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Near-threshold Behaviour of Generalised Oscillator Strengths for 2p Ionisation

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Abstract

Near the ionisation threshold, the behaviour of the generalised oscillator strength (GOS) is sensitive to the choice of the model atomic potential. In electron-impact ionisation, 'delayed maxima' of the GOS often occur near the ionisation threshold due to centrifugal barrier effects. In the present work, the sensitivity of this important effect of the centrifugal barrier to the model atomic potential has been studied using two local density atomic potentials, namely, the nonrelativistic Hartree–Slater potential and the relativistic RCΞ potential. Calculations have been done for electron-impact 2p ionisation of atomic oxygen, neon, aluminium, argon, iron and zinc. The GOS results for 2p ionisation of oxygen and aluminium are presented to illustrate the results.

1. Introduction

Generalised oscillator strengths (GOS) of atoms are useful in estimating the stopping power and total scattering cross sections for fast charged particle collisions with atoms (Inokuti 1971). There has been substantial recent interest in the determination of this quantity using refined experimental techniques (Li *et al.* 1988; Takayanagi *et al.* 1990). We have recently made use of a relativistic atomic model potential, known as the RCΞ potential, to determine the generalised oscillator strengths for electron-impact-induced atomic excitations (Padma and Deshmukh 1992) and for 2s subshell ionisation (Padma and Deshmukh 1994). We have also used the RCΞ potential to estimate the optical oscillator strengths from 'electron-impact photoionisation' (Padma and Deshmukh 1993).

In the low momentum transfer region, a phenomenon frequently observed in the ionisation of various subshells of several atoms (McGuire 1968; Manson 1972; Afrosimov *et al.* 1969; Amusia *et al.* 1970) is an increase in the GOS from threshold up to a maximum, and a subsequent monotonic decrease for higher values of the ionised electron energy. This 'delayed maximum' with respect to the ionised electron energy is a consequence of the centrifugal barrier effect (Manson 1972*a*) which prevents the continuum partial waves with nonzero angular momentum ($l \neq 0$) from penetrating the atomic core. In this paper, the sensitivity of the delayed maximum to the choice of the atomic potential employed in the GOS calculations is discussed.

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2. The Method

The GOS is an essential factor of the differential cross section in the first Born approximation (Inokuti 1971). The generalised oscillator strength for ionisation of an atom from an initial state $|0\rangle$ to a final continuum eigenstate $|\epsilon\rangle$ involving a momentum transfer $\hbar\boldsymbol{K}$ is given by (Manson 1972*a*)

$$f_{0\epsilon}(\boldsymbol{K}) = (E_\epsilon - E_0)\frac{1}{(Ka_0)^2}\,|F_{0\epsilon}(\boldsymbol{K})|^2\,, \tag{1}$$

where $F_{0\epsilon}(\boldsymbol{K})$ is the inelastic scattering form factor,

$$F_{0\epsilon}(\boldsymbol{K}) \;=\; \langle\epsilon|\sum_j \exp\,(\mathrm{i}\,\boldsymbol{K}\,.\,\boldsymbol{r}_j)|0\rangle\,, \tag{2}$$

$\boldsymbol{r}_j$ being the position vector of the jth atomic electron of the target. The energies considered in this relation are expressed in Rydberg units. The wavefunctions for discrete states are normalised to unity and the continuum functions are normalised per unit energy range. The absolute square of the form factor for the $nl \rightarrow \epsilon l'$ transition from a closed subshell, summed over final degenerate magnetic substates m' and averaged over initial substates m, is given by

$$|F_{nl,\epsilon l'}(K)|^2 = (2\ell'+1)\sum_\lambda (2\lambda+1)[R^\lambda_{nl,\epsilon l'}(K)]^2 \left|\begin{pmatrix} l' & \lambda & l \\ 0 & 0 & 0 \end{pmatrix}\right|^2, \tag{3}$$

where

$$R^\lambda_{nl,\epsilon l'}(K) = \int_0^\infty P_{nl}(r)\, j_\lambda(Kr)\, P_{\epsilon l'}(r)\,\mathrm{d}r\,.$$

The choice of values for the summation over λ in equation (3) is restricted to the range from $|l-l'|$ to $l+l'$ in steps of 2, since the $3j$ symbol vanishes for all other values of λ.

