Electronic excitations of C_{60} and $(C_{60})_N$ (N = 2, 3 and 4) clusters

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Abstract

Excitation properties of the isolated C_{60} and $(C_{60})_N$ model clusters (N = 2, 3 and 4) are studied using an approximate quantum mechanical method, the Complete Neglect of Differential Overlap considering the *l* azimuthal quantum number method (CNDOL)[1]. After solving self-consistently the ground state, the excited states are calculated in the framework of configuration interaction of singles (CIS). We show that this method describes properly the electron excitations of the isolated C_{60} with accuracy comparable to time-dependent density functional theory. Figure 1 shows the experimental spectra in n-hexane solution [2], in gas phase at 973 K [3], as well as the spectra calculated with CNDOL and the popular methods of TDDFT [4] and CNDO/S [5]. The error bars indicate the positions of band maxima and bandwidths obtained by extrapolation of these magnitudes measured in a series of solvents with different dielectric constants, to the dielectric constant of vacuum [6]. It can be noticed that the CNDOL method, not including any fitting parameter, provides quit good values of the peak positions and intensities.

Next, we have studied the optical response of clusters of C_{60}. As an approximate method, CNDOL allows to study large molecular systems with a modest computational framework. Figure 2 shows the absorption spectra of the C_{60} clusters. The changes of the spectrum due to cluster effect are visible in the low energy part, where an additional peak appears for N=2 and 3. The high-energy part of the spectrum is rather unchanged. Despite the relatively reduced cost of CNDOL calculations, the huge size of the CIS matrix has limited this study to small clusters of four fullerenes. We expect to overcome these limitations using a Lanczos methods that do not require o storage the full matrix in the random access memory.

References

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Figures

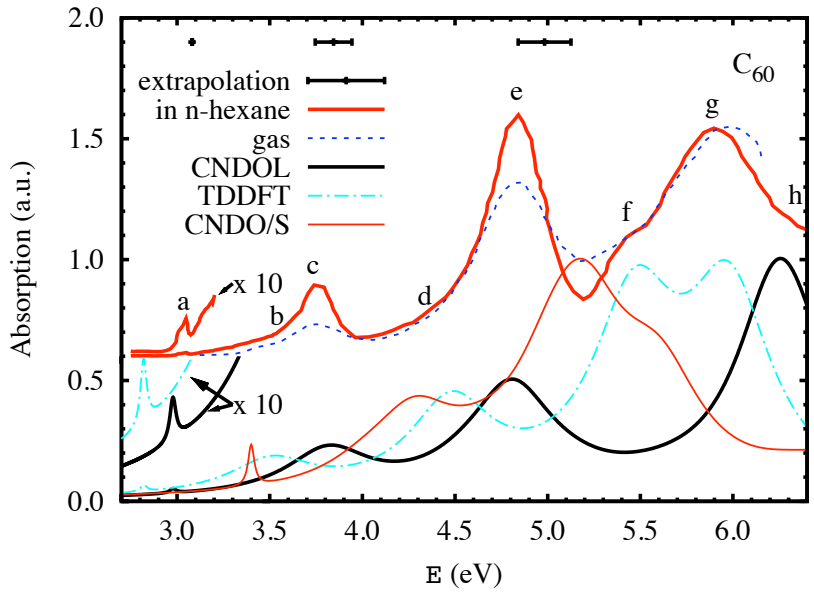

Fig. 1. Experimental and theoretical absorption spectra of C_{60}. The experiments are in n-hexane solution at 300 K [2] and in a molecular beam at estimated temperature 973 K [3]. The calculations are with CNDOL (this work), TDDFT [4], and CNDO/S [5]. Horizontal error bars (extrapolation) represent the extrapolated peak energies and line widths estimated for cold gas phase spectrum [6].

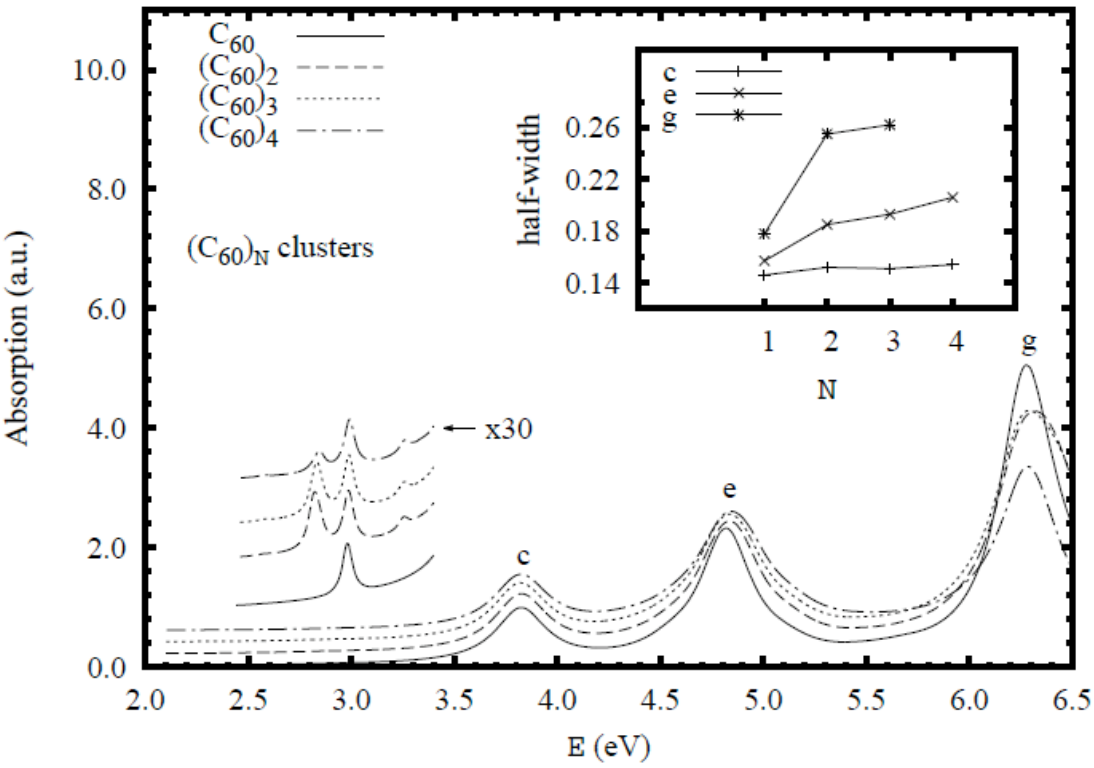

Fig. 2. CNDOL theoretical absorption spectra of $(C_{60})_N$ clusters. The inset shows the relation between the half-widths of the c, e and g bands with N in $(C_{60})_N$ models.