Inelastic X-ray Scattering Study of Exciton Dynamics of a Small Organic Molecule

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For the first time, inelastic x-ray scattering (IXS) technique is used to study the exciton dynamics of an organic molecule Py-So at SP12U1. Combined with INDO/CI method, we found that excitons are of Frenkel type, and their sizes or range of their center-of-mass motion are confined to a small fraction of the molecule. Our results provide a comprehensive and clear picture of exciton and their dynamics in such molecules, which is crucial for understanding their optical properties, and will eventually benefit the design of materials of desired optical properties. On the technical side, our work may open up a new subfield of the application of this technique.

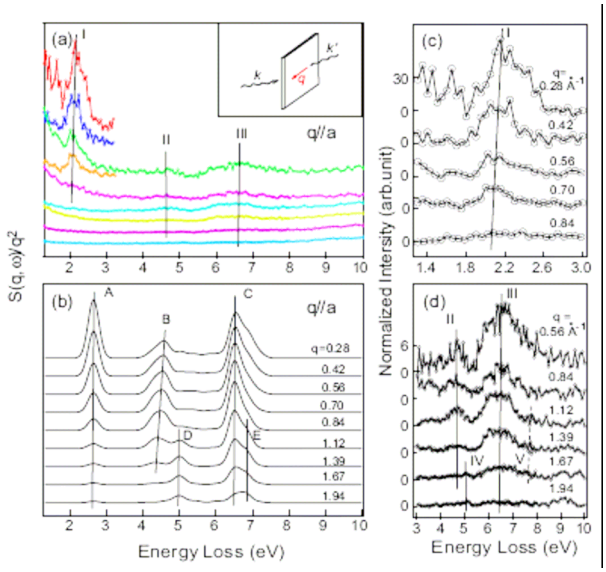

Figure 1. (a) Raw data got from IXS experiment. The spectra intensity is divided by q^2. For all spectra, momentum transfer q is parallel to a axis and in the ac plane of the sample. (b) INDO/CI simulated S(q,ω) divided by q^2 for Py-So. c) and d) Enlargements of spectra shown in (a) with background removed.

The IXS measurements were carried out at room temperature in transmission mode. The photon energy is 7908.75 eV. Energy resolution is 170meV. Experimental data of the Py-So molecular crystal measured by IXS are shown in Fig. 1a,c and d with momentum transfer q along the a axis. There are five distinct features, labelled as I~V respectively. The IXS spectra calculated for Py-So based on INDO/CI along the a axis are shown in Fig. 1b. Five distinct features are labeled as A~E. These theoretically spectra match the experiments qualitatively very well. There are one to one correspondence between the experimental feature I-V and A-E respectively.

For the lowest energy feature, A is only consisted of contributions from single exciton excitation. Its energy position, 2.6eV, is slightly higher than the experimental feature I. This because of the intermolecular hopping of the excitons, which is not considered in our single molecule calculation. Energy position of feature B~D are almost perfectly match experiment. These probably imply that the intermolecular interaction is not important for them. We note that the seeming dispersion of B is due to the intensity variance of its multiple components.

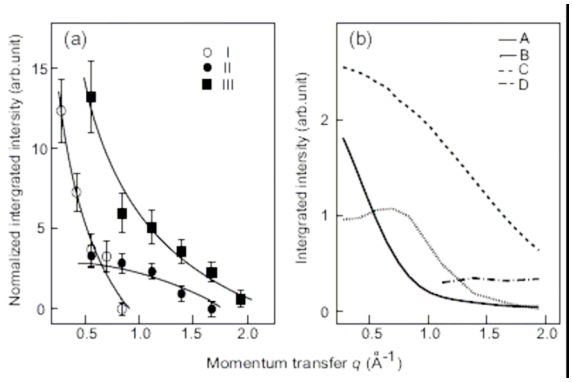

Figure 2. (a) Integrated intensities of peaks I, II, III in the spectra vs momentum transfer q. (b) Integrated intensities of the peaks A, B C and D shown in Fig.1b as function of momentum.

The momentum dependence of the integrated weight for the three main features are shown in Fig. 2a and b for experimental data and theory respectively. We can estimate the range of exciton center of mass motion. For lowest exciton, it is about 2.1Å which means its motion range is localize in a small region or molecule. Other two features also have same trend.

In the theoretical calculation, we also studied the exciton size of this system (Fig. 3). We find that for several main exciton, their size are about 2~3 Å. They are mainly confined in a fraction of the molecules.

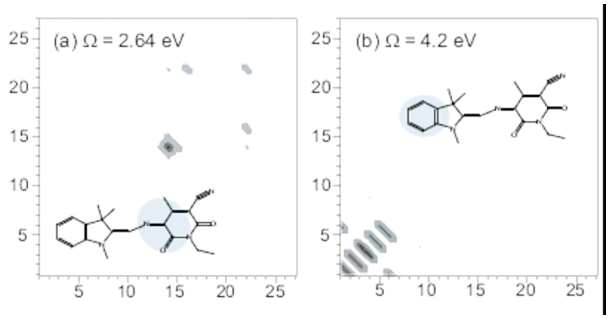

Figure 3. Contour plot of the absolute value of the exciton wavefunction calculated for Py-So molecule shown for excitation into:(a) peak I, 2.64eV. (b) peak II, 4.2eV. The axes are labelled by atoms at which electron/hole stay.