Electric field vector measurements via nanosecond electric field induced second harmonic generation

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Electric field induced second harmonic generation, or E-FISH, has received renewed interest as a nonintrusive tool for probing electric fields in gas discharges and plasmas using ultrashort laser pulses. An important contribution of this work lies in establishing that the E-FISH method works effectively in the nanosecond regime, yielding field sensitivities of about a kV/cm at atmospheric pressure from a 16 ns pulse. This is expected to broaden its applicability within the plasma community, given the wider access to conventional nanosecond laser sources. A Pockels-cell-based pulse-slicing scheme, which may be readily integrated with such nanosecond laser systems, is shown to be a complementary and cost-effective option for improving the time resolution of the electric field measurement. Using this scheme, a time resolution of ~3 ns is achieved, without any detriment to the signal sensitivity. This could prove invaluable for nonequilibrium plasma applications, where time resolution of a few nanoseconds or less is often critical. Finally, we take advantage of the field vector sensitivity of the E-FISH signal to demonstrate simultaneous measurements of both the horizontal and vertical components of the electric field. © 2019 Optical Society of America

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Electric field induced second harmonic (E-FISH) generation [1] is a nonlinear optical effect which has received significant attention since the 1970s for its widespread applicability. Examples of its extensive use range from measurements of gas hyperpolarizabilities [2], detection of terahertz radiation [3-4], enhancement of second harmonic yield [5-6], to the probing of electrical pulses in transmission lines [7]. Most recently, this method has been redeveloped for making point-based and 1D electric field measurements in plasmas [8-10]. Picosecond and femtosecond pulses have been the preferred mode of excitation, since the measurement time resolution is fundamentally limited only by the laser pulse duration. Furthermore, the intrinsically high intensity of these ultrashort pulses also augments the signal generation (proportional to the square of the laser intensity). To date, time-resolved electric field measurements using picosecond E-FISH have been successfully conducted in surface dielectric barrier discharges [11,12], and in nanosecond pulsed discharges [9] for understanding the fast ionization wave development [13]. Simultaneous species density and electric field measurements using femtosecond pulses have also been achieved by combining coherent anti-Stokes Raman scattering with E-FISH [14]. While the above emphasizes the utility of subnanosecond excitation for E-FISH measurements, it also raises a natural question as to whether this diagnostic may find application in the nanosecond regime. The payoffs are compelling since such an advance would readily find favor in numerous applications such as DC or AC discharges, where the lower time resolution may be acceptable. More importantly, the ability to resolve electric fields with a time resolution of a few nanoseconds could be vital for nonequilibrium plasma applications, where high-voltage pulses with a similar characteristic rise time are commonly employed [15-16]. These high-voltage pulses alter the chemical reactivity of a system through the production of strong and transient electric fields, and have been extensively applied in combustion [17], aerodynamics [18], and medicine [19]. Finally, demonstrating the viability of E-FISH with nanosecond pulses could potentially profit a wider audience, since picosecond and femtosecond laser systems are often less accessible due to their higher costs. To this end, we test two different nanosecond pulse durations: 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 time-extracting part of the original 16 ns pulse using a Pockels-cell-based pulse slicer.

The experimental setup consists of 3 parts – the high voltage (HV) pulser 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 pulse HV generator. An FID model number FPG 1.2-1NM 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, separated by a gap of 4 mm, as shown in Fig. 1a. The applied voltage is calculated via a summation of the incident and reflected waveforms sampled by a high bandwidth back current shunt (BCS) installed in the coaxial cable which connects the HV generator to the HV electrode. These BCSs have been used extensively in previous work as a HV probe [20].

The nanosecond laser source is a Quanta-Ray Lab-170, 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 energy to about 35 mJ, to avoid optical breakdown. Attenuating the energy in this way, rather than directly at the source, helps to achieve pulse stability, both in terms of energy and timing. To reduce the pulse width down to several nanoseconds, 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 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 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 Pockels 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 laser pulse, with a temporal profile which closely mirrors that of the input HV waveform [21]. 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. To mitigate this problem, a double-pass arrangement is implemented to effectively double the polarization rotation. Compared with a single pass arrangement, double-passing the 1064 nm beam through the cell increases the pulse energy without virtually any detriment to the pulse width. This is provided 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 related to the off-optimum design of the Pockels cell, resulting in losses from the antireflective coatings on the crystal surfaces and absorption within the crystal. The extinction ratio of the Pockels cell is measured to be less than 1%. The optical layout 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. By simply rotating PBS2 by 180°, pulse widths of either 16 ns or 3 ns may consequently access the E-FISH setup given in Fig. 1a. This setup is essentially similar to that described in [9]. 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 25 cm lens (FL2). Based on a Gaussian beam approximation, the confocal beam parameter (or measurement spatial resolution) 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 an 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 to facilitate simultaneous measurements of both these components of the electric field vector. Each of these signal components is 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 (H7422-50P) PMT captures the horizontally polarized signal while the vertical component is measured by a second Hamamatsu (H11526-20-NF) gated PMT. Both PMTs have a rated response time of about 1 ns and are operated with a gain of about 10^5. The entrance of both units are fitted with an iris and a 532 nm centered bandpass filter (10 nm FWHM) 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 is estimated to be less than 1 ns. The intensity of the 532 nm, E-FISH signal, $I_i(2\omega)$, is given by

