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# Phonon polaritons impact atmosphere transparency in the TeraHertz range

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**Abstract**—Atmospheric aerosols impact the climate by modifying the radiative balance of the planet and the microphysical properties of clouds and by altering the biogeochemical balance of the atmosphere. Above all, some aerosols exhibit fascinating effects due to the phonon-polaritons that can form in their structure. In this study, we investigate the impacts of phonon-polaritons in the atmosphere by comparing TeraHertz Time Domain Spectroscopy (THz-TDS) experiments on resuspended and pelletized calcite to Finite Difference Time Domain (FDTD) simulations and Mie theory. The contributions made here should be of wide interest in THz research applied to natural sciences.

**Index Terms**—Calcite, Aerosol, TeraHertz Time Domain Spectroscopy, Polariton.

## I. INTRODUCTION

Aerosol particles are ubiquitous in the atmosphere and exert a significant influence on global climate. First and foremost, atmospheric aerosols impact climate by directly scattering and absorbing incoming solar radiation and Earth surface radiation [1]. Therefore, a direct effect of atmospheric aerosols is aerosol-induced radiative forcing, which will alter the planet's radiation budget. Aerosols have also profound indirect effects that will influence the micro-physical properties of clouds and the biogeochemical stability of the atmosphere. During transport, the particles will alter the physicochemical environment of the atmosphere by mixing internally with secondary compounds such as ammonium sulfate, ammonium nitrate, carboxylic acids, organic materials containing nitrogen and sea salt by coagulation, cloud processing and heterogeneous reactions.

Furthermore, some of these particles will exhibit peculiar properties due to the fascinating quasi particles called

phonon-polaritons they support. The phonon-polariton is a type of polariton that form in a solid due to the coupling of phonons of the lattice and photons. Such strong coupling between light and matter has been investigated for their role in controlling chemical reactions in the so-called polaritonic chemistry/photochemistry or used for instance as radiative cooling materials [2].

Therefore, the polaritonic states found in some atmospheric aerosols raise a myriad of questions regarding the consequences that strong light-matter coupling could have on the climate : What are their impacts on radiative budget calculations ? Do they interfere with atmospheric reactions ? To what extent would their presence disrupt the prevailing examination procedure of the climate system ?

The overall goal of this work is thereby to study the possible effects of phonon-polaritons in the atmosphere by measuring calcite aerosol response experimentally and comparing it to simulations and theoretical calculations based on Mie theory. This thesis makes a number of valuable contributions to the field of natural sciences. An important contribution is also made to research on THz waves as the atmospheric phonon polaritons make THz frequencies truly relevant in radiative balance calculations and environmental studies.

## II. EXPERIMENTS AND SIMULATIONS

In this study, the optical response of resuspended calcite (calcium carbonate) particles is measured by TeraHertz Time Domain Spectroscopy (THz-TDS) in a controlled environment with an aerosol measuring bench set-up in the IEMN laboratory. Calcite aerosol is a valuable case study since it shows a

phonon-polariton at 3.4 THz [3]. Additionally, calcite particles are emitted in the atmosphere from several anthropogenic and natural sources including certain large deserts of which it is one of the major components (e.g. the Gobi Desert). The sizes and concentrations of the resuspended particles are recorded simultaneously by an optical particle size analyzer during the acquisition of the time traces. The aerosol data are paired to experiments carried out on pellets of 10 mg calcite powder diluted in docosane [4].

A morphological and chemical characterization of the sample is realized as well by Scanning Electron Microscopy - Energy dispersive X-rays on the collected resuspended particles and X-ray diffraction on powder in order to study the link between the optical response, morphology and chemical composition of the sample.

The experimental study is combined with Finite Difference Time Domain (FDTD) simulations, widely used in the nanophotonics community to effectively simulate the interaction of light with materials (Ansys Lumerical software) by solving a modified form of Maxwell's differential equations [5]. The simulations are run for sphere radii ranging from 0.1 to 20 microns. Additionally, a Mie theory calculator python code is implemented within our group to directly compare simulation results to Mie theory.

### III. RESULTS

Figure 1 compares the effective experimental absorption cross sections of aerosol and pelletized calcite with the simulated absorption cross sections of a single isotropic calcite sphere (ordinary indices) of 2.65  $\mu\text{m}$  and 12.35  $\mu\text{m}$  radius.

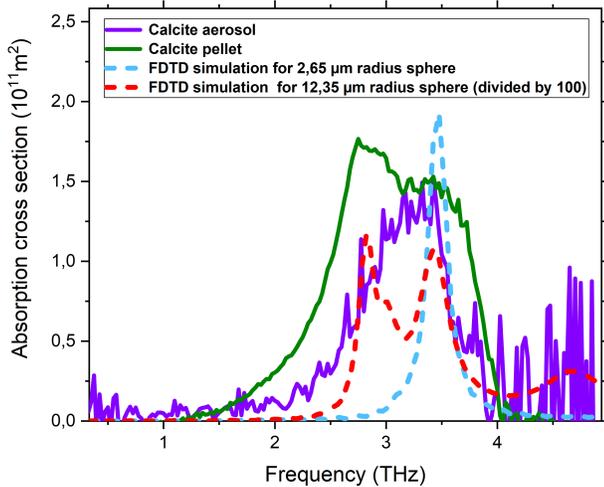


Fig. 1. Calculated effective cross section of calcite aerosol (purple) and calcite pellet (green) compared to simulated absorption cross section of a single calcite sphere of 2.65 microns radius (dotted, blue) and of 12.35  $\mu\text{m}$  radius (dotted, red) in the 0.2-5 THz range. The cross section of 12.35  $\mu\text{m}$  has been scaled by dividing by 100.

Simulations were performed on 2.65 and 12.35  $\mu\text{m}$  radii spheres as the signal contribution of larger particles is the

greatest although they are less among resuspended particles. In this case, simulated cross section for the 12.35  $\mu\text{m}$  sphere is around 100 times greater than that for the 2.65  $\mu\text{m}$  sphere. Larger particles will therefore be of interest when analyzing phonon-polariton modes of atmospheric aerosols.

Moreover, small calcite particles exhibit a peak at 3.4 THz which demonstrates a strong absorption characteristic of polariton modes. When the simulation is carried out without the polariton, no absorption or scattering can be observed at this scale. With increasing particle size, the initial peak shift to lower frequencies and other modes begin to contribute, notably at 2.8 THz for the 12.35  $\mu\text{m}$  sphere. The simulations show therefore an effect of particle size on the polariton mode as well as on the Mie modes. These effects also highlight the broad measured spectrum generated by the experimental size distribution and the differences that can be observed between pellet and aerosol calcite measurements.

### IV. CONCLUSION

The experimental particle size distribution will be applied subsequently in order to simulate the absorption cross sections comparable to an atmospheric aerosol. The present achievements confirm that phonon polaritons reveal a major effect that can multiply atmospheric aerosol losses. The results of this study indicate that these effects will be much greater in the infrared region regarding the consequences in atmospheric chemistry/photochemistry or in the planet's radiation balance calculations. During the conference, we will compare our results to Mie and Rayleigh theories in order to better understand the underlying photonic mechanisms. Finally, our study suggests that the polariton is much more than an object of interest for the physicist/engineer: the polariton is present in nature and its impacts in climate change must be studied from THz to infrared.

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