Interferometric-Spectroscopy With Quantum-Light; Revealing Out-of-Time-Ordering Correlators

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We survey the inclusion of interferometric elements in nonlinear spectroscopy performed with quantum light. Controlled interference of electromagnetic fields coupled to matter can induce constructive or destructive contributions of microscopic coupling sequences (histories) of matter. Since quantum fields do not commute, quantum light signals are sensitive to the order of light-matter coupling sequence. Matter correlation functions are thus imprinted by different field factors, which depend on that order. We identify the associated quantum information obtained by controlling the weights of different contributing pathways, and offer several experimental schemes for recovering it. Nonlinear quantum response functions include out-of-time-ordering matter correlators (OTOC) which reveal how perturbations spread throughout a quantum system (information scrambling). Their effect becomes most notable when using ultrafast pulse sequences with respect to the path difference induced by the interferometer. OTOC appear in quantum-informatics studies in other fields, including black holes, high energy, and condensed matter physics.

I. INTRODUCTION

The quantum nature of light can affect and be utilized to steer optical signals in many ways [1]. First, unique properties such as photon entanglement show nonclassical bandwidth characteristics, offering new ways to study many-body correlations. Multi-photon collective resonances [2] excited by entangled light sources give access to matter information not available with classical sources. Second, low-intensity quantum light sources are useful for various sensing applications. An entangled pair can be generated such that each photon has a very different frequency regime. This provides a convenient way to probe matter information in less accessible frequency ranges (e.g. IR, XUV), while measuring visible photons [3]. Another property of quantum light is the larger parameter space which enables sensing applications such as phase imaging [4], quantum sensing networks , and spectrally resolved optical phase profiles [5]. Third, quantum light allows to extend nonlinear spectroscopic signals down to the few-photon level where the quantum nature of the field must be taken into account. Observed effects include the strong light-matter coupling in cavities [6], the enhancement of the medium's nonlinearity [7] and linear pump-signal scaling of the two-photon absorption processes [8]. The parameter-set of the photon wave-function offers novel control knobs that supplement classical parameters, such as frequencies and time delays [9]. Quantum light opens a possibility to shape and control excitation pathways of matter in a way not possible by shaped classical pulses, and results in steering exciton relaxation in molecular systems [10]. Fourth, quantum light sources enhance phase measurements beyond the shot-noise limit and have been recently shown experimentally to enhance the performance of imaging schemes [11]. The spatial resolution may be enhanced in quantum imaging applications, quantum-optical coherence, as well as in quantum lithographic applications. Quantum imaging with entangled light can achieve enhanced resolution, and quantum metrology can overcome the shot noise limit [12].

In this perspective, we survey emerging novel spectroscopic techniques made possible by interferometric setups. Each setup includes three components: an incoming quantum light source (preparation), field-matter coupling, and detection. Interference of optical fields has a rich history of experimentally unraveling illusive physical phenomena. Due to its linear dispersion, path differences of light are associated with time delays, rendering controlled interference setups (interferometers) valuable sensitive phase evaluation devices. Quantum probes are more complex and potentially carry additional information [13]. This can be used to outperform purely classical schemes in precision measurements, due to higher Fisher information and corresponding lower Cramér-Rao bound [14]. Setups based on Mach-Zehnder Rarity *et al.* [15], Hong-Ou-Mandel [16], and Franson [17] interferometers with quantum light are sensitive to the change in photon statistics of quantum light upon coupling to matter, and can be revealed by coincidence detection with multiple detectors [17–19]. Quantum-enhanced interferometers – such as the ones used for the observation of gravitational waves [20, 21] – indeed demonstrate unprecedented phase estimation precision with high loss tolerance at lower photon flux [22–26], and in wide-field imaging [27]. Generally, interferometers shuffle the time ordering of the input fields, creating a superposition of possible histories related to different paths. When the input field is a com-

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position of well separated pulses, this effect is expressed in the output of time- resolved signals. This superimposed re-ordering can be described via linear transformations, and further classified into symmetry groups (Sec. II), suggesting a systematic classification of experimental setups. Here, we consider coupling a quantum material system of interest, to auxiliary electromagnetic fields under such conditions. The probe may propagate through known interference at any stage – prior, during or after the coupling with matter – and finally detected. Our approach [28] is closely related to the space-time tomographic mapping of superdensity operators [29]. We connect quantum information contributions to matter quantities.

