國立臺灣大學理學院物理學研究所

碩士論文

Department of Physics

College of Science

National Taiwan University

Master’s Thesis

電介質系統於時間調和電磁場下之共振與極化電荷及其熱效應，能量傳輸與交互電場增強之探討

Dielectric Systems under Time Harmonic Electromagnetic Field - the effect of resonances and polarization charges, and the resulting heating, energy transport and mutual enhancement of electric field

楊鈞禹

Chun-Yu Yang

指導教授: 朱國瑞 博士

Advisor: Kwo-Ray Chu Ph.D.

中華民國 113 年 6 月

June, 2024

doi:10.6342/NTU202400923

國立臺灣大學碩士學位論文
口試委員會審定書
MASTER'S THESIS ACCEPTANCE CERTIFICATE
NATIONAL TAIWAN UNIVERSITY

電介質系統於時間調和電磁場下之共振與極化電荷及

其熱效應，能量傳輸與交互電場增強之探討

Dielectric Systems under Time Harmonic Electromagnetic

Field - the effect of resonances and polarization charges,

and the resulting heating, energy transport and mutual

enhancement of electric field

本論文係 楊鈞禹 (姓名) R09222045（學號）在國立臺灣大學 物理 (系/所/學位學程)完成之碩士學位論文，於民國 113 年 6 月 24 日承下列考試委員審查通過及口試及格，特此證明。

The undersigned, appointed by the Department / Institute of Physics on 24 (date) 6 (month) 2024 (year) have examined a Master's thesis entitled above presented by Chun-Yu Yang (name) R09222045 (student ID) candidate and hereby certify that it is worthy of acceptance.

口試委員 Oral examination committee:

朱國瑞

（指導教授 Advisor）

姜雁元　劉偉強　陳仕宏

doi:10.6342/NTU202400923

Acknowledgements

總之，先謝謝給予我幫助過的大家，人實在太多了而我只能列舉其中一部分。謝謝我的親人無條件地支持我，臺灣的科技部（現改制為國家科學及技術委員會）與教育部的部分財務支持。感謝我的指導老師朱國瑞的指導，以及在各個層面被我雷過卻展現極大包容力的人們。我要特別感謝陳俞安，楊順福，吳玫瑩，吳建昌，楊州池，楊黃柑仔，陳邑鳳，楊榮杰，白秀棻，楊芷卉，楊芷珊，陳永芳，王喬萱，管希聖，蔣正偉，陳宗恩，張峻維，林敏聰，朱啟丹，陳品翰，劉立中，許展毓，梁如青，蔡有方，Michael Raab(德國)，Paulo von Peterson(德國)，林仕晉，葉宸睿，吳祐綾，葉宸宇，葉智琳，顏宏秋，葉青蒼，葉友，李芳銓，鄭正浩，王祥旭，朱士維，石琳，陳惠琳，陳惠娟，陳張明花，以及我沒能回想到的人們，還有國立台灣大學學務處，國立台灣大學學務處學生心理輔導中心，國立台灣大學學務處學生心理輔導中心資源教室，國立台灣大學醫學院附設醫院，國立台灣大學物理學系，國家同步輻射研究中心，國立成功大學太空與電漿科學研究所，法國艾克斯-馬賽大學，國際熱核融合實驗反應爐。本論文使用彭新翔在 Overleaf 平台上以 Creative Commons CC BY 4.0 (http://creativecommons.org/licenses/by/4.0/) 授權方式提供的國立臺灣大學碩博士學位論文 LaTeX 模板 (https://www.overleaf.com/latex/templates/national-taiwan-university-thesis-template/hvfybyfxgztt) 排版，並做了些許修改。

楊鈞禹 2024 年 6 月於臺北，臺灣

doi:10.6342/NTU202400923

Thanks to everyone who has helped me. Thanks to my family members who had supported me unconditionally, and Ministry of Science Technology (now National Science and Technology Council) of Taiwan and Ministry of Education of Taiwan for partial financial support. Thanks to my advisor Kwo-Ray Chu (朱國瑞) for advising me. Thanks to my peers for helping me. I want to, especially, thank Yu-An Chen (陳俞安) *et alia* who I don't know their English name spellings, and also people who are not listed because of my poor memory. Their Chinese or German names are listed above in the Chinese passage. Supporting institutions such as Office of Student Affairs and Student Counseling Center, Disability Support Services and the Hospital of National Taiwan University are also acknowledged for their support during my study. Department of physics in NTU, NSRRC, National Cheng Kung University Institute of Space and Plasma Sciences, Aix-Marseille University in France and International Thermonuclear Experimental Reactor (ITER) are also acknowledged. This thesis was written by Chun-Yu Yang, using the LaTeX template provided by Hsin-Hsiang Peng on Overleaf[1] under Creative Commons CC BY 4.0 (http://creativecommons.org/licenses/by/4.0/) with some minor modifications.

Chun-Yu Yang

ORCiD: 0009-0006-1160-387X

Taipei, Taiwan, June 2024

[1] https://www.overleaf.com/latex/templates/national-taiwan-university-thesis-template/hvfybyfxgztt

doi:10.6342/NTU202400923

摘要

給定一個系統（我們想描述的客體），例如一顆在真空中的電介質球，然後我們用某個從遠處發出的單頻電磁波持續照射它，它會持續散射這些電磁波；這是一個光與物質交互作用的例子，古典電動力學的彈性散射模型（如米理論）讓我們可以描述這個系統中各個位置和時間的電磁場到一定的精確度。然而，本論文將指出，古典彈性散射模型會違反調和場的坡印廷定理以及背後可能的原因，例如物質的電磁性質會隨加熱改變，因此穩態電磁性質的假設不成立，或是能量有可能透過非彈性散射進入其他頻率域，以及因為熱傳需要的溫度梯度違反均質假設。最後，本文會回顧兩個受電磁波照射的雙體系統的中央間隙電漿生成機制的理論，並指出極化電荷造成的電場熱點促使了電漿的生成。

關鍵字：極化電荷、雙體、電場熱點、米理論、坡印廷定理

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

Abstract

Given a system (what we want to describe), for example, a dielectric sphere in vacuum; if we illuminate this sphere continuously using a monochromatic plane wave, the sphere will keep scattering the plane wave. This is an example of light-matter interaction, and classical elastic scatter model such as Mie theory can describe this system to a certain accuracy. However, this thesis will show that the classical elastic scatter model will violate Poynting's theorem for harmonic fields and discuss the probable cause, such as the violation of steady-state assumption of electromagnetic property because of heating, or the energy may enter other frequency through inelastic scatter, and the temperature gradient needed for heat transfer violates the homogeneity assumption. Finally, this thesis will discuss two theories regarding the mechanism of plasma formed between the gap of dimer system under electromagnetic irradiation, and favor the theory that the electric field hot-spot due to mutual enhancement of polarization charges causes the plasma formation.

Keywords: polarization charges, dimer, electric field hotspot, Mie theory, Poynting's theorem

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

Contents

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

List of Figures

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

List of Tables

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

Denotation

AC	Alternating Current, which means the field is oscillating.
C	Coulomb. The elementary charge, e, is defined to be 1.602 176 634 $\times$ 10^{-19} C in SI.
c	Speed of light in vacuum (free space). c is *defined* to be exactly 299 792 458 m/s in SI. The time is defined by defining the unperturbed ground state hyperfine transition frequency of the caesium 133 atom, $\Delta\nu_{\mathrm{Cs}}$, is 9 192 631 770 Hz in SI. It can be derived that $c = 1/\sqrt{\epsilon_0\mu_0}$. In this thesis, I assume the speed of light in air is also c.
$\mathbb{C}$	The set of all complex numbers. $\mathbb{C} = \{a + bi \,\vert\, a, b \in \mathbb{R}\}$.
CE	Common Era. For example this thesis is finished in 2024 CE.
DC	Direct Current, which means the field is static.
EM	Electromagnetic.
EMW	Electromagnetic waves.

doi:10.6342/NTU202400923

E	Electric field, which is a vector field, and the field strength is in units of V/m.
E	Energy of a photon, $E = hf = \hbar\omega$.
E_{ext}	Static far electric field.
E_{gap}	Gap electric field.
f	Frequency of electromagnetic field (EMW), or the frequency of the photons.
FEM	Finite Element Method
H	Magnetic field.
h	Planck Constant. h is defined to be 6.626 070 15 $\times$ 10^{-34} J s in SI.
$\hbar$	Reduced Planck Constant ($\hbar = h/2\pi$).
$\in$	is an element of, or belongs to.
MD	Molecular Dynamics
MDRs	Morphology dependent resonances (Mie resonances).
ω	Angular frequency of electromagnetic field ($\omega = 2\pi f$), or a photon ($\omega = E/\hbar$).
$\mathbb{R}$	The set of all real numbers. $\mathbb{R}$ is a subset of $\mathbb{C}$. ($\mathbb{R} \subset \mathbb{C}$).

doi:10.6342/NTU202400923

SI	*SI Brochure: The International System of Units*. In this thesis I use the 9th edition of SI published in 2019 CE.
T	Temperature.
ϵ	Permittivity, usually contains imaginary part since mediums are in general lossy. The constitutive equation requires $\mathbf{D} = \epsilon \mathbf{E}$.
ϵ_0	Permittivity in free space (in this thesis, in vacuum or air). ϵ_0 = 8.854 187 8188(14) $\times$ 10^{-12} F $\cdot$ m^{-1} from CODATA 2022.
ϵ_r	Relative permittivity ($\epsilon_r = \epsilon/\epsilon_0$).
$\epsilon^{'}$	Real part of permittivity.
$\epsilon^{''}$	(Minus) Imaginary part of permittivity. Note that $\epsilon = \epsilon^{'} \pm i\epsilon^{''}$ if the time-dependence of the harmonic field is set to be exp$(\mp i\omega t)$.
λ_0	Wavelength of EM waves in free space. $f\lambda_0 = c$
λ_d	Wavelength of EM waves in dielectrics. $n_d\lambda_d = \lambda_0$
μ	Permeability of the material. The constitutive equation requires $\mathbf{B} = \mu \mathbf{H}$. In this thesis everything is assumed to be non-magnetic and therefore $\mu = \mu_0$, so **H** in this thesis is continuous everywhere like **B**.
μ_0	Magnetic permeability in free-space (in vacuum or air in this thesis). μ_0 = 1.256 637 061 27(20) $\times$ 10^{-6} N $\cdot$ A^{-2} from CODATA 2022.
μ_r	Relative permeability ($\mu_r = \mu/\mu_0$). For non-magnetic medium $\mu_r = 1$

doi:10.6342/NTU202400923

n Complex index of refraction. $n = \sqrt{\mu_r \epsilon_r}$

n_d Real part of n in non-magnetic dielectrics. $n_d = \mathrm{Re}(n) = \sqrt{\frac{|\epsilon_r| + \mathrm{Re}(\epsilon_r)}{2}}$

doi:10.6342/NTU202400923

Chapter 1 Introduction to Polarization Charges

In this chapter I discuss the basic idea of *polarization charges* and formalism of *permittivity*. Through out this thesis I assume that the observer is in the same rest frame with the dielectric medium (which is our system) initially. I exploit the 9th edition of *SI Brochure: The International System of Units* (SI)[1] published in 2019 Common Era (CE) by *Bureau International des Poids et Mesures*; the length is in unit of meters (m); the mass is in units of kilograms (kg); the time is in unit of seconds (s), and the charge is in unit of coulombs (C). The seven defining constants of the SI and the seven corresponding units they define are ascribed in [Table 1.1]. An interesting thing worth noted is that SI actually defines 1 meter by defining speed of light in vacuum c to be 299 792 458 m/s and define the time by defining the unperturbed ground-state hyperfine transition frequency of Caesium-133 $\Delta\nu_{\mathrm{Cs}}$ to be 9 192 631 770 Hz.

