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Quantitative analysis of the energy distributions of electrons backscattered elastically from polyethylene

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ABSTRACT

We present results of theoretical and experimental studies of the spectra of electrons backscattered elastically from polyethylene in the primary energy range between 1 and 5 keV. The experiments were performed using a high energy resolution electron spectroscopy. The theoretical interpretation is based on a Monte Carlo simulation of the recoil and Doppler effects. The separation between the carbon and hydrogen peak in the energy distributions is shown as a function of the primary electron energy. The simulations give many partial distributions separately, depending on the number of elastic scatterings (single, and multiple scatterings of different types). We show our results for intensity ratios, peak shifts and broadenings. We also present detailed analytical calculations for the main parameters of a single scattering. Finally, we present a qualitative comparison with the experimental data. We find our resulting energy distribution of elastically scattered electrons to be in good agreement with our measurements.

1. Introduction

Observation of the hydrogen peak is either a challenging or impossible task for the conventional electron spectroscopy. Hydrogen was observed earlier in electron scattering experiments at high energy using transmission geometry and formvar film [1].

In this work we present detailed theoretical as well as experimental studies for the detection of hydrogen by analyzing the spectra of electrons backscattered elastically from polyethylene ($(CH_2)_n$). We take advantage of the fact that the peak due to electrons backscattered elastically in this case splits into two component peaks which can be associated with the electrons backscattered mainly from hydrogen and carbon atoms of the sample, respectively. The energy of the elastically backscattered electrons is shifted from the primary values due to the energy transfer between the primary electron and the target atoms (recoil effect). Furthermore, the thermal motion of the scattering atoms causes broadening in the primary electron energy distribution, usually referred to as Doppler broadening. Monte Carlo calculations were carried out in order to simulate the spectra of backscattered electrons having primary energies between 1 and 5 keV. We note, that many X-ray photoelectron spectrometers also have an electron gun. And using this technique the hydrogen content of various samples can be determined. This can not be seen in the photoelectron spectra.

This technique was used for the determination of the hydrogen content at surfaces [2–5]. These investigations help us improve our understanding of the processes that involve the presence of H atoms at surfaces in polymers, carbon based hard coatings or new H storage materials. These later studies can be closely related to discovering new energy cells for new generation cars operating with hydrogen and producing simple water instead of toxic environment depleting gases. This procedure has also been used to check surface degradation in several polymers [6]. Filippi and Calliari extended this strategy of the quantification of H at more complex surfaces, containing other atoms like O [7].

2. Experiments

A polyethylene sample (Goodfellow ET311251) was used at room temperature in the elastic peak electron spectroscopy (EPES) experiments. The cleanliness of the sample surface was monitored by XPS analysis. As a reference specimen Cu polycrystalline metal was used. The Cu surface was cleaned by Ar^+ ion sputtering with an ion flux of 120 μA × min/cm^2 and 2 keV kinetic energy. XPS and EPES measurements were performed using the ESA-31 type electron spectrometer developed in ATOMKI [8]. In the EPES measurements a LEG 62 (VG Microtech) type electron gun was used. The energy width of the primary electron beam was 0.6 eV. During

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measurements the vacuum in the analysis chamber was better than 3×10^{-9} mbar.

3. The Monte Carlo simulation

3.1. Characterization of the model

If the thickness of the sample significantly exceeds the inelastic mean free path, the 30–40% of the intensity of the quasi-elastic peak arises from the multiple scattering in reflection mode [9]. In our recent studies the sample can be treated as a "semi-infinite sample". In extreme cases the contribution from the multiple scattering should even exceed 80%. For the case of multicomponent samples, the estimation of the multiple scattering effects is a very hard task. This fact is especially true when we are also interested in the energy distribution of the backscattered electrons. Namely, if the sample contains atoms with different masses, one part of the multiple scattering is come from the mixed collisions, i.e. the electron scattered on atoms with different masses and the resultant energy loss distribution can not be described with the single Gauss distribution. Therefore for mimicking the prospective electron energy distributions backscattered from a polyethylene (PE) sample we used Monte Carlo simulations. The applied model used simple impulse approximations. The main aspects of the model are as followings:

