On the Possibilities of Detecting Surface Charges with Second Harmonic Generation and Optical Diagnostics of Polymeric Insulators

Bengtsson, Magnus

2003

Link to publication

Citation for published version (APA):
On the possibilities of detecting surface charges with second harmonic generation and optical diagnostics of polymeric insulators

Magnus Bengtsson

Lund Reports on Atomic Physics
LRAP-303

Licenciate Thesis
Department of Physics
Lund Institute of Technology
June 2003
Contents

Abstract 3

List of Papers 5

1 Introduction 7
   1.1 Motivation 7
   1.2 Objective 7
   1.3 Outline 8

2 Some Existing Methods of Measuring Surface Charge 9
   2.1 The Pockels' Effect 9
   2.2 Electrostatic Field Probe 12
   2.3 Two-photon Photoelectric Emission 15

3 Field Calculations 17

4 Introducing Second Harmonic Generation 25
   4.1 Basic Principles. Second Harmonic Generation 25
   4.2 SHG Theory. Second Harmonic Generation on Surfaces 27

5 Experiments 35
   5.1 The Terawatt Laser System 40
   5.2 Experimental Arrangements 41
   5.3 Results and Discussion 41

Outlook 47

Comments on the papers 50

Acknowledgements 51

References 52
Abstract

Optical second-harmonic generation can be a sensitive probe of atoms and molecules at a surface or interface. For this reason, the method was chosen to study surface charges. Our measurements have been limited by large fluctuations in the frequency-doubled light and the mean value of the signals did not give any consistent changes due to changes in the surface charge. A solid metal constituted the sample but if the goal of the experiment had been to study any material configuration a silver-mica capacitor would have been a good choice due to earlier research.

As the original intention of the project was to detect surface charges on polymeric insulators a natural generalisation of the project has been to characterise polymeric insulator materials and we have started this by studying biological growths, like algae and fungi. Both are crucial for the performance of the material. In cooperation with Andreas Dernfalk at the Chalmers University of Technology the presence of algae and fungi could readily be remotely detected.
List of papers

The following papers are included in the thesis.

I  A. Larsson and M. Bengtsson,  
"The Sequence of Events in Mid-Gap Laser-Triggered Electrical Breakdown in Liquid Dielectrics"  
IEEE Transactions on Dielectrics and Electrical Insulation 9 (2002) 349.

Chapter 1

Introduction

1.1 Motivation

Within the programme High Performance Outdoor Electrical Insulation (ELIS) supported by the Swedish Foundation for Strategic Research (SSF) a major goal is to develop polymeric insulators with good flashover resistance. Surface discharges are one of the most serious problems of outdoor insulators. Surface charging of the insulator and how this affects the flashover withstand capability is one of the major issues for insulator performance. It would therefore be highly relevant to monitor the surface charges that are present on the surface prior to the discharges, and to couple the surface charge density of the material to its macroscopic surface discharge/flashover resistance.

1.2 Objective

An insulator is characterised by an energy gap between the conduction band and the valence band. For a metal the two bands overlap and there are many electrons in the conduction band, which implies that surface charges can easily move if an external electric field is applied.

The objective of the project is to develop techniques for remote and non-perturbing determination of the charge density on a surface. The amount of induced surface charge should arise from a weaker electric field than the threshold for electric breakdown in air. At normal temperature and pressure this threshold is approximately 3 MV/m and corresponds to a net surface charge of 27 \( \mu \text{C/m}^2 \). This can be interpreted roughly as a \( 10^{-4} \) change in the surface electron density. We believe knowing the surface charge density is important for understanding surface discharges which is a limiting factor in many electrical insulation systems. Specifically we consider non-linear optical laser measurement techniques, which can be designed such that they are sensitive only to the surface properties. The ability of remote and non-perturbing charge measurements would be a powerful tool for monitoring insulators during operation as well as for the development of new insulators.
1.3 Outline

Chapter 1 gives an introduction to the project and describes its background. Chapter 2 contains some techniques for measuring surface charge. Some field calculations have been made during the project and are described in chapter 3. Chapter 4 contains the theory to the second harmonic generation. Chapter 5 provides information about the Terawatt laser system, the experimental arrangements, the results and the discussion. An outlook is also presented.
Chapter 2

Some existing Methods of Measuring Surface Charge

In this chapter three techniques for measuring surface charge are explained. The “two-photon photoelectric emission” method is described since it is more sensitive to surface charge than second harmonic generation [Tomas et al., 1999]. Two other methods are also described, one which is based on the Pockels’ effect and are based on an electrostatic field probe where the surface charge is induced electrostatically. I have chosen to describe the method based on the Pockels’ effect since it is a laser method and the field probe due to its simplicity. Several other techniques exist to measure surface charge, e.g. Atomic force microscopy [Raiteri et al., 1996], Kelvin probe [Nalbach and Kliem, 2000], and Surface Enhanced Raman Scattering [Weiss and Haran, 2001].

2.1 The Pockels’ Effect

Nowadays, experimental devices for optical measurement of surface charge have become less expensive than before. Highly sensitive electro-optical crystals, powerful and highly stable single wavelength laser sources, high-quality optical elements, high-speed highly sensitive charge-coupled device (CCD) cameras, high-capacity frame memories and computers have all become available at lower prices.

Information about the surface charge distribution is obtained by studying the polarisation of light transmitted through a crystal, that exhibits the Pockels’ effect. This effect is a dielectric anisotropy caused by an electric field, which in this case is due to the surface charge.

One example of utilising the Pockels’ effect as a means for measurement of surface charge is presented in Fig. 2.1 [Zhu et al., 1995].
Surface charge is deposited on the insulating film by the application of a high voltage impulse to a needle electrode. The charge is attached to one side of a Bi₁₂SiO₂₀ (BSO) single crystal plate that has an earthed transparent indium tin oxide (ITO) electrode on the opposite surface.

A light beam from a He-Ne laser is expanded in diameter from 1 mm to 50 mm by a beam expander. A polarised beam splitter (PBS) polarises the light beam linearly. The optical phase modulation is described later. The light beam is then transmitted into the BSO crystal and reflected from the interface between the insulating film and the BSO crystal. The reflected light beam has a retardation (delay) between the two polarisation components (ordinary ray and extraordinary ray) of the electro-magnetic field and this retardation is proportional to the electric field due to the Pockels’ effect. The surface charge causes the electric field. The Pockels’ effect is an electro-optical effect in which the application of an electric field produces a birefringence (also known as double refraction) which is proportional to the field. The reflected light intensity is recorded by a high-speed CCD camera connected to an image lock-in amplifier computer system for image processing to get the surface charge distribution. The light intensity is recorded and by using two images it is possible to derive the retardation distribution \( \theta_{\sigma}(x,y) \), i.e. the delay between the two polarisation components as a function of \( x \) and \( y \).

The light intensity is detected as images. The light intensity distribution is expressed as

\[
I(x,y) = I_0(x,y) \sin^2 \theta(x,y) + I_{off-set}(x,y) \tag{2.1}
\]
where $\theta(x,y)$ is the total retardation distribution, $I_0(x,y)$ is the maximum light intensity distribution of the incident light and $I_{\text{off-set}}(x,y)$ is the DC off-set component due to the dark current in the camera and the leakage light intensity of the PBS. Transmission and reflection ratio distributions of the optical devices and the sensitivity of the CCD camera's imaging sensor matrix are incorporated by $\gamma(x,y)$.

All optical devices have a certain amount of natural birefringence caused by the residual mechanical stress. The total retardation $\theta(x,y)$ is expressed as a sum between the natural retardation distribution and the retardation distribution due to the surface charge distribution $\sigma(x,y)$ and the Pockels' effect.

$$\theta(x,y) = \theta_{\text{nat}}(x,y) + \theta_\sigma(x,y) \quad (2.2)$$

The DC off-set term $I_{\text{off-set}}$ is eliminated in the following way: Between the PBS and the test cell there is a spatial optical phase modulator (OPM). It modulates in pulsed mode the optical phase difference between the two components of light parallel and perpendicular to the fast axis of the optical phase modulator. A square wave drives the OPM with an amplitude of $V_m$ and two light intensity images are recorded during the positive and negative amplitude of the pulse. The recorded images, $I_{s+}(x,y)$ and $I_{s-}(x,y)$, may be written as

$$I_{s+}(x,y) = \gamma(x,y)I_0(x,y)\sin^2[\theta(x,y) + \theta_m] + I_{\text{off-set}}(x,y) \quad (2.3)$$

$$I_{s-}(x,y) = \gamma(x,y)I_0(x,y)\sin^2[\theta(x,y) - \theta_m] + I_{\text{off-set}}(x,y) \quad (2.4)$$

where $\theta_m$ describes to what extent the optical phase is modulated during the positive and negative amplitude of the modulating pulse voltage.

The light intensity difference between the two images is

$$\Delta I(x,y) = I_{s+}(x,y) - I_{s-}(x,y) = I_M(x,y)\sin[2\theta(x,y)] \quad (2.5)$$

$$I_M(x,y) = \gamma(x,y)I_0(x,y)\sin(2\theta_m) \quad (2.6)$$

where $I_M(x,y)$ is called the equivalent maximum light intensity distribution. Now the total retardation distribution can be calculated as

$$\theta(x,y) = \frac{1}{2} \arcsin[\Delta I(x,y) / I_M(x,y)] \quad (2.7)$$

In this way the DC off-set term $I_{\text{off-set}}(x,y)$ due to the dark current of the CCD camera is removed. The natural retardation distribution is affecting the total retardation distribution as

$$\theta_\sigma(x,y) = \theta(x,y) - \theta_{\text{nat}}(x,y) \quad (2.8)$$

and it is also possible to derive $\theta_{\text{Nat}}$.
\[ \theta_{nat}(x,y) = \frac{1}{2} \arcsin[\Delta I(x,y)/I_M(x,y)] \]  

(2.9)

where \( \Delta I(x,y) \) is the light intensity distribution of the difference image between two images but when there does not exist any surface charge on the film. The surface charge \( \sigma(x,y) \) is proportional to the retardation distribution \( \theta_{\sigma}(x,y) \) and hence

\[ \theta_{\sigma}(x,y) = K \cdot \sigma(x,y) \]  

(2.10)

where \( K \) is the optical constant of the BSO crystal. The equations (2.8), (2.9), and (2.10) determine the surface charge \( \sigma(x,y) \).

The lower limit of the sensitivity of the surface charge density is 10 \( \mu \text{C/m}^2 \) [Zhu et al., 1995]. This electro-optical method has the advantage of simultaneous visualization of electric charge and it is feasible to quantify the surface charge density. It is also possible to study the temporal development of the surface charge. Since the charge image data are stored in a computer, the spatial noise can be reduced to achieve adequate information. A minor disadvantage with this technique is the necessity of the lock-in amplifier to observe the dynamic of the surface charge. It is also difficult to apply this method to outdoor field experiments.

