

A study of electron momentum density distributions in polyethylene polypropylene and polybutene

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Introduction

Theoretical studies of electronic band structure of polyethylene have been made with both semi-empirical molecular orbital theories and the non-empirical Hartree-Fock-Slater approximation on an idealized periodic polyethylene chain. It is natural to find in these studies that the band structure is sensitive to the method employed and the choice of the basis set. Thus, experimental information about the ground state wave functions is mandatory to the development of the theoretical studies.

We aim to obtain information about the ground state wave functions in highly oriented polyethylene (PE), polypropylene (PP) and polybutene (PB) via Compton profile measurements. Compton profile is the twice integrated electron momentum density and defined as

$$J(p_z) = \iint dp_x dp_y \sum_b \left| \int \psi_b(\mathbf{r}) \exp(i\mathbf{p}\cdot\mathbf{r}) d\mathbf{r} \right|^2$$

where ψ_b is a wave function of an electron in state b , the summation runs over all occupied states and z is taken along the scattering vector of x-rays. Thus $J(p_z)$ carries direct information about the ground state wave functions.

Experimental

The measurements were carried out by a Compton scattering spectrometer with a momentum resolution of 0.13 a.u. installed at ARNE1 beam line of the Photon Factory, KEK. Almost 100% oriented PE was obtained from Toyobo Co. (Dyneema) in a form of fiber. The oriented PP and PB specimens were prepared by stretching semi-oriented film at a proper temperature.

Results and Discussion

Figure 1 shows the difference $\Delta J(p_z)$ between $J(p_z)$ measured with p_z along the direction of the carbon chain axis and that measured with p_z perpendicular to the axis for PE, PP and PB. It is seen that replacing one of H in PE with CH_3 (PP) and $\text{CH}_2\text{-CH}_3$ (PB) modifies $\Delta J(p_z)$ greatly.

It can easily be shown [1] that $B(\mathbf{r})$ is the sum of autocorrelation functions of the one-electron configuration space wave functions $\psi_i(\mathbf{r})$; along one specific direction:

$$B(\mathbf{r}) = \sum_i B_i(\mathbf{r}) = \sum_i \iiint \Psi_i(\mathbf{s}) \Psi_i(\mathbf{s} + \mathbf{r}) d\mathbf{s}$$

$$B(z) = \int J(p_z) e^{-ip_z z} dp_z$$

Positive or negative signs of $B(\mathbf{r})$ are related to the correlation of the electron wave functions. Figure 2 shows the difference $\Delta B(z)$ between $B(z)$ measured with z along the direction of the carbon chain axis and that measured with z perpendicular to the axis for PE, PP and PB. The minimum at 0.14nm and the maximum at 0.3nm are ascribed to the projection of the C-C bond length along the direction of the scattering vector [1]. This suggests that $\Delta B(z)$ probe one-dimensional electronic structure which is modified by branch of the C-C bond chain.

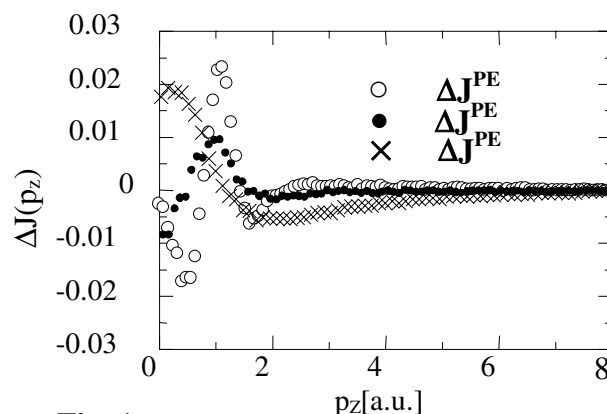


Fig. 1

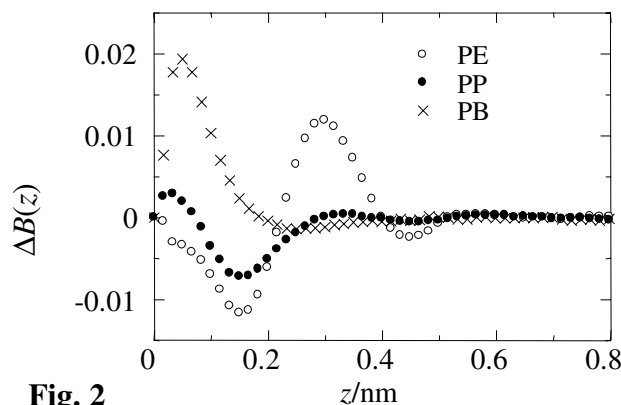


Fig. 2

Reference

[1] E.P. Mazarakiotis, D.L. Anastassopoulos, A.A. Vradis, G.D. Priftis, Ch. Bellin and G. Loupias, Physica B318 (2002) 382–386

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