In the present calculations, the one-electron wavefunctions for the initial ground state $P_{nl}(r)$ have been obtained from the RCΞ method (Vijayakumar *et al.* 1989). The radial wavefunctions for the final state corresponding to an ionised electron in the continuum are obtained by solving a one-electron differential equation:

$$\left(-\frac{\mathrm{d}^2}{\mathrm{d}r^2} + \frac{l(l+1)}{r^2} + V_{\epsilon l}(r)\right) P_{\epsilon l}(r) = \epsilon P_{\epsilon l}(r)\,. \tag{4}$$

The atomic potential $V_{\epsilon l}(r)$ in (4) is determined using the RCΞ method. For each value of ϵ, $P_{\epsilon l}(r)$ is obtained by integrating equation (4) for the SCF-RCΞ potential once (Cowan 1981).

The GOS for an initial state nl may be written as

$$f_{nl,\epsilon}(K) = \frac{\epsilon - \epsilon_{nl}}{(Ka_0)^2}\sum_{l'=0}^{\infty} |F_{nl,\epsilon l'}(K)|^2\,. \tag{5}$$

As is well known, the summation series in (5) is rapidly convergent since the higher partial waves are excluded due to centrifugal barrier effects. In the small momentum transfer region, the major contribution to the GOS comes from the optically allowed continuum partial wave channels. With an increase in energy, these partial waves overcome the centrifugal barrier and penetrate the atomic core deeper, resulting in a greater overlap with the initial state wavefunction. Hence the GOS increases, causing a 'delayed maximum' (Manson 1972*a*). For still higher values of the ionised electron energy, the GOS decreases due to cancellations in the transition matrix element resulting from rapid oscillations of the continuum wavefunctions.

The accuracy of the GOS results in the Born approximation calculations depends on the choice of the atomic model used for describing the initial and final states of the target atom. In the present work, the GOS has been calculated using the nonrelativistic Hartree–Slater (HS) potential (Herman and Skillman 1963) and the relativistic RCΞ potential (Vijayuakumar *et al.* 1989).

Calculations have been done for 2p ionisation of several atoms: O, Ne, Al, Ar, Fe and Zn. Illustrative results of the GOS are presented for oxygen and aluminium 2p ionisation.

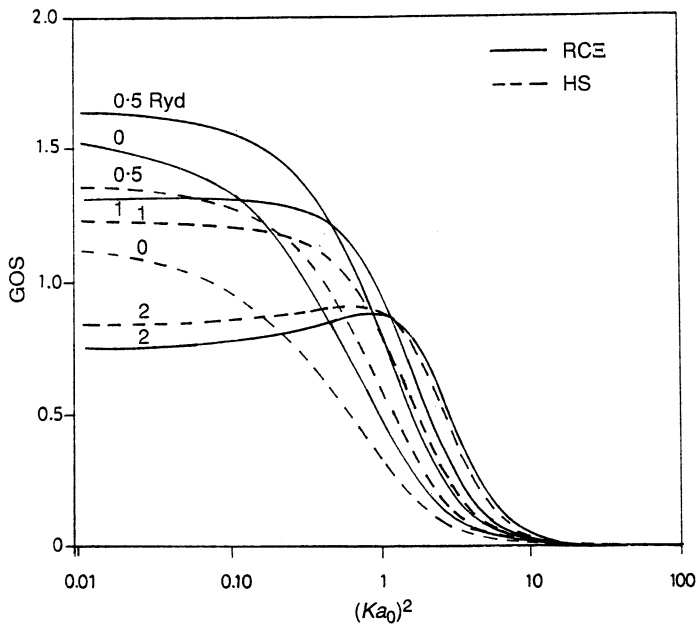

Fig. 1. Generalised oscillator strength per unit energy (in Rydberg) for continuum transitions from the 2p subshell of oxygen for ionised electron energies of 0, 0·5, 1 and 2 Ryd.