From CODATA 2022 provided on NIST website, vacuum electric permittivity[2] $\epsilon_0 = 8.854\ 187\ 8188(14) \times 10^{-12}\ \mathrm{F} \cdot \mathrm{m}^{-1}$ and vacuum magnetic permeability[3] $\mu_0 = 1.256\ 637\ 061\ 27(20) \times 10^{-6}\ \mathrm{N} \cdot \mathrm{A}^{-2}$. [(..) denotes the uncertainty in the final two digits. (88$\pm$14 and 27$\pm$20)].

[1] https://www.bipm.org/en/publications/si-brochure/
[2] https://physics.nist.gov/cgi-bin/cuu/Value?ep0|search_for=permittivity
[3] https://physics.nist.gov/cgi-bin/cuu/Value?mu0|search_for=permeability

doi:10.6342/NTU202400923

Defining constant	Symbol	Numerical value	Unit
hyperfine transition frequency of Cs	$\Delta\nu_{\text{Cs}}$	9 192 631 770	Hz (s^{-1})
speed of light in vacuum	c	299 792 458	m s^{-1}
Planck constant	h	$6.626\,070\,15 \times 10^{-34}$	J s
elementary charge	e	$1.602\,176\,634 \times 10^{-19}$	C
Boltzmann constant	k	$1.380\,649 \times 10^{-23}$	J K^{-1}
Avogadro constant	N_{A}	$6.022\,140\,76 \times 10^{23}$	mol^{-1}
luminous efficacy	K_{cd}	683	lm W^{-1}

Table 1.1: The seven defining constants of SI and the seven corresponding units they define, directly ascribed from Table 1 in English version of *SI Brochure* published in 2019 CE by *Bureau International des Poids et Mesures*. (https://www.bipm.org/en/publications/si-brochure/), distributed under the terms of the Creative Commons Attribution 3.0 IGO License (https://creativecommons.org/licenses/by/3.0/igo/).

1.1 Polarization P and the displacement electric field D

When immersed in a static electric field, a dielectric object will generate internal field against the external field (See [Figure 1.1], which is taken from [1, Fig. 1(a)][4]). This is the consequence of spatial variation of charge density (e.g., electron cloud around atomic nuclei) due to external field. Such effect is called polarization. Vivid figures demonstrating these ideas were provided by John David Jackson in his 3rd edition of *Classical Electrodynamics* (see [2, Figure 4.2 and Figure 4.7]).

For a dielectric medium, the polarization effect can be described with the formula,

$$\mathbf{D}(\mathbf{r}, t) \equiv \epsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}(\mathbf{r}, t), \tag{1.1}$$

where $\mathbf{D}(\mathbf{r}, t)$ is the electric displacement field at position $\mathbf{r}$ and time t, and from now on the $(\mathbf{r}, t)$ suffix denotes the same meaning, ϵ_0 is permittivity in free space (vacuum), $\mathbf{E}(\mathbf{r}, t)$ is the local electric field, and $\mathbf{P}(\mathbf{r}, t)$ is the polarization density. The electric displacement field $\mathbf{D}(\mathbf{r}, t)$ considers only free charges in the system, and thus the electric displacement

[4] https://doi.org/10.1063/5.0062014

doi:10.6342/NTU202400923

field must be continuous if there are no free charges in the system (If charges are not free charges, they are said to be bound charges, meaning that they have opposite charges binding them in close vicinity). Note that boldface indicates that the quantity is a three-dimensional (3D) vector, and **D**, **E**, **P** are generally time-dependent vector fields in 3D. Taking the divergence of Equation (1.1) gives:

$$\nabla \cdot \mathbf{D}(\mathbf{r}, t) = \epsilon_0 \nabla \cdot \mathbf{E}(\mathbf{r}, t) + \nabla \cdot \mathbf{P}(\mathbf{r}, t) \tag{1.2}$$

where

$$\nabla \cdot \mathbf{D}(\mathbf{r}, t) = \rho_f(\mathbf{r}, t) \tag{1.3}$$

$$\epsilon_0 \nabla \cdot \mathbf{E}(\mathbf{r}, t) = \rho(\mathbf{r}, t) \tag{1.4}$$

$$\nabla \cdot \mathbf{P}(\mathbf{r}, t) = -\rho_b(\mathbf{r}, t) \tag{1.5}$$

ρ_f , ρ and ρ_b are charge density of free charges, total charges and bound charges, respectively. It clearly follows that

$$\rho = \rho_f + \rho_b. \tag{1.6}$$

In short, the displacement field allows us to consider the field without bound charges first, and then add the effect of bound charges. Also, by this formulation, we are able to transform the problem of solving electric field into solving the permittivity discussed in Section 1.2.

doi:10.6342/NTU202400923

1.2 Permittivity of a material

1.2.1 Fourier Transform

Fourier transform (F.T.) converts the original scalar function $\Psi(\mathbf{r}, t)$ in time t domain into a complex valued function $\hat{\Psi}(\mathbf{r}, \omega)$ in angular frequency ω domain by the formula (see Jackson [2, p.243]),

$$\hat{\Psi}(\mathbf{r}, \omega) = \int_{-\infty}^{\infty} \Psi(\mathbf{r}, t) e^{i\omega t} dt \tag{1.7}$$

with the inverse transformation being the Inverse Fourier Transform (Inverse F.T.),

$$\Psi(\mathbf{r}, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \hat{\Psi}(\mathbf{r}, \omega) e^{-i\omega t} d\omega \tag{1.8}$$

or in short,

$$\begin{cases} \Psi : \mathbb{R}^3 \otimes \mathbb{R} \to \mathbb{R} \\ \hat{\Psi} : \mathbb{R}^3 \otimes \mathbb{R} \to \mathbb{C} \\ \Psi \xrightarrow{\text{F.T.}} \hat{\Psi} \\ \Psi \xleftarrow{\text{Inverse F.T.}} \hat{\Psi} \end{cases} \tag{1.9}$$

1.2.2 Frequency domain of permittivity and fields

The displacement field $\mathbf{D}(\mathbf{r}, t)$ can be decomposed (called *Fourier Decomposition*, it's actually an Inverse F.T. from *frequency domain*) using Equation (1.8),

$$\mathbf{D}(\mathbf{r}, t) = \frac{1}{2\pi} \int \mathbf{D}(\mathbf{r}, \omega) e^{-i\omega t} d\omega. \tag{1.10}$$

doi:10.6342/NTU202400923

Define the **constitutive equation** in **frequency domain**

$$D^i(\mathbf{r},\omega) = \sum_j \epsilon^i_j(\mathbf{r},\omega)E^j(\mathbf{r},\omega) \doteq \epsilon^i_j(\mathbf{r},\omega)E^j(\mathbf{r},\omega), \tag{1.11}$$

where $\omega = 2\pi f$ can now represent the *angular frequency* of electromagnetic waves (EMW), where f is the usual *frequency* we use in the unit of Hz (1/s). Note that ω is also called the *frequency* in much literature depending on the context, but in this thesis I call f the *frequency* and ω the *angular frequency*. $i = 1, 2, 3$ and $j = 1, 2, 3$. E^j is the j-th component of electric field **E** while D^i is the i-th component of displacement field **D**. The repeated index j in the final representation means sum over, which is the Einstein summation convention. ϵ^i_j is a second rank tensor called permittivity. It is a thermodynamic state function (i.e., it depends only on current equilibrium thermodynamic state but not on it's path to the current state, and therefore can be described as function of thermodynamic variables such as temperature and pressure) and can also depend on the magnitude, direction, and frequency of the applied field.

Note that Equation (1.11) is is an ω-space relation. Therefore, in general[5],

$$\mathbf{D}(\mathbf{r},t) \neq \epsilon(\mathbf{r},t)\mathbf{E}(\mathbf{r},t) \tag{1.12}$$

The SI unit of permittivity is farad per meter (F/m).

[5]Note that under some conditions the equality may hold. See course handout of K.R. Chu, in Classical Electrodynamics Ch.7.

doi:10.6342/NTU202400923

1.2.3 Nomenclature of the properties of permittivity of a medium

1.2.3.1 Isotropic versus anisotropic

If the permittivity of a medium is independent of the the direction of the electric field **E**, the medium is said to be isotropic, otherwise it is said to be anisotropic. The permittivity of an isotropic medium in frequency domain reduces to a scalar ϵ that can be a complex number ($\epsilon \in \mathbb{C}$).

1.2.3.2 Linear versus nonlinear

If ϵ depends on the field strength $|\mathbf{E}|$ of the external electric field, it is said to be nonlinear, otherwise it is said to be linear.

1.2.3.3 Homogeneous versus inhomogeneous

If ϵ is a constant throughout the material, the material is said to be homogeneous, otherwise the material is said to be inhomogeneous.

1.2.3.4 Dispersive versus nondispersive

If ϵ depends on the frequency f of the external electric field, the material is said to be dispersive, otherwise it is said to be non-dispersive. It can be shown that non-dispersive property leads to violation of causal relations.

The terms describing the nature of the permittivity of a material are summarized in Table 1.2.

doi:10.6342/NTU202400923

$\epsilon(\omega)$ is independent of	$\mathbf{r}$	$\mathbf{E}(\mathbf{r},\omega)/\lvert\mathbf{E}(\mathbf{r},\omega)\rvert$	$\lvert\mathbf{E}(\mathbf{r},\omega)\rvert$	f
Yes	homogeneous	isotropic	linear	nondispersive
No	inhomogeneous	anisotropic	nonlinear	dispersive

Table 1.2: Terms describing the nature of the permittivity of a material.

1.2.3.5 Complex permittivity (loss or gain in the medium)

For an isotropic homogeneous material, We can further define the quantities $\epsilon^{'} \equiv \mathrm{Re}(\epsilon)$ and $\epsilon^{''} \equiv \pm\mathrm{Im}(\epsilon)$ for harmonic time-dependence of field $\exp(\mp i\omega t)$, or equivalently:

$$\epsilon = \epsilon^{'} \pm i\epsilon^{''} \text{ for field dependece } \exp(\mp i\omega t). \tag{1.13}$$

In this thesis we assume $\exp(-i\omega t)$ field dependence, and therefore,

$$\epsilon = \epsilon^{'} + i\epsilon^{''}, \tag{1.14}$$

where $i \equiv \sqrt{-1}$; $\epsilon^{'}$ and $\epsilon^{''}$ are the real and imaginary part of ϵ , respectively. Using this definition, if $\epsilon^{''} > 0$ in the medium, the electromagnetic wave (EMW) passing the medium will loss energy after passing the medium. If $\epsilon^{''} < 0$ the EMW will gain energy.

1.2.3.6 Relative permittivity

The relative permittivity ϵ_r is defined as,

$$\epsilon_r \equiv \frac{\epsilon}{\epsilon_0}, \tag{1.15}$$

where ϵ is the complex permittivity and ϵ_0 is the permittivity of vacuum.

doi:10.6342/NTU202400923

1.2.3.7 Loss tangent

It is also a common practice to define a dimensionless quantity called electric loss tangent $\tan(\delta)$ to represent the dielectric loss:

$$\tan(\delta) = \frac{\epsilon^{''}}{\epsilon^{'}} \tag{1.16}$$

note that δ is often called the loss angle. The loss tangent is an ubiquitous way to parameterized the inherent dissipation of electromagnetic energy.

