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ANGULAR AND ENERGY DISTRIBUTIONS OF δ-RAYS EJECTED FROM LOW-Z MOLECULAR TARGETS BY INCIDENT PROTONS AND α PARTICLES

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The double differential cross-section mixed treatment (DDCS–MT), which has been developed in order to calculate the relatively energetic δ-rays measured along 4 to 12 MeV α particle tracks recorded in ionographic emulsions of various compositions, reproduces quite rigorously experimental DDCS for medium energy protons and α particles crossing not only helium and argon, but also molecular gases like water vapour, hydrocarbons, etc. In the latter case, a special procedure is required to describe the molecular target in terms of atomic quantities, notably by taking into account the binding energies of the molecular orbitals, as well as the breaking up of the molecular orbitals in terms of atomic orbitals.

1. Introduction

Linear density distributions of secondary electrons ejected by α particles have been determined in nuclear emulsions of various compositions, which had been submitted to the so-called activation treatment [1]. Good agreement with these data over the whole incident particle's energy range (4–12 MeV) could not be obtained by using single differential cross-sections (SDCS) of the Rutherford or the Binary Encounter Approximation (BEA) type, while a better accord could be reached by means of a so-called mixed treatment (SDCS–MT) [2]. In order to describe more realistically the heavy charged particle's track pattern, we have also tried classical double differential cross-sections (DDCS–BEA) [3], as well as quantum mechanical DDCS [4]; the calculations could however not be made to agree with our experimental δ-ray distributions in the energy range considered [5]. We have therefore developed a new approach, the so-called DDCS mixed treatment (DDCS–MT) [5]. This formalism led to a satisfactory reproduction of our ionographic results, as well as of experimental DDCS in helium and argon, without adjustment of the theoretical values by means of arbitrary fitting parameters. Promising results were also obtained by direct application of our formula to molecular targets like methane.

The present paper deals with the extension of the DDCS–MT to the calculation of molecular DDCS, by means of a suitable procedure taking into account the in this case more complicated target–projectile configurations. Water vapour ionized by protons and α particles, as well as methane and benzene traversed by protons will be presented as examples.

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2. Calculation of molecular DDCS

Let us briefly recall that the DDCS–MT [5] consists of two complementary formulae, a quantum mechanical formula for the K-, L- and M-shells, and a classical formula (DDCS–BEA) [3] for the N and higher shells. The quantum mechanical part was derived from the DDCS–Plane Wave Born Approximation (DDCS–PWBA) expression [4], notably by inserting the K-, L- and M-form factors established by Merzbacher [6]. A supplementary positive charge being introduced by the passing ion, we corrected the binding energies with respect to the energy of the incident ion following an approach proposed by Basbas et al. for the K-shell [7] and by Brandt et al. in the case of the L-shell [8]. Furthermore, the results could still be improved at the small ejection angles, θ, by taking into account the final state interaction between the ejected electron and the projectile, as proposed by Salin [9]. Nevertheless, in the case of nuclear emulsions, however, as already described elsewhere [5], the detectable energies T were of the order of some keV, i.e. high enough to render the influence of the molecular structure as well as of these corrections negligible.

In order to extend the field of applicability of the formalism to the case of molecular targets at smaller ejection energies, we had to take into account the binding energy of the molecular orbitals, and to express each of these molecular orbitals as a weighted sum of atomic orbitals.

2.1. Water vapour

For the water molecule, the binding energies of the five molecular orbitals have been taken into considera-

