B. 2*p*-3*s* excitations

The absorption spectra of the sodium atom in the 30–40-eV region give rise to several autoionization processes. This energy region has been studied experimentally both through photoabsorption[26,27] and ejected electron[28,29] spectroscopy. Theoretical calculations concerning the autoionization processes induced by electron impact[30,31] and the total cross section for the 2*p*-3*s* excitation have already been published.[32–35]

Within our present knowledge no absolute scattering differential cross section for the excitation of 2*p* electrons in the sodium atom has been previously published.

We do not consider in the present work the interaction between the 2*p*-3*s* excitation and the underlying continuum. This is, of course, a limitation in the present analysis, which may perhaps be justified considering the small intensity of the continuum due to the direct ionization of the 3*s* electron in the 30-eV energy-loss region.[27,31] The experimental results were therefore obtained by discounting the background due to ionizations of 3*s* electrons, and the theoretical calculations concern the 2*p*-3*s* excitation only.

Figure 5 shows our experimental and theoretical FBA results for the 2*p*-3*s* GOS. The agreement between the theoretical and experimental values is only reasonable.

One possible reason for this disagreement may be related to the FBA description of the collision process. One should expect, in principle, a poorer description of this process compared to the 3*s*-3*p* case, since in the former case the ratio impact-energy and/or excitation energy is

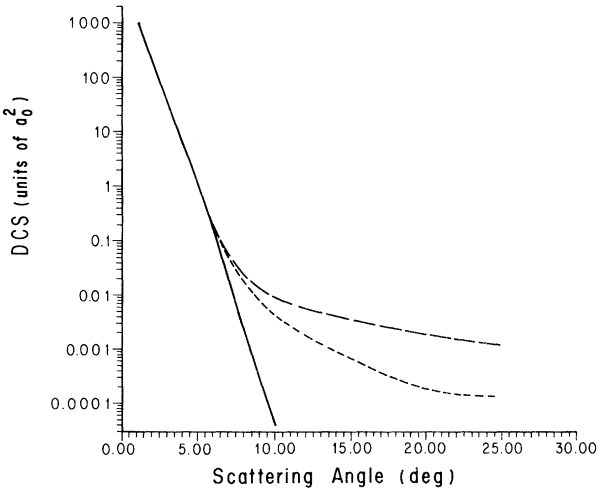

FIG. 4. Differential cross section for the 3*s*-3*p* excitation of the Na atom (all values from this work). ——, FBA; - - - -, Glauber IFC; — — —, Glauber YFC.

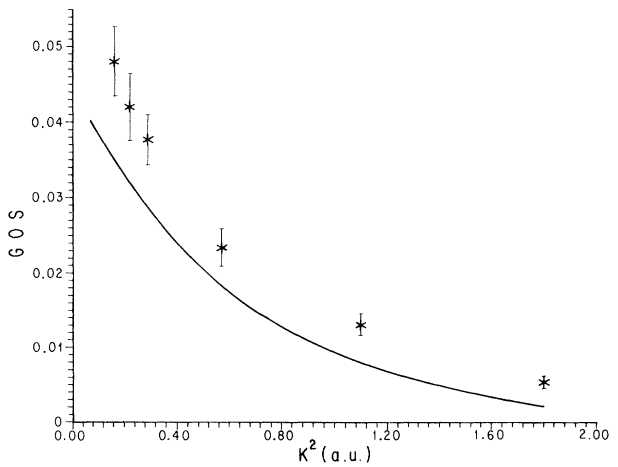

FIG. 5. Generalized oscillator strength (dimensionless units) for the 2*p*-3*s* excitation of the Na atom (all values from this work). *, experimental; ——, FBA.

about 15 times lower than in the 3p-3s case.

Another possibility is related to the Hartree-Fock frozen-core approximation used in the calculation of the atomic wave functions. In an inner-shell excitation process, one should expect relaxation effects to be more important than in the 3s-3p case. Correlation effects, which were not considered in the Hartree-Fock approximation, may also contribute. In effect, angular correlations should be important in the description of the $1s^2 2s^2 2p^5 3s^2$ final state. We have recently[36] shown, for the N_2 molecule, that correlation and relaxation effects can be very important in the calculation of the GOS for inner-valence electrons excitation. Finally, the fact that the interaction with the continuum was not considered could also contribute to the observed discrepancies between the experimental and calculated values.

V. CONCLUSIONS

Experimental values for the GOS of the 3s-3p excitation process have been determined and no minimum is observed in the momentum-transfer region considered. The Glauber and first Born approximations appropriately reproduce the experimental results for $K^2 < 0.6$ a.u.; for larger K^2 values the inclusion of higher-order corrections to these approximations is necessary.

A good agreement with the experimental results of Buckman and Teubner[2] for the 3s-3p process is found, but both sets of experiments do not show a minimum in the GOS around $K^2 = 0.4$ a.u. as predicted by Shuttleworth, Newell, and Smith.[1]

The elastic differential cross section has also been measured in the angular range of 0° to 30°, using 1-keV electrons. A comparison with theoretical results shows a general good agreement except for small scattering angles which might be due to the lack of inclusion of polarization effects in the calculations.

Theoretical and experimental values for the GOS for the 2p-3s excitation process have been determined for the first time and the results show a disagreement of about 25%. This is probably related to use of the FBA and of the Hartree-Fock frozen-core method used to calculate the atomic wave functions, as discussed in the text.

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