



Vibration Effect On The Elastic Differential Cross Section In The Gas-Phase Electron Diffraction Within The Multiple Scattering Model

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Abstract

It is well-known that the laser-induced electron diffraction (LIED) contains molecular structural information which can be extracted with a spatial resolution of angström and time resolution of few femtoseconds [1, 2]. The retrieval is based on the quantitative rescattering method (QRS) allowing the LIED signal to be split into two components [3], one of which is a laser-free differential cross section (DCS) containing molecular structure. The method based on fitting the experimental DCS extracted from the LIED spectra to the theoretical DCS calculated with assumed initial structure parameters then allows one to reveal the real molecular structures. The theoretical DCS of molecules is treated within the independent atoms model (IAM) [1, 4] or the more advanced model based on the multiple scattering theory (MS) [2, 5].

In this report, we talk about how to consider the molecular vibration effect to the MS model and examine this effect of molecular vibrations on the DCS by comparing the oscillation component with the component of the MS second order describing the interference of the scattering waves. We perform an application of the developed theory for some diatomic molecules.

1 Introduction

The collision of electrons with matter was first studied in 1893 [6]. A useful expression of the interaction between electrons and atoms was demonstrated in 1930 by Mott [7]. The diffraction pattern from compounds in the gaseous state interacting with high velocity electrons to investigate the molecular structures was shown by Wierl in 1931 [8]. This work shows that electron diffraction by gas molecules

is a useful method to explore information molecules. Gasphase electron diffraction (GED) has many advantages, such as explaining the nature of chemical bond by Linus Pauling [9]. The gas-phase electron diffraction has been an effective method for determining the structure of molecular [10]. For retrieval of the molecular structure, several simple models such as the independent-atoms model (IAM) [11] and the multiple scattering model (MS) [12].

GED were applied for calculating the electron-molecule differential cross section (DCS). Moreover, the molecular scattering intensity $M(s)$ and the radial distribution function $D(r)$ [10]. Within the IAM, molecular vibration distorts the DCS, $sM(s)$, and $D(r)$ of molecules; its inclusion corrects the molecular retrieval structure [13,14]. However, the MS developed for intermediate and low energies of the electron-molecule collision has not considered the vibration effect [12]. In this work, we calculate the DCS, $sM(s)$ and $D(r)$ of O₂ using IAM and MS accounting for molecular vibration. Then we analyze the difference of these quantities regarding or disregarding the vibration of O₂ molecules to answer for the questions: How does vibration affect on these terms using IAM and MS? When the second order MS be less than vibration contribution of IAM?

2 Methods

In order to retrieve the information of molecular structure, IAM and MS are two efficient models to calculate the theoretical DCS fast and precisely. Disregarding molecular vibration, the IAM generates molecular DCS from molecular structures by simply adding pre-calculated atomic DCS together [6]. The averaged molecular scattering intensity of electron waves in two cases

$$I(\theta) = \sum_i^n |f_i(\theta)|^2 + \sum_{i \neq j}^n \sum_{i \neq j}^n \left\{ |f_i(\theta)| |f_j(\theta)| \cos(\eta_i - \eta_j) \frac{\sin sr_{ij}}{sr_{ij}} \right\}$$

Considering anharmonicity of molecular vibration

$$I(\theta) = \sum_i^n |f_i(\theta)|^2 + \sum_{i \neq j}^n \sum_{i \neq j}^n \left\{ |f_i(\theta)| |f_j(\theta)| e^{-\frac{r_{ij}^2}{2}} \cos(\eta_i - \eta_j) \frac{\sin(r_a - \kappa s^2)}{sr_a} \right\}$$

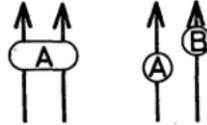
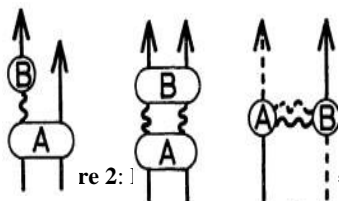


Figure 1: Diagram of IAM model

MS is a model splits scattering into paths in which electron interact with one or two atoms [7]. The averaged molecular scattering intensity of electron waves is

$$I = I_S + I_{SS} + I_{SD_1} + I_{SD_2} + I_{DD} + I_{DD_1}$$

I_S is sum of single-scattering waves; I_{SS} is interference single-scattering waves; I_{SD_1} is interference single & double scattering waves (one common atom); I_{SD_2} is interference single- and double-scattering waves (no common atom); I_{DD} is sum of double-scattering waves; I_{DD_1} is interference double-scattering waves.