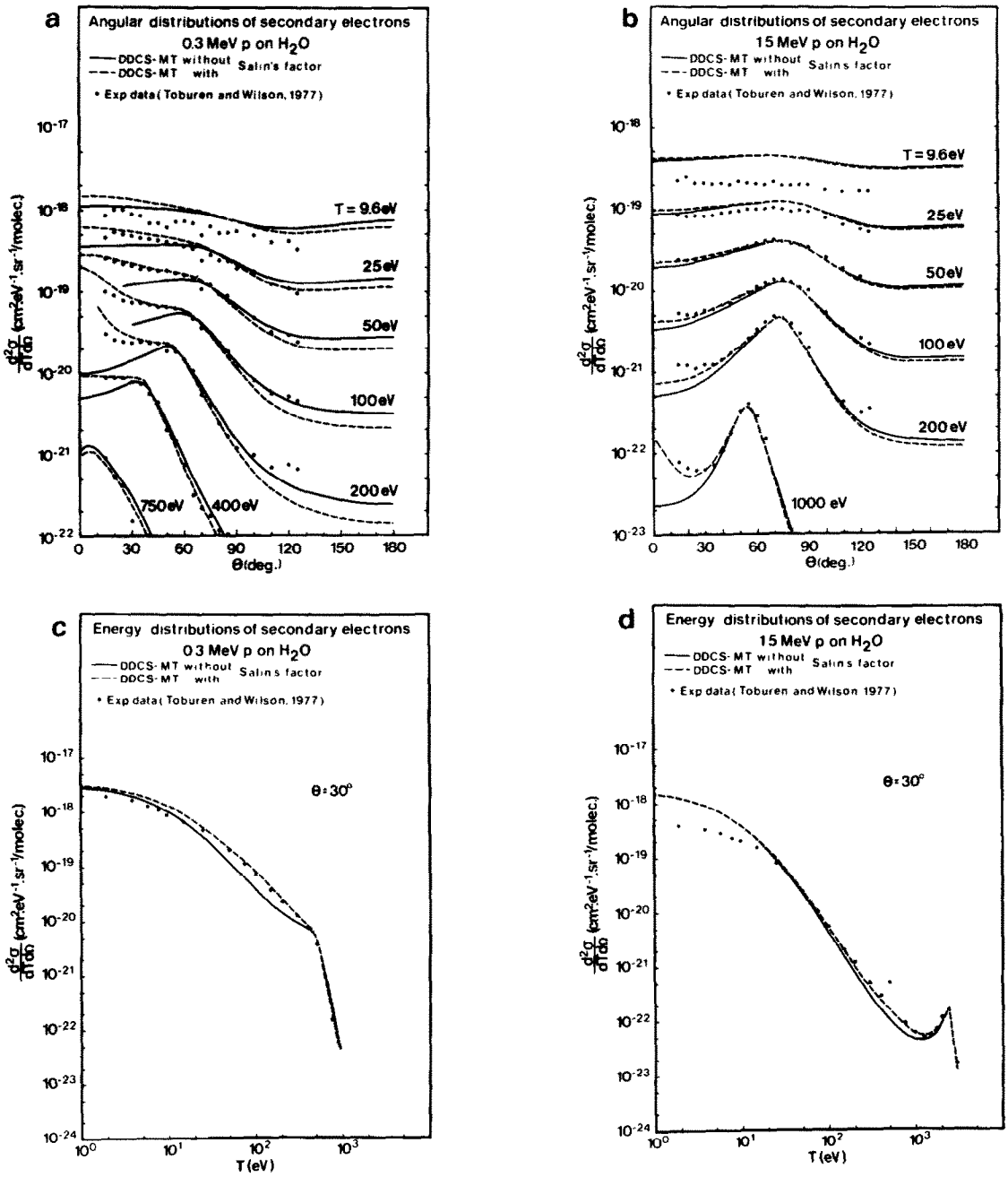

Fig. 1. (a) and (b): Angular distributions of δ-rays for various electron ejection energies, T. (c) and (d): Energy distributions of δ-rays ejected at $\theta = 30°$.

tion, as well as the breaking up of these orbitals in terms of H 1s, O 1s, O 2s and O 2p atomic orbitals [10]. Nine "atomic" levels, each characterized by a specific binding energy and/or specific quantum numbers were then treated separately; the partial DDCS were finally added up in order to obtain the DDCS for the whole molecule. These DDCS and the corresponding measured values [11] for 0.3 and 1.5 MeV incident protons are represented as a function of θ for different values of T (figs. 1a,b), and as a function of T for $\theta = 30°$ (fig. 1c,d). It appears that the accord between the calculated and the experimental data stays within the experimental uncertainties for ejection energies as low as $T \simeq 10$ eV. Since the Auger electrons have not been included in the

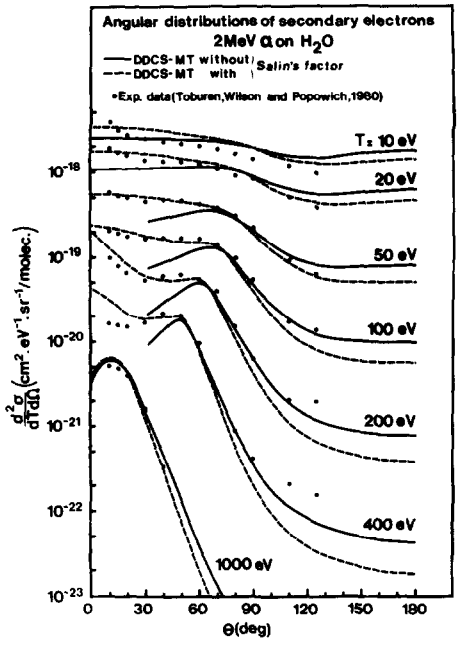

Fig. 2. Angular distributions of δ-rays for various electron ejection energies, T.

present calculations, the theoretical curve does not reproduce the experimental peak at $T \simeq 500$ eV (fig. 1d).