For further reading, study for example “Dynamic Observation of Needle-plane Surface Discharge using the Electro-optical Pockels Effect” [Zhu et al., 1996].

### 2.2 Electrostatic Field Probe

Interpretation of potential probe measurements has been discussed greatly since the early 80’s.

The electrostatic field probe makes it possible to obtain the charge distribution on a solid insulator. Charges are electrostatically induced on the sensor plate of the probe by the surface charge on the insulator. A low-loss capacitor is connected to the probe and the charge induced on the probe can yield the potential across the capacitor. To be more specific, as the probe is moved parallel to the surface it is the variation in probe potential which is the measured parameter.

A long cylindrical shaft at earth potential with a circular conducting-disc constitute the field probe, see Fig. 2.2. The disc is insulated from the shaft and coaxially mounted.
Figure 2.2 The probe/dielectric geometry.

Evaluation of Probe Measurements

Three methods have emerged to evaluate probe measurements: The direct method, the capacitive approximation and the indirect method.

The Direct Method

Basically, this method is utilising the solution of Poisson’s equation where interface surface charge and all other charges in the space between the electrodes and on the electrodes represent unknown sources. Known parameters are the probe potential and its location. This approach demands a lot of numerical analysis and therefore very few studies have adopted this method.
The Capacitive Approximation

This method assumes that the probe response can be related to an equivalent capacitive circuit. Due to the simplicity of the method it has been used in the majority of the probe investigations. For further information e.g. see reference 9-19 of [Crichton and McAllister, 1992].

The Indirect Method

A probe function named the $\lambda$-function connects the charge induced on the probe to the surface charge density at the dielectric interface [Rerup et al., 1994]. If the volume charge density inside the solid dielectric is assumed to be zero, the relationship can be expressed as

$$q_i = -\int_{A_0} \lambda \sigma dA$$  \hspace{1cm} (2.11)

where $q_i$ is the Poissonian induced charge on the sensor plate [Pedersen et al., 1993], and $\sigma$ is the surface charge density on the surface element $dA$ of the solid dielectric surface $A_0$. The dimensionless parameter $\lambda$ is shown to be a solution of the general Laplacian equation for the complete measuring-system geometry:

$$\nabla \cdot (\varepsilon \nabla \lambda) = 0$$  \hspace{1cm} (2.12)

At the probe sensor-plate the boundary condition is $\lambda = 1$, and at all other electrodes $\lambda = 0$. Further, at the dielectric interface the normal component of the derivatives of $\lambda$ must satisfy the condition

$$\varepsilon_+ \left( \frac{\partial \lambda}{\partial n} \right)_+ = \varepsilon_- \left( \frac{\partial \lambda}{\partial n} \right)_-$$  \hspace{1cm} (2.13)

where the positive and negative signs are the opposite sides of the interface. Eq. (2.12) can be solved numerically to evaluate the variation of $\lambda$ across the surface.

Practically, a scanning probe technique is employed to monitor the variation in $\sigma$ across the dielectric surface. Since $\lambda$ is geometry dependent it is necessary to find $\lambda$ as a function of probe location. Due to the variation of probe location a system of integral equations similar to Eq. (2.11) have to be solved. The solution determines the interface charge distribution.

The lower limit of the sensitivity of the surface charge density for the field probe is 0.1 nC/m² or less for the electric field of 50 Hz [ELFA, 2002]. However the probe technique is only effective in limited situations. There are measurement errors due to the following factors: thickness of the insulator affects $\lambda$, area of surface charging, distance between probe and solid insulator, coexistence of positive and negative charges, and bulk charging. Hence, the analysis of the probe potential outputs is the crucial problem that has
to be solved to obtain a method to measure surface charge more accurately. On the other hand, it is experimentally fairly uncomplicated.

2.3 Two-photon Photoelectric Emission

Photoemission spectroscopy is a powerful and an accurate tool for investigating the electronic structure of metals. Photoemission spectroscopy techniques can be used to study adsorbates, oxide formation, surface roughness, and laser cleaning.

By applying an external electric field to a surface, surface charge can be accumulated. The basic idea is that a laser beam impinging the surface will generate photoelectrons. By collecting the photoelectrons it is possible to determine the net amount of surface charge that is accumulated on the surface.

A picosecond Nd:YAG, i.e. Neodymium:Yttrium Aluminum Garnet, laser system is used, see Fig. 2.3, delivering pulses at the wavelength of 1064 nm with a repetition rate of 10 Hz [Tomas et al., 1999]. This fundamental wavelength is frequency-doubled to 532 nm (green light) by a potassium dihydrogen phosphate (KDP) crystal. The laser beam enters an ultrahigh vacuum chamber, and hits a sample made of gold, which constitutes of a photocathode. The anode is placed 5 mm above the photocathode and the attracting (to the electrons) electric field is delivered by a high voltage system (0 to 30 kV). The experiment is performed in an ultrahigh vacuum chamber with a pressure in the order of 10^{-17} Pa. A digital multimeter (used as a coulombmeter) measures the emitted photoelectrons.

![Diagram](image)

*Figure 2.3 A part of the experimental arrangement for two-photon photoelectric emission from Au.*

The photoemitted charge increases with the voltage and seems to limit as a plateau-like saturation. Photoelectrons are generated as a thin disk due to the incoming light and with a diameter that depends on the laser spot size on the target. Image charge and space charge must be considered. Due to coulomb forces, the electrons are attracted by the image
charge that is created in the cathode but repulsive space charge forces also exist between the electrons. Thus the electric field must be stronger than these forces that pull back the electrons to the material. Hence the collected charge increases with voltage and reaches the plateau when the voltage is large enough.

High-energy X-rays could be a problem and one could argue that a high voltage together with electrons might generate high-energy X-rays, which would hit the cathode and induce photoemission of high-energy electrons. If the high voltage increases both intensity and maximum energy of these X-rays might increase and thus the photoemitted charge too from the cathode. It is possible that such phenomena take place but the photoelectric signal is not enhanced once saturation is reached. Therefore these effects seem to be very small. This is also put up with the square law dependence of the photoelectric signal with incident laser intensity and serves as a fingerprint.

A lower limit of the sensitivity of the surface charge density is about 5 μC/m² [Tomas et al., 1999]. In the first approximation the photoelectric signals show quadratic-law dependence with the incident laser intensity (two-photon thermally assisted process) in accordance to a two-photon process. For higher intensities the process does not assume quadratic law-dependence. Unfortunately, the experimental set-up is complicated and intrusive. Further, the high vacuum makes the method very difficult to use outdoors for field experiments. It cannot be ascertained that the contribution to the photoemission is only a surface effect; the bulk may contribute [Tomas et al., 1999]. Otherwise the method seems to render measurement data of good resolution.
Chapter 3

Field Calculations

The finite element method (FEM) has been applied to electrostatic problems, containing no sources and with specified boundary conditions, solved by using the Laplace's equation which is given by

\[ \nabla \cdot (\varepsilon \nabla V) = 0 \]  \hspace{1cm} (3.1)

where \( \varepsilon \) is the permittivity and \( V \) the electric potential, i.e. the voltage. FEM utilises the minimum principle meaning that the solution to the Laplace’s equation is equivalent to minimise the electrostatic energy. The mathematical expression for this is expressed as

\[ W(V) = \frac{1}{2} \int_{\Omega} \varepsilon |\nabla V|^2 \, d\Omega. \]  \hspace{1cm} (3.2)

FEM has been used on a two-dimensional problem that resulted in a paper containing electric field calculations and titled 'The Sequence of Events in Mid-Gap Laser-Triggered Electrical Breakdown in Liquid Dielectrics'. Larsson et al. [Larsson et al., 2001] had earlier studied pre-breakdown and breakdown phenomena in oil and combined laser-triggering and the laser shadow method together with pre-breakdown current measurements. By doing this, the physical mechanisms and the timing sequence of the dielectric breakdown were revealed.

The electrode gap consisted of two hemispherically-tipped brass electrodes and was placed in oil. The electric field strength in oil was 12 MV/m which was achieved by using either 50 or 55 kV depending on the adjustable electrode distance that ranged between 4 and 5 mm. Ace [Ace version 3.0], a user friendly FEM software package, rendered the electric field calculations. The boundary conditions were 50 kV or 55 kV applied on the anode and a grounded cathode.

Calculations of the development of the electric field distribution in the gap with an expanding and perfectly dielectric cavity are shown in Fig. 3.1 and Fig 3.2. In the figures, the electric field strength is represented by the field enhancement factor, which is defined as the quotient between the field strength and the applied voltage.
The development of the maximum electric field strength in the electrode gap for a perfectly conducting cavity is shown in Fig. 3.3 and 3.4.
Development of the maximum field strength

Figure 3.3 The development of the maximum electric field strength gap for a perfectly conducting cavity.

Figure 3.4 The development of the maximum electric field strength gap for a perfectly conducting cavity.

A typical example of breakdown conditions is an applied voltage of 50 kV with a cavity radius of 1 mm for a 4 mm-gap. Fig. 3.5 gives that the breakdown voltage is approximately 30 kV. Hence, the breakdown voltage in Fig. 3.5 is too low. This information reveals that the cavity has not been a perfect conductor. By calculating the
electric field strength along the central axis for a cavity radius of 1 mm the critical field for breakdown (22.5 MV/m) was

![Radius of the cavity at breakdown](image)

**Figure 3.5** For a given applied voltage, the radius of the cavity is shown that gives rise to breakdown in oil. A perfect conducting cavity is assumed.

exceeded in the whole oil gap. When the cavity was assumed to be a perfect dielectric (non-conducting) the electric field strength in oil made this electric field strength too low to trigger a discharge in the oil between the cavity and the electrodes. The conclusion is that the cavity is not a perfect conductor but some charge redistribution must occur within the cavity to render the required field enhancement. The electric field calculations led to the explanations of the physical processes.

In the second harmonic generation experiments the FEM software package ANSYS [ANSYS release 5.7] was applied to find the electric field strength at the gold mirror target and specifically across the whole area where the laser light hit the mirror. This has been performed in collaboration with Mose Akyuz at the Division of Electricity and Lightning Research at Uppsala University.

The electrode gap consisted of an anode (rod and sphere) and the cathode (circular mirror) and was placed in air. The distance between the anode and the cathode (grounded gold/silver mirror) was approximately 3 cm and the applied potential was 4 kV on the anode. The geometry is shown in Fig. 3.6.
It can be seen in Fig. 3.7 that the surface charge was approximately $1.1 \, \mu\text{C/m}^2$ where the laser light interacted with the mirror. A more detailed study is shown in Fig. 3.8.