(1) The sample is semi-infinite, homogeneous and amorphous.
(2) The electron motion in the sample is treated as zigzag trajectory. If the electron collides with a moving atom, after the elastic collision its energy exchange can be calculated by the following formula:

$$E_{0r\ell} = \frac{2m}{M_\ell} E_0 \left[1 - \cos\theta_0 + \sqrt{\frac{M_\ell \varepsilon_\ell}{mE_0}} (\cos\vartheta_\ell - \cos\theta_0 \cos\vartheta_\ell - \sin\theta_0 \sin\vartheta_\ell \cos\varphi_\ell) \right], \quad (1)$$

where m, E_0 and θ_0 are the mass, the energy and the scattering angle of the electron respectively. ℓ characterize the atom where the elastic scattering happened. In our present case ℓ is either C or H. M_ℓ, ε_ℓ are the mass and energy of the given atom, and the angles ϑ and φ are characterizing the direction of the velocity of the scattering atom, relative to the direction of the velocity of the primary electron and to the plane of the scattering, respectively.
(3) The partial expansion method was used to describe the differential and total cross sections for elastic scattering [10]. In our calculations atomic target and Dirac–Hartree–Fock–Slater wave functions were used. This is the more general combination in similar calculations and energy range like EPES method obtaining the inelastic mean free path [11,12].
(4) The mean free path for inelastic scattering of electrons within a solid was described with the predictive formula of Tanuma et al. [13].
(5) For a given scattering event, random numbers defined the nature of the collision (elastic or inelastic), the type of the atom where the electron scatters (C or H), the scattering angles, the kinetic energy of the selected target (recoiled) atom, and the instantaneous direction of the selected target atom $(\vartheta_\ell, \varphi_\ell)$. The selection of collision type is based on the atomic concentration and cross sections. The instantaneous kinetic energy of the target atom is selected from the Maxwell-Boltzmann type distributions, where the average kinetic energies $(\bar{\varepsilon}_\ell)$ are described independently for the two components.
(6) The velocity distributions of the atoms are isotropic.
(7) For the case of the investigated angular range, (in our preset studies this is $\theta_0 = 130^\circ \pm 5^\circ$) the energy loss of the elastically scattered electrons determined and the energy loss spectra is stored.
(8) Beside the partial loss functions suffered 1, 2…50 elastic collisions, many other distributions can be analyzed based on the stored data of the Monte Carlo code. For example the partial loss functions are saved as a function of the number of elastic scatterings on C or H. We note, however, that for the study of partial distributions with suitable energy resolution and accuracy even for the peak of H, the evaluation of a huge number of primary trajectories was required.

3.2. Calculation procedure

Monte Carlo simulation of electron transport in solids is based on the stochastic description of scattering processes. Electron penetration is approximated by a classical zigzag trajectory. Details of the calculations are given elsewhere [14]. The scattering point is where the electron changes its direction and/or energy. In our calculations both the elastic and inelastic scattering events were taken into account. For the case of the first inelastic collision, the calculations were stopped. Particular values of scattering angles of electrons in an individual event are realized by random numbers following the angular differential elastic cross sections of carbon and hydrogen. After each elastic scattering the recoil energy was calculated according to Eq. (1), depending on whether the elastic scattering occurred on carbon or hydrogen. In this way, the energy distribution of elastically backscattered electrons was determined as a function of the number of elastic events only on carbon, only on hydrogen, and both on carbon and hydrogen (mixed scattering). Fig. 1 shows the geometric configuration used in the calculations.

With the help of our Monte Carlo code we are able to calculate the angular distributions of backscattered electrons in the xz-plane as a function of the angle of emission α at the given initial angle of incident beam (θ). In our recent experiment $\alpha = 115^\circ$ and $\theta = 75^\circ$, and accordingly the scattering angle is $\theta_0 = 130^\circ$.

3.3. Elastic scattering

The partial expansion method was used to describe the differential and total cross sections for elastic scattering. The elastic scattering of relativistic particles is described by the direct $f(\theta)$ and spin-flip $g(\theta)$ scattering amplitudes. The relativistic differential cross section per unit solid angle is given by

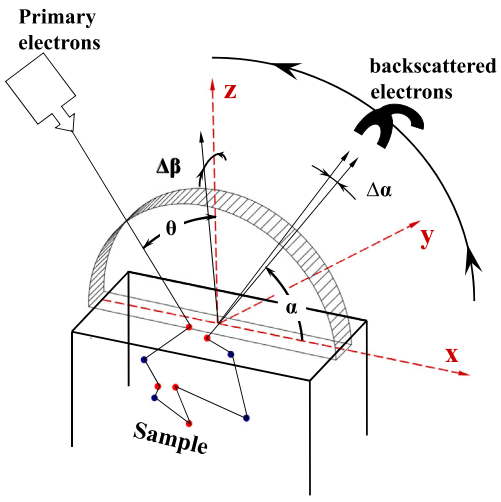

Fig. 1. (Color online) Schematic view of the geometric configuration of the calculation.