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Electric Field Vector Measurements Via Nanosecond Electric Field Induced Second Harmonic Generation

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This letter describes the development of an electric field vector measurement technique compatible with conventional nanosecond laser sources. The optical method is based on the principle of second harmonic generation in non-polar gases and is expected to be applicable at moderate to above atmospheric pressures. Nanosecond probe beams of two different pulse widths (FWHM) are evaluated: a 16 ns pulse and a shorter 3 ns pulse, both at an operating wavelength of 1064 nm. The shorter pulse is realized via a Pockels-cell-based pulse slicer which time-extracts a portion of the energy from the original 16 ns pulse. Both pulses are tested in a time-varying Laplacian field with a peak field strength of 23 kV/cm and are found to be capable of generating good signals. Comparable field sensitivities of 1.5 kV/cm and 0.6 kV/cm are obtained for the 16 ns and 3 ns pulse respectively. However, an evident benefit of using a shorter pulse is the enhanced temporal resolution since the time resolution of the measurement is effectively determined by the duration of the probe laser. The lower energy requirements and the shorter pulse width also imply a reduced probability of optical breakdown and laser-plasma interaction. Using the 3 ns pulse, we take advantage of the field vector sensitivity of the signal and demonstrate for the first time, the ability to make simultaneous measurements of both the horizontal and vertical components of the electric field. Combined with the above merits, the versatility and cost effectiveness of the current pulse slicing approach presents a powerful tool for making localized and potentially 1D measurements of electric field using nanosecond laser systems.

An optical method based on the principle of second harmonic generation in gases has been recently re-developed for the purpose of electric field measurements in plasmas [1-2]. Commonly known as Electric Field Induced Second Harmonic (E-FISH) generation [3], this method involves probing a non-polar gas sample with laser light, and quantifying its second harmonic response to an externally applied electric field that is to be measured. This second harmonic signal varies proportionally with the applied field strength squared, and is otherwise absent if no field is imposed. Calibration may be performed in a known electrostatic field so as to obtain absolute field information, for instance in a plasma.

Key advantages of the E-FISH method include its versatility, excellent signal localization in time and space, and relatively straightforward implementation. The versatility of the method is demonstrated by its non-resonant nature - the technique is in theory applicable to any non-polar gas or gas mixture and does not demand a specific probe laser wavelength. The temporal resolution of the measurement is only physically limited by the laser pulse duration since the signal generation is effectively instantaneous, with pulses as short as 50 fs being shown to yield good signals [1]. The relevant spatial scale is given by the confocal beam parameter (or twice the Rayleigh range), provided that the confocal beam parameter is much smaller than the coherence length [1]. Sub-millimeter scale spatial resolution can therefore be easily achieved. Finally, the method only involves a single probe beam and thus circumvents the need for complex and time-consuming alignment procedures. This makes it amenable to sophisticated geometries where optical access is limited or where the overlapping of multiple laser beams may be challenging.

As described below, the E-FISH signal exhibits a quadratic dependence on the probe laser intensity. It is likely for this reason that the first electric field measurements were demonstrated using a fs laser system. Subsequent work has noted that the occurrence of multiphoton ionization at the beam focus limits the probe beam energy and thus second harmonic signal that may be obtained with these fs pulses. However, the high intensity and broad spectral bandwidth of these ultrashort pulses may still prove crucial for performing 1D E-FISH measurements [4] or simultaneous species and electric field measurements using fs coherent anti-stokes raman scattering (CARS) in tandem with E-FISH [5]. Extending the E-FISH capability to include longer probe pulses on the order of 100 ps has met with considerable success. Stronger signals are produced since higher laser energies (several 10's of mJ) may be accessed without the onset of multiphoton ionization while still maintaining sub-ns time resolution [2]. To date, time-resolved electric field measurements using ps E-FISH have been successfully conducted in surface dielectric barrier discharges [6,7] and in a ns tube discharge for tracking the ionization wave development [8].