**Figure 3.6** The high-voltage geometry. Anode to the left and cathode to the right. The white spot indicates where the laser hit the mirror. The diameter of the spot was 0.5 cm.

**Figure 3.7** Surface charge along a diameter on the mirror (see the line in Fig. 3.6) and parallel to the rod of the anode. The x-axis is defined as negative along the line under the rod of the anode.
The surface charge varied across the laser beam when it hit the mirror. The radius of the laser beam was 2.5 mm. The laser spot went from $-2.5$ mm to $+2.5$ mm in this graph. The arrows in the picture indicate the endpoints of the laser light on the mirror.

Fig. 3.7 shows that the surface charge along the line in Fig 3.6 is quite large below the rod of the anode and decreases as the distance between the mirror and anode increases. Fig. 3.8 shows that the surface charge did not vary very much where the laser hit the mirror. The excess surface charge on the mirror ranged from $1.020$ to $1.063 \mu$C/m$^2$.

Fig. 3.9 shows the equipotential surfaces. The blue colour denotes a potential interval close to zero and the red colour close to 4 kV. The large circle defines the design space that encapsulates the region in space where the high-voltage system existed.
Figure 3.9 The equipotential surfaces.
Chapter 4

Introducing Second Harmonic Generation

Second harmonic generation (SHG) is a well-known and widely used process in nonlinear optics and was e.g. demonstrated on surfaces by Bloembergen et al. [Bloembergen et al., 1968] in the end of the sixties. A comprehensive theoretical description of the various contributions to SHG is for example given by Guyot-Sionnest et al. [Guyot-Sionnest and Shen, 1987] [Guyot-Sionnest et al., 1986]. Some basic principles introduce this chapter, which is then devoted to the SHG theory on surfaces.

This chapter is quite esoteric. The reader should be able to understand the basic principles of second harmonic generation, which is described in chapter 4.1. Chapter 4.2 deduces the important polarisation terms. The derivation of these terms is completely based on [Bloembergen et al., 1968] and less important. In the section “Intensity Relationship between Surface Charge Induced SHG and the Fundamental Wave” are also some important relations found.

4.1 Basic Principles of Second Harmonic Generation

Second harmonic generation or frequency doubling is present at scattering or reflection at surfaces and in bulks that do not display inversion symmetry. If a spatial asymmetry is present a second harmonic (SH) wave can be generated. Reflection at a surface and surface charge fulfil this spatial asymmetry. The polarisation, \( P \), is in classical linear optics expressed by the linear relationship

\[
P = \varepsilon_0 \chi^{(1)} \cdot E
\]  

(4.1)

where \( \chi^{(1)} \) is the linear susceptibility, \( \varepsilon_0 \) the electric permittivity of free space and \( E \) the electric field strength of the applied optical field. However, in nonlinear optics a more general description of the polarisation is utilised. The polarisation of a medium is expanded as a power series in the electric field strength.

\[
P = \varepsilon_0 (\chi^{(1)} E^1 + \chi^{(2)} E^2 + \chi^{(3)} E^3 + ...)
\]  

(4.2)

where \( \chi^{(2)} \) and \( \chi^{(3)} \) are the second and the third-order nonlinear optical susceptibilities [Boyd, 1992]. In classical linear optics only the first term is considered such that Eq. (4.2) simplifies to Eq. (4.1) but nonlinear effects commonly have to be incorporated into the theory when dealing with laser beams of high power. If a laser beam with the electric field \( E = E_0 \cdot \cos \omega t \) is reflected at a surface and if the intensity of the beam is high enough
it is possible to observe the SH wave at $2\omega$. This is illustrated in Fig. 4.1. The incident wave at the fundamental angular frequency $\omega$ creates a polarisation including a component at $2\omega$ at the surface as can be understood from the second term in Eq. (4.2) since

$$E^2 = (E_0 \cos \omega t)^2 = E_0^2 \cos^2 \omega t = E_0^2 \left( \frac{1}{2} + \frac{\cos 2\omega t}{2} \right).$$

(4.3)

The polarisation is a driving term in the general wave equation (for wave propagation) that is given by

$$\nabla \times \nabla \times E + \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = -\mu_0 \frac{\partial^2 P}{\partial t^2}. $$

(4.4)

where $\mu_0$ is the magnetic permeability of free space.

The solution to the general wave equation renders the electric field vector at the harmonic angular frequency $2\omega$.

For example, if a laser beam with infrared light of 800 nm is frequency-doubled a small fraction of this light is converted into blue light of 400 nm.

![Figure 4.1](image)

**Figure 4.1** Second harmonic generation from a surface. Incoming infrared light of 800 nm is visualized by the gray beam. After the reflection in the surface two beams with the same reflection angle emerge, the infrared and the SH wave of 400 nm visualized by the black beam.

An alternative description of SH generation can be seen in Fig. 4.2. This can be viewed in terms of converting two photons of 800 nm to one photon of 400 nm. This takes place in a single quantum mechanical process.

SH generation depends on the medium, in a noncentrosymmetric medium the contribution from the bulk is non-vanishing. For example a ruby laser beam can be converted in a quartz crystal to a blue second harmonic wave [Maker et al., 1962]. Phasematching has also to be considered and the rotation angle of the crystal, i.e. the angle between the optic axis and the propagation vector, is of utmost importance [Boyd, 1992]. Second-order processes are forbidden in centrosymmetric media due to symmetry considerations, but they are never forbidden at the surface layers. Liquids, gases, amorphous solids (such as glass), and many crystals with inversion symmetry have vanishing bulk susceptibilities.
Figure 4.2 Energy-level diagram describing the second-harmonic generation. Incoming infrared light of angular frequency \( \omega \) to the surface is frequency-doubled to blue light of \( 2\omega \). The solid line represents the atomic ground state and the dashed lines are virtual levels i.e. energy levels of the atom and one or more photons of the electromagnetic radiation field. "Light in" refers to the infrared light in to the surface and the "light out" refers to the produced blue light out from the surface.

Since the SH generation is a two-photon process it displays a quadratic-law dependence between the fundamental wave intensity at \( \omega \) and SH wave intensity at \( 2\omega \). This can be expressed as

\[
I_{2\omega} = \text{const} \cdot I_{\omega}^2
\]

(4.5)

where the constant is specific for the material. The constant depends on many parameters e.g. the surface charge. Eq. (4.5) can be deduced from the solution to the general form of the wave equation (4.4) and substituting the polarisation with Eq. (4.2) [Boyd, 1992]. \( I_{2\omega} \) depends also on the surface roughness, the pressure, and if the sample is a thin film or a solid material.

4.2 SHG theory. Second Harmonic Generation on Surfaces

N. Bloembergen et al. studied the optical second-harmonic generation in reflection from media with inversion symmetry [Bloembergen et al., 1968]. A brief introduction to the theory of the second harmonic generation at surfaces is presented in this chapter. A more in depth presentation can be obtained from different books and articles in nonlinear optics,
for example "The principles of nonlinear optics" [Shen, 1984], "Nonlinear Optics in Metals" [Bennemann, 1998], and "Electronic Excitations at Metal Surfaces" [Liebsch, 1997].

Fig. 4.3 shows a sketch of a centrosymmetric medium where the inversion symmetry is broken at the surface.

Figure 4.3 A schematic view of second harmonic generation at a surface.

It is possible to derive various terms of the nonlinear polarisation. The interaction Hamiltonian is given by

\[ H_1 = -\bar{P}_1 \cdot \bar{E}(\bar{R},t) - \bar{M} \cdot \bar{B}(\bar{R},t) - \bar{Q} : \nabla_{\bar{R}} \bar{E}(\bar{R},t) \] (4.6)

with

\[ \bar{P}_1 = -e\bar{x} \] (4.7)

\[ \bar{M} = -\frac{e}{2m}(\bar{x} \times \bar{p}) = -\frac{e}{2m}\bar{L} \] (4.8)

\[ \bar{Q} = -\frac{1}{2}e\bar{x} \bar{x} \] (4.9)

where \( \bar{p} \) is the momentum, \( \bar{P}_1 \) is a dipole operator, \( \bar{Q} \) is a quadrupole operator, \( \bar{M} \) is a magnetic dipole operator, \( \bar{R} \) is the origin of the unit cell of the crystal and \( \bar{x} \) is the relative displacement operator. The double dot notation denotes a tensorial contraction.

The total current density, \( \bar{J} \), due to all electrons is given by

\[
\bar{J} = N \left[ \frac{\partial}{\partial t} \left\langle \bar{P} \right\rangle_{\text{cell}} + \nabla_{\bar{R}} \times \left\langle \bar{M} \right\rangle_{\text{cell}} - \frac{\partial}{\partial t} \nabla_{\bar{R}} \cdot \left\langle \bar{Q} \right\rangle_{\text{cell}} \right],
\] (4.10)

where \( N \) is the number of unit cells per unit volume and \( \langle \rangle_{\text{cell}} \) is the expectation value for all the electrons in the cell. The first term can be understood from
\[- Ne \frac{\partial \vec{v}}{\partial t} = - Ne \vec{v} \quad (4.11)\]

and thus a current. The second term is also a current and appears in the classical electrodynamics. The third term is a quadrupolar term that exists if the electric quadrupole moment depends on time and similar to the first term which is non-zero if the dipole moment depends on time.

E and B are expanded by

\[ E(R,t) = \sum_\omega E(R,\omega)e^{-i\omega t} \quad (4.12) \]

and

\[ B(R,t) = \sum_\omega B(R,\omega)e^{-i\omega t}. \quad (4.13) \]

Quadrupolar Contribution from Localized Orbitals

The lowest-order nonlinear contribution from bound electrons in non-magnetic crystals with inversion symmetry can be estimated following the procedure in reference [Bloembergen et al., 1968]. From bound electrons for a cubic or isotropic material in the low-frequency approximation the nonlinear source term is

\[ \chi_{\omega \rightarrow 0}^{L,L} (2\omega, \omega \rightarrow 0) \approx \frac{3}{4 \Pi e} \cdot (\chi^L)^2 (E_i \nabla_j E_j - E_j \nabla_i E_i). \quad (4.14) \]

\Pi is the density of valence electrons and Einstein summation is used to sum over j=x,y, and z. The linear susceptibility is given by

\[ \chi_{\omega \rightarrow 0}^{L,L}(\omega) = \frac{2\Pi e v^2}{\hbar \omega_0} \langle 0| x^2 |0 \rangle \quad (4.15) \]

where \( |0> \) denotes the ground state in the unperturbed system and \( x^2 \) comes from the quadrupolar operator. The factor \( \hbar \omega_0 \) is an average energy (closure approximation) [Bloembergen et al., 1968]. The SHG of light in a medium with inversion symmetry was first detected in calcite by Terhune and co-workers. The contribution to the SHG signal came from a quadrupolar type of interaction.

