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Raman spectroscopy of natural carbons: from amorphous-like materials to graphite and diamond, from theory to experiments

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Raman spectroscopy is widely used to characterize carbonaceous materials (CM) from various natural contexts. From metamorphic rocks to archaeology or from soils to meteorites: scientific purposes are extremely diverse, but the studied objects are the same. Because of the unique properties of the C atom, among which its ability to create different hybridization with its atomic orbitals, there is a quasi-unlimited number of structure for disordered CM, generally in association with traces of heteroatoms (H, O and N). Raman spectroscopy is powerful not only to identify crystalline forms (graphite/diamond), but also to investigate the structure of these disordered materials. The first-order Raman spectrum of graphitic-like CM is composed by a G band (1580 cm^{-1}, E_{2g2} mode) and several defects bands and features in the second order due to overtone and/or combination scattering. Except for the G band, the significance of these bands is still a matter of debate and there is a huge and still rapidly growing literature on this issue. Although there is not a direct connection between the respective evolution of these bands and that of CM structure, the former is generally used as a marker of the process leading to the latter. Concerning diamond, the spectrum is apparently simple with one main mode at 1332 cm^{-1}, but any spectral change concerning this mode systematically generates a far-reaching interpretation.

Analyzing CM by Raman spectroscopy is not straightforward from the experiment to the interpretation stage. The question is what can we really say from the Raman spectrum of CM? From the literature and our own studies, both theoretical aspects and technical advices concerning spectra acquisition and treatment will be discussed and illustrated from various examples concerning graphitic-like materials as well as diamond.