Fist principles molecular dynamics study of Carbon-ferromagnetic metal based composites

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Since their discovery more than twenty years ago [1], carbon nanotubes (CNT) have attracted a growing interest because of potential applications in a wide range of next-generation electronic devices. Their acknowledged efficiency in terms of electronic and thermal transport, the relatively low production cost, and their controllable environmental impact [2] at least to some extents are counterbalanced by the intrinsic difficulty in growing a specific nanotube *in situ*. The physical properties of a CNT can be tuned by adding heteroatoms or clusters at the surface or by substitution of C sites directly on the CNT network. This can lead to new optical, electronic or magnetic properties according to the chemical nature of these heteroatoms [3-6]. Specifically, in the hyper-frequency domain, Sui and coworkers [7] have shown that Co atoms deposited on CNTs allow to modulate the frequency response of composite polymer/CNT systems resulting in a shift of the adsorption peak.
Despite the intensive experimental activity of the last ten years, a detailed atomic-level knowledge of the mechanisms regulating the interaction of heteroatoms and clusters with CNTs is still lacking. Nonetheless, this fundamental knowledge is instrumental to design and tune the resulting macroscopic properties, and in particular the dielectric response, of these composite materials. This calls for advanced simulations tools and methods suitable to realize reliable virtual experiments able to reproduce the behavior of CNT/Metal systems at finite temperature which, ultimately, determines the sought macroscopic properties. To date,, such a tool is represented by first principles molecular dynamics (FPMD) and related enhanced methods [8,9], which have already shown their maturity in predicting and quantifying the complex dielectric response of realistic materials [10]. In this context, preliminary studies have been realized at the IPCMS in the context of a M2 Master project, in collaboration with Dassault-Aviation. These FPMD studies have shown how it is possible to extract permittivity spectra in the case of a (10,10) CNT either pristine or carrying Co nanoclusters at the surface (Fig. 1).

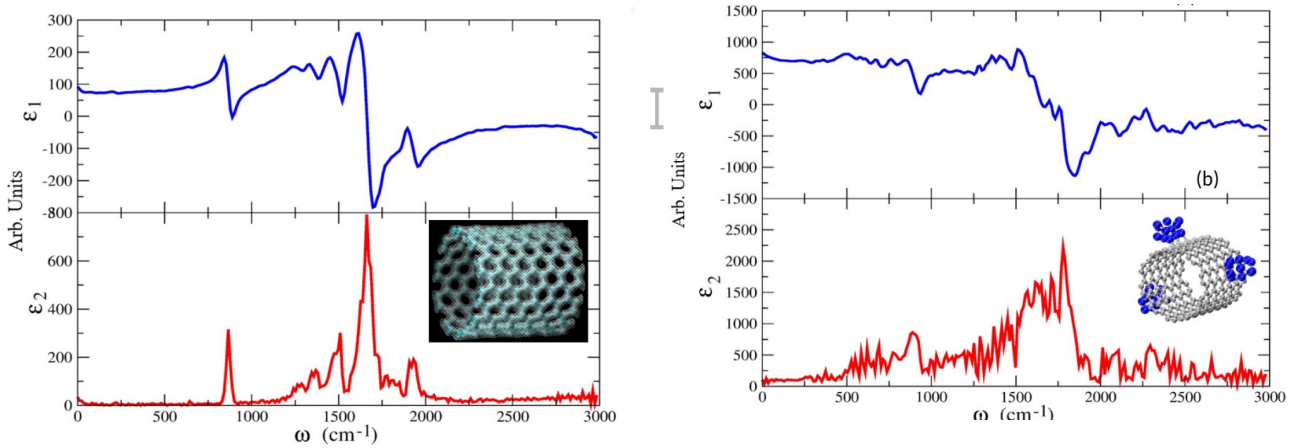

Figure 1. Adsorption spectrum and dielectric response (imaginary and real part of the dielectric function) of a (10,10) CNT without (left) and with (right) Co nanoclusters decorating the surface.

The present Ph.D project, scheduled for the academic years October 2022 - September 2025, will exploit FPMD methodologies for applications to CNT and carbon-based systems (mainly graphene) interacting with atoms, clusters and nanoparticles of ferromagnetic metals, such as Co, to tune and modulate the dielectric response in strict synergy with experiments. An accurate survey of the system size required, location and/or mobility of the metallic nano-objects at finite temperature and stability of the substrate will be the prerequisite to enable and extend the study toward the desired properties. The case of two-dimensional carbon-based systems such as graphene will be also targeted in view of their promising features as forefront materials for micro-wave adsorption [11]. The scope of this project is to unravel the underlying electronic structure in the temperature range where these systems are expected to be operational and to sort out the resulting complex dielectric function [10], directly obtained by dynamical simulations as a Fourier transform of the dipole autocorrelation function. Quantitative comparisons with experimental results will provide a solid background to test ad benchmark the computational results.
Massive parallel resources on high-performance computing (HPC) architectures are available for this study at the local HPC center (https://hpc.pages.unistra.fr/) of the Strasbourg university (Tier-2 level) and on national centers (Tier-1 level) under the GENCI (www.genci.fr) call for grants.

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