Contribution from the Free Electrons

Interband transitions, e.g. in KBr where the 3d and 4s bands overlap with the 3p band, complicate the agreement between theory and experiments. This is because the effective density of electrons in valence bands is higher. Thus \( \omega \) or \( 2\omega \) are close to an interband
transition, a correction term has to be added to the polarisation. If there are no interband
transitions of the valence electrons then it is possible to have a classical approach. The
low-frequency approximation above is not applicable here. In addition the magnetic-dipole
contribution (from the motion of the electrons) cannot be neglected. The nonlinear source
terms of the polarisation that are going to be derived are in a form appropriate for
comparison with the bound-electron contribution. The symmetry properties of the core
electrons are the same as for the conduction electrons.

The free electrons in the conduction band are treated as an electron gas where the
individual electrons have an average effective mass $m^*$. The hydrodynamic equation of
motion for the average velocity $\bar{v}$ can be written as

$$\frac{\partial \bar{v}}{\partial t} + \bar{v} \cdot \nabla \bar{v} = -\left(\frac{e}{m^*}\right)(\bar{E} + \bar{v} \times \bar{B}).$$  \hspace{1cm} (4.16)

There is further the Maxwell equation

$$\nabla \cdot \bar{E} = -\frac{e(n - n_0)}{\varepsilon_0}$$  \hspace{1cm} (4.17)

and the continuity equation

$$\frac{\partial n}{\partial t} = -\nabla \cdot (n\bar{v})$$  \hspace{1cm} (4.18)

where $n$ is the number density of electrons and $n_0$ is related to $n$ by the expansion

$$n = \sum_{k=0}^{\infty} n_k e^{-ik\omega t}.$$  \hspace{1cm} (4.19)

The electric current density is given by

$$\bar{J} = -ne\bar{v}.$$  \hspace{1cm} (4.20)

By expanding the average velocity $\bar{v}$ (and $\bar{E}$ and $\bar{B}$ in the same way) as

$$\bar{v} = \sum_{k=0}^{\infty} \bar{v}_k e^{-ik\omega t} + \text{complex conjugate terms}$$  \hspace{1cm} (4.21)

and solving Eqs. (4.16), (4.17) (together with Eq. (4.10), Eqs. (4.12-4.13) and (4.18-4.21))
by successive approximations the nonlinear part of the second order polarisation created in
the electron gas is formed as

$$\bar{P}_{\text{conducting \ media \ NL}}^{\text{NL}}(2\omega) = -\frac{e}{2i\omega}(n_0 \bar{v}_2 + n_1 \bar{v}_1) =$$

$$(\delta - \beta)(\bar{E}(\omega) \cdot \nabla)\bar{E}(\omega) + \beta\bar{E}(\omega)(\nabla \cdot \bar{E}(\omega)) + \alpha\bar{E}(\omega) \times \bar{B}(\omega)$$  \hspace{1cm} (4.22)
according to [Bloembergen et al., 1968]. The relations between the coefficients are given below. The second term in Eq. (4.22) stems from both electric and magnetic contributions from the bulk (see Eq. (4.25)). Normally the electric polarisation \( \overline{P} \) is defined as

\[
\overline{P} = \lim_{\Delta V \to 0} \sum_{\Delta V} \frac{\varepsilon \overline{F}}{\Delta V},
\]

(4.23)

and includes the volume of consideration and \( \overline{J} \) is given by

\[
\overline{J} = -ne\overline{V} = -\frac{\partial (ne\overline{F})}{\partial t} = -\frac{\partial \overline{P}}{\partial t}
\]

(4.24)

where \( n \) is the number density. Eq. (4.24) together with Eq. (4.23) are similar to the first term in Eq. (4.10). The denominator, \( 2i\omega \), in Eq. (4.22) originates from the time derivative of the polarisation in Eq. (4.24). To obtain the third expression in Eq. (4.22) \( \overline{J} \) is expanded in \( \overline{V} \) and the second order terms are collected.

Four different source terms contribute to SHG according to Eq. (4.22) together with the non-centrosymmetric contribution from the surface (adsorbates can also be included into the surface term). The nonlinear polarisation can be expressed as

\[
\overline{P}^{\text{NL}} (2\omega) = (\delta - \beta - 2\gamma)(\overline{E}(\omega) \cdot \overline{\nabla})\overline{E}(\omega) + \beta \overline{E}(\omega)(\overline{\nabla} \cdot \overline{E}(\omega)) + \gamma \overline{\nabla}(\overline{E}(\omega) \cdot \overline{E}(\omega))
\]

\[+ \chi^{(2)}_{\text{surface}} : \overline{E}(\omega) : \overline{E}(\omega) \]

(4.25)

or equivalently

\[
\overline{P}^{\text{NL}} (2\omega) = (\delta - \beta)(\overline{E}(\omega) \cdot \overline{\nabla})\overline{E}(\omega) + \beta \overline{E}(\omega)(\overline{\nabla} \cdot \overline{E}(\omega))
\]

\[+ \alpha \overline{E}(\omega) \times \overline{B}(\omega) + \chi^{(3)}_{\text{surface}} : \overline{E}(\omega) : \overline{E}(\omega) \]

(4.26)

The transformation from Eq. (4.25) to Eq. (4.26) is seen from the identity

\[
\overline{E} \times (\overline{\nabla} \times \overline{E}) = \frac{1}{2} \overline{\nabla}(\overline{E} \cdot \overline{E}) - (\overline{E} \cdot \overline{\nabla})\overline{E}
\]

(4.27)

and the Maxwell equation

\[
(\overline{\nabla} \times \overline{E}) = j\omega \overline{B}.
\]

(4.28)

The coefficients \( \alpha, \beta, \gamma \) and \( \delta \) are called nonlinearities and \( \alpha \) is given by

\[
\alpha = (2i\omega)\gamma.
\]

(4.29)

For an isotropic (or cubic) insulating (valence electrons) medium, in the low-frequency limit [Bloembergen et al., 1968] (see Eq. (4.14) and Eq. (4.25)),
For free electrons (plasma abbreviated 'pl') the nonlinear plasma contribution must be added. The nonlinearities are [Bloembergen et al., 1968]

\[ \beta_{pl} = \frac{e}{2m \cdot \omega^2} \]

\[ \gamma_{pl} = \frac{1}{2i\omega} \alpha_{pl} = \frac{n_0e^3}{8\varepsilon_0m^2\omega^4} = \beta_{pl} \left( \frac{\omega_{pl}^2}{4\omega^2} \right) \]

The nonlinearities are additive for free electrons and bound electrons [Bloembergen et al., 1966].

\[ \alpha = \alpha_{pl} + \alpha_b \]

\[ \beta = \beta_{pl} + \beta_b \]

\[ \delta_{pl} = \beta_{pl} + 2\gamma_{pl} \]

where 'b' for bound electrons, and

\[ \omega_{pl}^2 = \frac{n_0e^2}{\varepsilon_0m^*} \]

is the angular plasma frequency.

The first two terms in Eq. (4.26) are of electric quadrupole character and the contribution is determined by \( \delta \) and \( \beta \). The third term corresponds to the magnetic dipole contribution and determined by \( \alpha \). The fourth term describes the electric dipole source from the broken symmetry at the surface. The terms in Eq. (4.26) depends on the polarisation of the second harmonic output and sometimes a term exists in the surface but not in the bulk. This is one good reason for separating the bulk from the surface as depicted in Fig. 4.3.

The nonlinear polarisation can always be expressed as Eq. (4.26) regardless of the mechanism and model for the nonlinearity. These phenomenological relationships are valid irrespective of whether the electrons are free or bound.

For metals usually only the first atomic layers contribute to SHG [Weber and Liebsch, 1987]. Bloembergen et al. [Bloembergen et al., 1966] showed that the electrons of the 4d-valence band make a comparable contribution to that of the conduction electrons to SHG in silver and gold. For insulating materials and semiconductors (using below-bandgap excitation) the bulk contribution can be sizeable and may even dominate the contribution to SHG.

For metal surfaces the plasma contribution to the nonlinearity is usually larger than the nonlinear contribution arising from interband transitions of the valence electrons. The only
way of finding the correct $\delta$ and $\beta$ is to calculate the actual potential at the boundary. This should be done for appropriate wave functions for the surface state rather than the bulk and then determine $J_{NL}$ for these states.

### Intensity Relationship between Surface Charge Induced SHG and the Fundamental Wave

The SHG signal due to surface charge can be obtained from the nonlinear polarisation [Corn et al., 1984]. Electric field induced SHG is deduced from a dc field independent nonlinear polarisation $\overline{P}_0$ and a dc field dependent nonlinear polarisation $\overline{P}$:

\[
\overline{P}_0(2\omega) = (\delta - \beta)(\overline{E}(\omega) \cdot \nabla)\overline{E}(\omega) + \beta \overline{E}(\omega)(\nabla \cdot \overline{E}(\omega)) + \alpha \overline{E}(\omega) \times \overline{H}(\omega) + \chi_{\text{surface}}^{(3)} \cdot \overline{E}(\omega) \cdot \overline{E}(\omega) \tag{4.37}
\]

\[
\overline{P}(2\omega) = \chi_{a}^{(3)} \overline{E}_{dc}[\overline{E}(\omega)]^2 + \chi_{b}^{(3)} \overline{E}(\omega)[\overline{E}_{dc} \cdot \overline{E}(\omega)] \tag{4.38}
\]

The total polarisation is the sum of $\overline{P}_0$ and $\overline{P}$ [Corn et al., 1984] where the latter polarisation is a third order nonlinear optical effect. The polarisation at $2\omega$ is the result of the application of two optical fields at $\omega$ and a static field. This effect is for example utilised in electric field induced second harmonic in the field of nonlinear optics and in nonlinear electroreflectance measurements.

The signal is proportional to the square of the total nonlinear polarisation and given by

\[
I_{2\omega}(E_{dc}) \propto a + b \cdot |(E_{dc} - c)^2 | \tag{4.39}
\]

where $a,b,$ and $c$ are given by $|\text{Im} P_0|^2$, $(\chi_{a}^{(3)} + \chi_{b}^{(3)})|E(\omega)|^2$, and $\text{Re} \ P_0 / b$, respectively.

