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Inverse Problem Method: A Complementary Way for the Design and the Characterization of Nanostructures

Dominique Barchiesi Automatic Mesh Generation and Advanced Methods (GAMMA3), University of Technology of Troyes (UTT) - Institut National de Recherche en Informatique et Automatique (INRIA), Troyes cedex, France

Thomas Grosges Automatic Mesh Generation and Advanced Methods (GAMMA3), University of Technology of Troyes (UTT) - Institut National de Recherche en Informatique et Automatique (INRIA), Troyes cedex, France

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Nanotechnology has reached a level of maturity that allows them to take advantage of the computer-aided design. The inverse problem methodologies can help to achieve this by exceeding the simple explanation of phenomena and the direct optimization of devices. The only condition is that the scientific community takes ownership of these methods and considers them to be effective development tools.

Context of Nanotechnology

Nanotechnology experienced massive development since the 80s. During the first decades, development was mainly directed towards the production of laboratory results, for understanding purposes. This phase has allowed the development of efficient experimental techniques of production of samples and of their characterization, as well as accurate models and computational codes, while reducing the necessary calculation time. However scientists who contributed to this development were mainly physicists and opticians, and were little aware of the technology in its dimension of computer-assisted development, which is well known in mechanics and materials sciences. Numerical optimization was used in classical optics but remains infrequent in nanotechnologies [1]. However models were generally not used to solve the inverse problem, to optimize fabrication process, nor to get abacus for complex phenomenon in nanotechonology. The following examples of resolution of the inverse problem, in order to get some heuristic laws for improving the engineering of nanostructures, show that this crucial step can be overtaken in nanotechnology.

Inverse Problem, Modeling and Optimization

The color control and sometimes dichroism of glass embedding nanoparticles is known since ancient times [2]. Nevertheless, the physical characterization of such media requires either chemical or physical analysis of a small piece of glass [3] [4]. The non-destructive analysis of such an artefact is of great interest even if spectroscopy is obviously tedious on the whole artefact and requires hazardous handling. To overcome these practical problems, the resolution of the inverse problem of the origin of color formation is an alternative or complementary way. The necessary tools are threefold: first, a model of the diffusion of light by nanoparticles (the Mie theory [5], the Finite Element Method [6], the S method for multilayers [7]); second, a model of color encoding in photographs (sRGB encoding [8]) and thirdly, an inverse problem method which is able to solve multivalued problems (the Particle Swarm Optimization (PSO) method among all [9]).

The PSO is based on the mimic of bees' swarms flying in the space of search of unknowns. The random search, the memory of

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the best position of each bee (the parameters set of the model), the memory of the very best position over the swarm and over all generations of the swarm make the method efficient to find families of possible solutions, leading to a controlled approximation of a target. This method was used to explain strange results in experiments by Turbadar [10] In this case the target was the experimental records obtained with Surface Plasmon Resonance (SPR) experiment. The oxidation of a thin aluminum layer was shown to be the source of the puzzling shape of records [11] The same method applied to the SPR but with more recent multilayered settlement of metals was shown to be able to recover the thicknesses [12] The dichroism of the Lycurgus cup was also explained by the presence of copper in nanoparticles [13].

The model and the inverse problem method being validated by using known data, the numerical process can be used to predict the behavior within its domain of validity. For example, the transmission coefficient was necessary but the dipole approximation of the Mie theory was shown to fail to describe the phenomenon. The possible sets of input parameters for the model are a source of data for statistics.

