



RESEARCH ARTICLE

REVISED **Nonlinear field-control of terahertz waves in random media for spatiotemporal focusing [version 2; peer review: 2 approved]**Vittorio Cecconi ¹, Vivek Kumar¹, Alessia Pasquazi^{1,2},
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<https://doi.org/10.12688/openreseurope.14508.3>**Abstract**

Controlling the transmission of broadband optical pulses in scattering media is a critical open challenge in photonics. To date, wavefront shaping techniques at optical frequencies have been successfully applied to control the spatial properties of multiple-scattered light. However, a fundamental restriction in achieving an equivalent degree of control over the temporal properties of a broadband pulse is the limited availability of experimental techniques to detect the coherent properties (i.e., the spectral amplitude and absolute phase) of the transmitted field. Terahertz experimental frameworks, on the contrary, enable measuring the field dynamics of broadband pulses at ultrafast (sub-cycle) time scales directly. In this work, we provide a theoretical/numerical demonstration that, within this context, complex scattering can be used to achieve spatio-temporal control of instantaneous fields and manipulate the temporal properties of single-cycle pulses by solely acting on spatial degrees of freedom of the illuminating field. As direct application scenarios, we demonstrate spatio-temporal focusing, chirp compensation, and control of the carrier-envelope-phase (CEP) of a CP-stable, transform-limited THz pulse.

Keywords

Scattering, terahertz, time-domain spectroscopy, random medium, spatiotemporal focusing, superfocusing, genetic algorithm

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REVISED Amendments from Version 1

- 1) We addressed some typos related to the ability to control the Carrier-Envelope-Phase (CEP) of the THz pulse, previously referred to as Carrier-Envelope-Offset (CEO).
- 2) Inclusion of a new affiliation for the authors Alessia Pasquazi, Juan Sebastian Toterogongora, and Marco Peccianti.
- 3) Change of the email address of the corresponding author.

Any further responses from the reviewers can be found at the end of the article

Plain language summary

Multiple scattering of light is a common phenomenon in everyday life. The opaqueness of fog, milk, and clouds are excellent examples of how light is scrambled when travelling through materials composed of thousands of particles scattering light in all directions. While scattering is generally perceived as unwanted, researchers have shown that complex but inexpensive substances like white paint or frosted glass can be adapted to behave as expensive optical devices, such as high-resolution lenses or optical computing devices. This surprising result is achieved by using particular projectors on the incident light to impress specific patterns yielding a desired shape at the output. While manipulating the spatial profile of light is now well-understood, controlling how scattering affects ultra-short light pulses remains exceptionally challenging, primarily due to the difficulty in measuring the effects of scattering at the ultrafast timescales of the pulse (quadrillionths of a second).

In this theoretical work, we propose a new way to tackle this challenge by leveraging the unique properties of terahertz light waves. Terahertz light lies between microwaves and infrared in the electromagnetic spectrum and is highly sought in research and industry. It can easily reveal the material composition of an object and penetrate common materials like paper and plastic as X-rays do, but without being harmful. More importantly, we can experimentally detect the individual oscillations in a terahertz pulse and extract an unprecedented understanding of how scattering affects it.

We show that this added knowledge gives us the ability to finely manipulate the properties of the pulse, compensate the pulse-broadening effects of scattering, and “engineer” specific pulse shapes, a highly-sought ability in photonics. In the long term, this ability could be applied to reveal the internal composition of complex samples, such as biological tissue, or extract images of samples placed behind (or within) scattering materials.

Introduction

Multiple scattering is generally perceived as detrimental in photonics, as it is commonly associated with unpredictability and loss of information. For instance, in microscopy or astronomy, scattering is widely known to severely affect the resolution and fidelity of an imaging system^{1,2}. As a result, a large body of research has historically focused on compensating or

straight-out eliminating scattering effects³⁻⁵. In a stark paradigm shift, researchers have recently demonstrated that disordered media can become essential ingredients in developing photonic devices with sophisticated optical performances⁶⁻⁹. These include, among others, scattering-assisted super-resolution imaging lenses, wavefront-shaping components, or optical neuro-computing devices¹⁰⁻¹⁸. At their heart, these demonstrations rely on wavefront shaping techniques, i.e., on the identification of an optimal incident field distribution, or pattern, yielding a desired intensity distribution at the output of a scattering medium^{19,20}. While the ability to control monochromatic beams’ spatial properties has matured recently, manipulating the temporal and spectral properties of the scattered field remains challenging, particularly when considering broadband ultrafast illumination²¹⁻²⁵. Multiple scattering is an inherently dispersive phenomenon that naturally leads to the broadening of optical pulses^{26,27}. Recent works have shown that the intrinsic coupling between spatial and temporal dimensions in scattering media can be leveraged to manipulate the temporal properties of pulses by controlling the spatial features of the illumination^{28,29}. As a result, spatial wavefront shaping can be applied to achieve spatiotemporal focusing, corresponding to a simultaneous focusing in space and pulse re-compression in time, either through iterative approaches or by measuring the frequency-dependent transmission matrix of the sample^{21,30-32}. These approaches have shown how to control the envelope of the transmitted waveform successfully (e.g., to adjust the centre of the transmitted pulse³²⁻³⁵), but they remain unsuitable for manipulating the carrier-wave properties (e.g., the carrier-envelope offset of the transmitted pulse). Further advances in this area are fundamentally hindered by the inability to directly measure the full-wave properties of the scattered field, most notably the absolute spectral phase^{23,35}. An interesting question is whether a direct measurement of the electric field oscillations could enable controlling the coherent features of the transmitted pulse and devising advanced forms of waveform synthesis and spectral shaping currently out-of-reach at optical frequencies. In this context, field-sensitive detection is well-established in terahertz (THz) photonics, where time-domain spectroscopy (TDS) grants access to the time-resolved detection of the electric field of single-cycle THz pulses³⁶. Leveraging this ability, THz-TDS has been applied to study the broadband properties of scattering samples, with a particular emphasis on the role of multiple scattering in biomedical applications^{26,27,37-40} and the effects of resonant excitation of the scatterers composing the medium⁴¹⁻⁴⁵. However, the implementation of a complete, field-based wavefront control methodology to manipulate broadband pulses is essentially unexplored in the THz frequency band. In this work, we provide a first theoretical exploration of the potential advantages offered by time-resolved, field-sensitive detection in manipulating broadband THz pulses using scattering media. We combine the nonlinear generation of THz patterns from structured optical beams with an evolutionary optimisation feedback-loop targeting the coherent, full-field properties of the transmitted field, showcasing the ability to achieve spatiotemporal focus and control the absolute phase of the transmitted pulse. Quite interestingly, access to the absolute spectral phase of the pulse enables the definition of effective spectral shaping strategies, such as the ability to

compress an incident chirped-pulse or control the carrier-envelope phase (CEP) of an incident pulse, a significantly challenging task at optical frequencies.