Matter does not affect the state of coherent light, thus, all light-matter coupling histories (Liouville pathways) contribute with the same weight of the field. Their sum defines the classical response function. In contrast, other states of light (e.g., fixed number of photons) may carry different amplitude for each possible pathway. Each light-matter interaction sequence is then associated with a unique generalized response, which constitutes the classical (nonlinear) response [30–32]. From this point of view, the excess (quantum) information carried by the probe, allows one to open the measurement black box and closely observe the triggering sequence. Interferometric transformations of such states of light, correspond to altering between superimposed pathways judiciously (in lossless transformations). The interference of classical probes results in an output modulated by the classical response. The algebraic-geometric view of interferometry, implies that invariant observables transform as scalars (e.g., total photon number) and thus detected as constant flux. Others, (e.g., single polarization after basis transformation) are sensitive to rotations and thus show oscillations in measurements [33, 34]. The latter may carry useful information and should be studied in more detail.

The response to classical light is given by correlation functions of the dipole operator $V(t_n)\,V(t_{n-1})\ldots V(t_1)$ with a specific prescription of time ordering, we label them time ordered correlators (TOC). Multiphoton interferometric signals can give rise to generalized response functions, composed of light-matter coupling sequences in irregular time-ordering. These are broadly denoted out-of-time-ordering correlators (OTOC). This terminology will be precisely defined in Sec. III A. OTOC are attracting considerable attention in other fields, connected to quantum information dynamics of interacting many-body (closed) systems [35–52]. They provide useful signatures of quantum information scrambling, motivated by the quantum analogue of the "butterfly effect". In chaotic quantum systems, they grow exponentially fast (in time) prior to the Ehrenfest time (timescale in which quantum effects dominate) [35, 36, 40, 53, 54]. Otherwise, it follows a powerlaw at most [42, 44, 45]. Computation of multipoint space-time correlations in such setups can be carried out using the density operator formalism in Liouville space, introduced in [28] and more

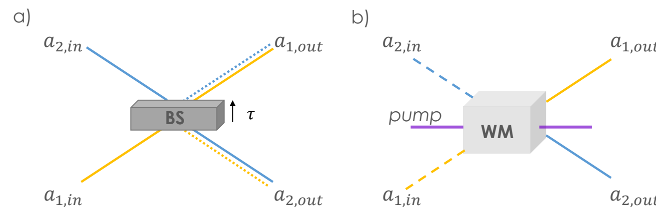

Figure 1. Interferometric elements. (a) Passive interferometric element, used to combine two incoming modes to superposed outgoing modes with the input output relation given by Eq. 1, where $\phi = \omega\tau$. (b) An active interferometric element, involves a nonlinear parametric process such as three-wave mixing, and four-wave mixing. Dashed lines represent modes that are initially in the vacuum state. In four-wave mixing, one of the inputs is populated, e.g., attenuated pump as in Eq. 4.

recently in [29]. Alternatively, it can be done using the wave-function (Hilbert space) approach [40]. The latter circumvents perplexing paradoxes one would inevitably encounter in *time symmetric* formulations to quantum mechanics [55, 56].

In Sec. II we describe the building blocks of linear and nonlinear interferometry. In Sec. III we present a general expression for the observable in Liouville space. In III A we discuss the contributions of OTOC obtained by post-coupling interferometry (detection). We then introduce novel pathway selection protocols such as exchange-phase cycling in Sec. III B, and time domain sorting in Sec. III C – both enabled by state-preparation in interferometric setups. We discuss an approach for harnessing Einstein-Podolsky-Rosen (EPR) correlations for enhanced joint time-frequency resolutions in Sec. III D. Finally we summarize our results in Sec. IV.