Behavior Laws for Engineering, Uncertainty and Tolerance

The model being established, the behavior laws can be deduced from the statistics of results and therefore they can help for engineering processes. The careful determination of the domain of validity of models is required ahead. For this, reference data should be produced from the same experiment, in very close conditions. Afterward, the inverse problem method can be used blindly with the model, and statistics of results can be considered as relevant. Obviously the selection of the results which fall below a given threshold of tolerance compared to specifications can help to design fabrication process in terms of uncertainty management [14] or tolerance [15] Indeed, the identification of critical parameters in physical phenomenon helps to design the experimental setup of characterization as well as the engineering process which is dedicated to the fabrication Figure 1 shows a schematic of the whole numerical process for engineering. The three color boxes distinguish the requirements for the elaboration of a model which can be used in an evolutionary loop to solve the inverse problem but also to recover statistics of possible solutions. This step gives not only the best parameters (inputs of the model) for a given nanostructure according to its application (biosensor, telecom device…), but also a set of acceptable alternatives that can be statistically processed to get heuristic behavior laws. The characterization of the critical parameters consists in the evaluation of their relative influence on the results given by the model. The associated study gives the information on the propagation of uncertainties. This information can be used to design the fabrication setup and specifically to choose the precision of each element of the chain of fabrication.

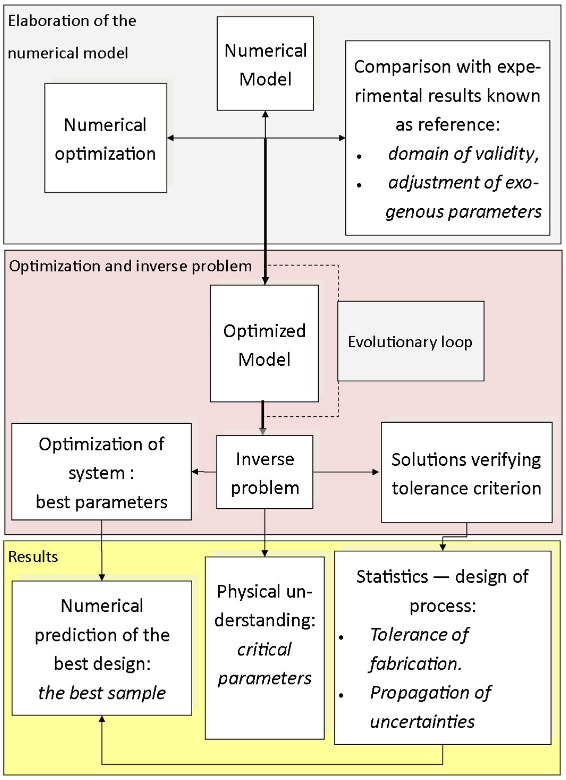

Figure 1. *Schematic of the numerical approach for engineering.*

An example of application of such a method using the same model as that used in Ref. [13] is following. Considering the photographs of colloidal dispersion of AuAg alloy nanoparticles of solutions [16] the resolution of the inverse problem gives a set of possible solutions. The best spectrum and the minimum and maximum values of possible spectra are shown in Figure 2. The original photograph from Ref. [16] is also shown as the reference color as well as the color obtained from the recovered parameters for the model (PSO).

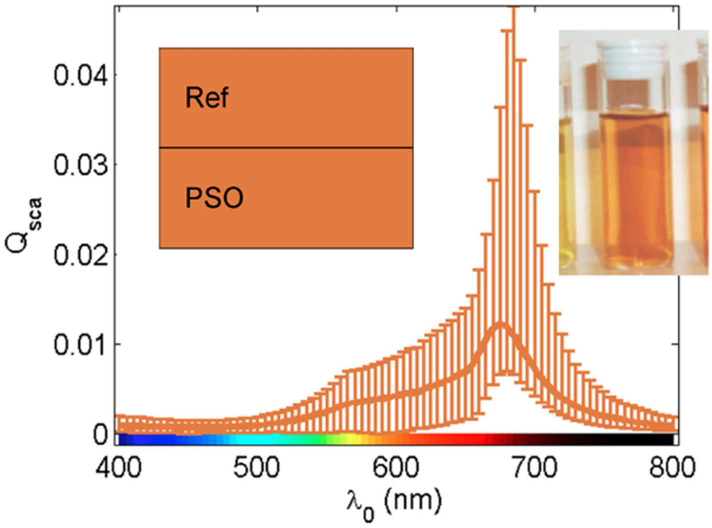

Figure 2. *Results of the inverse problem leading to the same color as in photographs.*

The basic statistic of recovered parameters as inputs of the model reveals the sensitivity of color to the radius of particles, to the proportion of silver and to the optical properties of the solution (effective refractive index). Hundred realizations of the inverse problem reveal the following. In this example the three results obtained from the evolutionary loop (Figure 1) are indicated.