1.3 Single dielectric sphere in vacuum

Now consider a dielectric sphere of an isotropic homogeneous linear medium immersed in an uniform static electric field $\boldsymbol{E} = E_{ext}\hat{\mathbf{x}}$ and vacuum space, the polarization charge is only on the surface of the sphere and is prescribed in [2, Equation (4.58)].

$$\sigma_{pol} = 3\epsilon_0 \left(\frac{\epsilon/\epsilon_0 - 1}{\epsilon/\epsilon_0 + 2} \right) E_{ext} \cos\theta \tag{1.17}$$

Where σ_{pol} is the polarization charge density on the surface of the sphere, θ is the polar angle. ϵ_0 is the permittivity in vacuum (free space). Figure 1.1 (taken from [1, Fig. 1(a)]) demonstrates the polarization charge and field lines of a single dielectric orb of isotropic medium immersed in an uniform electric field $\boldsymbol{E} = E_{ext}\hat{\mathbf{x}}$ and vacuum. Figure 1.2 shows the field strength distribution (taken from [1, Fig. 1(b)].)

doi:10.6342/NTU202400923

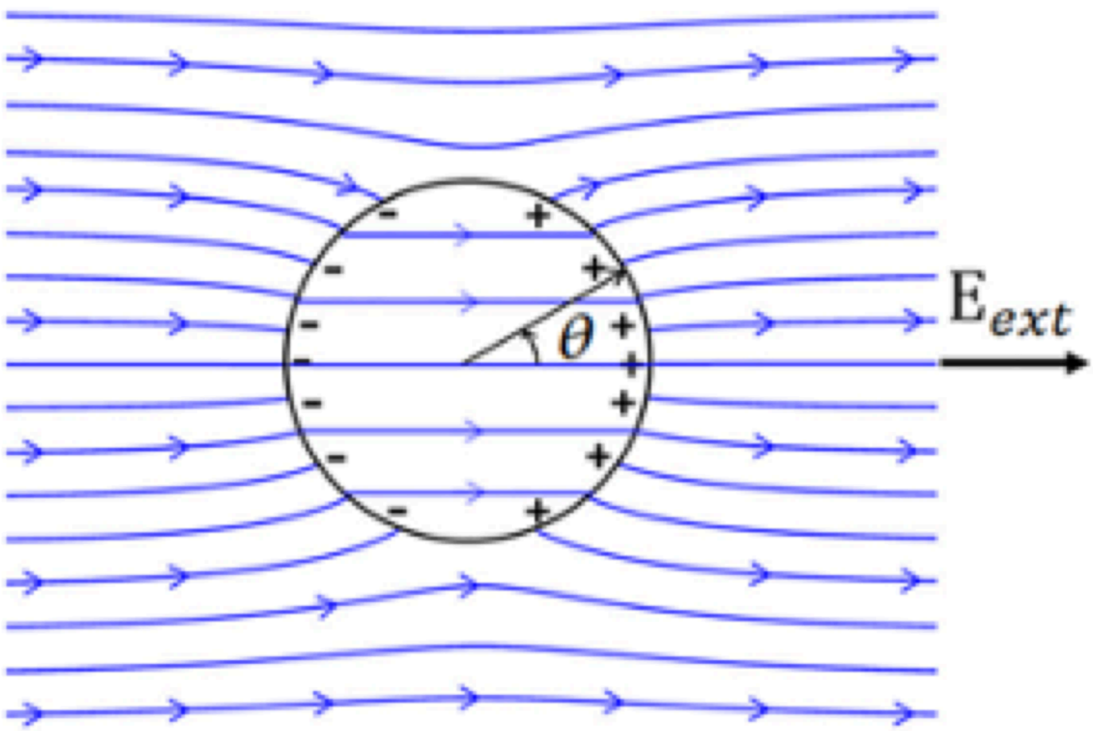


Figure 1.1: Polarization charge and field lines of a sphere immersed in uniform electric field, taken from Figure 1(a) of M. S. Lin, L. C. Liu, L. R. Barnett, Y. F. Tsai, and K. R. Chu. On electromagnetic wave ignited sparks in aqueous dimers. Physics of Plasmas, 28(10):102102, 2021. (https://doi.org/10.1063/5.0062014), licensed under a Creative Commons Attribution (CC BY 4.0) license (http://creativecommons.org/licenses/by/4.0/).

doi:10.6342/NTU202400923

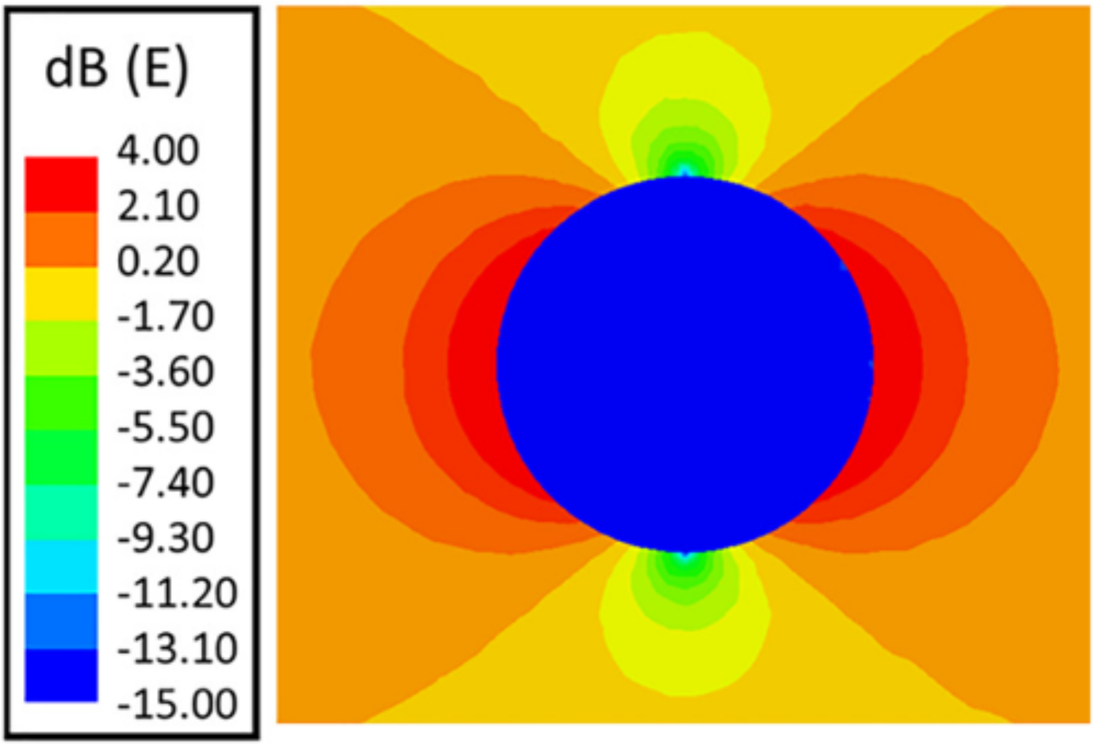


Figure 1.2: Polarization field strength of a sphere immersed in uniform electric field. dB(E)=$\log_{10}(|\mathbf{E}|/E_{ext})$, taken from Figure 1(b) of M. S. Lin, L. C. Liu, L. R. Barnett, Y. F. Tsai, and K. R. Chu. On electromagnetic wave ignited sparks in aqueous dimers. Physics of Plasmas, 28(10):102102, 2021. (https://doi.org/10.1063/5.0062014), licensed under a Creative Commons Attribution (CC BY 4.0) license (http://creativecommons.org/licenses/by/4.0/).

doi:10.6342/NTU202400923

Chapter 2 Dielectric Dispersion and Relaxation

2.1 Dielectric dispersion

Dielectric dispersion is the dependence of the permittivity of a dielectric medium on the frequency of the applied field. We can therefore write:

$$\epsilon = \epsilon(\omega) \tag{2.1}$$

where ω is the angular frequency of the applied electric field. By definition $\omega \equiv 2\pi f$. Oscillation

2.2 Dielectric relaxation

When exposed to a time-varying electric field, the permittivity of a dielectric material will vary with the electric field but with a time delay (usually called relaxation time, denoted by τ). This is usually the result of delayed molecular response. Usually, the delayed permittivity response can be described as a function of frequency of applied electric field. In an ideal system, such response can be described by Debye function [Equation 2.2].

doi:10.6342/NTU202400923

2.2.1 Debye function

In an ideal (linear, isotropic, homogeneous) system, the permittivity ϵ and relaxation time τ of certain external electric field of angular frequency ω can be described by the following function [3, 4],

$$\epsilon_r(\omega) = \frac{\epsilon(\omega)}{\epsilon_0} = \epsilon_r(\omega = \infty) + \frac{\epsilon_r(\omega = 0) - \epsilon_r(\omega = \infty)}{1 - i\omega\tau}, \tag{2.2}$$

where $\epsilon_r(\omega = \infty)$ is the relative permittivity at infinite frequency (optical regime) and $\epsilon_r(\omega = 0)$ is the electro-static relative permittivity. Note that due to different notation, the complex conjugate of my ϵ_r is the ϵ in [4, Equation (4)]. If we rewrite the equation above in form of Equation (1.14), we get[1],

$$\frac{\epsilon^{'}(\omega)}{\epsilon_0} \equiv \text{Re}[\epsilon_r(\omega)] = \epsilon_r(\omega = \infty) + \frac{\epsilon_r(\omega = 0) - \epsilon_r(\omega = \infty)}{1 + \omega^2\tau^2}, \tag{2.3}$$

$$\frac{\epsilon^{''}(\omega)}{\epsilon_0} \equiv \text{Im}[\epsilon_r(\omega)] = \frac{[\epsilon_r(\omega = 0) - \epsilon_r(\omega = \infty)]\omega\tau}{1 + \omega^2\tau^2}. \tag{2.4}$$

2.3 Dielectric constant of water at atmospheric pressure and different temperatures.

Dielectric constant of water is widely studied [4–7] in the past few decades. It is also interesting that the dielectric relaxation of water in terahertz (THz) domain domain which had been investigated using terahertz reflection spectroscopy and molecular dynamics (MD) simulation [7] seem to have an additional relaxation time apart from simple Debye model. The MD simulation is also used to calculate the dielectric constant of water

[1] An interesting question may be raised here: Is Debye Model consistent with Kramers-Kronig relation?

doi:10.6342/NTU202400923

under constant **D** [8]. The temperature and pressure dependence of water had also been investigated through MD simulations [9]. The general dependency of complex permittivity and relaxation time of water at frequency range of 1.1 gigahertz (GHz) to 57 GHz and temperature between $0\,°\mathrm{C}$ and $50\,°\mathrm{C}$ had been proposed [4]; they are summarized below [Equation (2.5-2.8)]:

$$\epsilon_r(\omega, T) = \epsilon_r(\omega = \infty, T) + \frac{\epsilon_r(\omega = 0, T) - \epsilon_r(\omega = \infty, T)}{1 - i\omega\tau(T)} \tag{2.5}$$

$$\epsilon_r(\omega = 0, T) = 10^{1.94404 - 1.991\times 10^{-3}\mathrm{K}^{-1}(T - 273.15\mathrm{K})} \tag{2.6}$$

$$\epsilon_r(\omega = \infty, T) = 5.77 - 2.74 \times 10^{-2}\mathrm{K}^{-1}(T - 273.15\mathrm{K}) \tag{2.7}$$

$$\tau(T) = (3.745 \times 10^{-15}\mathrm{s})[1 + (7 \times 10^{-5}\mathrm{K}^{-2}) \times (T - 300.65\mathrm{K})^2] \exp\left(\frac{2.2957 \times 10^3\mathrm{K}}{T}\right) \tag{2.8}$$

where $\epsilon_r(\omega, T)$ and $\tau(T)$ resemble $\epsilon_r(\omega)$ and τ in Equation (2.2), but add temperature T (in units of Kelvins) into consideration.