The foregoing discussion emphasizes the utility of sub-ns excitation for E-FISH measurements but at the same time, raises an interesting question as to whether this diagnostic tool may find application in the ns regime. The main arguments against such a proposition would be the risk of laser-induced avalanche breakdown as the laser energy is increased, as well as the corresponding penalty in temporal resolution. However, it is conceivable that there exists an appropriate pulse duration which may provide sufficient field sensitivity and at a laser energy low enough to avoid optical breakdown. The payoffs are compelling since such a measurement method would readily find favor in numerous applications such as DC or AC discharges where the drop in time resolution may be acceptable. Furthermore, this would pave the way for profiting a wider audience since ps and fs laser systems are less accessible compared with ns laser sources, in part due to their significantly higher costs.

The main objective of the present work is to assess the viability of the E-FISH method with a ns probe. Two different ns pulse durations are tested: the first is a 16 ns (FWHM) pulse from a conventional Nd:YAG laser and the other is a shorter 2.8 ns pulse (FWHM) obtained by extracting part of the original 16 ns pulse using a Pockels-cell-based pulse slicer. Our results generally support the use of the shorter pulse and hence underscores the potential of this pulse-slicing approach as a cost-effective way towards making useful E-FISH measurements with ns laser sources.

The experimental setup consists of 3 parts. The high voltage (HV) pulses and electrode configuration used for conducting the E-FISH measurements, the optical layout (including the Pockels cell) for the pulse-slicing, and the corresponding optical layout for the E-FISH measurements.

A uniform electrostatic field is sustained between two parallel plate electrodes under ambient conditions using a nanosecond pulsed HV generator. An FID model number FPG 12-1NM HV generator delivers positive polarity ~ 4.7 kV amplitude voltage waveforms with a rise time of ~ 8 ns and a FWHM of ~ 30 ns at a repetition rate of 10 Hz. This voltage is applied across two 80 mm long copper electrodes which are separated by a gap of 4 mm shown in Fig. 1a. The applied voltage is calculated via a summation of the incident and reflected waveforms sampled by a calibrated, high bandwidth back current shunt (BCS) installed in the coaxial cable which connects the HV generator to the HV electrode. The BCS represents a ~ 2 cm long insertion in the cable shielding comprising of several low inductance resistors soldered in parallel, and has been used extensively in previous work as both a HV and electric field probe [9].

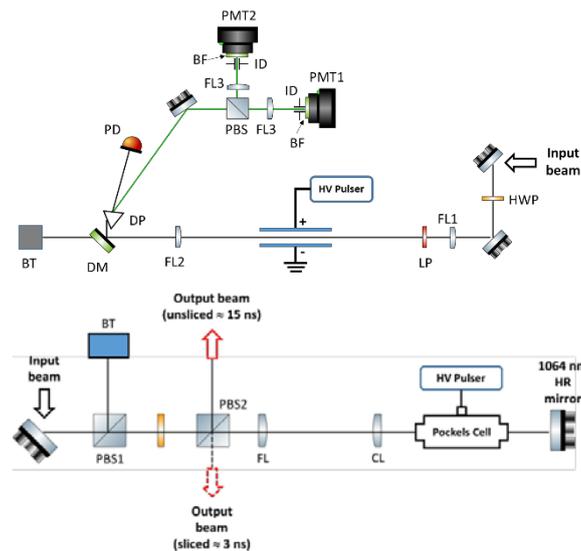


Fig. 1. (a) Experimental schematic for the E-FISH measurements. (b) Optical layout for the pulse slicer. (Legend – BF: bandpass filter, BT: beam trap, CL: plano-concave spherical lens, DM: dichroic mirror, DP: equilateral prism, FL: plano-convex spherical lens, HWP: half-wave waveplate, ID: iris, LP: longpass filter, PBS: polarizing beamsplitter, PD: photodiode, PMT: photomultiplier.)