- The most probable radius of particles is 15 nm (with standard deviation less than 1 nm) and the proportion of Ag is 0.05 (0.14). The most probable recovered effective refractive index is complex number: 2.67+0.06i (0.14 + 0.04i). The best values recovered from the inverse problem are: 13.2 nm for the radius, 0.61 for the proportion of silver, 2.69+0.20i for the effective refractive index of surrounding glass. The most probable values and the mean values (respectively 15 nm, 0.19, 2.64+0.11i) differ from the optimum and the standard deviation is an indicator of the sensitivity of the model to the input parameters.
- The sensitivity of the results to the radius is higher than that to the proportion of silver. The ratios of the standard deviation to the mean value are 3.4% for the radius, 76% for the proportion of silver, and 1% for the real part of the refractive index and 38% for its imaginary part. Therefore, the control of particles size seems to be more critical than its composition in the engineering process of fabrication if a given color must be produced.
- Moreover, the analysis of correlations between parameters gives behavior laws: the increasing of radius corresponds to a decrease of the proportion of silver to obtain the same color in photographs. For example, a proportion of silver p=0.6 corresponds to a radius R=13nm, and p=0.27 to R=15nm.

Obviously the analysis of colors in photographs cannot be considered as an accurate analysis of matter. However, the results are coherent with those obtained from spectroscopy and transmission electron micrographs [16] They could be considered as a first approach of characterization of nanoparticles embedded in transparent medium. Indeed, the radius of spherical particles can be considered as the equivalent mean radius of nanoparticles with different shapes. Any further information obtained from methods of analysis can be used to restrict the domain of possible solutions.

Another interesting application of the proposed method of the analysis of sensitivity of models that may impact the process of fabrication is devoted to the use of optical constants in numerical models. These constants (refractive index, relative permittivity) were measured for bulk materials. However, physics experiments shown that these values are modified in nanostructures. Therefore, their use as inputs in numerical models should be subject to a preliminary analysis of sensitivity to assess the numerical results, especially if the optimization of resonances is the goal.

Conclusion

This approach is classical in industry but is in its infancy in nanotechnologies where models are often reduced to assess or explain experiments by simple comparison of shapes. The reliability of models as well as the improvement of speed of calculation of numerical algorithms, allow their repeated use inside loops of heuristic methods for solving the inverse problem in nanotechnologies. The environment could benefit from an economy of costly and polluting experiments by using these methods to optimize nanosensors for medical and biological applications [17].

Even multiphysics phenomena in nanostructures with complex shape, can be modeled in a reasonable time [18], although an effort must still be done to further reduce the computational time and the accuracy of algorithms, especially in the case of Multiphysics problems. Moreover, these complementary approaches could lead to lower costs of development from industrial point of view. What remains to do is to promote this type of methodology so that scientists can appropriate it as a support tool for their studies. ■

Dominique Barchiesi

AASCIT member. Dominique Barchiesi is full professor at the University of Technology of Troyes (UTT), France and topical editor at the Optical Society of America. Research interests include theoretical physics, applied mathematics and statistics and processes of multidisciplinary science teaching. D. Barchiesi conducts active research in numerical modelling, optimization and advanced methods with applications to engineering of nanotechnologies and plasmonics. The result of his research has been published in over one hundred eighty articles, conferences, and book chapters since 1992, in the fields of optics, electromagnetism, signal processing, operational research and finite element methods.
dominique.barchiesi@utt.fr (Corresponding author)

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