For a perfect conductor Eq. (4.39) can be written as

\[
I_{2\omega}(\sigma) = a + b \cdot \left(\frac{\sigma}{\varepsilon_0} - c\right)^2 \tag{4.40}
\]

### Polarisation ($\phi$) and Incident Angular ($\theta$) Dependence

Imagine the surface and the incident plane in Fig. 4.1. When the polarisation-direction $\phi$ equals $\pi/2$ the light is perpendicular to the incident plane (s-polarised) and when $\phi$ is zero the laser light is parallel to the incident plane (p-polarised). P-polarised light is sensitive to the breaking of the centrosymmetry at the surface. The frequency-doubled light generated by s-polarised laser light arises only from higher order effects and surface roughness [Tomas et al., 1999].
Nonlinear dominant surface terms are responsible for a $\cos^4 \phi$ dependence of the SH intensity where $\phi$ is defined as the angle between the electric field vector of the laser and the plane of incidence. The incidence angle ($\theta$) dependence (to the surface) is roughly determined by $\sin^2(\theta)\cos^4(\theta)$. 
Chapter 5

Experiments

5.1 The Terawatt Laser System

A brief description of the terawatt laser system, which has been used in the experiments on SHG, is presented here. Svanberg et al. [Svanberg et al., 1994] provide a good description of the terawatt laser which is the main system of the Lund high-power laser facility. Although this paper is 9 years old and much has been changed since then the principle is still the same.

The research carried out using the terawatt laser generally requires high optical field strengths, for example generation of X-rays produced from a laser produced plasma and white-light generation with applications in medicine, biology and chemistry.

The terawatt laser relies on chirped pulse amplification (CPA), see Fig. 5.1, to achieve high optical intensities from a system that fits on top of a few optical tables. The fundamental idea is to stretch a short pulse in time before amplification. This decreases the peak intensity in the amplifier stage and does not damage the optical components. In this way large beam diameters and consequently large laser systems are avoided.

![Chirped pulse amplification](image)

**Figure 5.1** Chirped pulse amplification. The oscillator delivers a short pulse, A, that is stretched in time by the stretcher. The stretcher decrease the pulse peak intensity through temporal stretching, B. The pulse is then amplified, C, and finally compressed into a pulse with a shorter time duration and a higher peak power, D. [Svanberg et al., 1994].
A diagram of the laser system is shown in Fig. 5.2 and the components are explained below. The gain medium in the terawatt laser system is a titanium doped sapphire (Ti:S) crystal. Ti:S can easily be pumped by green light at 532 nm from a frequency-doubled Nd:YAG laser. Due to the short lifetime of Ti:S in its excited state (3 μs), flashlamp pumping is unsuitable. The gain profile of Ti:S is spectrally broad and enables amplification of very short laser pulses (50 fs); the minimum duration of a laser pulse is inversely proportional to the spectral bandwidth.

Figure 5.2 Block diagram of the Lund terawatt laser system [Svanberg et al., 1994].
The terawatt laser system uses an Ar⁺-laser pumped Kerr-lens mode locked Ti:S oscillator that produces pulses with a time duration of about 100 fs. The repetition rate of the oscillator is 80 MHz and the average power approximately 350 mW (4 nJ/pulse). The group velocity dispersion in the oscillator limits the pulsewidth, but the spectral bandwidth allows for much shorter pulses. The mode-locking is obtained by exploiting the Kerr-effect. The change in the refractive index is proportional to the square of the electric field (the Kerr effect). Due to the self-focusing that follows from this effect the Ti:S crystal can act as a lens. Three adjacent longitudinal laser modes with different frequencies

![Electric field vs Time](image)

**Figure 5.3** Mode-locking can be described by superposition of waves, in this case three modes are superposed and at one time constructive interference takes place and yields a short pulse [Roos, 2001].

and the sum of these are shown in Fig. 5.3. [Roos, 2001]. Fig. 5.4 illustrates Kerr-lens mode locking. Constructive interference occurs at one time and the high-intensity light is focused due to the Kerr-effect. Unfocused continuous light from modes with random phases can be blocked.
The 100 fs pulses from the oscillator are stretched approximately 2500 times. This is done in a grating and lens arrangement shown in Fig. 5.5a. It is a double-pass arrangement which means that the light is going through the system and is then being reflected and passes the optical components a second time on its way back. The low-frequency components emerge first after the reflection back through the system due to a shorter optical path.

In the compressor, see Fig. 5.5b, the opposite happens. The stretched pulse is converted to an ultra-short pulse. It is important that all frequency components experience the same amount of amplification to obtain a short pulse duration.
The stretched, horizontally polarised oscillator pulse is injected into a regenerative Ti:S amplifier, see Fig. 5.2. By switching the transmitted polarisation, using Pockels cells, only ten pulses are allowed to be amplified. This effectively lowers the repetition rate from 80 MHz to 10 Hz.

The final power enhancement is performed in a four-pass Ti:S amplifier crystal to attain 450 mJ. Two high-energy Nd:YAG lasers with frequency-doubled green light pump this amplifier.

At this stage a part of the pulse is sent to a second multi-pass amplifier and a vacuum compressor to obtain pulses of 35 fs and 1.2 J. This part is not shown in Fig. 5.2 because it was not used in the experiment presented herein. The SHG experiments have been performed in the so-called application lab in the basement at the Lund High Power Laser Facility. A reflection from a mirror in the terawatt laser was sent to the application lab.

Finally, the pulse is compressed. After the compressor the high frequency part of the pulse has caught up with the low frequency part, and the pulses are short (~50 fs). The pulse energy is usually not allowed to exceed 100 mJ (2 TW) in order to keep the effects of the non-linear interaction with air small.
Presently, the system has been upgraded so it is possible to use the system for experiments, which demand more than 30 TW. The new system consists of two beams; one of the beams has a maximum energy per pulse of 250 mJ (standard level is 100 mJ) and the other beam has a maximum energy per pulse of 1.2 J. Both beams produce pulses with a duration less than 50 fs.

5.2 Experimental arrangements

The experimental set-up (Fig. 5.6) has been optimised during the project and the final set-up is described here. To avoid laser irradiation damage of the plate (the measurement object) a collimated laser beam was utilised and the energy density was kept at 20 J/m². However, in the beginning of the project some experiments were performed with an additional lens (not shown in the figure) to focus the laser light into the plate. The time duration of the pulses was approximately 100 fs and the wavelength approximately 800 nm, i.e. infrared light.

Pulses with 10 Hz were delivered from the terawatt laser system. The beam size was determined with a circular aperture. The terawatt laser radiation was horizontally polarised. P-polarised (parallel to the incident plane) infrared light (800 nm) was reflected e.g. on a polished gold mirror. Due to nonlinear interaction between the light and the mirror, light that has frequency equal to twice the input frequency was created. Two photons of the infrared light at 800 nm were converted to one photon with the wavelength 400 nm, blue light. A pellin-broca prism, i.e. a wavelength-separating prism, separated the two wavelengths almost completely. A mirror (both silver and gold have been used) reflected the blue light (400 nm) and a bandpass filter (Schott BG 38, i.e. a blue green optical glass-filter) transmitted only this wavelength. A monochromator (Bausch & Lomb) was used to remove all the infrared light. The blue light was detected by a photomultiplier (type 9558, cathode material S20, i.e. Na-K-Sb-Cs) connected to an oscilloscope (Tektronix TDS 540). The beam path for

![Figure 5.6 Experimental set-up. HV denotes a high voltage.](image)

the blue light was aligned using a frequency-doubling crystal. The crystal, placed between the first and the second mirror, generated an easily detectable blue light beam. When the
light from the crystal hit the photomultiplier, the crystal was removed. After this procedure the blue light that was generated from the plate also hit the photomultiplier. The crystal was removed during the measurements. To ascertain that no blue light came from the other optical components the filter, that removed the blue light from the laser system, was placed after the mirror. In that case there was not any SH signal detected on the oscilloscope.

A photodiode was inserted into the experimental arrangement to measure the laser intensity fluctuations. The photodiode and the photomultiplier were connected to the oscilloscope with shielded coaxial cables. The oscilloscope was connected to a computer with a GPIB-cable, i.e. general purpose interface bus (GPIB). Labview, a data measurement system installed on the computer, collected the signals from the oscilloscope.

Labview can for example be programmed to measure the whole curve from the oscilloscope, but it is also possible for Labview to make calculations when collecting the data. Both options have been tested.

A high voltage electrode was placed close to the plate (grounded) and a net surface charge was induced on the plate due to an applied high-voltage on the anode. One of Maxwell’s postulates gives the surface charge as the divergence of the net electric field. If the electric field lines are parallel then the surface charge is given by

\[ \varepsilon_0 E = \sigma \]  

(5.1)

In the experiment which was shown in Fig. 5.6 the electric field lines were not totally parallel, see chapter 3, and hence one has to apply a numerical method to calculate the surface charge more accurately. The goal has been to detect a change in the blue light when the surface charge was varied. The corresponding applied electric field must not exceed the breakdown voltage in air, \(~3\text{MV/m}\). Therefore the detection limit for the electric field induced SHG was difficult to obtain.

### 5.3 Results and Discussion

A typical incident laser pulse (see Fig. 5.6) to a gold mirror is shown in Fig. 5.7 (a) measured by a photodiode. The generated SHG signal captured by a photo multiplier tube is seen in Fig. 5.7 (b). For the SHG signal the area of the graph in (b) was taken as \(I_{2\omega}\), and the height of the graph in (a) as \(I_\omega\). The oscilloscope used a sample rate of 250 MS/s per channel.
In Fig. 5.7a the height of the graph corresponding to \( I_{\omega} \) has been taken as the difference between point A and B. It has been checked with some optical density filters that the height of the graph is proportional to \( I_{\omega} \). Since the use of a focusing laser light deteriorated the mirror a collimated laser beam was utilised. The quotient between the standard deviation and the mean value is a useful measure for the fluctuations. The fundamental wave intensity \( I_{\omega} \) showed small fluctuations. The fluctuations \( F \), defined as two times the standard deviation divided by the mean, for \( I_{\omega} \) and \( I_{2\omega} \) were typically 7.6 % and 15.4 % respectively for a gold mirror with a \( \lambda/10 \) surface roughness. An aluminium plate that was only polished down to a surface roughness of some microns showed \( I_{2\omega} \) fluctuations of 50.8 %.
Fig. 5.8 shows a plot (log-log scale) of the SHG signal, $I_{2\omega}$, from a gold mirror as a function of the fundamental field intensity, $I_{\omega}$. By attenuating the laser power it was possible to obtain various magnitudes for the incident field intensities. The correlation coefficient $C$ defined as

$$C = \frac{\langle I_{\omega}I_{2\omega} \rangle - \langle I_{\omega} \rangle \langle I_{2\omega} \rangle}{\sigma(I_{\omega}) \cdot \sigma(I_{2\omega})}$$

(5.2)

where $\sigma$ is the standard deviation and $\langle \rangle$ is taken as the expectation value. The correlation coefficient of shot-to-shot fluctuations between $I_{2\omega}$ and $I_{\omega}$ was calculated for one optical density filter to 0.06 and the slope for the mean values of each $I_{\omega}$ to 1.7, i.e. approximately 2. The low value of the correlation coefficient means that there is no strong connection between fundamental beam and the SH light for a constant $I_{\omega}$. Obviously, if $I_{\omega}$ is changed due to the use of different optical density filters, and the correlation coefficient is calculated for the mean values of the three measurements in Fig. 5.8, the value of $C$ is close to one. The presence of shot-to-shot fluctuations could be easily inferred from the figure and this determined the experimental accuracy.