The ns laser source is a Quanta-Ray Lab-230 Nd:YAG Q-switched system which provides 1064 nm, ~ 16 ns, 200 mJ pulses at a repetition rate of 10 Hz for these experiments. A half-wave plate-polarizer pair (HWP & PBS2) attenuates the laser pulse energy to about 35 mJ, to avoid optical breakdown. Attenuating the energy in this way, rather than directly at the source, helps achieving stabilized output from pulse to pulse, both in terms of energy and time. To reduce the pulse width down to several ns, a pulse-slicing scheme shown in Fig. 1b is used. The optical layout comprises mainly a Pockels cell utilized in a double-pass configuration, achieved with the aid of a highly reflecting 1064 nm mirror. A positive-negative lens pair (FL & CL) with respective focal lengths of 300 mm and -150 mm reduces the initial beam diameter by half, to match the entrance aperture of the Pockels cell.

The Pockels cell (supplied by Leysop Ltd) consists of a UV BBO crystal driven by a second FID HV generator (model number FDS 3-1NM1) capable of delivering up to 3 kV pulses with a nominal pulse width of 2 ns (FWHM). Applying a voltage to the cell induces a birefringence in the BBO crystal (i.e. Pockels effect) and produces a corresponding rotation in the polarization of the transmitted light. Since this response occurs almost instantaneously, supplying a HV pulse to the cell generates a sliced laser pulse, with a temporal profile which closely mirrors that of the input HV waveform [10]. The idealized response of this Pockels cell is approximately 2 ns (FWHM), determined by passing a CW He-Ne laser beam through the cell and measuring the width of the output

pulse. Maximum energy and conversion efficiency from the Pockels cell are attained when half-wave rotation can be effected. Unfortunately, being originally designed for 205 nm operation, this limits the polarization rotation that can be obtained at 1064 nm. Furthermore, the possibility of crystal damage at this off-optimum wavelength also restricts the energy input into the cell. To mitigate this problem, a double-pass arrangement is implemented to effectively double the polarization rotation seen by the 1064 nm light. Compared with a single pass arrangement, this double-pass configuration produces about 50 times more pulse energy, without virtually any detriment to the pulse width. This finding is anticipated to hold true as long as the round trip time of the light through the cell is significantly shorter than the duration of the applied HV pulse. The pulse slicer reduces a 16 ns, 50 mJ input beam to a 2.8 ns, 2 mJ pulse, realizing a fairly modest conversion efficiency of about 4%. This low efficiency is likely to be a consequence of the off-optimum design of the Pockels cell, resulting in losses from the anti-reflective coatings on the crystal surfaces as well as absorption within the crystal. The extinction ratio of the Pockels cell is measured to be less than 1%. The optical layout given in Fig. 1b provides a convenient means of generating a 10 mm diameter, vertically polarized beam of differing pulse duration on either side of PBS2.

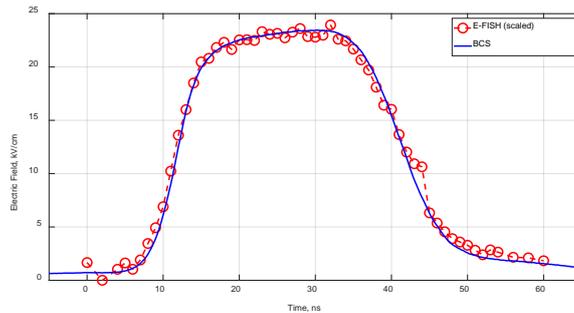
These two respective beams, with pulse widths of either 16 ns or 3 ns, consequently utilize the same E-FISH setup given in Fig. 1a. This setup is essentially similar in principle to that described in [2]. The input laser beam is focused with a 30 cm plano-convex spherical lens (FL1) into the center of the electrode gap and collimated with a second 25 cm lens (FL2). Based on a Gaussian beam approximation, the confocal beam parameter is calculated to be about 2.5 mm. A 665 nm longpass filter (Thorlabs FGL665S) is inserted after the focusing lens to remove any second harmonic light generated by the surfaces of the upstream optics. A dedicated 532 nm reflecting, 1064 nm transmitting dichroic mirror together with a dispersive equilateral prism separate the residual 1064 nm beam and the 532 nm second harmonic signal. The horizontal and vertical polarization components of this 532 nm light are then isolated with a 532 nm polarizing cube so as facilitate simultaneous measurements of both components of the electric field vector perpendicular to the laser beam. Each of these signal components is then focused with a 15 cm lens onto a photomultiplier (PMT) and sampled with a 600 MHz bandwidth LeCroy WaveRunner 64Xi-A oscilloscope with a resolution of 200 ps. A Hamamatsu (model number H7422-50P) PMT captures the horizontally polarized signal while the vertical component is measured by a second Hamamatsu (model number H11526-20-NF) gated PMT. Both PMTs have a rated response time of about 1 ns and are operated with a gain of about 10^4 . The entrance of both units are fitted with an iris and a 532 nm centered bandpass filter (10 nm FWHM) in order to reject stray light. The laser intensity of the probe beam is monitored by picking off part of the residual 1064 nm beam reflected off the dichroic mirror and directing this light onto a 1 ns rise time Thorlabs DET10A photodiode. Following Eq. (1), the electric field may be obtained by taking the square root of the time-integrated PMT signals and normalizing these by the corresponding time-integrated photodiode signals. The PMT and photodiode signals are acquired and averaged over 300 laser shots to improve the signal-to-noise ratio (SNR) of the measurements.