II. BUILDING BLOCKS OF NTERFEROMETRIC SIGNALS

Interferometry can be classified into two main types: passive-linear or active-nonlinear wave-mixing. Introducing a group-theoretic description of the interferometric elements, reveals clear notions regarding available information in terms of conserved currents. Matter degrees of freedom may introduce broken symmetries, altering otherwise-invariant quantities in terms of photon flux. We consider optical modes described by boson annihilation (creation) operators a_i $\left(a_i^\dagger\right)$, satisfying $\left[a_i, a_j^\dagger\right] = \delta_{ij}$ and $[a_i, a_j] = 0$. In order to discuss their transformations under interferometric setups, we adopt the vector notation $\boldsymbol{a} \equiv (a_1, a_2)^T$.

A. Linear-passive interferometric elements

A generic linear interferometric setup is realized by arrays of beam-splitters (BS) as depicted in Fig. 1a, mirrors and phase elements. The input-output relations corresponding to the two-port device (BS) is described by the unitary transformation $\boldsymbol{a}_{\text{out}} = \hat{\mathcal{R}}_\phi \boldsymbol{a}_{\text{in}}$ and

$$\hat{\mathcal{R}}_\phi = \begin{pmatrix} T & iRe^{i\phi} \\ iRe^{-i\phi} & T \end{pmatrix} \tag{1}$$

Here T and R are the reflection and transmission coefficients such that $T^2 + R^2 = 1$, and ϕ is a relative phase employed e.g., by shifted BS or mirrors (assuming lossless BS). Such elements are employed in many interferometric schemes – historically highlighting different physical realizations(i.e., Mach-Zehnder, Franson, Michelson Sagnac, relying on combinations of optical modes). The symmetry group of such transformations becomes more apparent by introducing the Hermitian operators [57]

$$J_x = \frac{1}{2}\left(a_1^\dagger a_2 + a_2^\dagger a_1\right), \tag{2a}$$

$$J_y = -\frac{i}{2}\left(a_1^\dagger a_2 - a_2^\dagger a_1\right), \tag{2b}$$

$$J_z = \frac{1}{2}\left(a_1^\dagger a_1 - a_2^\dagger a_2\right), \tag{2c}$$

satisfying the commutation relations of the Lie algebra of $SU(2)$, $[J_i, J_j] = i\epsilon_{ijk} J_k$, where ϵ_{ijk} is the antisymmetric tensor and and $i, j, k \in x, y, z$ (the number operator $N = a_1^\dagger a_1 + a_2^\dagger a_2$ is proportional to the identity). Clearly, two-mode passive rotation corresponds to an $SU(2)$ transformation with the invariant (Casimir) $J^2 = \frac{N}{2}\left(\frac{N}{2}+1\right)$. Coupling to matter degrees of freedom in the interaction picture, is represented by a relative shift in the unitary evolution. This stems from the fact that for each path, the joint light-matter system is evolved for different duration, giving rise to the out-of-time-ordering matter correlators (OTOC) and discussed in Sec. III A. The evolution operator is given by $\hat{U}(t, t') \equiv \exp\left\{-\frac{i}{\hbar} H (t - t')\right\}$, where $H = H_\phi + H_\mu + H_{\mu\phi}$. In the absence of matter, the path difference merely yields a linear phase $\phi = \omega\tau$ corresponding to time translations of the combined modes.

B. Nonlinear-active interferometric elements

Active interferometric elements constitute nonlinear combinations of fields, e.g., n-wave mixing processes. Three-wave mixing generates entangled photon pairs through parametric down conversion. A pump photon is down converted to a pair of spontaneously generated entangled photons. Four-wave mixing (FWM) induces further quadrature squeezing Reid and Walls [58], which attracted considerable attention from the early days of quantum enhanced metrology, aiming to improve the detection of gravitation wave Caves [20].

Nonlinear interferometric techniques present several merits. They utilize remarkable bandwidth extension with narrowband probes Shaked *et al.* [26], improved contrast in phase measurements with sub-shotnoise scaling – while maintaining these enhanced features with impressive loss tolerance Hudelist *et al.* [22], Li *et al.* [23], Anderson *et al.* [24], Manceau *et al.* [25], Frascella *et al.* [27], Du *et al.* [59]. It can be employed in the detection process to characterize time-domain light-matter pathways as further discussed in Sec. III C.