$$I_i(2\omega) \propto \left[ \chi_{ijk}^{(3)}(2\omega, 0, \omega, \omega) N E_i^{\text{ext}} E_k^{(\omega)} E_l^{(\omega)} \right]^2 \left[ \sin \left( \frac{\Delta k L}{2} \right) \right]^2.$$  

(1)

In Eq. (1), $E^{\text{ext}}$ represents the electric field to be measured, $E^{(\omega)}$ is the electric field of the 1064 nm probe laser, $\chi_{ijk}^{(3)}$ is the third-order nonlinear hyperpolarizability, $N$ is the gas number density, $L$ is the confocal beam parameter and $\Delta k = (2k_\omega - k_{\omega})$ 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 implies that $E_k^{(\omega)} E_l^{(\omega)}$ may be simplified as $E_{y}^{(\omega)}$, 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 $\chi_{zzxy}^{(3)}$ and $\chi_{zyyy}^{(3)}$, which apply respectively to the horizontal and vertical components of the second harmonic signal [22]. It should be added that Eq. (1) assumes a sample which consists only of centrosymmetric gases, i.e. there is no background signal in the absence of an applied field.

![Figure 2](image1.png)

**Fig. 2.** (a) Peak applied field strength as a function of the square root of the E-FISH signal obtained using 16 ns pulses. (b) Comparison of the peak-normalized electric field evolution measured by the BCS versus the evolution of the peak-normalized, (square rooted) E-FISH signals.

To evaluate the time resolution obtained using these laser pulses, the electric field is probed in a time-varying field by stepping through the delay of the laser Q-switch relative to the applied HV pulses, in constant intervals of 1 ns. A comparison with the field values measured by the BCS data in Fig. 2b reveals that these ~16 ns pulses are unable to effectively track the temporal profile of the HV pulse. It should be emphasized however, 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 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. The above notwithstanding, these pulses – especially given their associated millimeter scale (~2.5 mm in our case) – spatial resolution – could still find value 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).

![Figure 3](image2.png)

**Fig. 3.** (a) Time evolution of (square rooted) E-FISH signals produced by 28 ns pulses, appropriately scaled and plotted together with the field evolution measured by the back current shunt (BCS). (b) Calibration curve obtained by plotting the measured field values from the BCS against the square root of the E-FISH signals.

On the contrary, Fig 3a shows that the same experiments performed with the shorter ~3 ns, 2 mJ pulses demonstrate a distinct capability of these pulses to accurately follow the fast rise and fall of the applied electric field. The measured field values from the BCS are plotted against their corresponding (square rooted) E-FISH signals, as displayed in Fig. 3b. 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 minimum field sensitivity is about 0.6 kV/cm. Apart from the superior time resolution afforded by the sliced pulses, the lower laser energy also means that the probability of optical breakdown is significantly reduced. Such breakdown events not only invalidate the field measurement, but can catastrophically damage 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 inbuilt within the device. Another benefit of shorter pulses is that the lower laser energy implies they are less susceptible to inducing laser-plasma interactions, which could affect the measurement accuracy.

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, $\theta$, is varied. Here $\theta$ is defined as the angle between the field vector and the $x$-axis. Instead of rotating 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 1.064 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 [8,22]. This could be addressed by using a more sensitive PMT on the polarization arm with the weaker signal or by rotating the laser polarization to balance the magnitudes of both signals. Finally, a direct consequence of measuring the signal intensity is that both the phase information, and therefore the ability to identify the sign of the electric field vector, is lost. A similar approach avoids this problem by incorporating homodyne detection, and has been used to image electric fields on a silicon-on-sapphire substrate [23].

In concluding, we point out that even shorter (~100 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. The excellent control authority over the pulse width and energy allows the resulting output to be tailored for a given application, and could possibly even reduce the sharp intensity fluctuations associated with mode-beating in multimode lasers. It is envisioned that this versatile and cost-effective approach may thus facilitate 1D electric field measurements with standard nanosecond lasers.

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Full reference list