3 Results and Discussion

3.1 The DCS for electrons collide with O₂ for different energies

The DCS for electrons collide with O₂ for different energies is shown in the figure 1. It shows vibration effect insignificant to molecular DCS. The vibrating DCS coincide with the nonvibrating DCS of both models. There are different DCSs at 50eV. The higher incident energies of electron, the more similar the results of two models are. Therefore, when the energy of scattering electron higher than 200 eV, we can use IAM instead of MS in calculating DCS of molecules.

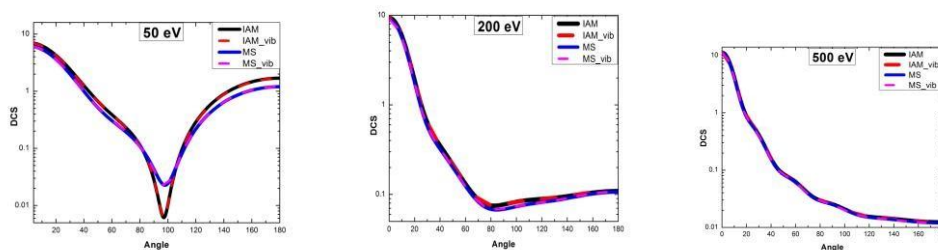


Figure 1: DCS of O₂ with different energies of electron 50, 200 & 500 eV.

3.2 Comparison the vibrational contribution IAM and the second order MS of the DCS

The vibrational contribution IAM is compared to the second order MS of the DCS (Figure 2). It points out the first term is lower than the second one. They are quite distinct at 50 eV. The higher energy rise, the smaller gaps between these terms are. Consequently, IAM can be used if the energy is above 200eV at the large angles.

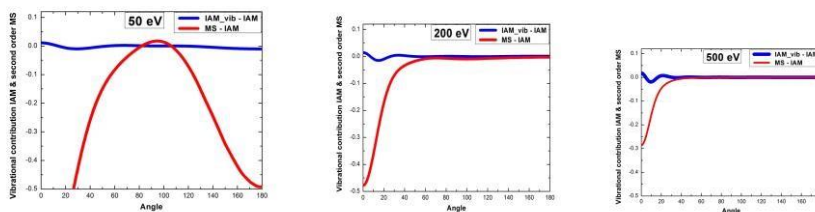


Figure 2: Comparison the vibrational contribution IAM and the second order MS of the DCS.

3.3 The molecular interference term $sM(s)$ of O₂

We show the molecular interference term $sM(s)$ of O₂ using IAM and MS accounting for vibration effect or not (Figure 3). The above figures point out the higher energy electrons scatter, the more difference between vibration or non-vibration in the $sM(s)$ of molecule are shown. In the energy below 50 eV, the momentum transfer s is small. Therefore, we should use the fitting method instead of Fourier transform the $sM(s)$.

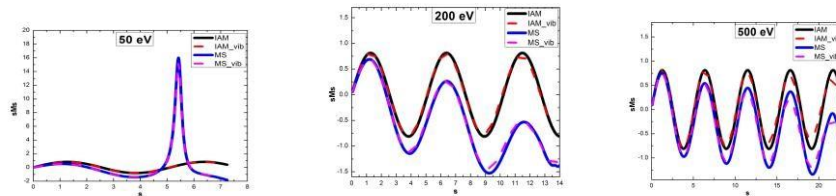


Figure 3: The molecular interference term $sM(s)$ of O₂

3.4 The $D(r)$ of O₂

The $D(r)$ of O₂ are shown below (Figure 4). We can see there is a peak at around 1.23 Å, which represent the internuclear distance for O=O. When the energy electron increases, the height of $D(r)$ rises. Below 200eV, we can easily distinguish the $D(r)$ calculating by IAM accounting for vibration or not as well as MS. The peak of $D(r)$ calculating by IAM is distinct from MS while they are coincided in 500eV. This result confirms the conclusion that IAM works quite well in the large energy range.

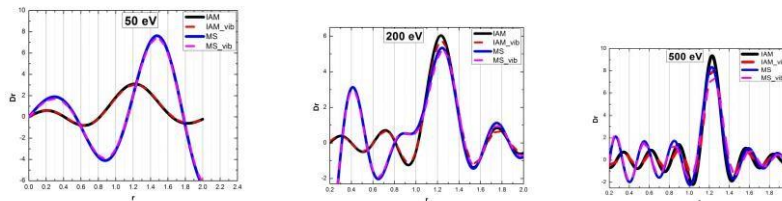


Figure 4: The $D(r)$ of O₂

4 Conclusions

We include the molecular vibration to the independent atoms model (IAM) and the multiple scattering model (MS) for calculating the electron-molecular differential cross section (DCS), the molecular scattering intensity and the radial distribution function, applying these calculation to the radial distribution function, applying these calculation to oxygen molecules, we compare the applicability of the two models, IAM and MS. Furthermore, we analyze the energy region of electron scattering where the MS is highly accurate and, consequently, the included molecular vibration effect can be noticeable.

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