To characterize a two-photon FWM operation in terms of a transformation, we introduce the operators

$$K_x = \frac{1}{2}\left(a_1^\dagger a_2^\dagger + a_1 a_2\right), \tag{3a}$$

$$K_y = -\frac{i}{2}\left(a_1^\dagger a_2^\dagger - a_1 a_2\right), \tag{3b}$$

$$K_z = \frac{1}{2}\left(a_1^\dagger a_1 + a_2 a_2^\dagger\right), \tag{3c}$$

satisfying the of the Lorentz group $SU(1,1)$; $[K_x, K_y] = -iK_z$, $[K_y, K_z] = iK_x$ and $[K_z, K_x] = iK_y$, and the Casimir (invariant) $K^2 = J_z(J_z + 1)$. To demonstrate their effect, we consider a realization of this transformation in which one of the inputs in Fig. 1b is populated by an attenuated pump, with a relative delay δ with respect to the activating pump. The scattering matrix is then given by

$$\begin{pmatrix} a_1 \\ a_2^\dagger \end{pmatrix}_{\text{out}} = \begin{pmatrix} \cosh\beta & e^{-i\delta}\sinh\beta \\ e^{i\delta}\sinh\beta & \cosh\beta \end{pmatrix} \begin{pmatrix} a_1 \\ a_2^\dagger \end{pmatrix}_{\text{in}}, \tag{4}$$

where β is related to the reflectivity of the FWM Yurke *et al.* [57]. The J operators transform under the passive elements using $SU(2)$ rotations which are (almost) equivalent to manifold-preserving rotations in 3D. In contrast, the active elements impose Lorentz boosts on the K operators which corresponds to quadrature squeezing (manifold-shearing). In addition to the benefits derived from narrowband pump operation (above), high-order mixing generate particularly useful set for sorting through individual spontaneous processes (Sec. III C).

III. INTERFEROMETRIC QUANTUM SPECTROSCOPY – AN OPEN FRONTIER

Quantum fields are represented by operator quantities, in contrast to classical fields, which are c-numbers. Pertubative expansion of field-matter interactions yields optical signals expressed as multi-point correlation functions. The relative order of the dipole operators impacts the detected time ordering of the field-matter interactions and their expectation values. For non commuting fields, each arrangement of matter correlation function corresponds

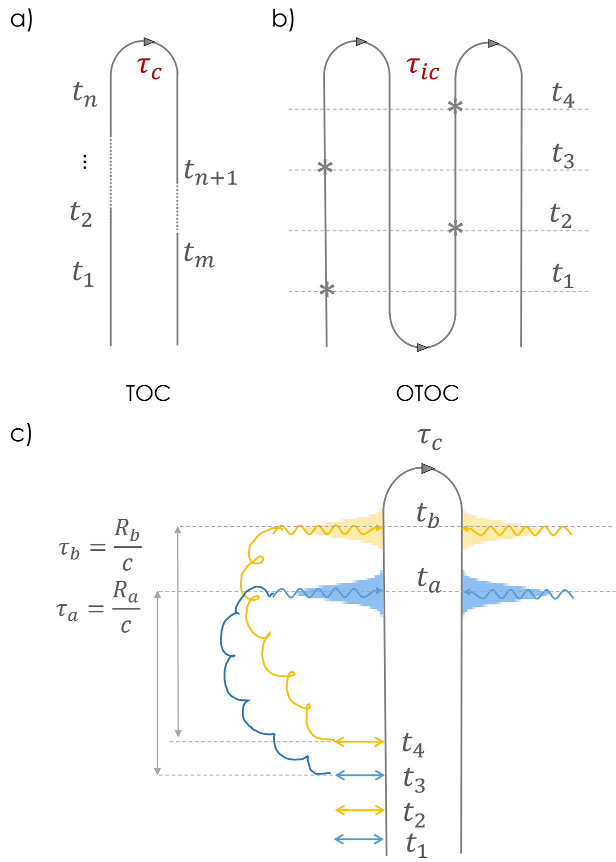

Figure 2. **Time-ordered Vs. out-of-time-ordering matter correlators**. (a) A fully time-ordered matter correlator (TOC) computed along a typical closed time contour τ_c. Such contribution compose the nonlinear response of matter upon coupling to classical light. (b) An OTOC diagram. Different optical paths in the interferometer re-arrange the radiative trajectories in the detection plane. The resulting OTOC are computed along an irregular wiggling time contour τ_{ic}. (c) A fully time-ordered loop diagrammatic representation of a possible process that contributes to the 4^{th} order optical signal. Two photons interact with a sample, then detected in coincidence following free propagation duration of τ_a and τ_b. The correlators are computed along the closed time contour τ_c from the distant past to the present back to the past.

to a different field correlation function. Thus, various detection schemes, can provide different information regarding the many-body dynamics.