![Figure 5.8](image)

**Figure 5.8** Correlation between SHG signal and incident laser beam.

In Fig. 5.9 SHG experiments on a silver mirror have been carried out by alternating a zero electric field and a field of 0.12 MV/m (corresponds to a net surface charge density, $\sigma$, of 1.1 $\mu$C/m$^2$). The time elapsed between the experiments was less than a minute. Any change of SHG signal was difficult to reproduce.
Several experiments have been carried out to establish how the relationship between $I_o$ and $I_{2\omega}$ might change due to increased surface charge at the mirror. The conclusion of the SHG experiments was that the increased surface charge did not give any reproducible change in $I_{2\omega}$. From these experiments, it has also been verified that the surface roughness of the mirror strongly affected the fluctuations of $I_{2\omega}$. Considering shot-to-shot ($I_o$ approximately constant), it has been shown that there was a bad correlation between $I_o$ and $I_{2\omega}$ and this was the major difficulty. Further the relationship between $I_{2\omega}$ and $I_o$ fulfilled a square dependence, see Eq. (4.5).

During the project we have improved the experimental set-up and the last one is presented in chapter 5.2. In the beginning we focused the laser light by a lens and the beam waist reached a minimum at the aluminium plate. Several gold mirrors were also used to direct the laser light to the plate. Two dikroic mirrors were placed after the pellin-broca prism, see Fig. 5.6 in chapter 5.2, to completely remove scattered infrared light in the direction of the frequency doubled beam. The second harmonic light was rather weak and both blue
and infrared light were detected by the photomultiplier. The laser light intensity was initially too large and deteriorated the aluminium plate.

The focusing lens was removed and collimated laser light was used. This enhanced significantly the detected blue light. The metal plate was not deteriorated any longer. Every gold mirror created SH light and we had to change room for the experiment to remove the gold mirrors in the set-up. The dikroic mirrors were also removed. A gold mirror replaced the aluminium plate. We found that the laser contained quite a lot of blue light. It was comparable to the SH light that was created from one gold mirror. By using the experimental set-up in Fig. 5.6, we detected only blue SH light that was created at the gold mirror target. Second harmonic light was easily detectable and all infrared light had been removed.

The conclusion of this experiment was that the detection sensitivity was limited by the laser intensity fluctuations. There were fluctuations both in the fundamental and the SH signal. The fluctuations made it impossible to get any valuable information and a much better correlation from shot-to-shot between the fundamental and SH pulses would have been desirable. In SHG experiments there will often be at least 1-3 % fluctuations that cannot be easily removed, e.g. if the time duration of the pulse is changed from 50 fs to 52 fs. The choice of sample material is crucial for the outcome of the experiment. The sample was changed during the experiments from a solid aluminium plate to a thin film (gold mirror) since it reduced the fluctuations. However, the goal of the project aimed to study a solid metal by applying an external electric field strength less than 3 MV/m. The constants a, b and c are unknown in eq. 4.39. The relative change, \( \Delta l / l \), depends on these constants. Thus it has been difficult to predict the outcome of the experiment.

In [Corn et al., 1984] the SH signal on a silver-mica capacitor was studied as a function of applied electric field. They showed that the required electric field strength range 0-100 MV/m has a dependence of SHG. By studying a silver-mica interface it might be possible to detect a change in the SHG (created from plasmon surface polaritons (PSPs) and thus not a pure surface charge effect) if the electric field is less and changed less than 3 MV/m [Corn et al., 1984]. The SH fluctuations F of 15 % should be compared to the expected ~11 % (maximum) change of the signal.

What would happen if the silver-mica (a polymeric material can substitute mica) experiment had been performed with an oscillator with a repetition rate of 80 MHz? The optimal data acquisition rate ought to be studied since it may depend on the sample heating and a signal-to-noise ratio (SNR) of \(~10-100\) is realistic [Dadap et al., 1995]. The SNR depends on the quantum efficiency of the photomultiplier, e.g. 20\%, and the dark current, e.g. 5000 counts per second. The SNR depends also on the input of signal light to the photomultiplier. By using a fluence of the fundamental beam of 3 J/m\(^2\) the SHG count rate could be \(3 \cdot 10^6\) photons per second according to this reference. A frequency of some kHz can modulate an applied external electric field strength of 100 MV/m and can be achieved by using an electron tube. The anode of the electron tube should be connected to a resistance that is connected to a high voltage. The potential between the anode and the resistance is given to the anode of the silver-mica capacitor. The cathode of the electron tube and the capacitor are grounded. One has to search for the modulated frequency in the signal. It can probably not be found by using a lock-in based computer analysis due to the fluctuations of the SH light. I believe that the only way to succeed with this silver-mica
capacitor experiment is to analyse and quantify every single parameter that can affect the signal strength and thus disturb the measurement.
Outlook

Frequency doubling has turned out to be difficult to use as a method for quantifying surface charge on a solid metal. The project will in the continuing work focus on characterisation of insulator materials with different optical methods. Fluorescence, which can be performed remotely, has shown to be a promising method. A mobile Light Detection And Ranging (LIDAR) system [Weibring et al., 2003] can in future work be used for multi-wavelength excitation and detection in fluorescence imaging of insulators. This can be done remotely.

The mobile Light Detection And Ranging (Lidar) system, housed in a Volvo F610 truck, is based on all solid-state laser technology, and can conveniently be used for multi-wavelength excitation and detection in fluorescence imaging, gas concentration- and flux measurements. The light source is a rapidly tuneable Nd:YAG pumped optical parametric oscillator (OPO) combined with a doubling- and difference frequency generating system that makes it possible to produce narrow band light in the wide range from UV to mid IR, i.e., 220-1880 nm and 2900-4300 nm with maximal pulse energies of 100 mJ and 20 mJ, respectively. In the truck, a receiving and detecting system is integrated as well as computers for control, surveillance and evaluation. The system is self-contained, with a diesel powered electrical generator, making it well-suited for field measurements. The dome and light receiving unit in the Lidar truck is shown in Fig. 1. Light from the laser, in this case 355 nm, hits the folding mirror and is directed to the target under consideration. Fluorescence light from the target is emitted in all directions and a part of this is sent back to the folding mirror and collected by the detection system.

![Figure 1 Dome and light receiving unit.](image-url)
A successful experiment for remote detection of biological contamination on high-voltage outdoor insulators has been performed in co-operation with the department of high voltage engineering at Chalmers University of Technology [Demfalk et al., 2003], see Fig. 2. Measurements were made outdoors on a number of clean, as well as, biologically contaminated insulators. Several types of biological contamination were included, as five of the studied insulators had become covered when installed in Sweden, and another three had been contaminated by fungal growth in laboratory. Additional work has been performed. Fungal growth has been studied separately, see Fig. 3, in the laboratory and the signature for fungus has been distinguished from the fluorescence peak of chlorophyll, which is significant, for regions covered by algae. A successful multivariate analysis has also been performed. A model was based on fungus and validated data were classified correctly. This means that it is possible to measure the fluorescence from an insulator and obtain the information if the surface is contaminated with fungi or not. This information can be obtained even if it sometimes is impossible to see any fungi on the insulator.

![Figure 2](image_url)  
*Figure 2 Typical spectra obtained from a region with green growth on one of the insulators.*
Figure 3 Two typical spectra (not calibrated). A clean surface and a surface coated by fungus. The laser excitation wavelength was 337 nm.

In the future multi-wavelength excitation studies will be performed on contaminated insulators and the spectral information will be analysed. A multivariate analysis will also be performed. The goal of the multivariate analysis techniques is to predict output variables (called response variables) by input variables to the process (called explanatory variables) using as few independent characteristics as possible. One technique, which has been extensively used to reduce the number of explanatory variables, is Principal Component Analysis (PCA).

Remote break-down spectroscopy (LIBS) has been investigated and several remote LIBS spectra on metals have been obtained. LIBS spectra of salt on insulators will be measured and the technique will be studied for possible contaminant ablation.
Comments on the papers

Paper I: A. Larsson and M. Bengtsson
"The Sequence of Events in Mid-Gap Laser-Triggered Electrical Breakdown in Liquid Dielectrics"
I carried out the field calculations and Anders Larsson prepared the manuscript.

Paper II: A.D. Dernfalk et al.
"Laser-induced fluorescence spectroscopy for detection of biological contaminations on composite insulators"
I performed the experiments together with Andreas Dernfalk and the molecular spectroscopy group. Mikael Sjöholm and I wrote the section about the experimental set-up. Rasmus Grönlund and Andreas Dernfalk were also involved in the writing process.
Acknowledgements

I wish to thank my supervisor Stefan Kröll for good guidance, rational explanations, nice conversations, great patience and never-ending enthusiasm. Many thanks to my vice-supervisor Anders Larsson for clear-sighted guidance, stimulating and informative discussions, and very good support. Special thanks to my vice-supervisor Sune Svanberg for a great collaboration and all your professionalism.

Thanks to Anders Persson for all your help in the laboratory. It is very appreciated.

Thanks to Hans Edner and Petter Weibring for the fruitful experiments with the Lidar system and the invaluable support. Thanks to the rest of the molecular spectroscopy group for your productive help and experimental skill. Thanks to the medical group for introducing me to the OMA system.

Thanks to Anders Sjögren for all information about the terawatt laser system.

My sincere gratitude to Bertil Hermansson and Åke Bergquist for taking care of my never-ending computer problems and for the electric devices. Special thanks to Göran Werner for nice conversations and all your help in the workshop. Thanks to Lennart Nilsson, Lars Engström and Ulf Gustafsson for nice conversations. Thanks Britt-Marie, Laila and Marie for your help.

Thanks to all people at the division of Atomic Physics and especially the photon echo group for all interesting chats.

Thanks Mose Akyuz for your help to solve my finite element problems.

Thanks Andreas Dernfalk and Stanislaw Gubanski at Chalmers University of Technology for lending us your electrical insulators and materials. Special thanks for your contribution to the conference paper.

Thanks Tomas Metz for your excellent introduction and German precision to the second harmonic generation. Thanks Joachim Walewski for your statistical program and support. Thanks Axel Franke for your support concerning Unix and Ace.