Physical framework and methodology

Problem definition: full-wave control of THz pulses in complex media

We model the linear transmission properties of the scattering medium through a dispersive transmission operator $TM(x, y, \omega)$ ^{33,46}. For the sake of simplicity, we focus on a scalar description of the scatterer, but our approach can be easily extended to a full-vector formulation⁴⁷.

In the presence of an input field distribution $E^-(x, y, \omega)$, the field transmitted through the scatterer $E^+(x, y, \omega)$ is expressed through a (spatial) convolution relation:

$$E^+(x, y, \omega) = \int dx' dy' TM(x-x', y-y', \omega) E^-(x', y', \omega). \quad (1)$$

Following Ref. 48, we discretize the output and input planes in M and N two-dimensional, square pixels, respectively. In such a formulation, the spectral components of the fields are re-defined as column arrays, and Equation (1) is rewritten in terms of a frequency-dependent transfer matrix $T_{mn}(\omega)$ as follows:

$$E_m^+(\omega) = \sum_{n=k}^N TM_{mn}(\omega) \cdot E_n^-(\omega), \quad (2)$$

where E_m^+ E_n^- represents the field in the m -th (n -th) pixel of the output (input) field distributions. Following standard approaches, we express the transmission matrix as a complex-valued, random Gaussian matrix:

$$TM_{mn}(\omega) = \exp[i\phi_{mn}(\omega)]/N, \quad (3)$$

where $\phi_{mn}(\omega)$ is a random phase distribution uncorrelated in space and gaussian-correlated along the frequency axis. The degree of spectral correlation, generally expressed in terms of the spectral correlation bandwidth $\Delta\nu_c$, is commonly employed to characterise the spectral response of a scattering medium, and it is generally defined as the inverse of the Thouless time²⁴. In practice, a frequency-correlated transfer matrix can be modelled numerically by applying a Gaussian spectral filter of width $\Delta\nu_c$ to a white-noise random matrix^{49,50}.

In general terms, a wavefront control approach aims to identify the optimal incident field distribution $E_{optimal}^-(x, y, t)$ yielding the desired output field $E_{target}^+(x, y, t)$ at the output facet of the scatterer⁵¹. However, when operating at THz frequencies, the ability to control the incident electric field distribution is hindered by the limited availability of spatial light modulator (SLM) devices. To overcome this limitation, we employ the nonlinear conversion of structured optical beams, an approach we have recently developed within the framework of time-resolved nonlinear ghost imaging (NGI)⁵²⁻⁵⁴. With this approach, any optical pattern generated through a standard SLM device can act as a direct source of broadband THz patterns. By considering a nonlinear quadratic process (e.g., in a $\chi^{(2)}$ crystal

such as ZnTe), the relation between incident optical intensity and generated THz field is linear, i.e., $E_{THz}(x, y, t) \propto I_{pump}(x, y, t)$, enabling the precise control over the THz field profile by simply shaping the incident optical pulse. A key advantage is that the spatial resolution of the THz patterns can be pushed way below the standard THz wavelength scale, as it is only bound by the diffraction limit of the optical beam. Besides, the nonlinear conversion ensures that the spatial pattern is well-defined across the whole THz band, enabling the generation and control of single-cycle, THz structured beams. We can assume that the incident THz patterns are expressed as:

$$E^-(x, y, \omega) \propto A_0 P(x, y) f_{THz}(\omega), \quad (4)$$

where A_0 is an amplitude, $P(x, y)$ is the (all positive) spatial profile, and $f_{THz}(\omega)$ is the spectrum of the THz pulse, which is assumed to be the same for all input points. Figure 1 outlines a possible implementation of our wavefront-control methodology⁵²⁻⁵⁴. The spatial pattern, impressed by an optical SLM on the pump field, is transferred to the THz pulse through nonlinear conversion in a quadratic crystal. The patterned THz field impinges on the scatterer, that is placed in close proximity (i.e., in the near-field or direct contact) of the generating crystal. Upon propagation, an electro-optical sampler collects the transmitted field in the typical fashion of standard TDS detection⁵⁵. As our objective is to manipulate the full-field properties of the transmitted THz pulse in a specific pixel of the output plane, we introduce a spatial pinhole at the output facet of the scatterer, corresponding to the desired focal spot (x_0, y_0) . To optimise the wavefront of the transmitted pulse, the collected THz field from a predetermined ensemble of patterns is analysed and ranked through an evolutionary optimisation algorithm (e.g., a genetic algorithm (GA)) that provides the feedback required to iteratively optimise the transmitted field. This configuration enables establishing an optimisation feedback loop relying on the temporal field properties instead of average intensity, as is generally the case when operating at optical frequencies. In our configuration, the combination of near-field coupling and generation of THz patterns from structured optical fields provides a significant advantage in terms of available degrees of freedom. The THz field can be densely sampled in space, with patterns reaching sub-wavelength spatial resolution in the near-field region. This is a drastic difference from typical optical embodiments, where the spatial sampling is limited by the numerical aperture of the illumination⁵⁶. On the contrary, in our case, the sampling can exceed the density of the modes accessible from the scatterer input facet, and the spatial density of the transfer matrix can be as high as required to represent the scattering medium accurately⁴⁸.

