Polarizability of Ferroelectric Particles

Daniel Sjöberg[(1)]
(1) Lund University, Lund, Sweden (daniel.sjoberg@eit.lth.se)

Extended abstract

Ferroelectric materials are the electric counterpart of ferromagnetic materials, where a remanent polarization is maintained by a local minimum in the free energy. They attract current interest in electronics as transistor gate materials and memory technology, but are also interesting on their own as a fundamental example of nonlinear materials with tunable scattering properties. In this paper, we focus on properties relevant for Rayleigh scattering, that is, the polarizability of a ferroelectric particle. In the Landau-Ginzburg-Devonshire dynamic model of ferroelectricity, the free energy density density G is

$$G = a_{ij}^{(1)} P_i P_j + a_{ijkl}^{(2)} P_i P_j P_k P_l + a_{ijklmn}^{(3)} P_i P_j P_k P_l P_m P_n + \frac{1}{2} g_{ijkl} P_{k,i} P_{l,j} - P_i E_i - \frac{1}{2}\varepsilon_0 \kappa_{ij} E_i E_j + \frac{1}{2} c_{ijkl} (\varepsilon_{ij} - Q_{ijmn} P_m P_n)(\varepsilon_{kl} - Q_{klmn} P_m P_n) \quad (1)$$

where the Einstein index summation convention is used (repeated indices are summed over and $P_{i,j}$ means P_i is differentiated with respect to the jth coordinate), P_i is the polarization, E_i is the electric field, and ε_{ij} is the strain. It was demonstrated in [1] how this free energy can be simulated by finite elements using the Landau-Ginzburg-Devonshire dynamics (where $\boldsymbol{D}$ and $\boldsymbol{\sigma}$ are the electric flux density and mechanical stress, respectively)

$$\frac{1}{L}\frac{\partial \boldsymbol{P}}{\partial t} = -\frac{\partial G}{\partial \boldsymbol{P}}, \quad 0 = \nabla\cdot\boldsymbol{D} = -\nabla\cdot\frac{\partial G}{\partial \boldsymbol{E}}, \quad \boldsymbol{0} = \nabla\cdot\boldsymbol{\sigma} = \nabla\cdot\frac{\partial G}{\partial \boldsymbol{\varepsilon}} \quad (2)$$

We apply a small signal analysis to this equation, that is, we assume $\boldsymbol{P} = \boldsymbol{P}_0 + \Delta\boldsymbol{P}$ and $\boldsymbol{E} = \boldsymbol{E}_0 + \Delta\boldsymbol{E}$ where $\boldsymbol{P}_0$ and $\boldsymbol{E}_0$ correspond to the static remanent condition, whereas $\Delta\boldsymbol{P}$ and $\Delta\boldsymbol{E}$ are small signals with time harmonic time dependence $\mathrm{e}^{\mathrm{j}\omega t}$, where ω is the angular frequency of the incident wave. It is then seen that the small signal susceptibility $\boldsymbol{\chi}$ is given by

$$\Delta\boldsymbol{P} = \left[\frac{\mathrm{j}\omega}{L}\mathbf{I} + \left.\frac{\partial^2 G}{\partial \boldsymbol{P}^2}\right|_{\boldsymbol{P}_0}\right]^{-1}\cdot\Delta\boldsymbol{E} = \varepsilon_0\boldsymbol{\chi}\cdot\Delta\boldsymbol{E} \quad (3)$$

where the spatial variation of $\boldsymbol{\chi}$ depends on the solution for the remanent polarization $\boldsymbol{P}_0$, which is a nonlinear problem. Given the function $\boldsymbol{\chi}(\boldsymbol{r})$ of a ferroelectric particle and considering Rayleigh scattering (particles small compared to wavelength), a linear electrostatic problem can be solved from which the particle's total small signal dipole moment can be computed (where $\boldsymbol{E}_0$ represents the amplitude and polarization of the incident wave, and $\boldsymbol{\kappa}(\boldsymbol{r})$ is the background permittivity of the ferroelectric in the expression for G)

$$\nabla\cdot[\varepsilon_0(\boldsymbol{\kappa}(\boldsymbol{r}) + \boldsymbol{\chi}(\boldsymbol{r}))\cdot(\boldsymbol{E}_0 - \nabla u)] = 0 \quad \Rightarrow \quad \boldsymbol{p} = \varepsilon_0\int(\boldsymbol{\kappa}(\boldsymbol{r}) + \boldsymbol{\chi}(\boldsymbol{r}) - \mathbf{I})\cdot(\boldsymbol{E}_0 - \nabla u)\,\mathrm{d}V = \varepsilon_0\boldsymbol{\gamma}\cdot\boldsymbol{E}_0 \quad (4)$$

which defines the small signal polarizability matrix $\boldsymbol{\gamma}$ of the particle. This single 3×3 matrix is the only information relevant for the small signal problem of Rayleigh scattering against the particle, and as such makes a significant reduction of modelling parameters for this problem, although the original nonlinear problem contains a lot of parameters and is challenging to solve. In the presentation, we go deeper into how this polarizability matrix $\boldsymbol{\gamma}$ can be tuned by varying the remanent polarization $\boldsymbol{P}_0$ which is the solution to the nonlinear problem.

References

[1] D. Sjöberg, "Analysis of nonlinear effects in ferroelectric materials using phase field simulations in open source finite elements software," in *URSI GASS 2023, Sapporo, Japan, 19 – 26 August 2023*, 2023.