A special thanks to Michael Harbst for fruitful discussions about quantum mechanics.

Thanks Zhang Zhiguo for fruitful discussions about everything.

This work was supported by ELFORSK.
References

Ace version 3.0, ABB common platform for field analysis and simulation (Västerås, Sweden: ABB Corporate Research Centre)

ANSYS (Release 5.7), ANSYS Inc., Southpointe 275 Technology Drive Canonsburg, PA 15317 USA


Crichton G.C. and McAllister I.W., "On the measurement of surface charge density using a field probe", Nordic Insulation Symposium, Västerås, June 15, 1992


Demfalk A.D., Bengtsson M., Grönlund R., Sjöholm M., Weibring P., Edner H., Gubanski S.M., Kröll S. and Svanberg S. "Lidar fluorescence measurements of algal growth on electrical insulators" have also been accepted as an oral presentation of the CLEO/Europe-EQEC Conference. The meeting will be held 23-27 June 2003 at the Laser 2003 World of Photonics in the International Congress Centre (ICM Munich, Germany)

ELFA (company), private communication


Zhu Y., Takada T., Inoue Y., and Tu D., "Dynamic Observation of Needle-plane Surface Discharge using the Electro-optical Pockels Effect", IEEE Transactions on Dielectrics and Electrical Insulation, 3 (1996) 460
Paper I
The Sequence of Events in Mid-Gap Laser-Triggered 
Electrical Breakdown in Liquid Dielectrics

Anders Larsson and Magnus Bengtsson
Division of Atomic Physics
Lund Institute of Technology, P.O. Box 118
SE-22100 Lund, Sweden

ABSTRACT
Recent experiments on mid-gap laser-triggered electrical breakdown in liquid dielectrics have been scrutinised. The sequence of events that leads to the disruptive discharge is described and accompanied by electric field calculations.

1. INTRODUCTION

PRE-BREAKDOWN and breakdown phenomena in liquid dielectrics have been studied extensively during the last decades. New diagnostic tools and techniques have rendered deeper understanding of these phenomena [1]. Recently, an elaborate technique presented by Larsson et al [2] combines laser-triggering and the laser shadow method. This technique, integrated with pre-breakdown current measurements, was used in experimental research on mid-gap laser-triggered dielectric breakdown in transformer oil, where the physical mechanisms and the timing sequence of this dielectric breakdown were revealed [2, 3]. The elicited information gives, as presented in this paper, the sequence of events in the breakdown process together with electric field calculations [4, 5].

Typical experimental conditions were as follows: The electrode gap, was placed in a cell filled with Nytro 10x transformer oil, consisted of two hemispherically-tipped brass electrodes with a tip radius of 10 mm. The adjustable electrode distance ranged between 4 and 5 mm. The oil was stressed with an electric field strength of 12 MV/m which was achieved by applying either 50 or 55 kV depending on the electrode distance. The self-breakdown electric field strength of the oil was 22.5 MV/m. A laser pulse (wavelength, 1064 nm; pulse duration, 8 ns) was focused in the middle of the electrode gap, and a pulse energy of 30 mJ was required for plasma formation ('breakdown threshold'). In our experiments, we varied the pulse energy between the breakdown threshold and 100 mJ. The experiments were performed at room temperature and at ambient pressure. The expansion of the gas-filled cavity formed by the laser-initiated plasma was monitored by the shadowing technique and the pre-breakdown current was measured simultaneously.

The filamentary structures observed in electrical discharges in liquid dielectrics are traditionally called 'streamers'. This is also the case in this paper. However, this term may give confusing association when compared to its namesake in gas discharges. Recent research points out numerous similarities between the discharge channel propagation in liquids and leader propagation in gases [6].

2. SEQUENCE OF EVENTS IN MID-GAP LASER-TRIGGERED DIELECTRIC BREAKDOWN

The sequence of events in mid-gap laser-triggered electrical breakdown in liquid dielectrics is depicted in Figure 1. The focused laser pulse creates a plasma in the electrode gap (step 1 in Figure 1). If the laser pulse energy is close to the breakdown threshold, the initial plasma appears as a spot. If the energy is increased, the plasma will be larger and will assume a cigar-like shape. The lifetime of the plasma is less than 100 ns, and it causes a high-pressure gas-filled cavity to form (step 2). The high pressure expands the cavity. The dynamics of an expanding cavity in a liquid can be expressed by a simple equation assuming that the liquid consists of an infinite mass of homogenous and incompressible fluid acted upon by no forces. With these assumptions, the relationship between the expansion time \( t \) of the cavity and the cavity radius \( R \) is [7]

\[
t = \frac{R_{\text{max}}}{2 \rho_{\text{p}}} \int_0^{R_{\text{max}}} \sqrt{\frac{\beta^3}{1 - \beta^3}} v_0 d\beta
\]  

where \( R_{\text{max}} \) is the maximum radius of the cavity, \( \rho \) is the mass density of the liquid and \( p_{\text{a}} \) the ambient pressure. The integral of (1) lacks a simple analytical solution, but can easily be calculated numerically. Figure 2 shows a calculation of the expansion and collapse of a cavity with \( R_{\text{max}} = 1.4 \text{ mm} \) at an ambient pressure of \( p_{\text{a}} = 1013 \text{ hPa} \) and a liquid density of \( \rho = 876 \text{ kg/m}^3 \). Strictly speaking, (1) is only valid for the expansion phase. However, it can easily be shown that the expansion and collapse are sym-
metrical in time around the instant of maximum size of the cavity. In the figure, measured data of expansion and collapse of a cavity are also included, for the case without applied electric field. The agreement is satisfying.

Several μs after the plasma initiation, partial discharges (PD) are observed to occur within the cavity, and these occur before the liquid starts to break down dielectrically (step 3). These PD can be explained by the following analysis. Calculations of the development of the electric field distribution in the gap with an expanding and perfectly dielectric cavity are shown in Figure 3. In the figure, the electric field strength is represented by the dimensionless field enhancement factor, which is defined as the quotient between the local field strength and the average field strength, that is, \( E(x)/\langle U/d \rangle \). The relative permittivity of the gas \( (\varepsilon_{\text{gas}} = 1) \) is less than the one for oil \( (\varepsilon_{\text{oil}} = 2.2) \) which leads to a field enhancement in the cavity, as shown in the figure. The dielectric strength of the oil is significantly greater than the dielectric strength of the gas in the cavity, which is a gas mixture of various hydrocarbons. Hence, the field inside the cavity as shown in Figure 3 will cause PD within the cavity, and these PD will deposit charge on the cavity walls facing the electrodes and thus limit the field strength within the cavity. Effectively, the charge redistribution within the cavity will limit the cavity electric field strength to be less than the breakdown field strength of the gas in the cavity. Figure 4 shows calculations of the development of the electric field distribution for a cavity where the cavity is assumed to be a perfect conductor (zero electric field within the cavity).

About 70 μs after the plasma initiation, positive streamers are observed to launch from the cavity towards the
Since the voltage is constant across the electrode gap, the limitation of the field strength in the cavity will give rise to increased field strength in the liquid, as evident from the calculations presented in Figure 4. This field enhancement will eventually trigger positive streamers that are launched from the cavity towards the cathode, since positive streamers have lower inception field strength than negative ones [1, 6]. The development of the field enhancement factor in the gap for a perfectly conducting cavity is shown in Figure 5. Note that the maximum field strength is at the cavity as evident from Figure 4, but that the maximum field strength is located at the electrodes for the case without any cavity.

When the positive streamers have bridged the gap between the cavity and the cathode, a charge transfer occurs between them. Consequently, the field strength in the gap between the cavity and the anode is further enhanced, resulting in negative streamers to be launched towards the anode (step 5). When a negative streamer has bridged the gap between the cavity and the anode, the disruptive discharge is imminent (step 6). The propagation of the positive streamers and the spark formation (step 4 and step 6) were not time-resolved (sub-microsecond processes), but the propagation of the negative streamers took a few microseconds (propagation speed \( \sim 250 \text{ m/s} \)).
Figure 6. Size of a perfectly conducting cavity necessary for liquid breakdown vs applied voltage.

A certain combination of cavity radius and applied voltage will give rise to the breakdown field strength of the oil in the gap (22.5 MV/m), for a certain condition for the electrical properties of the cavity. Figure 6 shows this combination for a perfectly conducting cavity, that is, for a given applied voltage, what is the radius of the cavity that give rise to a maximum field strength in the gap that is equal to the breakdown field strength of the oil? The breakdown voltage according to Figure 6 is too low since a typical example of breakdown conditions from the experimental studies is an applied voltage of 50 kV with a cavity radius of 1 mm for a 4 mm gap. Thus, the assumption of a perfectly conducting cavity is not appropriate. Figure 7 gives the calculated breakdown voltage for a 1 mm cavity in a 4 mm gap as a function of the voltage gradient inside the cavity. From this figure, it is suggested that this voltage gradient is about 7 MV/m.

3 CONCLUSION

We believe that the process, as illustrated in Figure 1, is qualitatively valid for mid-gap laser-triggered electrical breakdown in general where the expanding cavity has a lower dielectric strength than the surrounding medium. A gas-filled high-pressure cavity is formed from the laser-induced plasma in the focal region of the laser radiation. This cavity expands and distorts the electric field distribution in the electrode gap. Since the gas in the cavity has a lower dielectric strength compared to the surrounding liquid, PD will occur in the cavity before the liquid starts to break down dielectrically. These PD create charge redistribution within the cavity and, in effect, limit the within-cavity electric field strength to be less than the breakdown field strength of the gas in the cavity. The field limitation in the cavity gives a field enhancement in the liquid. When the field enhancement is sufficient, dielectric breakdown of the liquid is induced.

ACKNOWLEDGMENTS

The fruitful discussions with Professor Stefan Kröll are greatly acknowledged. This work has been performed within the frame of the research programme ELIS (High Performance Outdoor Electrical Insulation) supported by the Swedish Foundation for Strategic Research.

REFERENCES


Manuscript received on 8 August 2001, in final form 9 January 2002.
Laser-induced fluorescence spectroscopy for detection of biological contamination on composite insulators

A.A. Dernfalk¹, M. Bengtsson², R. Grönlund³, M. Sjöholm², P. Weibring², H. Edner², S.M. Gubanski¹, S. Kröll² and S. Svanberg²
¹Chalmers University of Technology, Gothenburg, Sweden
²Lund Institute of Technology, Lund, Sweden

Abstract: A new technique for remote detection of biological contamination on high-voltage outdoor insulators has been investigated. The technique, which is based on laser-induced fluorescence (LIF) spectroscopy, has been applied to study surfaces of real silicon rubber insulators from a distance of approximately 60 m. Measurements were performed outdoors on a number of clean, as well as, biologically contaminated insulators. Several types of biological contamination were included, as five of the studied insulators had become covered when installed in Sweden, and another three had been contaminated by fungal growth in laboratory. Fluorescence spectra obtained from the surfaces of the described insulators are presented and the applicability of the technique is discussed and compared with photographic methods.