Numerical implementation and optimisation strategy

To demonstrate this concept, we numerically simulated the configuration in Figure 1 using MATLAB 2021b. The plotting and simulation codes are fully compatible with Octave open-source software⁵⁷. The simulation codes require the “statistics” Octave package, freely available through the [Octave Forge repository](#). The plotting codes require the “signal” Octave package, freely available through the [Octave Forge repository](#). As

a THz input field, we considered a transform-limited THz pulse with a duration of 1 ps, a typical product of a bandwidth-limited optical rectification process in a nonlinear crystal. The incident pulse spectrum is centred at 1 THz ($\Delta\nu \cdot \tau_p \approx 0.44$, where τ_p is the full width half maximum (FWHM) pulse duration and $\Delta\nu$ the bandwidth of the power spectral distribution). In terms of spatial features, we considered grayscale spatial patterns composed of 33×33 square pixels of side $\Delta x = 100 \mu\text{m}$. The scattered field spectral correlation bandwidth is $\Delta\nu_c = 250 \text{ GHz}$, a value that is compatible with experimental literature and which places us in a non-monochromatic case^{38–40,58}. The optimisation is performed via a standard GA targeting different fitness functions, and corresponding to the genetic algorithm described in Ref. 59. Leveraging the ability to resolve in time the transmitted electric field, we define the fitness functions maximised by the GA in terms of the statistical moments of the electric field waveform, defined as follows:

$$\begin{aligned}
 E_0 &= \int dt |E(t)|, \\
 \mu[E(t)] &= \int dt t \frac{|E(t)|}{E_0}, \\
 \sigma[E(t)] &= \sqrt{\int dt (t-\mu)^2 \frac{|E(t)|}{E_0}}, \\
 S[E(t)] &= \frac{1}{\sigma^3} \int dt (t-\mu)^3 \frac{|E(t)|}{E_0},
 \end{aligned}
 \tag{5}$$

where μ , σ , and S are the first, second (standard deviation) and third moments (skewness) of the probability

distribution defined by $|E(t)|$. Practically speaking, these quantities correspond to the centre, duration, and symmetry

of the pulse waveform. We stress that all those quantities do not relate to the pulse envelope or to locally averaged intensity profiles, as in the common optical practice. Table 1 summarises the different types of fitness functions employed in our analysis.

Results

Spatiotemporal focusing and temporal shift of recompressed pulses.

As an initial test case, we targeted the ability to simultaneously focus the transmitted field in space and time (spatiotemporal focusing). Our simulation results are shown in Figure 2. As an input field, we considered a single-cycle, transform-limited THz pulse (Figure 2a). As illustrated in Figure 2b and c, the corresponding transmitted wavefront for a non-optimised pattern is generally spread in time (Figure 2b) and scattered in space (Figure 2c). To obtain a spatiotemporal focus, we applied our GA algorithm to maximise the cost function A, where $\max E_0(t) \equiv \max E(x_0, y_0, t)$ is the peak electric-field value in the desired focal spot (x_0, y_0) . Such a fitness function simultaneously maximises the transmitted field’s peak value and minimises the pulse profile’s standard deviation in time.

A typical optimised pulse is shown in Figure 2e and f, showcasing our ability to simultaneously focus and recompress the transmitted pulse in the desired focal spot. The ability to restore the spectral properties of the pulse is particularly evident by comparing the spectrum of the optimised pulsed (Figure 2d, solid green line) with the incident one (Figure 2e, blue dashed line). Quantitatively, the optimised spectrum has an FWHM band of 0.52 THz (input 0.8 THz). Quite interestingly, our approach is sensitive to the coherent field properties of the pulse, as illustrated in Figure 2g and h, where we demonstrate the ability

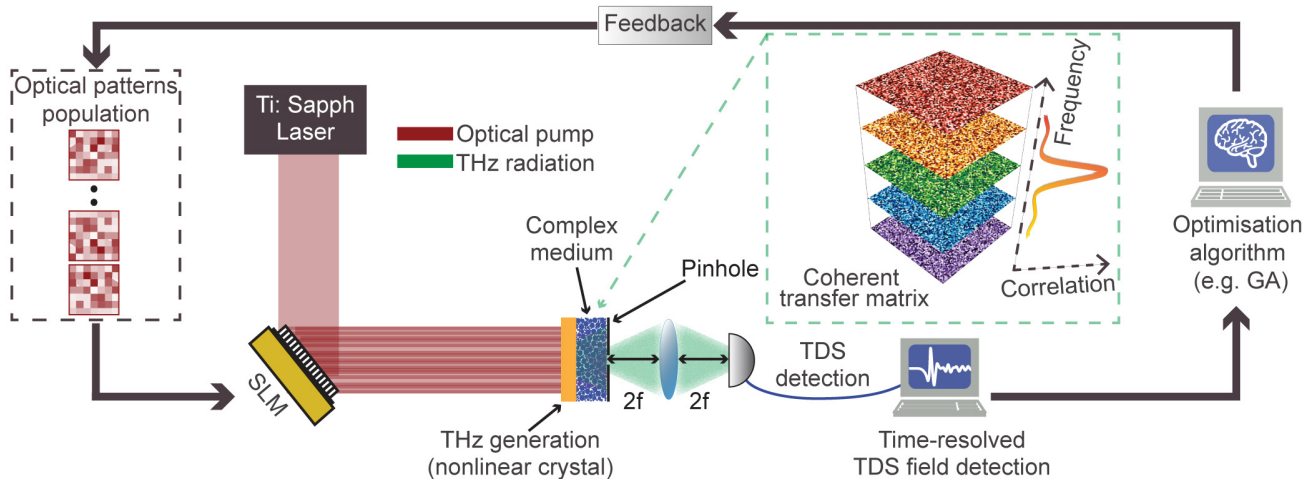


Figure 1. Nonlinear wavefront control of THz complex media and conceptual overview of the iterative algorithm. Illustrative imaging setup, including a nonlinear crystal (yellow) emitting structured THz waves (green) generated from optical patterns (red) through a standard spatial light modulator (SLM). The transmitted THz field is collected via time-domain spectroscopy (TDS). The generation crystal is placed close to the scattering medium. The black arrows describe the schematic loop of the iterative identification of an optimal pattern producing a pulse with the desired spatiotemporal properties via a genetic algorithm (GA) approach. In the green-dashed box, we show a conceptual picture of the hyperspectral transfer matrix and its frequency correlation.

Table 1. The set of cost functions used in the genetic algorithm. In all these expressions, the electric field is measured in the desired focal point, i.e. $E_0(t) \equiv E(x_0, y_0, t)$. The statistical moments of the electric field μ , σ , and S are defined in Equation (5). For the fitness function C, the variables α , β , γ and ξ are weights controlling the relative importance of each component of the multi-objective fitness function. For the fitness function D, c.c. stands for complex conjugate.