Interferometric setups typically mix several modes and thus the mapping between physical interaction occurrences and their detection time is not straightforward. Below, we survey several approaches to manipulate and distinguish between time ordered events, and show how OTOC show up in measurements.

A. Out-of-time-ordering correlators – order of arrival Vs. order of interaction

When a quantum system is coupled to a classical field the, response is given by correlation functions of the form $\langle V(t_m)\cdots V(t_{n+1})V(t_n)\cdots V(t_2)V(t_1)\rangle$ represented by the loop diagram in Fig. 2a. Time proceeds forward in the left branch from t_1 to t_n, then proceeds backward on the right branch from t_{n+1} to t_m. We denote these time ordered correlators (TOC). Interferometric measurements with quantum light are given by more complex objects where time proceeds forward and backward multiple times, as shown in Fig. 2b. These are denoted OTOC.

As an example we consider the loop diagram shown in Fig. 2c. Matter correlation function in this example can be read off the diagram as $\langle \mathcal{T}_c V(t_4)V(t_3)V(t_2)V(t_1)\rangle$, where $\mathcal{T}_c$ is a time ordering operator corresponding to the closed time contour τ_c. Light-matter interaction events are ordered along the loop, and propagated forward in time from the distant past on the left branch, and backwards in time to the past on the right branch. In this example, $t_4 > t_3 > t_2 > t_1$ such that the propagation is always forward in time as depicted in Fig. 2c. An OTOC appears when the time-flow may be inverted backwards in some intervals of the correlation function, e.g., $\langle \mathcal{T} V(t_4)V(t_2)V(t_3)V(t_1)\rangle$, which yields reverse evolution between the second and third operators and depicted in Fig. 2b. Two-photon input in an interferometer introduces several modified time-orderings, resulting in irregular time-flow at the output. The coupling of light with matter parametrizes the matter correlation functions along the wiggling contour such as the one introduced in Ref. [40] for computations of OTOC in closed systems. This can be interpreted as interference of past and future contributions of matter multipoint correlation functions. However, this terminology can be avoided. We next review the interferometric transformation in terms of the detected signal.

When the electromagnetic field propagation direction is known, the Jordan-Schwinger map (JSM) is described by Stokes operators which follow the Lie algebra of $SU(2)$ symmetry group [57, 60–62]. Thus, (passive) interferometric setups can be described using a sequence of $SU(2)$ rotations (see Sec. II). The Hong-Ou-Mandel (HOM) interferometer depicted in Fig. 3 [63], is the simplest setup that gives rise to interference between future and past matter events. It combines two optical modes on a movable BS, which are then detected in coincidence using two detectors. The shift of the BS with respect to the center introduces controlled path differences between four distinct trajectories. Observables in this setup are composed purely of field operators that evolve according to the free electromagnetic Hamiltonian H_ϕ. Measurements are described using annihilation of modes in the far-field basis (post-rotation) as described by Glauber [64].

The light-matter coupling $H_{\mu\phi}$ is generally composed of operators from the joint Hilbert space. Light and matter degrees of freedom thus become entangled upon energy exchange. We consider an optical signal, generated from a general multipoint correlation function of field operators, given by (see Sec. S2 of the SM)

$$\mathcal{C}\left(t_1,...,t_n\right) = \left\langle \mathcal{T}\mathcal{O}_{t_1,...,t_n}\left(\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{E}}^{\dagger}\right) e^{-\frac{i}{\hbar}\int_{t_0}^{t} du \mathcal{H}_{\mu\phi,-}(u)} \right\rangle . \quad (5)$$

