

Ionization of Molecules with a Generalized Sturmian Functions Approach

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Synopsis We are interested in the single ionization by electron impact of small polyatomic molecules such as CH₄, NH₃ or H₂O. Within a Sturmian approach, we expand the wave function describing the ejected electron in an appropriate set of Generalized Sturmian Functions. The obtained differential cross sections are compared with other theoretical calculations and with experimental data.

Ionization by electron impact is a fundamental process which tests our ability to describe the target electronic structure, the role of electron-electron interactions and to understand the collision mechanisms. Diverse experimental and theoretical advances of the last years have boosted the interest on molecular targets. Initially, most of the theoretical studies have been limited to small molecules such as H₂ or N₂. The extension to molecules of traditional atomic methods is limited by the complexity of the problem (multicenter formulation and generally highly noncentral). Recent (*e*, 2*e*) measurements on polyatomic molecules [1–4] motivated the development of theoretical methods to describe ionization cross sections in their most differential form.

We propose to use the Sturmian approach, using Generalized Sturmian Functions (GSFs) [5, 6] to study single ionization of molecules. The method was applied successfully to study single photoionization of atoms and small molecules [7, 8], as well as double ionization of atoms induced by photon [9], electron [10] or proton [11] impact. In the present investigation, we consider one-center expansions, and take the initial state wave function from Ref. [12]. In a one-active electron approach, the scattering wave function is expanded in a set of GSFs that have an appropriate asymptotic outgoing-type behavior; this property makes the method rather efficient. Moreover, the scattering amplitude can be extracted from the asymptotic behavior of the scattering solution (essentially the expansion coefficients), without the need of calculating a transition matrix element.

We present calculations of triple differential cross sections (TDCSs) for several coplanar asymmetric geometries. For the chosen experimental incident energies (ranging from 250 to 500 eV) we work within the first Born approximation. We compare our results with cross sections obtained with other theoretical methods (in an absolute scale), and with the available relative scale experimental data [1–4]. Specifically, we consider the electron impact ionization of the valence orbitals 1*t*₂ of CH₄, 3*a*₁ of NH₃, and 3*a*₁

and 1*b*₁ of H₂O.

Like other theoretical results, ours reproduce only partially the experimentally observed cross sections features. Important differences in the position and height of the recoil peak, in particular, clearly indicate an agreement breakdown between the measurements and the presently available theories including ours. The analysis of the binary to recoil ratio as a function of the momentum transfer illustrates that the agreement deteriorates for larger momentum transfer values.

Interestingly, under given kinematical conditions, we predict for methane a double peak structure in the cross section binary region, a clear signature of the *p*-nature of the molecular orbital; this is similar to what has been observed experimentally and theoretically for water [1, 8].

Partial results have been already presented in Ref. [13].

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