Fitness function	Definition	Type of Optimisation
A	$\frac{\max(E_0(t))}{\sigma[E_0(t)]}$	Spatiotemporal focusing
B	$-\frac{\min(E_0(t))}{\sigma[E_0(t)]}$	Spatiotemporal focusing and phase inversion
C	$\alpha \max(E_0(t)) - \beta \mu[E_0(t)] - t_0 - \gamma \sigma[E_0(t)] - \sigma_0 - \xi S[E_0(t)] - S_0 $	Achieving a desired time delay t_0 , temporal deviation σ_0 and skewness S_0
D	$\frac{\int d\omega E_0(\omega) E_{target}^*(\omega) + c.c.}{\sigma[E_0(t)]}$	Phase-sensitive spectral shaping, where the aim is to obtain a measured field $E_0(\omega)$ as close as possible to a target field $E_{target}(\omega)$

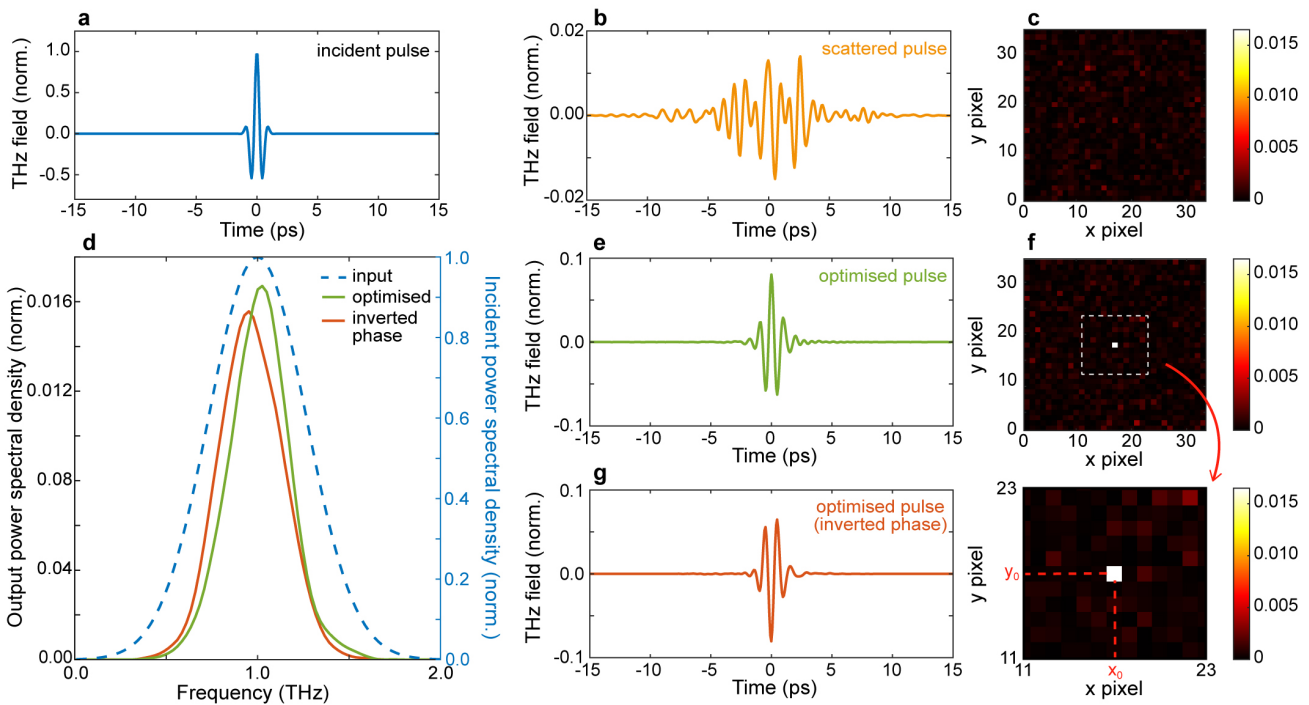


Figure 2. Spatiotemporal refocusing and pulse flip. **a.** Input temporal profile. **b.** Temporal profile of a scattered pulse at the output of the scattering medium; the pulse is normalised with respect to the incident pulse. **c.** Non-optimised spatial intensity distribution at 1 THz. **d.** Power spectral density (PSD) spectrum of the optimal terahertz pulse (green line), optimised flipped pulse (orange line) and incident pulse (blue dashed line, right y-axis). **e.** Temporal profile of an optimised pulse. **f.** Optimised spatial intensity distribution at 1 THz. **g.** Temporal profile of an optimised pulse with an inverted (flipped) phase. Numerical parameters: spectral correlation $\Delta\nu_c = 250$ GHz, number of generations 10000, size of population per generation 100.

to flip, or invert, the optical phase of the THz pulse by employing the fitness function B from Table 1. The corresponding spectral profile is shown in Figure 2d (orange line).

Figure 3 illustrates the ability to simultaneously optimise several temporal properties of the transmitted wavefront in a

single GA optimisation cycle. For this scenario, we employed the multi-objective optimisation fitness function C in Table 1, which maximises the peak field in time while simultaneously searching for incident patterns yielding an output wavefront with the desired centre t_0 , duration σ_0 and skewness S_0 . Specifically, we performed a series of simulations with a target

duration t_0 and skewness S_0 coinciding with the transform-limited incident pulse, while we varied the target time-shift within the interval $t_0 \in [-5\text{ps}, 5\text{ps}]$. The factors $\{\alpha, \beta, \gamma, \zeta\}$ multiplying each term are the cost function's weights. As illustrated in Figure 3a, the pulse can be simultaneously recompressed and controllably time-shifted in a similar fashion to the case studied in Ref. 25.

However, the ability to control the pulse centre comes at the price of a reduced ability to achieve a good degree of spatio-temporal focusing, as can be evinced by assessing the properties of the optimised wavefronts (Figure 3b–e). While in our simulations, we achieved a broad degree of control of the centre of the pulse μ (Figure 3b), we observed an overall decrease in the peak field (Figure 3b) and an increase in the pulse duration (Figure 3d). This behaviour is in good qualitative agreement with the experimental results of Ref. 25. However, in the framework of THz-TDS, these results suggest that one could ideally scan the THz pulse profile within a range of a few ps without mechanical time-delay devices (i.e., translation stages), commonly used in ultrafast optical setups.