Compare with [10, Table 1.1], which lists the complex permittivity of water at $20\,°\mathrm{C}$ at different external electric field frequency and is taken from [11], the absolute values of deviations of electric loss tangent ($|[\tan(\delta_1) - \tan(\delta_2)]/\tan(\delta_2)|$) are within 6.60% (0.271% - 6.60%).

doi:10.6342/NTU202400923

2.4 An example of the quasi-static limit in electrodynamics: Rayleigh Scatter

2.4.1 An elastic scatter model

The wavelength of the electromagnetic wave (EMW) of frequency f in the non-magnetic (permeability $\mu = \mu_0$) medium whose permittivity is ϵ is defined as $\lambda_d(f, \epsilon)$. The real part of complex index of refraction n of a non-magnetic medium can be derived from [2, Equation (7.5)]

$$\text{Re}(n) = n_d = \sqrt{\frac{|\epsilon_r| + \text{Re}(\epsilon_r)}{2}}, \tag{2.9}$$

and therefore,

$$\lambda_d = \lambda_0 / n_d, \tag{2.10}$$

where λ_0 is the free-space wavelength given by $\lambda_0 = c/f$. Consider a dielectric sphere of radius r and permittivity ϵ is placed in an open space (which may be air or vacuum, I assume the permittivity and permeability of the open space outside the dielectric sphere are ϵ_0 and μ_0, respectively), and we hit this sphere by an incident plane wave whose electric field is $\mathbf{E}_{inc}(\mathbf{r}, t)$, linearly polarized in x-direction and traveling in positive z-direction ($\mathbf{k} = \hat{z}$), assuming **monochromatic harmonic time dependence** $e^{-i\omega t}$ of field and therefore the system is in **steady-state** and the scattering process involved is **elastic scatter** (the frequency of the scatter field is the same as the incident field), we may write the total electric field $\mathbf{E}(\mathbf{r}, t)$ as the sum of $\mathbf{E}_{inc}(\mathbf{r}, t)$ and the scatter electric field $\mathbf{E}_{sc}(\mathbf{r}, t)$,

$$\mathbf{E} = \mathbf{E}_{inc} + \mathbf{E}_{sc} \tag{2.11}$$

doi:10.6342/NTU202400923

Solving Maxwell equations mentioned in section 3.2 with scattering boundary condition, we can get the spatial configuration of the electric field (For detailed analytical solutions, please see [10]). For simplicity, I put the center of the sphere on (0,0,0).

2.4.2 Elastic scatter of a water sphere hit by 2.45 GHz microwave: quasi-static case

Following subsection 2.4.1, suppose $\epsilon_r(f = 2.45 \text{ GHz}, T = 298.15\text{K}) = 77.4 + 9.48i$, and $r = 1.000$ mm $\ll \lambda_d \approx 13.88$ mm. The simulation[2] result (See Figure 2.1) of normalized time-averaged electric field strength

$$\langle|\mathbf{E}(x, y, z)|\rangle \equiv \frac{\int_{t=0}^{1/f} |\mathbf{E}(\mathbf{r}, t)|dt}{\int_{t=0}^{1/f} |\mathbf{E}_{inc}(\mathbf{r}, t)|dt} \tag{2.12}$$

configuration of the field near the sphere looks just like the field configuration when the sphere is immersed in the static electric field (See [Figure 1.1 and Figure 1.2]). This phenomena is called the **quasi-static limit** [12, p.298]. The $\lambda_d \gg r$ elastic scatter theory is called **Rayleigh scatter** in literature, and for $\lambda_d \approx r$ it's called **Mie scatter**; please see the next chapter for further detail.

[2] Simulation was done using Ansys HFSS software, and the figure was drawn using Python packages such as Matplotlib.

doi:10.6342/NTU202400923

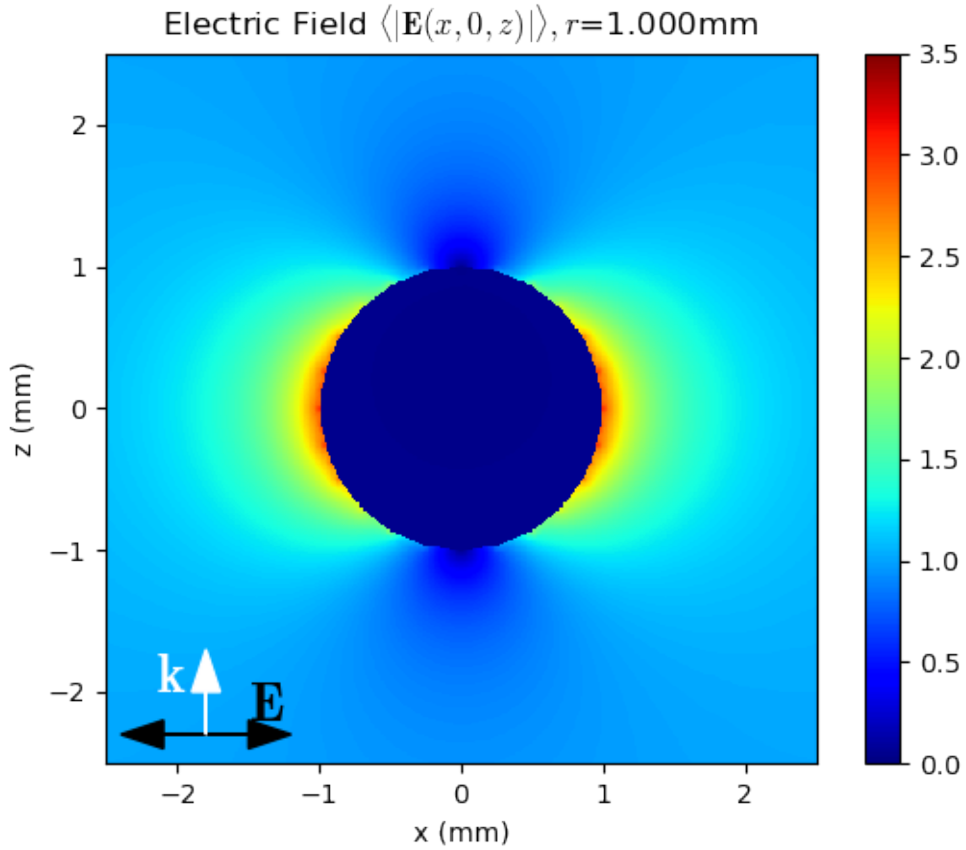


Figure 2.1: $\langle|\mathbf{E}(x,0,z)|\rangle$ of a linear isotropic homogeneous ($\epsilon_r = 77.4 + 9.48i$) water sphere of radus $r = 1.000$ mm hit by 2.45 GHz plane wave whose $\mathbf{k} = \hat{z}$ and linearly polarized in $\hat{x}$ direction.

doi:10.6342/NTU202400923

Chapter 3 Resonances

3.1 Theory

Mie resonances are a type of scatters that the scattering objects are in the same scale as the length of the incident wave, while the scattering objects can be seen as cavities (therefore resonance happens within it). Danish physicist Ludvig Lorenz[1] actually did the equivalent calculations before Gustav Mie [13, p.789], and therefore Mie scatter theory is also called Lorenz-Mie theory. Nevertheless, Mie scatter theory is still the most popular name. The illuminated objects can be in the shapes that support resonance to happen, such as spheres, cylinders and ellipsoids [14]. It is also called Morphology Dpendent Resonances (MDRs). MDRs address the near-field effects of resonant interactions of electromagnetic waves with wavelength-scale objects [15–17]. The objects can be conductive or dielectric and absorptive or transparent, depending on the complex dielectric permittivity of the material [15].

[1] His most famous work may be the Lorenz Gauge $\boldsymbol{\nabla} \cdot \mathbf{A} + \frac{1}{c^2}\frac{\partial V}{\partial t} = 0$ in electrodynamics; where $\mathbf{B} = \boldsymbol{\nabla} \times \mathbf{A}$ and $\mathbf{E} = -\nabla V - \frac{\partial A}{\partial t}$.

doi:10.6342/NTU202400923

3.2 Formulation

A group of widely recognized equations of electromagnetic fields are called Maxwell's equations. Formulation in free space, SI units convention and differential form is:

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\epsilon_0} \tag{3.1}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{3.2}$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \tag{3.3}$$

$$\nabla \times \mathbf{B} = \mu_0 \mathbf{J} + \mu_0 \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} \tag{3.4}$$

where **E** and **B** are the electric and magnetic field, respectively. μ_0 is called the permeability of free space. ρ and **J** are charge and current density, respectively, and subject to conservation of charge [Equation (3.5)].

$$\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} = 0 \tag{3.5}$$

Equation (3.1-3.4) are called microscopic Maxwell's equations, for their elucidation of electromagnetic field (EM field) in terms of current and charge densities presented(which can be very small), as well as the interaction between electric and magnetic field.

In mediums, there are charge distributions which are too sophisticated to model. Nuclei and surrounding electrons distributed through out the medium require large amount of calculations if we do it using *ab initio* method without simplification. However, for a highly homogeneous medium, we can use macroscopic Maxwell equations to model the

doi:10.6342/NTU202400923

macroscopic electromagnetic field within such system.

3.2.1 Macroscopic Maxwell equations

The four macroscopic Maxwell equations are:

$$\nabla \cdot \mathbf{D} = \rho_f \tag{3.6}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{3.7}$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \tag{3.8}$$

$$\nabla \times \mathbf{H} = \mathbf{J}_f + \frac{\partial \mathbf{D}}{\partial t} \tag{3.9}$$

where **D** field is introduced in Chapter 1, and **H** is simply the magnetic counterpart of **E** (**E** and **H** are not continuous amid surfaces of mediums with different ϵ and μ). $\mathbf{J}_f$ is the current density formed by free charges.

3.2.2 Wave equations

Consider in free space without charge distributions, if we take the curl of each curl equation of microscopic Maxwell equations, and substitute another curl equation into it, we will have two wave equations of electromagnetic field, and they are:

$$(\nabla^2 - \mu_0\epsilon_0\frac{\partial^2}{\partial t^2})\mathbf{E} = \Box\mathbf{E} = 0 \tag{3.10}$$

$$(\nabla^2 - \mu_0\epsilon_0\frac{\partial^2}{\partial t^2})\mathbf{B} = \Box\mathbf{B} = 0 \tag{3.11}$$

doi:10.6342/NTU202400923

where ∇^2 and $\Box$ are Laplace and d'Alembert operator, respectively. This shows the wave-like nature of electromagnetic fields.

