Non-Dipole Effects in Atomic and Molecular Photoemission

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Much of our understanding of the interaction between radiation and matter is based on the electric-dipole approximation (DA). Significant departures from the DA are commonly thought to occur only at wavelengths comparable to or smaller than the spatial dimensions of the absorbing electronic orbital, typically at a few KeV photon energy and higher. However, recent experiments and theoretical calculations have shown that significant non-dipole effects occur in atomic [1,2] and molecular [3] photoemission at much longer wavelengths than expected. In the soft-x-ray regime, the first-order effects are expected to be the major deviations from the DA. In photoemission, these first-order effects only affect the angular distribution of photoelectrons, not the cross section, and manifest themselves in a forward/backward asymmetry relative to the photon direction. Even so, experimental and theoretical findings show that also second-order effects occur for soft-x-ray photoionization, which may affect both cross section and angular distribution [2].
Here we review some of these experimental findings, and also report new observations of significant deviations from the DA in core-level photoemission from small molecules. In contrast to the situation for atomic targets, large deviations from the DA are found at surprisingly low photon energies, just a few eV above the core-level ionization threshold, for many small molecules such as CO, N_2, CO_2, CF_4 and SF_6. Particularly SF_6 has a complex behavior, with rapid variations in the non-dipole parameters over the t_{2g} and e_g resonances above the S $L_{2,3}$ edges. The measurements were performed at the Advanced Light Source synchrotron radiation facility at the Lawrence Berkeley National Laboratory. Four time-of-flight spectrometers, positioned both in and out of the plane perpendicular to the photon beam to make the non-dipole-effect measurements possible, were used to record the photoelectron spectra [4].

References

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