3. Results and Discussion

We have performed spin-polarised calculations using the RCΞ method. For open-shell atoms, this method provides slightly different wavefunctions for spin-up and spin-down electrons. Hence for $2p_{1/2}$ ionisation of aluminium, for example,

the GOS has been obtained for $2p_{1/2\uparrow}$ and $2p_{1/2\downarrow}$ electrons from two separate calculations. Similarly, the GOS for $2p_{3/2}$ ionisation has been obtained from two separate calculations for the spin-up and spin-down electrons in the $2p_{3/2}$ subshell. Since the one-electron GOS results were almost identical in all these calculations, they were summed to get the total GOS for 2p ionisation. The GOS has been calculated for various values of momentum transfer from 0·1 to 20 atomic units. To study the GOS behaviour as a function of the ionised electron energy, the GOS has also been calculated for various values of ionised electron energy from threshold to 10 Ryd.

The GOS results for 2p ionisation of oxygen are given in Fig. 1 as a function of the square of the momentum transfer $(Ka_0)^2$. The results are plotted for four different values of the ionised electron energy, 0, 0·5, 1 and 2 Ryd. Results of the RCΞ and HS calculations are shown for comparison. In the low momentum transfer region, the GOS at 0·5 Ryd is higher than that at the ionisation threshold, in both RCΞ and HS calculations. This is due to the centrifugal barrier effect.

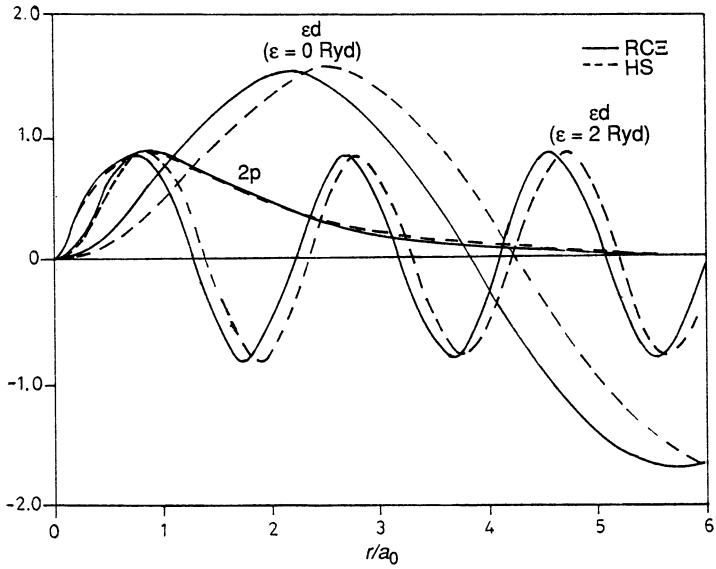

Fig. 2. Normalised 2p and ϵd wavefunctions in oxygen.

At threshold and also at 0·5 and 1 Ryd, the RCΞ value of the GOS is slightly higher than the corresponding HS value. This may be attributed to the fact that, while the bound state 2p orbital has an almost identical profile in the RCΞ and HS potentials, the ϵd orbital (which is the principal contributor to the GOS) is more compact in the RCΞ potential relative to that in the HS potential.

Fig. 2 shows the radial 2p and the continuum d orbitals at threshold ($\epsilon = 0$) and at $\epsilon = 2$ Ryd in oxygen. Only the radial $2p_{1/2}$ orbital in the RCΞ potential is shown. The $2p_{3/2}$ orbital is slightly more compact, but the difference is insignificant and not perceptible on the scale shown. The spin–orbit splitting of the continuum wave functions is ignored in the present work. At the ionisation threshold, the compactness of the continuum d orbital in the RCΞ potential,

relative to that in the HS, provides an increased overlap with the ground state 2p orbital and consequently a higher value of the GOS in the RCΞ potential compared to that in the HS approximation. However, at higher energies (e.g. 2 Ryd), cancellation of positive and negative parts of various elements of the integrand over the range $0 \leq r < \infty$, arising respectively from the positive and negative amplitudes of the continuum d wave, is more effective in the RCΞ model than in the HS model. This causes a lower value of the RCΞ–GOS than the HS–GOS. However, at higher momentum transfers, the GOS in the RCΞ model at $\epsilon = 2$ Ryd, is slightly higher than the GOS from HS calculations. This is due to the larger contribution from the $2p \rightarrow \epsilon f$ channel in the RCΞ model than in the HS model.