Here $\mathcal{C}\left(t_1,...,t_n\right)$ is an n-point correlation function where t is taken to be the latest time, $\mathcal{T}$ is the time-ordering superoperator, $\mathcal{O}_{t_1,...,t_n}$ is a superoperator composition of the electric field operators in Liouville space and $\langle\cdots\rangle \equiv \mathrm{tr}\left\{\cdots\rho\left(t_0\right)\right\}$ is the trace with respect to the initial state of the density operator. $\mathcal{H}_{\mu\phi,-}(t)$ is the interaction superoperator operating on a Hilbert space operator A as a commutator $\mathcal{H}_{\mu\phi,-}A \equiv \mathcal{H}_{\mu\phi}A - A\mathcal{H}_{\mu\phi}$ Mukamel [28]. $\boldsymbol{\mathcal{E}}\left(\boldsymbol{\mathcal{E}}^{\dagger}\right)$ is the positive (negative) frequency electric field components $\boldsymbol{E} = \boldsymbol{\mathcal{E}} + \boldsymbol{\mathcal{E}}^{\dagger}$. In the following illustration we are interested in expectation values of intensity correlations. To that end it is convenient to introduce the right-left superoperator notation, whereby left (right) superoperators $\mathcal{E}_L\left(\mathcal{E}_R\right)$, act on the density according to $\mathcal{E}_L\rho \equiv \mathcal{E}\rho$ $\left(\mathcal{E}_R\rho \equiv \rho\mathcal{E}\right)$. One can calculate the observables according to *order of arrival* at the detector, imposing the coupling description in this basis, $\boldsymbol{\mathcal{E}}_{\mathrm{detection}} = \hat{\mathcal{R}}\boldsymbol{\mathcal{E}}_{\mathrm{interaction}}$. Alternatively, the observable can be computed in the *order of interaction* with matter, in which $\mathcal{O}_{t_1,...,t_n}\left(\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{E}}^{\dagger}\right)$ will be expressed in basis of the interaction operators (rotation backwards, see Sec. S2 of the SM). In this description, the high dimensional space – accommodating both the electromagnetic field and the matter field of dim $H_\mu \oplus H_\phi$ – is unraveled by fully time-ordered correlation functions in the interaction picture. Since only part of the system (i.e. the electromagnetic field) is detected, path difference of the auxiliary degrees of freedom correspond to effective time-flow wiggling in the measured matter correlation function. When the observable $\mathcal{O}\left(\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{E}}^{\dagger}\right)$ is invariant under rotations (scalar e.g., $\boldsymbol{\mathcal{E}}^{\dagger}\cdot\boldsymbol{\mathcal{E}}$, or any function of the Casimir of the Stokes operators), no interference will be registered in the absence of matter. When the observable is basis dependent, interference between future and past matter pathways may show up in coincidence counting experiments. Notably, this is also where one would look for superior quantum performance with respect to phase-shift measurements [57].

Example of an OTOC contribution

Consider the setup sketched in Fig. 3. Two modes $\{E_a, E_b\}$ are prepared with a relative delay time τ, then interact with a sample. The modes are rotated by a BS and measured in coincidence respectively at space-time coordinates $\{\boldsymbol{r}_a t_a, \boldsymbol{r}_b t_b\}$. The recorded signal can be computed by Eq. 5, by simultaneous annihilation of two photons from both sides of the density operator

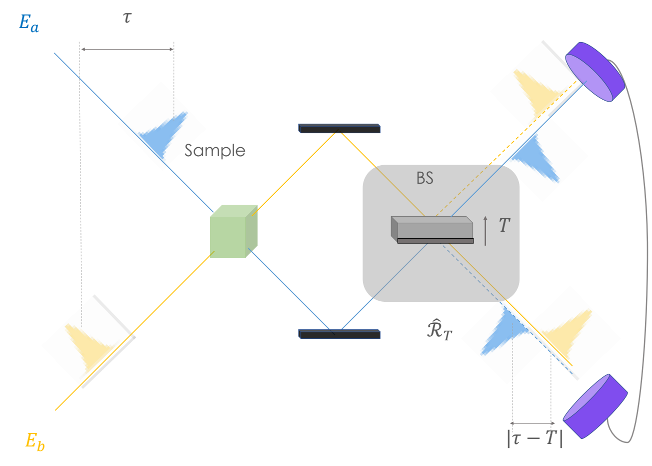

Figure 3. **OTOC detection via a Hong-Ou-Mandel interferometer**. Two quantum probes arranged in space-time wave-packets with relative delay time τ. Then the photons interact with matter (sample), and transformed using BS introducing several possible propagation trajectories of light-matter interaction in a Hong-Ou-Mandel interferometer configuration. The photons are time-resolved and measured in coincidence, forming matter correlation function on a wiggling time contour.

