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Title

Second-harmonic generation in ferroelectric thin films on metal substrates for isotropic and cubic symmetries

Source

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Abstract

The Landau-Ginzburg phenomenological theory of ferroelectrics and Landau-Khalatnikov equations of motion are used to model the interaction of electromagnetic radiation and a ferroelectric thin film on a metal substrate. The analysis starts with the minimization of a Gibbs free energy functional that accounts for the free energy cost of the boundaries of the thin film through a gradient term that is non-zero when the polarization in the film varies. The minimization procedure leads to an Euler-Lagrange equation that can be solved to find the equilibrium polarization in the film. Next the nonlinear dynamical equations that describe the response to an incident electromagnetic field are solved by a perturbation expansion about the equilibrium polarization. The second-harmonic generation term from the expansion is selected. We then go on to calculate a reflection coefficient for the harmonic generation term and investigate how the finite thickness of the film influences the reflection coefficient. In Landau theory the symmetry of the crystal making up the film in the paraelectric phase is reflected in the free energy expression. The analysis here is done for an isotropic symmetry that is often used as an approximation to the actual crystal symmetry and which is mathematically easier to handle. However it is also indicated how the free energy expression can be changed so that it is appropriate for cubic symmetry, and a discussion of how the second harmonic term can be calculated for this case is given. The theory presented is relevant to experimental studies using far-infrared or terahertz reflection measurements because the ferroelectric film is resonant in the far infrared and terahertz ranges.