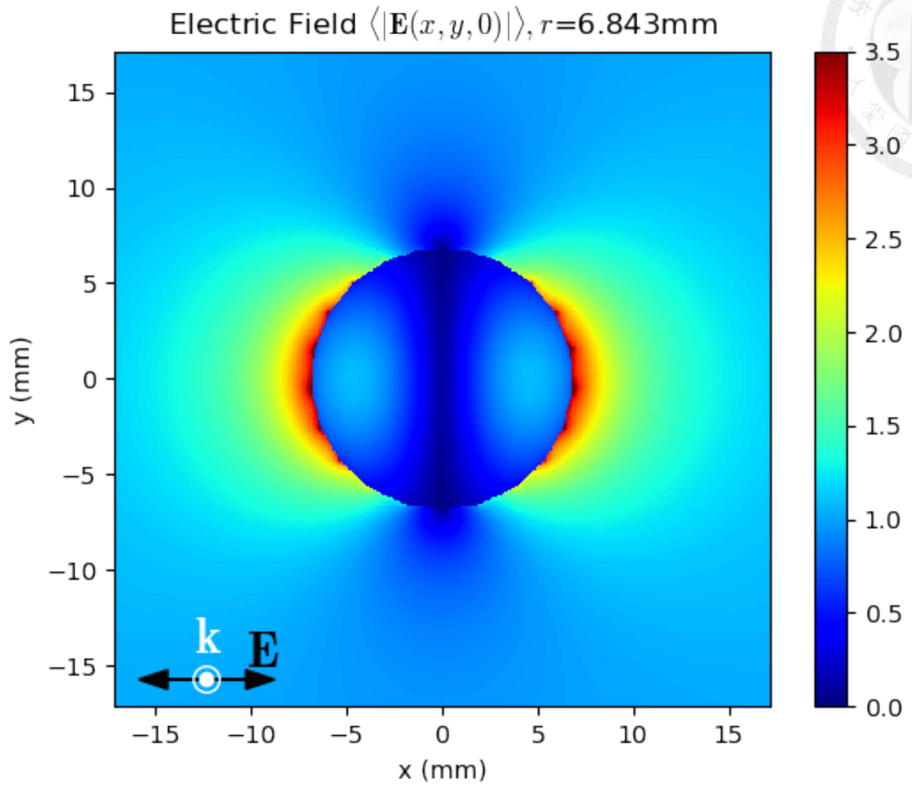
3.3 Mie resonance in a single water sphere

Let's consider a water sphere in air. According to [10], the first and second resonance of 2.45 GHz electromagnetic wave happen when the radii of the lossy water ($\epsilon_r = 77.4 + 9.48i$) sphere are 6.843 mm (this is close to $(\lambda_d/2)$ ≈6.94 mm) and 9.796 mm, respectively. Using Ansys High Frequency Structure Simulator (HFSS), which utilizes finite-element method (FEM), let's see spatial configuration of electric field intensity $\langle|\mathbf{E}(x, y, z)|\rangle$ [see Equation (2.12)] of radii (a) 6.843 mm and (b) 9.796 mm in three orthogonal cross-sections in Figure 3.1 (x-y cross-section), Figure 3.2 (x-z cross-section) and Figure 3.3 (y-z cross-section). These figures are drawn using Python packages. Note that these simulations are done in a single frequency, and therefore monochromatic fields (in other words, *elastic scatter*) at 2.45 GHz is assumed.

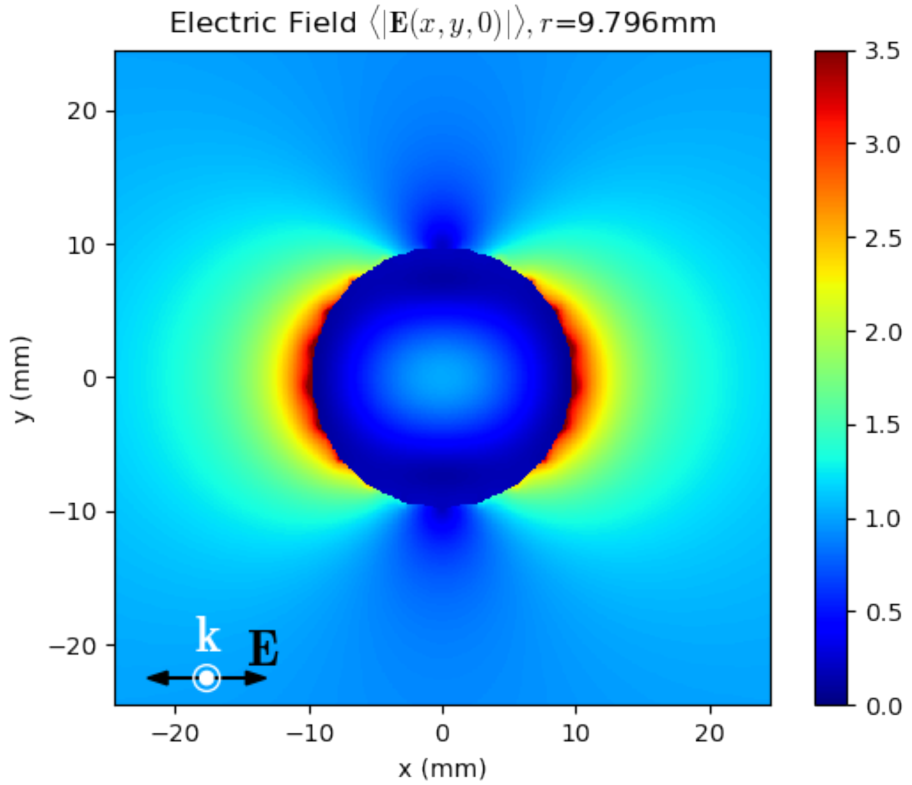
3.3.1 Analytical solutions

A detailed derivation of the analytical solutions of electromagnetic fields of the elastic scatter model of a dielectric sphere (as mentioned in subsection 2.4.1) and a dielectric cylinder was done by K.W. Chen in his master thesis [10].

doi:10.6342/NTU202400923



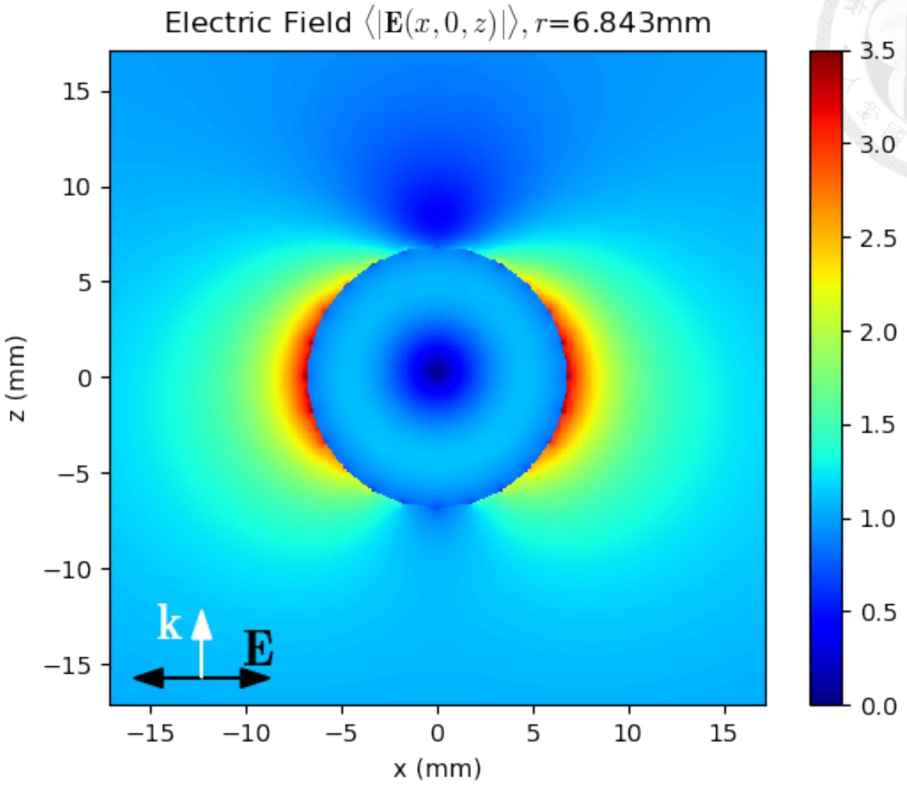
(a) $r = 6.843$ mm



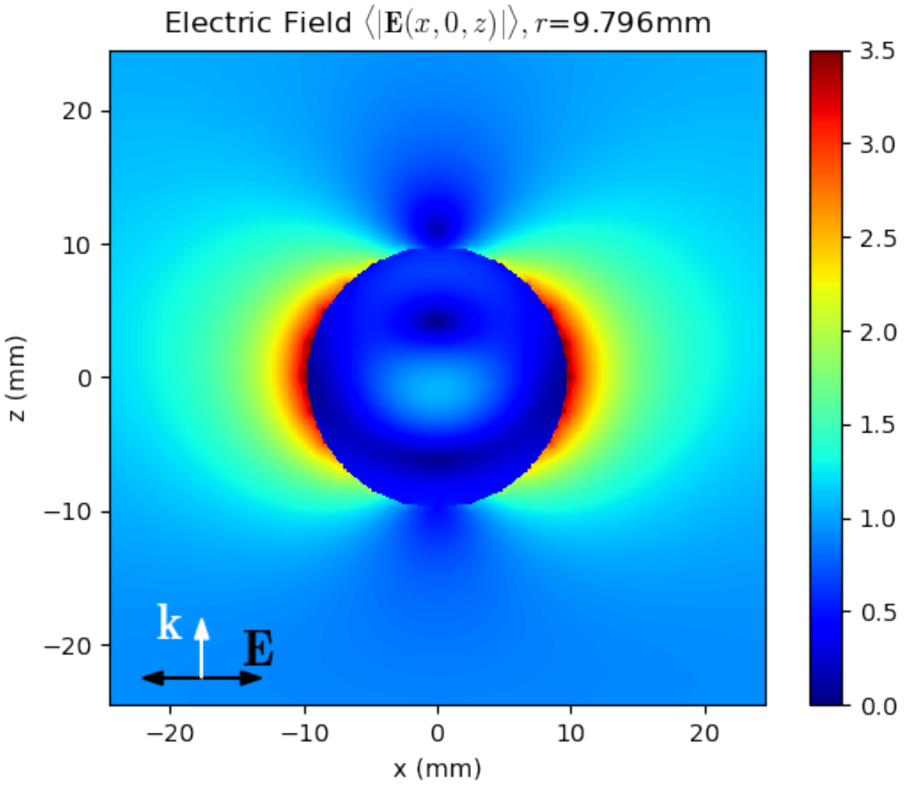
(b) $r = 9.796$ mm

Figure 3.1: $\langle|\mathbf{E}(x, y, 0)|\rangle$ of linear isotropic homogeneous ($\epsilon_r = 77.4 + 9.48i$) water spheres of radii (a) $r = 6.843$ mm and (b) $r = 9.796$ mm hit by 2.45 GHz plane wave whose $\mathbf{k} = \hat{z}$ and linearly polarized in $\hat{x}$ direction.