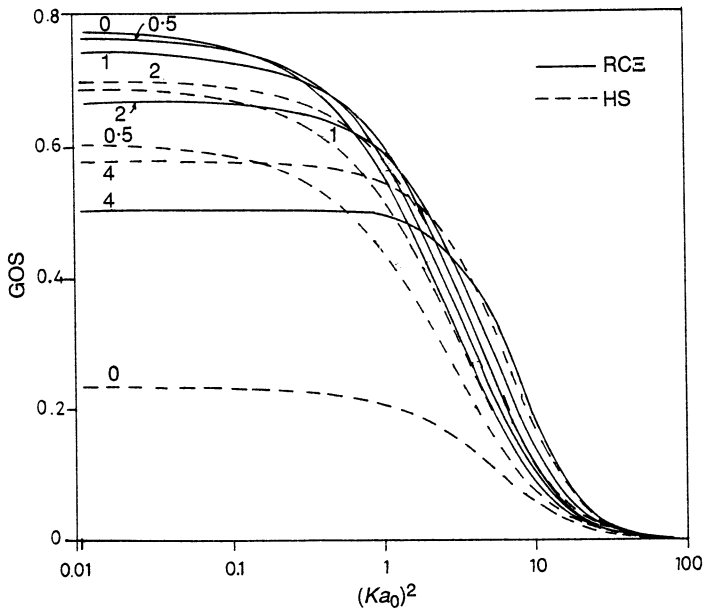

Fig. 3. Generalised oscillator strengths per unit energy (in Rydberg) for continuum transitions of the 2p subshell of aluminium for ionised electron energies of 0, 0·5, 1, 2 and 4 Ryd.

GOS results for 2p ionisation of Al are shown in Fig. 3. Results from both the RCΞ and HS calculations are shown for comparison. As can be readily inferred, the delayed maximum is a prominent feature of the GOS in the HS results, whereas the RCΞ results do not show such an effect. The difference in the RCΞ and HS results can be traced to the different energy dependence of the $2p \rightarrow \epsilon d$ channel which is the principal contributor in the two potentials, as can be seen in Fig. 4 which shows the partial wave contributions to the GOS.

The spectral shape of the GOS near the ionisation threshold is thus sensitive to the atomic potential. The RCΞ potential is more attractive than the HS potential for the following reasons:

(i) unlike the HS method, the self-interaction contribution to the total exchange interaction is calculated exactly in the RCΞ potential and the

local density approximation is made only for the residual part of the exchange interaction;

(ii) the inclusion of relativistic effects in the RCΞ potential results in stronger binding of the atomic electrons.

Consequently, the RCΞ continuum wavefunctions shown, for example, in Fig. 2 for oxygen are more compact than the HS continuum wavefunctions. However, this effect is not obvious in the 2p bound state wavefunctions obtained using the respective atomic potentials, as can be seen in the same figure. This may be attributed to the different boundary conditions satisfied by the bound and continuum wavefunctions. The bound state wavefunctions vanish at infinity and hence the difference between the RCΞ and HS potentials, which is mainly in their asymptotic behaviour (Vaidehi and Gopinathan 1984), does not alter the bound state wavefunctions. The asymptotic behaviour of the potential nevertheless affects the continuum wavefunctions which are oscillatory in that region.