The timing jitter between the HV pulses and the laser beam is estimated to be less than 1 ns. Synchronization between the two is realized with the help of two identical BNC Mod575-4C delay generators. The first delay generator triggers both the flashlamp of the laser and the HV generator which produces the required electrostatic field. This HV generator provides an output signal which is synchronized with the delivery of each HV pulse. This signal is in turn used to trigger the second delay generator responsible for the timing of the laser Q-switch and (where necessary) the HV generator which drives the Pockels cell. The HV pulses delivered to the Pockels cell are slaved to the laser Q-switch and timed such that they coincide with the arrival of the laser pulse to within less than a nanosecond. In this way, the output energy of the sliced pulse is optimized.

The intensity of the 532 nm, E-FISH signal, $I_i^{(2\omega)}$, is given by

$$I_i^{(2\omega)} \propto \left[\chi_{ijkl}^{(3)}(2\omega, \mathbf{0}, \omega, \omega) N E_i^{ext} E_k^{(\omega)} E_l^{(\omega)} L \right]^2 \left[\text{sinc} \left(\frac{\Delta k L}{2} \right) \right]^2 \quad (1)$$

In Eq. (1), E^{ext} represents the electric field to be measured, $E^{(\omega)}$ is the electric field of the 1064 nm probe laser, $\chi_{ijkl}^{(3)}$ is the third-order nonlinear susceptibility, N is the gas number density, L is the confocal beam parameter and $\Delta k = (2k_\omega - k_{2\omega})$ is the difference between the fundamental and the second harmonic wave vectors. Defining a coordinate system whereby x and y represent the horizontal and vertical axes, the use of a vertically (y) polarized probe beam in our case implies that $E_k^{(\omega)} E_l^{(\omega)}$ may be simplified as $I_y^{(\omega)}$, where I is the beam intensity. Furthermore, it follows based on symmetry considerations that the only independent, non-zero components of the nonlinear susceptibility are $\chi_{xxyy}^{(3)}$ and $\chi_{yyyy}^{(3)}$, which apply to the horizontal and vertical components of the second harmonic signal, respectively [11].



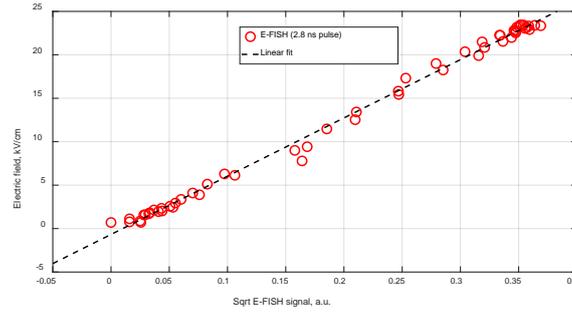


Fig. 2. (a) Time evolution of (square rooted) E-FISH signals produced by 2.8 ns pulses, appropriately scaled and plotted together with the field evolution measured by the back current shunt (BCS). The time-varying field is imposed by applying 30 ns (FWHM) HV pulses with a rise time of about 8 ns. (b) Calibration curve obtained by plotting the measured field values from the BCS against the square root of the E-FISH signals. All acquired signals are averaged over 300 laser shots.

