Spectra of charged solitons and temperature dependence of the mobility of neutral solitons in trans-polyacetylene. Forner, Wolfgang

Abstract

The geometry of trans-polyacetylene chains as a function of time was obtained on the basis of the dynamics of charged solitons within the Su-Schrieffer-Heeger SSH model. The SSH model was chosen for the simulation because this one-particle model is based on re-normalized parameters that essentially contain the effects of electron-electron interactions. At least for charged solitons, the theory gives quite correct soliton widths as compared to PPP calcns. The present study is also aimed to study whether the SSH model is able to yield reliable geometries in time simulations. With the help of the PPP model, Moller-Plesset perturbation theory of second order and the RPA, the spectra as a function of time were calcd. at two different doping levels and for an excited chain. Then these spectra calcd. at different times were superimposed. All features in the spectra are consistently shifted by about 0.5 eV higher than expected and the spectra show many more local min. and maxima than those obsd. exptl. This effect could be due to a chain length distribution present in the real material. Photogenerated charged solitons appear at lower energy than doping-generated solitons and this effect is not necessarily due to the presence of counterions in the doped material, as suggested previously. With the help of explicitly calcd. lattice and electron dynamics and within the Su-Schrieffer-Heeger Hamiltonian, it can be shown that neutral solitons in trans-polyacetylene start to become slightly mobile from 10 K and completely free above 100 K. This behavior agrees with exptl. data in the literature. The derivation of equations of motion is given. It is shown, that without explicit consideration of electron dynamics, the exptl. results cannot be reproduced.