$$\mathcal{O}_{t_a,t_b}\left(\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{E}}^{\dagger}\right) = \mathcal{E}^{\dagger}_{R,a}\left(\boldsymbol{r}_a t_a\right)\mathcal{E}^{\dagger}_{R,b}\left(\boldsymbol{r}_b t_b\right)\mathcal{E}_{L,b}\left(\boldsymbol{r}_b t_b\right)\mathcal{E}_{L,a}\left(\boldsymbol{r}_a t_a\right) . \quad (6)$$

Eq. 6 projects the two-photon subspace of the density operator as a function of time at two designated detection positions $\{\boldsymbol{r}_a, \boldsymbol{r}_b\}$, annihilating two photons from the left and right. Crucially, both modes (a, b) are measured at the detection plane, and require change of basis with respect to the interaction plane, using the coupling Hamiltonian $\mathcal{H}_{\mu\phi}(t) = \boldsymbol{V}(\boldsymbol{r},t)\cdot\boldsymbol{E}(\boldsymbol{r},t)$ in the interaction-regime basis, and $\boldsymbol{V} = \boldsymbol{\mu} + \boldsymbol{\mu}^{\dagger}$ is the dipole operator (see Sec. S1 and S2 of the SM). A nonvanishing signal is recorded only if two photons are detected. The lowest nonvanishing order contributing to the signal (apart from noninteracting background) includes four events, the two modes are annihilated then created as a result of the interaction with the sample. Multiple processes contribute to the overall signal, however, we are interested to demonstrate a contribution which results in OTOC. Such contributions appear for example by considering the process displayed in Fig. 2a, followed by interferometry from the sample to the detectors which reorders the correlation function as shown in Fig. 2b.

We consider the two-photon initial state of light $|\Psi_0\rangle = \int d\omega_a d\omega_b \Phi\left(\omega_a,\omega_b\right) a^{\dagger}\left(\omega_a\right) b^{\dagger}\left(\omega_b\right)|\mathrm{vac}\rangle$, describing creation of the two modes from the vacuum with amplitude $\Phi\left(\omega_a,\omega_b\right) = \phi_a\left(\omega_a\right)\phi_b\left(\omega_b\right)$,and compute the process amplitude (see Sec. S2 of the SM). When the temporal distribution of the wavepackets ϵ is narrow in comparison to their relative delay τ, and the matter response time, one obtains sufficient temporal resolution to study the OTOC explicitly. We show this by approximating the temporal envelope by delta distributions $\phi_a(t) \to \delta_\epsilon(t-\tau)$, $\phi_b(t) \to \delta_\epsilon(t)$ and obtain

$$\mathcal{C}_{ab}(\tau) \propto \langle V_a(\tau)\, V_b(0)\, V_a(\tau)\, V_b(0)\rangle\,, \tag{7}$$

for $\tau = 2T$. Eq. 7 clearly reflects the wiggling (forward-backward-forward) time-flow of matter correlation functions carried by an auxiliary probe through an interferometer [65]. We stress that such processes contribute even with no temporal resolution, the delta distributions are invoked purely for illustration purposes. To illustrate this, we have computed the OTOC contribution obtained by applying an entangled pair generated by a spontaneous parametric down-converter pumped using a narrowband beam in Sec. II.B of the SM. Matter information is imprinted in the EM field according to the incidence time. The interaction may occur at various times (which are integrated upon in the interaction picture). The interference of the detected beams depends on equally distributed times dictated by the BS relative displacement, thanks to the linear group velocity of the field. This can be viewed as interfering past and future from the matter point of view (in the absence of losses).