Field-sensitive spectral shaping: chirp and CEO control

A key advantage of our methodology is the possibility of detecting the coherent properties of the transmitted THz pulse. These correspond, in particular, to the spectral amplitude and absolute phase, which are generally not directly measurable quantities in experiments at optical frequencies.

The ability to coherently resolve the complex-valued spectrum of the transmitted waveforms is particularly suited to perform phase-sensitive spectral shaping of the incident pulse. For this type of task, we employ the cost function D in Table 1, where

$E_{target}(\omega)$ is the desired spectral profile, $E_{out}(\omega)$ is the output spectrum, and $\sigma(E(t))$ denotes the standard deviation of the pulse in the desired focal point.

This particular choice of fitness function selects patterns yielding output waveforms with a short duration and a high degree of spectral correlation with the desired waveform. As a first example, in Figure 4, we illustrate the possibility of ‘recompressing’ at the output of the scatterer an incident pulse characterised by a linear chirp. The field profile of the incident pulse is shown in Figure 4a, while the spectral amplitude (blue line) and absolute phase (orange dots) are shown in Figure 4c, respectively. In this scenario, we choose the transform-limited version of the incident pulse as the target waveform in Figure 4a. As can be readily evinced from the optimised pulse profile shown in Figure 4b, in our simulations, we were able to restore the symmetry of the optimised wavefront, albeit for a slightly longer pulse. The ability to flatten the spectral phase is clearly shown in Figure 3d, where we report the spectral amplitude and absolute phase of the optimised pulse. Interestingly, our approach to pulse shaping can be directly applied to achieve extremely fine control of the absolute phase of the output waveform. As a final example, we assessed the possibility of finely tuning the carrier-envelope-phase (CEP) of the optimised waveform. Our results are shown in Figure 5. In all four cases, we considered an input field corresponding to a transform-limited THz pulse (identical to the case in Figure 2a). As output wavefront targets, we considered waveforms with different CEP values (Figure 5a–c). The corresponding optimised output fields, illustrated in Figure 5d–f, demonstrate the ability to finely tune the CEP of the output waveform, showcasing a remarkable ability to manipulate the coherent properties of the incident pulse.

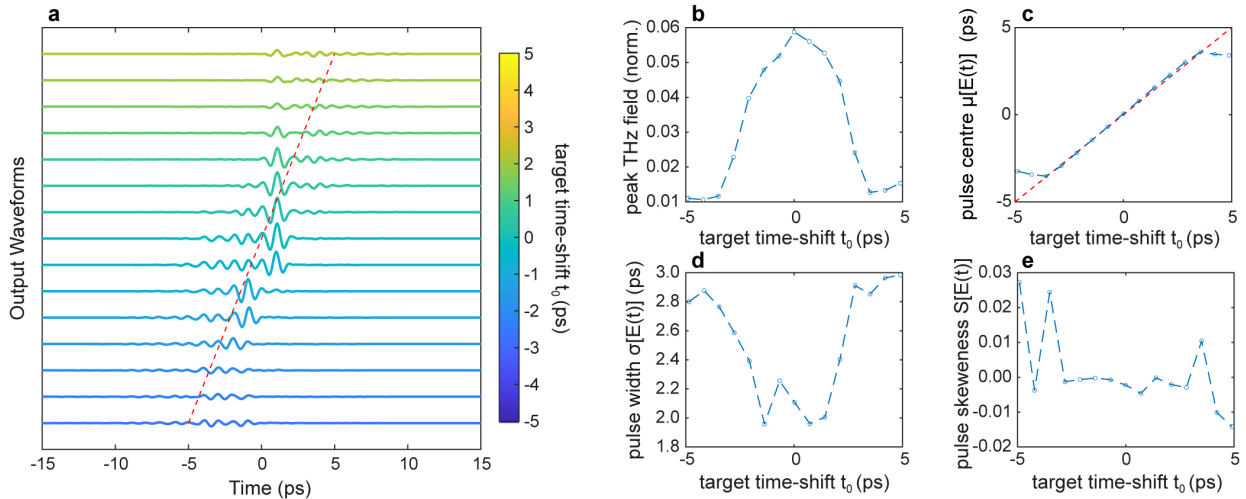


Figure 3. Time shift control of a THz pulse. **a.** Temporal profiles of the output THz pulses optimised for different values of time delay t_0 . **b.** THz peak field as a function of the time delay t_0 . **c.** First central moment μ , corresponding to the pulse centre, as a function of the time delay t_0 . The red dashed line represents the desired time-delay $\mu = t_0$. **d.** The second central moment σ , corresponding to the pulse width, as a function of the time delay t_0 . **e.** Third central moment S , corresponding to the pulse symmetry, as a function of the time delay t_0 . Numerical parameters: spectral correlation $\Delta\nu_c = 250 \text{ GHz}$, number of generations 20000, size of population per generation 50, and weight factors $\{\alpha, \beta, \gamma, \zeta\} \equiv \{10^2, 5 \cdot 10^{12}, 10^{12}, 1\}$.