doi:10.6342/NTU202400923



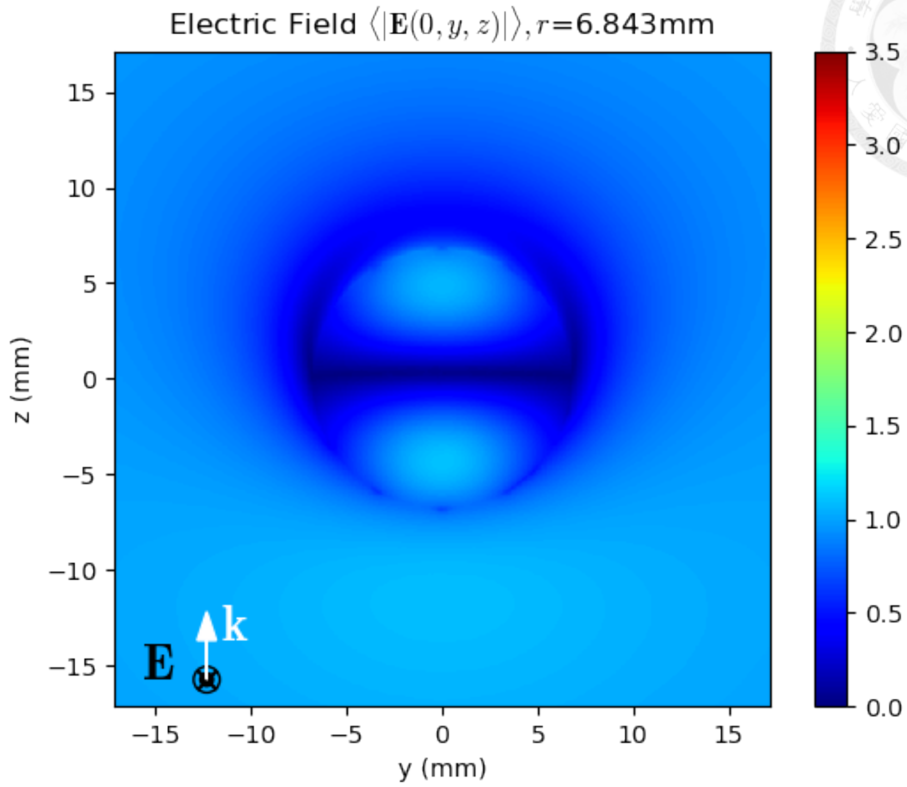
(a) $r = 6.843$ mm



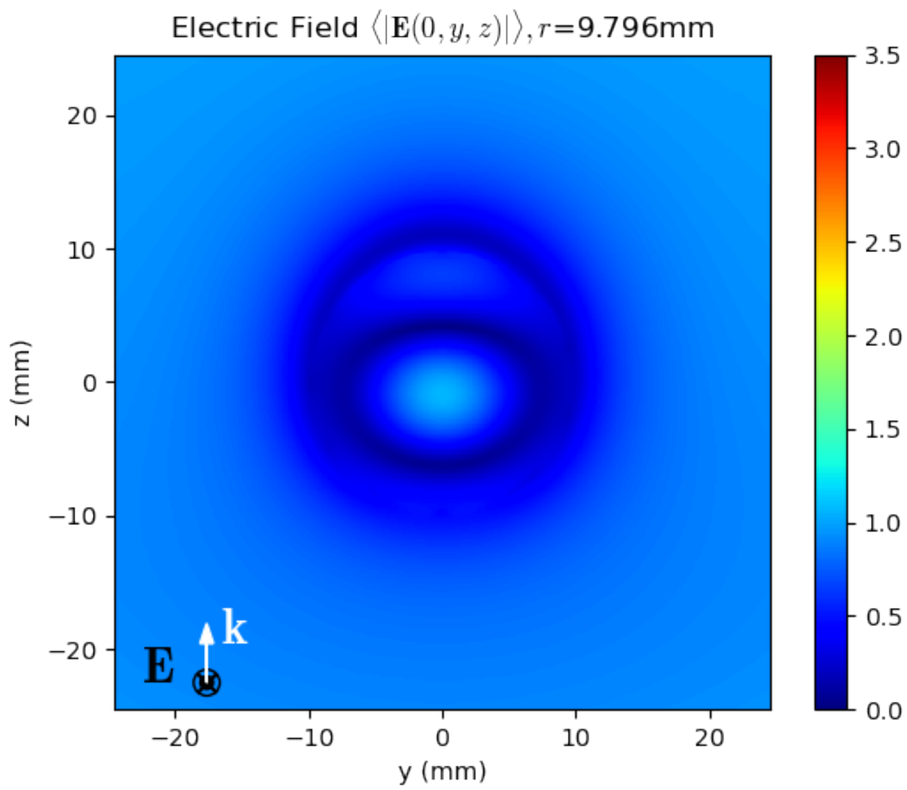
(b) $r = 9.796$ mm

Figure 3.2: $\langle|\mathbf{E}(x,0,z)|\rangle$ of linear isotropic homogeneous ($\epsilon_r = 77.4 + 9.48i$) water spheres of radii (a) $r = 6.843$ mm and (b) $r = 9.796$ mm hit by 2.45 GHz plane wave whose $\mathbf{k} = \hat{z}$ and linearly polarized in $\hat{x}$ direction.

doi:10.6342/NTU202400923



(a) $r = 6.843$ mm



(b) $r = 9.796$ mm

Figure 3.3: $\langle|\mathbf{E}(0,y,z)|\rangle$ of linear isotropic homogeneous ($\epsilon_r = 77.4 + 9.48i$) water spheres of radii (a) $r = 6.843$ mm and (b) $r = 9.796$ mm hit by 2.45 GHz plane wave whose $\mathbf{k} = \hat{z}$ and linearly polarized in $\hat{x}$ direction.

doi:10.6342/NTU202400923

3.4 Application

MDRs are widely studied and exploited in literature [15, 17, 18]. Bakker, Reuben M., *et al.* demonstrated both experimentally and theoretically that the enhancement of localized electric and magnetic fields can be achieved in a silicon nanodimer utilizing theory of MDRs [18]. Kuwata, Hitoshi, *et al.* proposed a simple analytical formula that can quantitatively predict resonant light scattering from metal nanoparticles of arbitrary shape, whose sizes are too large for Rayleigh approximation to be applicable [17]. Khattak, Hamza K., Pablo Bianucci, and Aaron D. Slepkov linked plasma formation in grapes to microwave resonances of aqueous dimers [15].

doi:10.6342/NTU202400923

Chapter 4 Dielectric Heating

4.1 Theory

We begin by considering the conservation of energy, and proceed to some particular results of dielectric heating.

4.1.1 Poynting's theorem (1884 CE)

Poynting's theorem is a theory that considers the conservation of energy, and is well documented [2, Section 6.7]. Specifically, Poynting's theorem, in time domain, assuming Newtonian equation of motion and Lorentz force [See Appendix A], is given by [2, Equation (6.105)],

$$\int_V \mathbf{J} \cdot \mathbf{E} \, \mathrm{d}^3 x = -\int_V \left[\boldsymbol{\nabla} \cdot (\mathbf{E} \times \mathbf{H}) + \mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} + \mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} \right] \mathrm{d}^3 x \,. \tag{4.1}$$

If we further assume:

1. The medium is linear, nondispersive and lossless.

doi:10.6342/NTU202400923

2. The total electromagnetic energy stored in the field can be represented by

$$u = \frac{\mathbf{E} \cdot \mathbf{D} + \mathbf{B} \cdot \mathbf{H}}{2}. \tag{4.2}$$

Choose V to be an infinitesimal volume element, we will have [2, Equation (6.108)],

$$\frac{\partial u}{\partial t} + \boldsymbol{\nabla} \cdot \mathbf{S} = -\mathbf{J} \cdot \mathbf{E} \tag{4.3}$$

where u is the energy stored in EM field (EM field energy), the $\mathbf{J} \cdot \mathbf{E}$ on the right hand side represents the conversion of electromagnetic energy into mechanical or heat energy. $\mathbf{S}$ is the Poynting vector representing the energy flow, and can be written as,

$$\mathbf{S} = \mathbf{E} \times \mathbf{H}. \tag{4.4}$$

The $\boldsymbol{\nabla} \cdot \mathbf{S}$ term describes the net energy transport out of certain point through EM field, and will be negative if there's net energy transport in certain point through EM field. $\partial u / \partial t$ is the accumulation rate of EM field energy, and will be negative if the EM field energy is decreasing. $\mathbf{J} \cdot \mathbf{E}$ is the rate of conversion of electromagnetic energy into mechanical or heat energy.

4.1.2 Poynting's theorem in linear dispersive medium with losses

Poynting's theorem in linear dispersive medium with losses can be quite complicated. It was recorded in [2, Equation (6.127)],

$$\frac{\partial u_{\text{eff}}}{\partial t} + \boldsymbol{\nabla} \cdot \mathbf{S} = -\mathbf{J} \cdot \mathbf{E} - \omega_0 \epsilon''(\omega_0) \langle \mathbf{E} \cdot \mathbf{E} \rangle - \omega_0 \mu''(\omega_0) \langle \mathbf{H} \cdot \mathbf{H} \rangle \tag{4.5}$$

doi:10.6342/NTU202400923

where ω_0 is the carrier frequency and $\mu''(\omega_0)$ is the imaginary part of the permeability of the medium at frequency ω_0. The effective potential u_{eff} is given by [2, Equation (6.126b)]

$$u_{\text{eff}} = \frac{1}{2}\left\{\text{Re}\left[\frac{\text{d}(\omega\epsilon)}{\text{d}\omega}(\omega_0)\right]\langle\mathbf{E}\cdot\mathbf{E}\rangle + \text{Re}\left[\frac{\text{d}(\omega\mu)}{\text{d}\omega}(\omega_0)\right]\langle\mathbf{H}\cdot\mathbf{H}\rangle\right\}. \tag{4.6}$$

Note that $\langle..\rangle$ means the time-averaged value.

The ohmic losses $\mathbf{J}\cdot\mathbf{E}$ and the heating of the medium (if $\epsilon'' \neq 0$ and/or $\mu'' \neq 0$) in realistic situation are now taking into consideration of local conservation of energy by Equation (4.5).

4.2 Dielectric heating in an uniform AC electric field

Lin, M. S., Lin, S. M., Chiang, W. Y., Barnett, L. R., and Chu, K. R. had investigated effects of polarization-charge shielding in microwave heating of spherical and cubical object [19].

Consider a dielectric medium whose permittivity is in the form of Equation (1.14). The medium is now immersed in an uniform AC electric field given by:

$$\mathbf{E} = \text{Re}[\mathbf{E}_c(\mathbf{r},\omega)\exp(-i\omega t)], \tag{4.7}$$

where $\mathbf{E}_c(\mathbf{r},\omega)$ is a constant electric field (i.e., $\mathbf{E_c}$ is a constant vector across all position), the electrical polarization density $\mathbf{P}$ within the medium can now be given by (Re on the right hand side of equation is omitted here in the following equation, as in [19]),

doi:10.6342/NTU202400923

$$\mathbf{P} = (\epsilon - \epsilon_0)\mathbf{E}_c \exp(-i\omega t). \tag{4.8}$$

The polarization current density $\mathbf{J}_p$ can now be given by:

$$\mathbf{J}_p \equiv \frac{\partial \mathbf{P}}{\partial t} = -i\omega(\epsilon - \epsilon_0)\mathbf{E}_c \exp(-i\omega t) \tag{4.9}$$

The power of heating in a unit volume of medium, $P_{heating}$, is then [19, Equation (1)],

$$P_{heating} = \frac{1}{2}[\text{Re}(\mathbf{J}_p \cdot \mathbf{E}^*)] = \frac{1}{2}\epsilon''\omega|\mathbf{E}_c|^2. \tag{4.10}$$

Note that $\frac{1}{2}[\text{Re}(\mathbf{J}_p \cdot \mathbf{E}^*)]$ is consistent with the first term of Equation (5.4a), since they are the real part of two conjugated quantities ($\text{Re}(\mathbf{J}^* \cdot \mathbf{E}) = \text{Re}[(\mathbf{J}^* \cdot \mathbf{E})^*] = \text{Re}(\mathbf{J} \cdot \mathbf{E}^*)$).

doi:10.6342/NTU202400923

Chapter 5 Violation of Poynting's Theorem in Frequency Domain in Mie Theory and Probable Causes

Poynting's theorem in frequency domain is prescribed by [2, Equation (6.134)],

$$\frac{1}{2}\int_V \mathbf{J}^* \cdot \mathbf{E} d^3x + 2i\omega \int_V (w_e - w_m) d^3x + \oint_S \mathbf{S} \cdot \hat{\mathbf{n}} da = 0; \tag{5.1}$$

where the real part of the first term is the time-averaged heating rate and $w_e = \frac{1}{4}(\mathbf{E} \cdot \mathbf{D}^*)$, $w_m = \frac{1}{4}(\mathbf{B} \cdot \mathbf{H}^*)$, $\mathbf{S} = \frac{1}{2}\mathbf{E} \times \mathbf{H}^*$ is the complex Poynting vector. Applying divergence theorem on Equation (5.1), we will have,

$$\frac{1}{2}\int_V \mathbf{J}^* \cdot \mathbf{E} d^3x + 2i\omega \int_V (w_e - w_m) d^3x + \int_V \nabla \cdot \mathbf{S} d^3x = 0, \tag{5.2}$$

or simply let V be the infinitesimal volume element, and we will get,

$$\frac{1}{2}\mathbf{J}^* \cdot \mathbf{E} + 2i\omega (w_e - w_m) + \nabla \cdot \mathbf{S} = 0, \tag{5.3}$$

doi:10.6342/NTU202400923

or explicitly,

$$\text{Re}\left\{\frac{1}{2}\mathbf{J}^* \cdot \mathbf{E} + 2i\omega(w_e - w_m) + \boldsymbol{\nabla} \cdot \mathbf{S}\right\} = 0, \tag{5.4a}$$

and

$$\text{Im}\left\{\frac{1}{2}\mathbf{J}^* \cdot \mathbf{E} + 2i\omega(w_e - w_m) + \boldsymbol{\nabla} \cdot \mathbf{S}\right\} = 0. \tag{5.4b}$$

Equation (5.1) actually can be derived from Equation (4.1), assuming harmonic time dependence $e^{-i\omega t}$ of fields $\mathbf{E}(\mathbf{r}, t)$ and $\mathbf{H}(\mathbf{r}, t)$ and therefore steady-state (time-invariant) of electromagnetic properties μ and ϵ. However, I will show that Equation (5.4a) didn't hold in classical Mie scatter theory, assuming linear homogeneous isotropic dielectric properties μ and ϵ. Equation (5.4a) can therefore be a useful tool in diagnosing classical models in energy-conservation aspect.