1. Introduction

Reports on biological growth on outdoor insulators reveal that microbiological colonization of ceramic as well as composite insulators can take place in all parts of the world [1-14]. Since the impact of biological growth on insulator performance is not fully understood, it makes utility engineers concerned [14, 15]. The fact that microorganisms grow on composite insulators is of special concern. Silicone rubbers are known to exhibit high resistance to biological degradation. One of the reasons is that the material consists of both organic as well as inorganic components, and that microorganisms like fungi cannot digest the inorganic parts [16]. However, it is believed that in service, composite insulators are mostly attacked by fungi [8, 17].

There is a number of reports on biological contamination of composite insulators [1, 2, 4-13, 15, 18], about half of them from tropical climate [2, 6-8, 11, 15, 18]. Different types of silicon rubber (SIR) insulators, epoxies and blends of silicone and ethylene-propylene-diene monomer rubber (EPDM) have been found to support growth of bacteria, algae, fungi and lichen. However, so far there is not much information published on the growth on EPDM insulators.

This study is a step in a process of investigating the influence of biological growth on insulator performance. More specifically, the aim is to develop a method that can be used to remotely detect and measure/characterize growth and growth distributions.

At close distance, presence of for instance algae on an insulator surface can be detected through visual inspection. However, in order to get a measure of its “severity”, more refined techniques have to be used. An example of such a diagnostic technique is photography followed by digital image analysis, which has been proposed by one of the authors [19, 20]. Using this technique, it is possible to get measures of area covered by the growth and its distribution. However, it is difficult to differentiate between regions covered by microorganisms and regions covered by other contamination.

The technique investigated in this study, laser-induced fluorescence (LIF) spectroscopy, has e.g. been utilized for imaging of historical monuments [21] and previously been suggested for insulator diagnostics [22]. In the latter report, the authors presented fluorescence spectra obtained in laboratory from different SIR and EPDM materials and from SIR surfaces where its hydrophobic properties had been reduced through water immersion. In the present study, LIF spectra have been obtained from real insulators partly covered by biological growth using a mobile measuring system. To simulate a real situation, i.e. studying insulators installed in a HV-system, measurements were performed outdoors at a distance of about 60 m.

2. Experimental set-up

The used mobile Light Detection And Ranging (LIDAR) system, housed in a Volvo F610 truck, is based on all solid-state laser technology, and can be used for multi-wavelength excitation and detection in fluorescence imaging, gas concentration- and flux measurements [23]. The system is self-contained, with a diesel powered electrical generator, making it well-suited for field measurements.

In the present experiment, only a part of the laser system, a frequency tripled Nd:YAG laser (Quanta-Ray) was used. This laser delivers pulses with a duration of 4-5 ns at a repetition rate of 20 Hz and a wavelength of 355 nm. The laser beam is passed through an aperture to produce a better mode, rendering a beam diameter of 5 mm and a pulse energy of around 25 mJ. After a totally reflecting degree prism, the laser beam is expanded in a vertically mounted Galilean quartz telescope to 3 cm in diameter. This gives sufficiently low laser intensity
on the computer controlled output folding mirror mounted in a dome, Figure 1, not to damage the mirror. The transmitted excitation light is directed towards a remote target, which in the present work was a number of insulators.

From a control computer, the dome can be rotated 360 degrees (resolution of 0.0035 degrees) with a stepper motor in the azimuthal angle and the folding mirror can be tilted from −10 to +55 degrees (resolution of 0.011 degrees) in the polar angle. At the target located 60 m from the laser source, this corresponds to a resolution of 3.7 mm and 12 mm respectively. However, if using this resolution the time for acquiring spectra from the complete insulator surface would be very long. Therefore, a beam diameter of ~20 mm and a separation between adjacent measurement points of ~20 mm at the insulator were used. The change of position of the laser beam at the target due to mechanical instability of the whole truck was estimated to 5 mm.

![Figure 1. Dome and light receiving unit.](image)

The remotely emitted fluorescence light is received by a 40 cm diameter Newtonian telescope (f=1 m), coaxially mounted with the Galilean telescope. By using a cut-off filter in front of the focus of the telescope, the excitation light is discriminated. An optical fibre, 600 μm core diameter and numerical aperture 0.22, guides the fluorescence light to a compact time-gated Optical Multi-channel Analyser system (OMA) designed for medical use. It consists of a spectrometer and a charge coupled device (CCD) camera. In the OMA system a beamsplitter and a cut-off filter selects only wavelengths longer than approximately 385 nm, effectively suppressing the dominating excitation light which is still of appreciable intensity.

A crossed Czerny-Turner spectrograph diffracts the fluorescence light to the image intensified CCD. The thermo-electrically cooled detector has a CCD array with 1024x128 pixels where the 128 pixels are binned. The resulting resolution for the OMA system, set by the slit width (100 μm), is 2.2 nm up to 805 nm. Ambient daylight is suppressed by using a time-gate of 100 ns, positioned in time by a properly delayed trigger signal from the laser. Spectra from the OMA system are acquired by a data collection computer that stores the spectrum together with information about the measurement coordinates.

**Studied insulators**

The present study included eight insulators partly covered by biological contamination and one clean reference insulator. The insulators have been divided into three categories, A-C, according to their design. Type A and B are of line suspension type, while insulators of type C are of apparatus type and have a hollow core. All insulators have sheds and sheath of SIR. However, the formulation differs between the three categories.

The three insulators A have been installed in a 10 kV distribution system in Sweden for approximately 12 years, where they have become covered by a rather thick green growth. However, at the time of the measurement, the growth had turned brown/grey, as the insulators had been kept in laboratory for 6 months. Three of the four insulators B had been exposed to fungal spores and kept in a climate chamber for approximately two years [19]. This treatment lead to colonisation of the insulator surfaces by fungi in spot-like manner. The insulators of the third type, C, have been installed in a test station 30 km south of Gothenburg, Sweden, since 1995. They had become covered by a greenish growth on the surfaces shaded from direct sunlight.

During the measurements, the insulators were placed about 60 m away from the truck. To get a better view of the growth, which mainly were located on the upper side of the sheds, the insulators had to be turned up-side-down, tilting away from the point of observation. The insulators C were supported by a wooden structure, and tilted an angle of 30 degrees. Type A and B were fixed using horizontal metal rods and tilted 60 and 50 degrees, respectively. As a safety precaution, a board covered with black paper was put behind the insulators, hindering the laser beam to propagate further.

**3. Results**

LIF spectra were obtained from characteristic points on the insulators. A typical region with thick, green growth located on a shed of one of insulators C gave the spectrum shown in Figure 2. As seen, there is a large peak located around 680-690 nm, corresponding to the well-known fluorescence peak of chlorophyll a at 685 nm [23]. Further, the shoulder on the right probably corresponds to another smaller fluorescence peak of chlorophyll at 735 nm.

Comparison of the obtained spectra gave that the fluorescence peak of chlorophyll around 685 nm is significant for the region covered by growth. The parts
of the insulator, which are not covered by algae, show a low and decaying fluorescence above 600 nm. The spectra obtained from measurements on insulators of type B were more difficult to interpret due to several reasons. First, these insulators were covered by fungi, which do not utilize photosynthesis, and thus not contain any chlorophyll. Second, the growth was more or less evenly distributed over the surface in the form of small isolated spots. As the spatial resolution of the measurements was in the order of 20 mm, spectra could not be obtained from regions completely without growth or regions totally covered by growth. However, comparison of typical spectra from a clean reference insulator and from insulators with growth shows that the total fluorescence of the contaminated insulator is lower, compared to the clean insulator, Figure 3. Further, if the amplitudes are normalized, it is revealed that the contaminated insulator has a slightly higher relative fluorescence in the wavelength band 520-600 nm.

Measurements on insulators A revealed that these had a low fluorescence compared to the other insulators studied. As the insulators of type B, the intensity of fluoresced light was much lower from regions covered by growth, than from clean regions, Figure 4. Further, regions covered by growth showed an increased fluorescence in an interval around 685 nm. However, the difference was very small compared to the insulators of type C. The reason for the low response was probably that the algae had died, due to desiccation and lack of sun radiation, when stored in laboratory.

Figure 2. Typical spectra obtained from a region with green growth on one of the insulators C.

Figure 3. Spectra from clean reference insulator B (dashed) and an insulator B with growth (solid).

In order to improve sensitivity and avoid incorrect classification of regions, the obtained spectral data were processed. Average intensities were calculated...
for two different wavelength bands, $B_1$ and $B_2$, and the intensity ratio $I(B_2)/I(B_1)$ were formed. The main advantage of these types of dimensionless ratios is that they eliminate the influence of geometrical artifacts, i.e. intensity differences due to different directions of studied surfaces [21]. An example is shown in Figure 6. Image a) shows a photograph taken from close distance, b) shows the average fluorescence intensity in the 410-500 nm band, while c) shows the average intensity in the 670-690 nm band. The ratio of the light fluoresced in the two bands is shown in d), where the dark areas are well corresponding to the regions covered by green growth.

![Figure 6](image-url)

Figure 6. a) Photograph of an insulator of type C, b) average intensity in 410-500 nm band, c) in 670-690 nm band and d) ratio of intensities in 670-699 nm band and 410-500 nm band.

4. Discussion and Conclusions

In comparison with photography, LIF can be used to get a higher sensitivity, especially when there is a large difference in fluorescence between insulator material and contamination. Further, as collection of fluorescence light only is performed under a short period of time, ~100 ns, measurements are not affected by sun radiation.

Comparison of the obtained spectra shows that the fluorescence peak of chlorophyll around 685 nm is significant for the regions covered by algae, while regions with no algae showed low fluorescence in the same wavelength interval. Presence of growth without chlorophyll, i.e. fungi and bacteria, did only cause small changes in the shape of the spectra, compared to the ones of clean surfaces. The most apparent difference here was an amplitude reduction. However, it is not unlikely that a good detection sensitivity of growth without chlorophyll can be obtained using other excitation wavelengths and more extensive data analysis. In conclusion, it has been shown that remote LIF measurements can be used to detect and measure distribution of biological contamination on composite insulators.

5. References


Author address: Andreas Dernfalk, Department of Electrical Engineering, Chalmers University of Technology, 412 96 Goteborg, Sweden. Email: andreas.dernfalk@elektro.chalmers.se