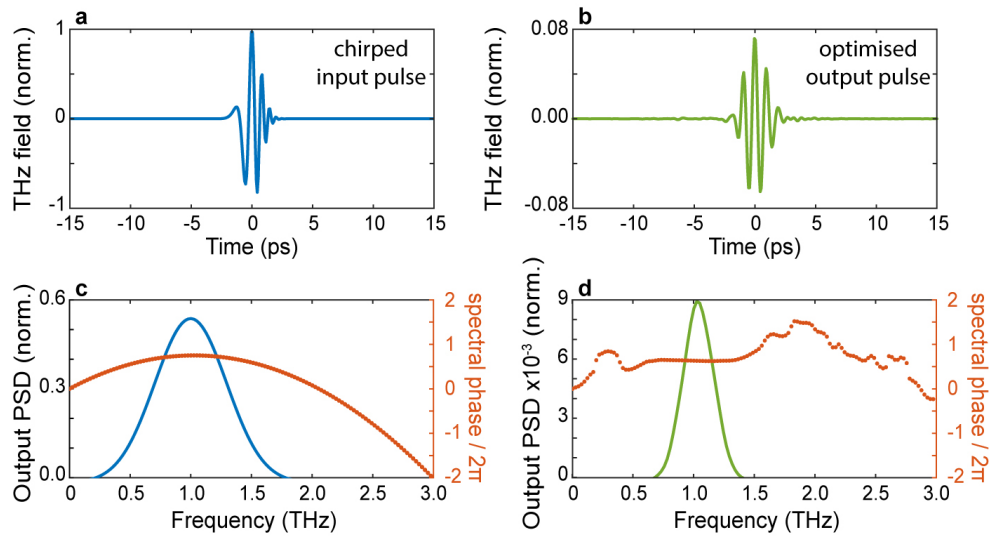


Figure 4. Chirped-pulse compression. **a.** The input temporal trace of an initially chirped pulse. **b.** Temporal trace of the optimised pulse. **c.** Power spectral density of the input pulse (blue line) and spectral phase (orange dots). **d.** Power spectral density of the optimised pulse (blue line) and spectral phase (orange dots). The algorithm tries to fit the output spectral phase as close as possible to the phase of a transform-limited pulse. Numerical parameters: spectral correlation $\Delta\nu_c = 250$ GHz, number of generations 10000, size of population per generation 300.

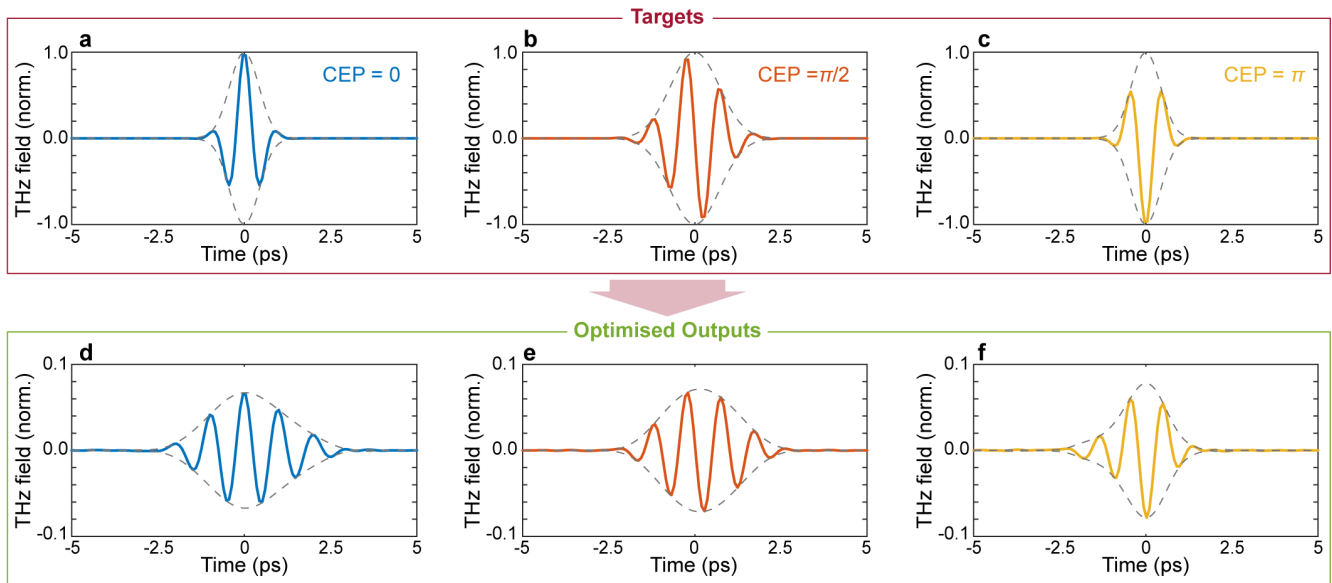


Figure 5. Carrier envelope phase (CEP) modulation. Temporal profiles of target and optimised pulses for CEP $\phi = 0$. (**a**, **d**), $\phi = \frac{\pi}{2}$ (**b**, **e**), and $\phi = \pi$ (**c**, **f**). For all cases, the input field is the transform limited pulse shown in Figure 2a. Numerical parameters: spectral correlation $\Delta\nu_c = 250$ GHz, number of generations 50000, size of population per generation 50.

Discussion and conclusions

In this work, we theoretically investigate a field-sensitive wavefront shaping methodology to control the spatiotemporal properties of terahertz pulses transmitted through a scattering medium. Our approach combines the field detection capabilities of TDS techniques with the generation of terahertz structured beams through nonlinear conversion of optical

beams. Our simulations show how the full access to the coherent properties of the transmitted terahertz field enables defining phase-sensitive evolutionary optimisation strategies to manipulate the carrier-wave properties of THz pulses as they travel through complex media. As in relevant case studies, we considered different wavefront control applications, including spatiotemporal focusing, phase inversion, temporal shifting control,

and re-compression of a chirped THz pulse. Quite remarkably, we demonstrated the ability to manipulate the CEP of a single-cycle, CP-stable incident pulse. Controlling the CEO of ultrafast pulses is a highly challenging task in photonics that usually requires the ability to independently control the group and phase velocity of CP-stable pulse or implementing advanced detection techniques such as f - $2f$ interferometry⁶⁰⁻⁶². Our results differ from traditional spatiotemporal focusing at optical and infrared frequencies as THz-TDS detection provides a direct, coherent measure of the properties of the transmitted electric field. Such a remarkable result would have a profound impact, especially for THz imaging, since time-resolved characterisation techniques are currently highly desired due to the broad spectrum of potential applications, including deep-tissue biological imaging and time-reversal control of optical waves^{63,64}. Although we did not provide specific modelling for near-field interactions in this work, the access to the near field modes is less challenging at terahertz frequencies. Hence, it is to be noted that if the THz source is located in near-field conditions with the sample and spatially controlled with NGI-methodology, the accessible modes within the scattering medium can potentially focus light under the diffraction limit, a potential route to subwavelength imaging resolution. Moreover, TDS-based wavefront control techniques could provide a practical approach to manipulating broadband terahertz pulses' spatiotemporal properties and design entirely new classes of wavefront and spectral shaping applications. It is also important to highlight that due to the scale of the wavelengths, working at THz frequencies offers a considerable advantage to have a precise and determinist approach in sample realisation (e.g., 3D printing)^{65,66}. Additionally, the size of each scattering particle would be bigger (i.e., hundreds of micrometre⁴⁵) compared to what is usually used in optical frequencies (e.g., nanometre-scale⁶⁷), therefore rendering samples intrinsically more robust to thermal and mechanical effects. Finally, the combination of THz technology with concepts and methodologies from complex photonics opens up the intriguing (and unexplored) possibility of performing time-resolved, full-wave scattering matrix retrieval experiments.