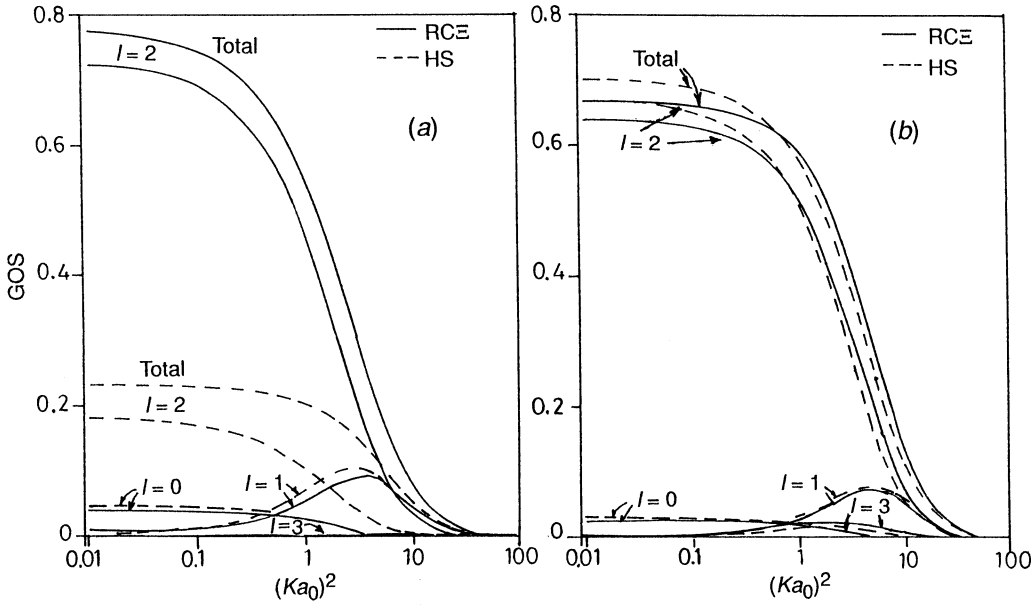

Fig. 4. Partial wave contributions to the Al 2p GOS from the various continuum angular momentum ($\epsilon l'$) channels for ionisation at (a) threshold ($\epsilon = 0$), and (b) $\epsilon = 2$ Ryd.

The stronger attraction of the RCΞ atomic potential permits the ϵd wave (the major contributor to the GOS for 2p ionisation) to penetrate the centrifugal barrier region even at threshold, resulting in a significant overlap with the ground state 2p wavefunction. The GOS results obtained using the RCΞ method are thus higher than the HS results at the threshold, and any increase in energy does not increase the overlap integral any further.

The GOS results for 2p ionisation of the other atoms mentioned above also support these findings. In Ar 2p ionisation, no delayed maximum was seen in the RCΞ calculations. In the case of oxygen, neon, iron and zinc, the RCΞ calculations did show a delayed maximum which was, nevertheless, far less prominent than

that found in the HS calculations. The stronger attraction of the RCΞ atomic potential is thus manifest in all these cases.

It may be recalled that the experimental results of Afrosimov *et al.* (1969) on rare gas atoms predicted a delayed maximum for argon 2p ionisation at $\approx 0 \cdot 44$ Ryd, whereas the HS results (Manson 1972*b*) predicted a delayed maximum at 2 Ryd above threshold. The RCΞ results for Ar do not show a delayed maximum, as mentioned earlier. It thus appears that the RCΞ potential may underestimate the delayed maximum effect and the HS potential may overestimate it.

As reported earlier (Padma and Deshmukh 1993), the GOS in the $K \rightarrow 0$ limit corresponds to the photoionisation cross section. A comparison of our results in the low momentum transfer region with the photoionisation cross section calculated by McGuire (1968) showed the two results to be in excellent agreement. The agreement between the photoionisation limit of the GOS obtained in the present calculation for O, Ne and Al 2p ionisation was found to be exact, which is not surprising since the same model potential (Hartree–Slater) is employed in both cases. The RCΞ results for oxygen agree with those of McGuire, whereas for the other atoms the centrifugal barrier effect is not very pronounced in the RCΞ model as noted earlier.

We find that it is not just the energy at which the delayed maximum occurs above the threshold that is different in the two potentials, but the very occurrence of this is not predicted by the RCΞ potential for some of the cases considered, contrary to the HS predictions.

4. Conclusions

The present results reveal that the delayed maximum for 2p ionisation is sensitive to the model potential employed in the calculation and the differences may be not merely quantitative but also qualitative. Further experimental studies of this phenomenon will be useful in developing a clear understanding of the reliability and limitations of the RCΞ and Hartree–Slater atomic models.

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