Fig. 2 shows the experimental DDCS values [12] and the corresponding DDCS–MT calculations in the case of the ionization of water vapour by 2 MeV α particles. Here also good agreement was obtained for all ejection energies larger than $T \simeq 10$ eV, i.e. the same limit as for incoming protons. It seems therefore that the approach proposed may be extended to ions heavier than the proton, at least as long as they act as bare particles.

2.2. Hydrocarbons

In the case of methane, the binding energies of the molecular orbitals have been determined by Hamrin et al. [13]. We have estimated the breaking up into atomic orbitals on the basis of the sp^3 hybridization of the carbon atom. In fig. 3 are shown experimental data [14] obtained with 2 MeV incident protons together with the corresponding results calculated by means of the DDCS–MT. It can be seen that the agreement is satisfactory even at $T = 1.4$ and 5.6 eV, an energy region where the relative experimental error becomes very important.

Similar calculations have been carried out in the case of ring compounds, for instance the benzene molecule, by introducing into the treatment the binding energies of the molecular orbitals determined by Fridh et al. [15] and by deriving the breaking up into atomic orbitals from the sp^2 hybridization of the carbon atom in this molecule. For 2 MeV protons, the data calculated by means of our treatment are in good agreement with the corresponding experimental values [16] over the whole

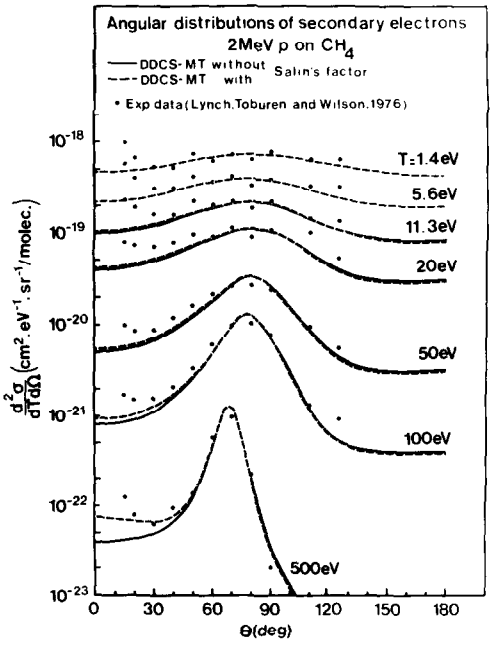

Fig. 3. Angular distributions of δ-rays for various electron ejection energies, T.

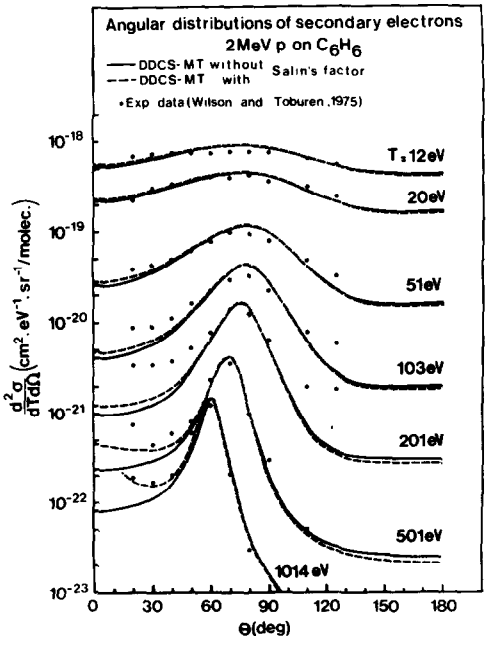

Fig. 4. Angular distributions of δ-rays for various electron ejection energies, T.

ejection energy interval considered, except for ejection energies of the order of 200 eV where Auger electrons contribute significantly to the measured DDCS (fig. 4).

3. Conclusion

In the light of the examples given in this paper, it appears that the DDCS–MT adapted by means of a specific procedure to molecular targets is able to predict within the experimental errors the values of the DDCS for hydrogen and helium ions traversing molecular media. This has been verified at the actual stage of this study for 0.3 to 2.0 MeV protons and for 2 MeV α particles.

We shall continue our investigations in order to extend the DDCS–MT to other projectiles and ejected electron energies, and to other molecules. The introduction of this DDCS–MT into a general model code intended to describe the track structure of heavy charged particles traversing matter, notably biological tissues, is foreseen.

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