Data availability

Underlying data

Figshare: Figure data for "Nonlinear field-control of terahertz waves in random media for spatiotemporal focusing". <http://doi.org/10.6084/m9.figshare.19096859>⁵⁷.

This project contains the following underlying data, organised by folder:

- Figure 2/
 - o mat/ (folder containing raw datasets (in .fig and .mat formats) for each panel in Figure 2)
- Figure 3/
 - o mat/ (folder containing raw datasets (in .mat format) for each panel in Figure 3)
- Figure 4/
 - o mat/ (folder containing raw datasets (in .fig and .mat formats) for each panel in Figure 4)

- Figure 5/
 - o mat/ (folder containing raw datasets (in .fig and .mat formats) for each panel in Figure 5)

Extended data

Figshare: Figure data for "Nonlinear field-control of terahertz waves in random media for spatiotemporal focusing". <http://doi.org/10.6084/m9.figshare.19096859>⁵⁷.

This project contains the following extended data, organised by folder:

- Figure 2/
 - o make_f2.m (MATLAB script that reads and processes the raw data and plots the figure)
 - o make_f2_octave.m (Octave script that reads and processes the raw data and plots the figure)
- Figure 3/
 - o make_f3.m (MATLAB script that reads and processes the raw data and plots the figure)
 - o make_f3_octave.m (Octave script that reads and processes the raw data and plots the figure)
- Figure 4/
 - o make_f4.m (MATLAB script that reads and processes the raw data and plots the figure)
 - o make_f4_octave.m (Octave script that reads and processes the raw data and plots the figure)
- Figure 5/
 - o make_f5.m (MATLAB script that reads and processes the raw data and plots the figure)
 - o make_f5_octave.m (Octave script that reads and processes the raw data and plots the figure)
- simulation_codes_octave/
 - o Main_costfunction_A.m (simulation code with cost function A from Table 1)
 - o Main_costfunction_B.m (simulation code with cost function B from Table 1)
 - o Main_costfunction_C.m (simulation code with cost function C from Table 1)
 - o Main_costfunction_D.m (simulation code with cost function D from Table 1)
 - o rsgeng1D.m (function to generate gaussian-correlated 1D data)
 - o TM_f.m (function to generate a frequency-correlated transmission matrix)
 - o fwhm.m (function to estimate the FWHM of a temporal pulse profile)
 - o compute_moment.m (function to calculate the statistical moments of temporal pulse profiles)

- `spectr.m` (function to calculate the Fourier spectrum of a temporal field profile)
- `erdc_fireice_h.m` (utility function for customised colormap)
- `fireice.m` (utility function for customised colormap)

Data and simulation codes are available under the terms of the [Creative Commons Attribution 4.0 International license](https://creativecommons.org/licenses/by/4.0/) (CC-BY 4.0).

Ethics and consent

Ethical approval and consent were not required.

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Silvia Gentilini 

Institute for Complex Systems, National Research Council (ISC-CNR), Rome, Italy

The manuscript "Nonlinear field-control of terahertz waves in random media for spatiotemporal focusing" presents a theoretical and numerical demonstration of a technique for the full-wave control of a Terahertz pulse propagating in a random media. In the last ten years, the spatial manipulation of an electromagnetic wave to control its propagation through turbid media has raised many researchers' interest in photonics. This has brought significant improvement in the techniques aimed at overcoming and eventually exploiting the structural disorder at the scale of optical wavelength. At these frequencies, the success of these techniques is mainly due to the availability of spatial light modulators (SLM) that, combined with optimizing algorithms, enabled an approach to the inverse scattering problem and obtained results such as super focusing and imaging through opaque media. In this framework, the novelty of the work authored by Ceconi et al. is to achieve spatio-temporal shaping of ultra-short pulse in the THz range, combining the well-established outcomes obtained to control the spatial distribution of optical field with the advantage of the full-field detection of time-domain-spectroscopy (TDS). Specifically, the Authors propose using an optical SLM to modulate the wavefront of the optical pump that, impinging on the nonlinear crystal that generates the THz pulse, transmits to this latter the modulation. The optimization of such a modulation is obtained by a scheme that foresees feedback, provided by a genetic algorithm, between the pulse transmitted and the pattern to impress on the optical pump

The Authors simulated some test cases to demonstrate the validity of their theoretical approach and of the proposed scheme to realize it.

Specifically by playing with the genetic algorithm (GA) chosen as optimization algorithm, they demonstrate a sub-wavelength focusing of the pulse and simultaneously the ability to control the pulse centre, keeping fix the focusing point. Remarkable is also that, leveraging the phase sensitive TDS detection and by suitable choice of the fitness function of the GA, they succeed in optimizing the spectral shape of the pulse transmitted by the disordered sample performing a control on the chirp of the pulse and of its carrier-envelope-phase.

The work is reported with accuracy and provides details helpful to interested researchers to implement the clever strategy proposed for other applications in the THz range.

The work also raised a series of interesting question to address in future as the existence of a disorder threshold to use the methodology. I'm also wondering if it was possible to implement the technique to obtain multi-point focusing by translating the collection system in order to obtain a scan of the output surface.

Below I report some minor revisions that the Authors could address in the text:

1. I don't understand the meaning of the infinity symbol in Eq.(4).

2. Some misreferences to the figures in the text.

- In the second paragraph of the Results, on page 5, the Authors refer to Figure 2e for the spectrum of the incident beam. I think that the correct reference figure is Figure 2d.

- In the second paragraph on page 7, the Authors refer to Figure 3b for the pulse centre. I think that the correct reference is Figure 3c.

3. In the title of the second subsection of the "Results", I think the CEO has to be changed in CEP.

Is the work clearly and accurately presented and does it cite the current literature?

Yes

Is the study design appropriate and does the work have academic merit?

Yes

Are sufficient details of methods and analysis provided to allow replication by others?

Yes

If applicable, is the statistical analysis and its interpretation appropriate?

Yes

Are all the source data underlying the results available to ensure full reproducibility?

Yes

Are the conclusions drawn adequately supported by the results?

Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Photonic of complex systems

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Author Response 27 Jan 2023

Juan Sebastian TOTERO GONGORA

We are glad that the reviewer appreciated the work we have performed, and we thank the reviewer for their positive assessment. In relation to the interesting suggestion to obtain multi-point focusing (e.g., to perform a raster scan of a sample), in our approach, it is possible to focus the THz pulse in different spatial target points by writing different cost functions related to different spatial positions. The ability to manipulate different points of the scattered field is indeed a key advantage of using an iterative optimisation algorithm, and it could enable us to find the optimal patterns producing foci in different points. We note, however, that in actual experiments finding an optimal solution far away from the centre of the optical path might be more challenging. In the presence of non-uniform illumination, for example, points close to the edge of the illumination region might be characterised by a lower signal-to-noise ratio. To clarify this point, we added the sentence "While our analysis was conducted by considering a target focal point placed in the centre of the optical path, the use of an iterative optimisation approach enables, in principle, to select different points on the output plan. In practical terms, this would correspond to modifying the cost functions in Table 1 to consider the scattered electric field at different positions. We note, however, that in real-life experiments, the ability to obtain an optimal solution at any point on the output plane might be restricted by the properties of the scattering medium and the uniformity of the optical illumination." in the Discussion and Conclusions section. Please find below our point-by-point answer to the further suggestion for revision:

1) Regarding the infinity symbol in equation (4), it is a rendering error when generating the PDF. We thank the reviewer for pointing it out and have amended it in the new version.

2, 3) Thanks for the note; we have corrected the references.

4) Regarding the title of the second subsection of the 'Results', the Referee is correct. We changed it to 'Field-sensitive spectral shaping: chirp and CEP control'.

Competing Interests: No competing interests were disclosed.

Reviewer Report 31 August 2022

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Romain Peretti 

IEMN - CNRS, Villeneuve d'Ascq, France

The manuscript "Nonlinear field-control of terahertz waves in random media for spatiotemporal

focusing" describes a theoretical and numerical work describing an innovative implementation of super focusing in scattering disordered media in the terahertz frequency band. Imaging and tight focusing directly inside disordered media was assumed to be impossible mostly due to the multiple scattering events making the photon path almost impossible to predict. However since the scattering processes remain coherent and time reversible, works of the past decades showed that thanks to an inverse problem approach one can retrieve information at a very high resolution. Such an approach had several theoretical outcomes and applied consequences as in Optical coherence tomography. One of the key levers to achieve such an approach is to control the shape of the wave front of the beam. The author of the papers propose a very smart way to do it using nonlinear generation of Terahertz radiation and instead of imprinting the pattern on the THz beam they proposed to imprint it on the infrared beam used to generate the THz through nonlinear optics effect. In this way, one only needs conventional spatial light modulators and not THz ones.

The manuscript provides numerical analysis based on simulated scattering samples showing subwavelength focusing on a broad band. Not only that but the authors managed to focus, but they also shifted the pulse to a desired time delay in a few picoseconds range and modulated the carrier envelope phase. The conclusions are well supported and the text is very clear.

This work is a real advance in the field and would suggest many questions among them : If scattering is more and more understood in the visible and near infrared with scatterers corresponding to this wavelength range, it is less the case for the scatterer in the THz frequency ranges. The model proposed are well known in the higher energy band but I wonder if they have already been applied in the THz since ref 37 to 40 of the papers does not discuss it. In other words, are there any arguments for this model to work in the THz range especially compared to the analysis in ref 37?

Is it possible to focus the THz spot elsewhere than in the middle of the optical path?

Is the work clearly and accurately presented and does it cite the current literature?

Yes

Is the study design appropriate and does the work have academic merit?

Yes

Are sufficient details of methods and analysis provided to allow replication by others?

Yes

If applicable, is the statistical analysis and its interpretation appropriate?

Not applicable

Are all the source data underlying the results available to ensure full reproducibility?

Yes

Are the conclusions drawn adequately supported by the results?

Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Terahertz Photonics

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Author Response 27 Jan 2023

Juan Sebastian TOTERO GONGORA

We thank the reviewer for the thorough analysis of our manuscript and their positive assessment of our results. In the following, we address the reviewer's comments:

1) Thanks for the interesting question. As correctly pointed out by the reviewer, there are no experimental studies on the broadband transmission matrix representation of a scattering medium at THz frequencies. In the state-of-the-art (cf. Refs. 37-40), researchers have mostly analysed the scattering response in terms of average diffusion models by collecting the total transmitted field at the output. A key challenge in obtaining a full transfer matrix representation of the scattering is the requirement of resolving the scattered electric field in space, which is extremely challenging in THz experiments. At optical frequencies, however, it has been shown that the transfer matrix representation is equivalent to the diffusion approximation results for a space-dependent response. The transfer matrix framework, in general, is derived and validated in accordance with the diffusion approximation [R1]. The experimental validation of our transfer matrix model will be the subject of our future work. Our objective in this manuscript is to review the theoretical implications of time-resolved, field-sensitive detection for manipulating broadband THz pulses, and until now, no complete, field-based wavefront control methodology has been developed for manipulating broadband pulses at THz frequencies.

2) We thank the author for raising this excellent point. In our approach, it is possible to focus the THz pulse in different spatial target points by writing an appropriate cost function. The ability to manipulate different points of the scattered field is indeed a key advantage of using an iterative optimisation algorithm. In practical terms, this would correspond to changing the desired focal point where E_0 is calculated or expressing different cost functions for different points at the output. We note, however, that in actual experiments finding an optimal solution far away from the centre of the optical path might be more challenging. In the presence of non-uniform illumination, for example, points close to the edge of the illumination region might be characterised by a lower signal-to-noise ratio. To clarify this point, we added the sentence "While our analysis was conducted by considering a target focal point placed in the centre of the optical path, the use of an iterative optimisation approach enables, in principle, to select different points on the output plane. In practical terms, this would correspond to modifying the cost functions in Table 1 to consider the scattered electric field at different positions. We note, however, that in real-life experiments, the ability to obtain an optimal solution at any point on the output plane might be restricted by the properties of the scattering medium and the uniformity of the optical illumination." in the Discussion and Conclusions section. [R1] S M Popoff, et al., 2011 New J. Phys. 13 123021. DOI 10.1088/1367-2630/13/12/123021

Competing Interests: No competing interests were disclosed.