Both the ~ 3 ns and ~ 16 ns laser pulses are tested in a time-varying Laplacian field sustained between the parallel plate electrodes. The electric field vector is oriented in the vertical (y) direction, and hence only the vertically polarized component of the E-FISH signal is measured. The electric field is probed at different time instants by stepping through the delay of the laser Q-switch relative to the applied HV pulses in constant time intervals of 1 ns. Figure 2 shows the results obtained with the sliced pulse and reveals that these ~ 3 ns, 2 mJ pulses are clearly capable of generating E-FISH signals which follow closely the fast rise and fall of the applied electric field. The Laplacian field strength is calculated by taking the voltage measured by the BCS and dividing this value by the inter-electrode gap distance of 4 mm. The measured field values from the BCS are plotted against their corresponding (square rooted) E-FISH signals, as displayed in Fig. 2b. This calibration curve shows that the predicted linear dependence between the applied field and the square root of the second harmonic signal is observed in the experiments. The peak field strength is about 23 kV/cm and the sensitivity of the measurement achieved is about 0.6 kV/cm, the latter being defined as the value of the field strength which generates a corresponding SNR of 2.

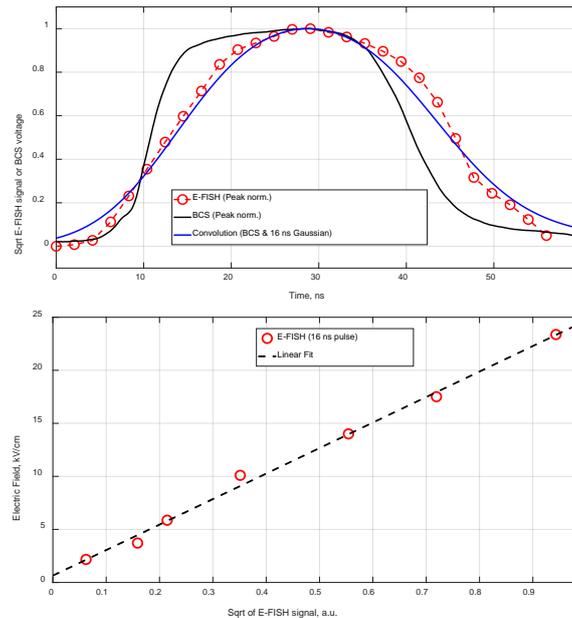


Fig. 3 (a) Comparison of the peak-normalized electric field evolution measured by the BCS versus the peak-normalized E-FISH signal evolution, using 16 ns pulses. (b) Peak applied field strength as a function of the square root of the E-FISH signal.

On the contrary, the longer ~ 16 ns, 35 mJ pulses are unable to effectively track the temporal profile of the same HV pulse. This is evident from Fig. 3, which presents the peak-normalized electric field evolution measured by the BCS compared against the corresponding evolution of the E-FISH signal. It should be noted that the poor agreement with the BCS data is purely a consequence of the inadequate time resolution of the probe laser, rather than an inherent inability to make an accurate field measurement. This point is reinforced by the relatively good agreement displayed between the E-FISH evolution curve and that obtained by a convolution between the BCS data and a 16 ns Gaussian representing the temporal shape of the laser pulse. As a further illustration, the peak applied field strength is plotted as a function of the square root of the E-FISH signal and shown in Fig. 3b. In these experiments case, the applied electric field is varied by adjusting the amplitude of the HV pulses delivered by the HV generator. The minimum field sensitivity achieved in this case is estimated to be about 1.5 kV/cm. The linear dependence of the signal on the applied field indicates that these longer pulses could still be useful in applications where the timescale of the electric field variation is longer than the duration of the probe laser pulse (viz. time resolution of the measurement).