5.1 Violation

Before continuing on the argument, I want to show you some evidence of the simulation results first. Let's see Figure 5.1 (which was done using COMSOL multiphysics software) and Figure 5.2 (which was done using Ansys HFSS software). We can see that Equation (5.1) indeed doesn't hold under assumptions of Mie scatter (amid different software, morphology, frequency and material).

doi:10.6342/NTU202400923

5.2 Evidence from other literature

It is also written in literature [20] explicitly that these assumptions in traditional Mie scattering problems are too strong.

5.3 Probable causes

5.3.1 Inelastic scatter

Note that Mie scatter is an elastic scatter mechanism, meaning that the energy E of the incoming and scatter photon doesn't change, and therefore the frequency $f = E/h$ doesn't change.[1] However, we do know that there exist some inelastic scatter mechanism such as Stokes and anti-Stokes Raman scatter. Therefore, from this aspect, violation in Equation (5.1) may make classical Mie theory better, because now we leave a room for inelastic scatterings in our theory. The violation of Eq. (5.1) actually allows net energy transport between different frequencies.

5.3.2 Non steady-state

Recall that we assume electromagnetic steady-state in Mie-scatter. However, we do know that a microwave of 2.45 GHz will heat up a water sphere in room temperature and atmospheric pressure (such as the working principle of commercial microwave ovens.) From Section 2.3 we know that the dielectric constant ϵ depends on temperature. This lead us to the violation of steady-state assumption.

[1] h is the Planck constant.

doi:10.6342/NTU202400923

5.3.3 Inhomogeneity

ϵ_r varies with temperature from Section 2.3. For heat to transfer from the inner part of the sphere to the outer part of the sphere, there should be temperature gradients. This violates the homogeneous assumption in classical derivation of Mie theory.

5.4 Short remarks

The deficiencies of monochromatic frequency-domain analysis in the energy-domain actually reminds us the underlying physics - our theories are just approximations of the real world. More precise theory may be built upon quantum field theory (*ab initio*) or molecular dynamics (MD) simulation, but this will exceed the current (2024 CE) computational ability of tabletop computers, since we are talking about approximately 10^{23} (a water sphere of radius 6.843 mm) of water molecules, unless some appropriate approximations are used. Nevertheless, Equation (5.4a) should still be a useful tool in diagnosing the energy-consistency in classical models.

doi:10.6342/NTU202400923

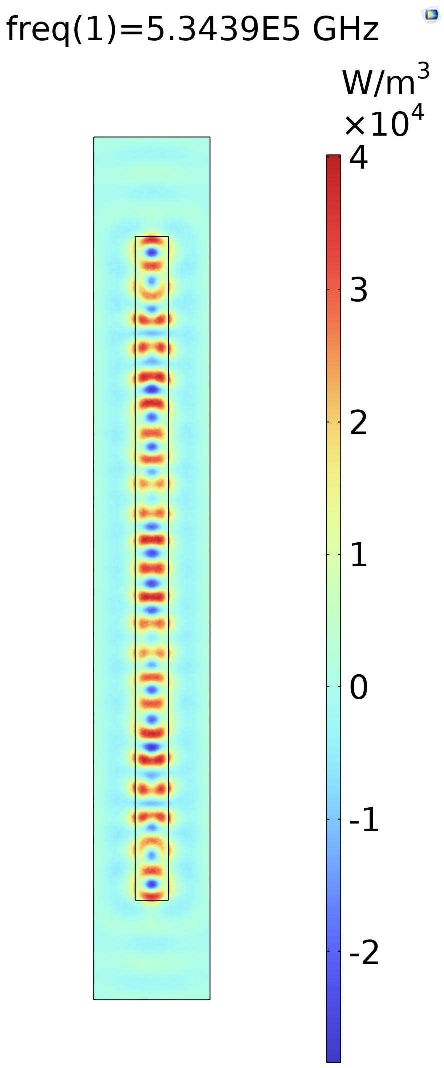


Figure 5.1: COMSOL simulation plot of Re $\left[\frac{1}{2}\mathbf{J}^* \cdot \mathbf{E} + 2i\omega(w_e - w_m) + \boldsymbol{\nabla} \cdot \mathbf{S}\right]$ on the central slice of a silicon nitride nano wire, immersed in air, whose long axis is along the $z-$ axis illuminated by a TE mode (linearly polarized in $y-$ direction) plane wave of electric field strength 1 $\mathrm{V/m}$ travelling in positive $x-$direction. The diameter of the silicon nitride nanowire is 500 nm and its length is 13 μm. The free-space wavelength of the incident plane wave is 561 nm. ϵ and μ are constant within wire. Courtesy of Yu-An Chen (陳俞安) (ORCiD: 0009-0009-4575-3114, E-mail: cyuan933gmail.com).

doi:10.6342/NTU202400923

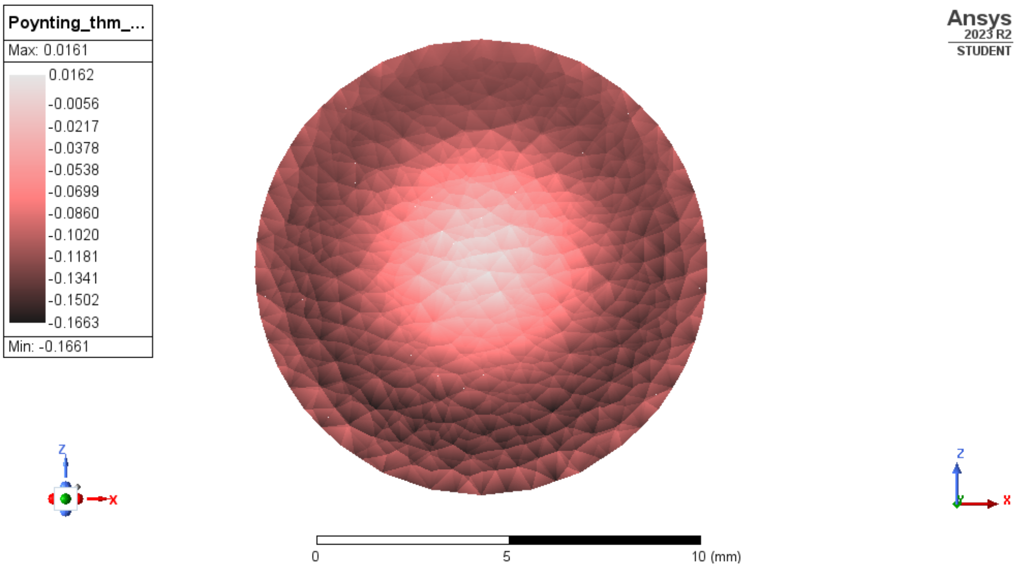


Figure 5.2: The central slice of $x - z$ plane of Re $\left[\frac{1}{2}\mathbf{J}^* \cdot \mathbf{E} + 2i\omega(w_e - w_m) + \boldsymbol{\nabla} \cdot \mathbf{S}\right]$ in W/m^3 of a water sphere of radius 6.0 mm and constant ϵ and μ illuminated by a linearly $\hat{x}$-polarized plane wave of frequency 2.45 GHz and peak intensity 1 V/m traveling in $\hat{z}$-direction in vacuum. The simulation is done by Chun-Yu Yang (楊鈞禹) (ORCiD: 0009-0006-1160-387X) using Ansys HFSS Student edition.

doi:10.6342/NTU202400923

Chapter 6 Two Theories Regarding the Etiology of Plasma Formation Between Aqueous Dimers

6.1 MDRs (2019 CE)

In 2019 CE, Hamza K. Khattak, Pablo Bianucci and Aaron D. Slepkov published their theories about the origin of plasma formation between spherical dimers [15]. They linked the phenomena to the cooperative interaction of MDRs in the individual spheres, and plenty of simulations and experiments were done. This research soon raised public attention after covered by some famous scientific journals as well as general media.

6.1.1 MDRs

They suggest that the enhancement of EM field is the result of combination of different MDRs patterns within the gap region of the dimer system. The simulations of EM field utilize finite element method (FEM). They observe several EM field hotspot in the

doi:10.6342/NTU202400923

gap and center part of the dimer system in the FEM simulations, and thus suggest that there may be interactions between MDRs of each sphere.

6.1.2 Thermal behavior of the system

As discussed in Chapter 4, the dielectric heating of the system can be modeled by Equation (4.5). This means that one can use thermal behavior of the system to infer the EM field strength. The authors take infrared (IR) images to show the thermal behavior of the system surface. They also do COMSOL simulations, and finally, they use thermal papers wrapped around the system to record the thermal behavior of the system. They find a subwavelength ($\lambda_0/80$) thermal hotspot in the gap axial region, and thus infer that there are cooperative interactions of MDRs of each sphere in the gap axial hotspot.

6.2 Polarization charges (2021 CE)

In 2021 CE, however, another group proposed another pourquoi story of plasma formation between aqueous dimers [1]. They think polarization introduced in Chapter 1 is the major cause, and I will discuss their results from now on.

6.2.1 Mutual enhancement of polarization between dimers

Quasi-static limits are mentioned in [12, p.298] and subsection 2.4.2. If the wavelength of the imposed EM wave is large compare to the size of the object, we can simplify our description of the system by mimicking electro-static conditions. Polarization is thus a valid model for the dimer system.

doi:10.6342/NTU202400923

The polarization of each sphere in the dimer system will enhance the polarization effect on each other, until they finally reach electro-static equilibrium.

To see whether polarization theory is correct, the authors performed the simulation and experiment at 27 MHz frequency. MDRs of the dimer system composed of two identical aqueous spheres of radius 7mm can not happen at this frequency. The result confirmed that polarization alone can trigger sparks between such system.

6.2.2 Forces between dimers

Recall that when disposing the real-time *in-situ* snapshots of aqueous dimer, the work [15] uses a concave instead of a horizontal plane to hold the dimer spheres together. This method, however, will make the dimer system subject to gravitational force, and thus we will be unable to see whether the initial external field causes attractive force or repulsive force between dimer spheres except when the resulting force is repulsive and strong enough to conquer the force exerted by gravitation and the concave. The experiments in [15] finally yield a oscillating result, but this should be the consequence of gravitational force and elastic Leidenfrost effect [21]. The experiment of [1], in contrast, is able to detect the force between the dimer spheres caused by external field. The attractive results strongly suggest the polarization nature of the dimer systems, since the force should be repulsive due to mutual radiation pressure if MDRs do dominate the system.

6.2.3 Electromagnetic fields between dimers

If the polarization theory is correct, because of the quasi-static nature of the system, it is expected to observe strong enhancement electric fields between dimers, while the

doi:10.6342/NTU202400923

magnetic field strength may be relatively unchanged. If the MDRs theory is correct, both electric and magnetic field strength may be enhanced simultaneously in scale. Comparison of [15] and [1] shows that the field profile suggests the polarization nature. The figures in [15] do not distinguish magnetic and electric field strength, while the enhancement of magnetic field strength is not in scale with the electric field in the simulations of [1].