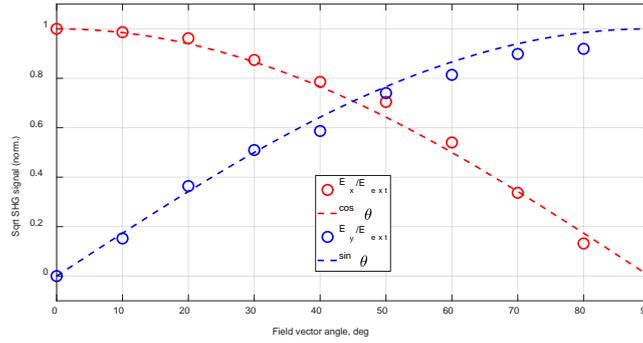


Fig. 4 Horizontal (x) and vertical (y) components of the peak-normalized, square-rooted E-FISH signals as a function of the field vector angle, θ . For a given angle θ , the horizontal and vertical components of an applied field E_{ext} vary as $E_{ext} \cos \theta$ and $E_{ext} \sin \theta$, respectively.

Apart from the superior time resolution afforded by the sliced pulses, the lower laser energy also means that the occurrence of optical breakdown is significantly reduced. It is worth highlighting that such breakdown events not only invalidate the field measurement, but can generate extreme levels of continuum light which may result in catastrophic damage to a PMT. During the experiments with the shorter pulses, no incident of laser-induced breakdown was recorded. However, though relatively infrequent, optical breakdown events did result from time to time with the longer pulses. Fortunately, any damage to our PMT was avoided by virtue of an excessive signal protection circuit in-built within the device. On a related note, the higher energy required by the longer laser pulses may bring into question the non-intrusiveness of the E-FISH method, given the increased probability of laser-plasma interactions.

Field vector measurements are made possible with the E-FISH method since the attendant signal polarization matches that of the applied field vector (see Eq. (1)). This capability is illustrated by simultaneously measuring both x and y components of the electric field vector, as the vertically (y) oriented Laplacian field is effectively rotated with respect to the laser polarization. Figure 4 shows that the expected changes to the vertical and horizontal components of the electric field are duly captured as the field vector angle, θ , is varied. Here θ is defined as the angle made between the field vector and the x -axis. Instead of opting to rotate the electrode system, which could potentially introduce precision errors, both the input laser polarization and the frame of reference of the polarization analyzer (viz. 532 nm polarizing cube in Fig. 1a) are rotated, with a 1064 nm and 532 nm half-wave plate, respectively. While such an optical arrangement to rotate the field vector would obviously be unnecessary in an actual experiment, it is envisaged that each component of the signal polarization will require its own separate calibration curve. An additional important consideration is that, to a first-order approximation, the component of signal polarization orthogonal to that of the laser polarization is attenuated by a factor of 1/9 relative to the parallel component [1,11]. This could be addressed by using a more sensitive PMT on the polarization arm with the weaker signal or by rotating the laser polarization in order to balance the magnitudes of both signals.

The development of a laser-based electric field measurement diagnostic compatible with standard ns laser sources has been demonstrated in this work. A Pockels-cell-based pulse slicer provides the option of realizing shorter duration ns pulses which are found to engender superior performance. The shorter pulse exhibits better prospects not just in terms of time resolution, but also requires less energy, which could aid in suppressing optical breakdown and laser-plasma interaction. Compared with a ps or fs probe, operation within the ns regime has the potential to produce signals with a narrower spectral bandwidth and may be beneficial for signal discrimination against broadband plasma emission.

In reality, even shorter (hundreds of ps) and higher energy pulses may be readily achieved with a well-designed, commercial Pockels cell package and at a fraction of the cost of a typical ultrashort pulse laser system. Integrating these cells with regular multimode YAG lasers for example, should be relatively straightforward and may even aid in reducing sharp temporal intensity fluctuations associated with lasing from multiple modes. The excellent control authority over the pulse width and energy means that the resulting output could in principle be tailored for any given application. It is envisioned that this cost-effective approach may therefore pave the way for performing 1D electric field measurements using a conventional ns laser.

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