6.2.4 Configuration that only MDRs occur

Interestingly, if we put the dimer system perpendicular to the external electric field, we will be able to keep MDRs, while there will be no mutual enhancement of polarization charge, as $\theta = \pm\frac{\pi}{2}$ in Equation (1.17) in the gap region. This enables us to test whether sparks occur in absence of mutual enhancement of polarization.

The results are shown in [1, (Figs. 3,7)]. As expected, no evidence of spark formation is found. The results suggest that MDRs are not the major cause of spark formation between dimer systems.

6.2.5 Evidence from other literatures

The polarization effect is well utilized in many areas such as electrostatic self-assembly of particles [22, 23], microwave assisted synthesis of quantum dots [24], etc. A recent paper in Physical Review Letters also used electric hot spot (See Figure 6.1, taken from [25, Fig. 1]). These literature suggest the electric nature of plasma formation of dimer systems exposed in oscillating EM field.

doi:10.6342/NTU202400923

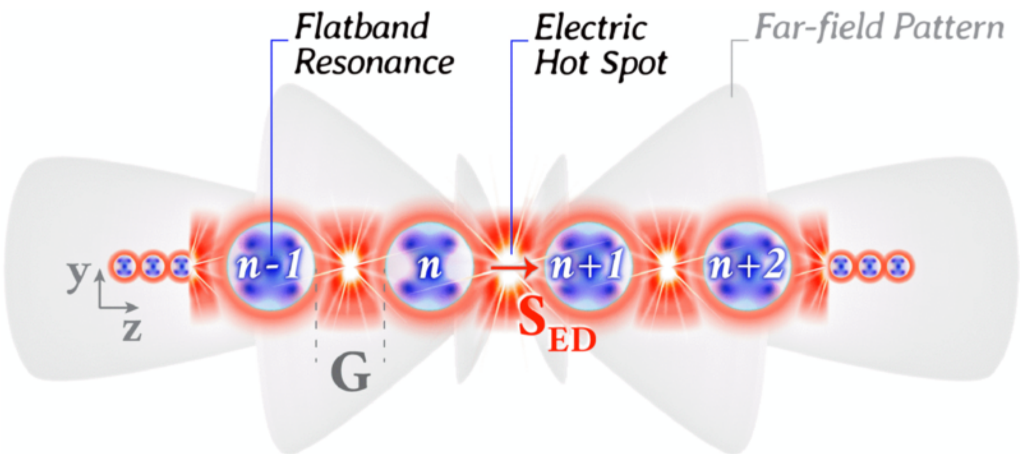


Figure 6.1: Schematic that showed the electric hot spot. Resonances happen within spheres. Taken from Thanh Xuan Hoang, Daniel Leykam, and Yuri Kivshar. Photonic Flatband Resonances in Multiple Light Scattering. Physical Review Letters, 132:043803, Jan 2024. (DOI: https://doi.org/10.1103/PhysRevLett.132.043803), licensed under CC BY 4.0 (https://creativecommons.org/licenses/by/4.0/).

doi:10.6342/NTU202400923

doi:10.6342/NTU202400923

References

[1] M. S. Lin, L. C. Liu, L. R. Barnett, Y. F. Tsai, and K. R. Chu. On electromagnetic wave ignited sparks in aqueous dimers. Physics of Plasmas, 28(10):102102, 2021.

[2] John David Jackson. Classical Electrodynamics. Wiley, New York, 3rd edition, 1999.

[3] Peter Josef William Debye. Polar molecules. Dover publications, 1929.

[4] Udo Kaatze. Complex permittivity of water as a function of frequency and temperature. Journal of Chemical & Engineering Data, 34(4):371–374, 1989.

[5] Jun Zhou, Xin Rao, Xiaoming Liu, Tao Li, Lin Zhou, Yanshun Zheng, and Zheng Zhu. Temperature dependent optical and dielectric properties of liquid water studied by terahertz time-domain spectroscopy. AIP Advances, 9(3):035346, 2019.

[6] U Kaatze and V Uhlendorf. The dielectric properties of water at microwave frequencies. Zeitschrift für Physikalische Chemie, 126(2):151–165, 1981.

[7] Cecilie Rønne, Lars Thrane, Per-Olof Åstrand, Anders Wallqvist, Kurt V Mikkelsen, and Søren R Keiding. Investigation of the temperature dependence of dielectric relaxation in liquid water by thz reflection spectroscopy and molecular dynamics simulation. The Journal of chemical physics, 107(14):5319–5331, 1997.

doi:10.6342/NTU202400923

[8] Chao Zhang and Michiel Sprik. Computing the dielectric constant of liquid water at constant dielectric displacement. Physical Review B, 93:144201, Apr 2016.

[9] Gabriele Raabe and Richard J. Sadus. Molecular dynamics simulation of the dielectric constant of water: The effect of bond flexibility. The Journal of Chemical Physics, 134(23):234501, 2011.

[10] Kuan-Wen Chen (陳冠文). An investigation on microwave dielectric heating : polarization charge shielding effect and microwave resonance phenomenon/ 介電質微波加熱之特性探討: 極化電荷之屏蔽效應與微波共振現象, 2019.

[11] Stuart Nelson and Andrzej Kraszewski. Dielectric properties of materials and measurement techniques. Drying Technology - DRY TECHNOL, 8:1123–1142, 01 1990.

[12] Roger F. Harrington. Time-Harmonic Electromagnetic Fields. IEEE-Press, 2001.

[13] Andrew Zangwill. Modern Electrodynamics. Cambridge University Press, 2012.

[14] Someone on wiki. Morphology-dependent resonance (https://en.wikipedia.org/wiki/Morphology-dependent_resonance), May 2021.

[15] Hamza K. Khattak, Pablo Bianucci, and Aaron D. Slepkov. Linking plasma formation in grapes to microwave resonances of aqueous dimers. Proceedings of the National Academy of Sciences, 116(10):4000–4005, 2019.

[16] Gustav Mie. Beiträge zur optik trüber medien, speziell kolloidaler metallösungen. Annalen der physik, 330(3):377–445, 1908.

[17] Hitoshi Kuwata, Hiroharu Tamaru, Kunio Esumi, and Kenjiro Miyano. Resonant light scattering from metal nanoparticles: Practical analysis beyond rayleigh approximation. Applied Physics Letters, 83(22):4625–4627, 2003.

doi:10.6342/NTU202400923

[18] Reuben M Bakker, Dmitry Permyakov, Ye Feng Yu, Dmitry Markovich, Ramón Paniagua-Domínguez, Leonard Gonzaga, Anton Samusev, Yuri Kivshar, Boris Luk'yanchuk, and Arseniy I Kuznetsov. Magnetic and electric hotspots with silicon nanodimers. Nano Letters, 15(3):2137–2142, 2015.

[19] Ming S Lin, Shih M Lin, Wei Y Chiang, LR Barnett, and KR Chu. Effects of polarization-charge shielding in microwave heating. Physics of Plasmas, 22(8):083302, 2015.

[20] Masud Mansuripur and Armis R. Zakharian. Maxwell's macroscopic equations, the energy-momentum postulates, and the lorentz law of force. Phys. Rev. E, 79:026608, Feb 2009.

[21] Scott R Waitukaitis, Antal Zuiderwijk, Anton Souslov, Corentin Coulais, and Martin van Hecke. Coupling the leidenfrost effect and elastic deformations to power sustained bouncing. Nature Physics, 13(11):1095–1099, November 2017.

[22] Eric B. Lindgren, Ivan N. Derbenev, Armik Khachatourian, Ho-Kei Chan, Anthony J. Stace, and Elena Besley. Electrostatic self-assembly: Understanding the significance of the solvent. Journal of Chemical Theory and Computation, 14(2):905–915, 2018. PMID: 29251927.

[23] Zhenguo Gao, Binghui Xu, Mingliang Ma, Ailing Feng, Yi Zhang, Xuehua Liu, Zirui Jia, and Guanglei Wu. Electrostatic self-assembly synthesis of znfe2o4 quantum dots (znfe2o4@c) and electromagnetic microwave absorption. Composites Part B: Engineering, 179:107417, 2019.

[24] R.K. Singh, R. Kumar, D.P. Singh, R. Savu, and S.A. Moshkalev. Progress in

doi:10.6342/NTU202400923

microwave-assisted synthesis of quantum dots (graphene/carbon/semiconducting) for bioapplications: a review. Materials Today Chemistry, 12:282–314, 2019.

[25] Thanh Xuan Hoang, Daniel Leykam, and Yuri Kivshar. Photonic flatband resonances in multiple light scattering. Phys. Rev. Lett., 132:043803, Jan 2024.

doi:10.6342/NTU202400923

Appendix A — Derivation of Poynting's Theorem in Time Domain and Time Harmonics

Starting from Newtonian Equation of Motion, the power P done by a force $\mathbf{F}$ on a particle moving with velocity $\mathbf{v}$ is,

$$P = \mathbf{F} \cdot \mathbf{v}. \tag{A.1}$$

For a charge particle of charge q moving in electric field $\mathbf{E}$ and magnetic field $\mathbf{B}$, the Lorentz force acting on it (in SI units) is,

$$\mathbf{F} = q\,[\,\mathbf{E} + \mathbf{v} \times \mathbf{B}\,]. \tag{A.2}$$

The power is therefore $P = q\mathbf{E} \cdot \mathbf{v}$, or in a volume integral form,

$$P = \int \mathrm{d}^3x \; p = \int \mathrm{d}^3x \; \rho\mathbf{E} \cdot \mathbf{v} = \int \mathrm{d}^3x \; \mathbf{J} \cdot \mathbf{E}, \tag{A.3}$$

where p is the power density, ρ is the charge density and $\mathbf{J}$ is the current (density). Use the macroscopic Maxwell equation $\boldsymbol{\nabla} \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t}$ to substitue $\mathbf{J}$ in the final term, we

doi:10.6342/NTU202400923

have

$$\int d^3x\ \mathbf{J}\cdot\mathbf{E} = \int d^3x \left[(\boldsymbol{\nabla}\times\mathbf{H})\cdot\mathbf{E} - \frac{\partial \mathbf{D}}{\partial t}\cdot\mathbf{E}\right]. \tag{A.4}$$

Use

$$\boldsymbol{\nabla}\cdot(\mathbf{E}\times\mathbf{H}) = \mathbf{H}\cdot(\boldsymbol{\nabla}\times\mathbf{E}) - \mathbf{E}\cdot(\boldsymbol{\nabla}\times\mathbf{H}), \tag{A.5}$$

and

$$\boldsymbol{\nabla}\times\mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \tag{A.6}$$

we will obtain,

$$\int d^3x\ \mathbf{J}\cdot\mathbf{E} = -\int d^3x \left[\boldsymbol{\nabla}\cdot(\mathbf{E}\times\mathbf{H}) + \mathbf{E}\cdot\frac{\partial \mathbf{D}}{\partial t} + \mathbf{H}\cdot\frac{\partial \mathbf{B}}{\partial t}\right], \tag{A.7}$$

or the most general form of macroscopic energy balance (in time domain),

$$\mathbf{J}\cdot\mathbf{E} + \boldsymbol{\nabla}\cdot(\mathbf{E}\times\mathbf{H}) + \left[\mathbf{E}\cdot\frac{\partial \mathbf{D}}{\partial t} + \mathbf{H}\cdot\frac{\partial \mathbf{B}}{\partial t}\right] = 0. \tag{A.8}$$

The frequency domain equation

1. assumes $e^{-i\omega t}$ dependence of above equation;
2. therefore assumes steady-state of ϵ and μ (assumes they are time-invariant).
3. evaluates the time-averaged quantities of Equation (A.8).

Following above steps, We will reach Equation (5.4a).

doi:10.6342/NTU202400923