Using this setup, one can measure rates of quantum information scrambling in molecules. Consider for example a two-color measurement whereby two wavepackets, each resonant with different bond (vibrational) or localized electronic state (core electrons) as depicted in Fig. 3. The signal will now carry quantum information regarding "cross talk" of these possibly far-apart channels as well as decoherence processes. Such measurements become particularly interesting for complex bio-molecules, since it can reveal the time and length-scales in which quantum dynamics are important.

The field rotation effect on the time-flow of matter correlation functions, bears a resemblance to the Keldysh rotation. The latter is used to benefit from the linear relations between different kinds of matter Green's functions [66]. Here, the left and right components of the field operators play a role corresponding to forward and backward evolution, by modifying the bra and the ket. Intuitively, the polarization degree of freedom transforms according to Pauli matrices, similar to the augmented Keldysh contour reported in [40]. The correspondence between augmented Keldysh contours and interference of auxiliary fields merits further study in a unified framework.

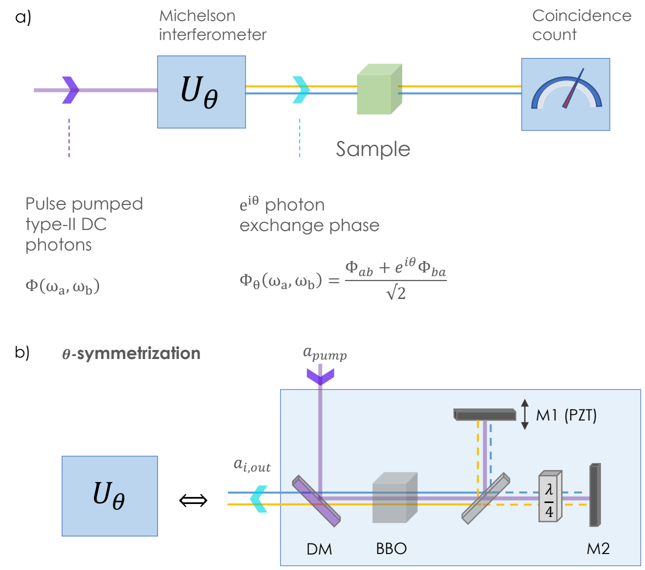

Figure 4. Exchange-phase-cycling scheme. (a) Two-photon exchange phase-cycling setup. The interferometric preparation process attributes θ phase difference with respect to photon exchange, denoted as θ–symmetrization prior to the coupling with the sample. Following the light-matter interaction, the photons are detected in coincidence, scanning the two-photon subspace of the density operator. (b) Implementation of θ-symmetrization via a modified Michelson interferometer (see text).

B. Exchange-phase-cycling protocols; quantum statistics and pathway selectivity

Quantum statistics is known to play a central role in shaping the interference patterns observed in coincidence detection of bosons [67], fermions [68] and fractional charges, e.g., quantum-hall quasiparticles [69]. Indistinguishable photon wavefunctions, such as (ideal) entangled photon pairs, are symmetric with respect to exchange. This is reflected in the generic form of wavefunction using a symmetrized pair amplitude $|\Psi_{\{2\}}\rangle = \frac{1}{\sqrt{2}}\int d\omega_a d\omega_b \left[\Phi(\omega_a,\omega_b) + \Phi(\omega_b,\omega_a)\right] a^\dagger(\omega_a)\, b^\dagger(\omega_b)\,|\mathrm{vac}\rangle$, where $a(\omega)$ $\left(a^\dagger(\omega)\right)$ and $b(\omega)$ $\left(b^\dagger(\omega)\right)$ are annihilation (creation) photon operators applied on the vacuum $|\mathrm{vac}\rangle$. In practice, entangled photon pairs are distinguishable owing to variations in the quantum channel responsible for their generation. Orthogonally polarized photon wave packets of entangled pair produced in a type-II parametric down conversion using an ultrashort pump pulse, may be rather distinguishable. Each polarization has a different bandwidth due to the dispersion characteristics of the birefringent crystal [70]. The time-frequency signature invokes some degree of distinguishability resulting in reduced interference contrast due to a nonvanishing exchange phase. Interestingly, the exchange phase of such entangled pair can be set in

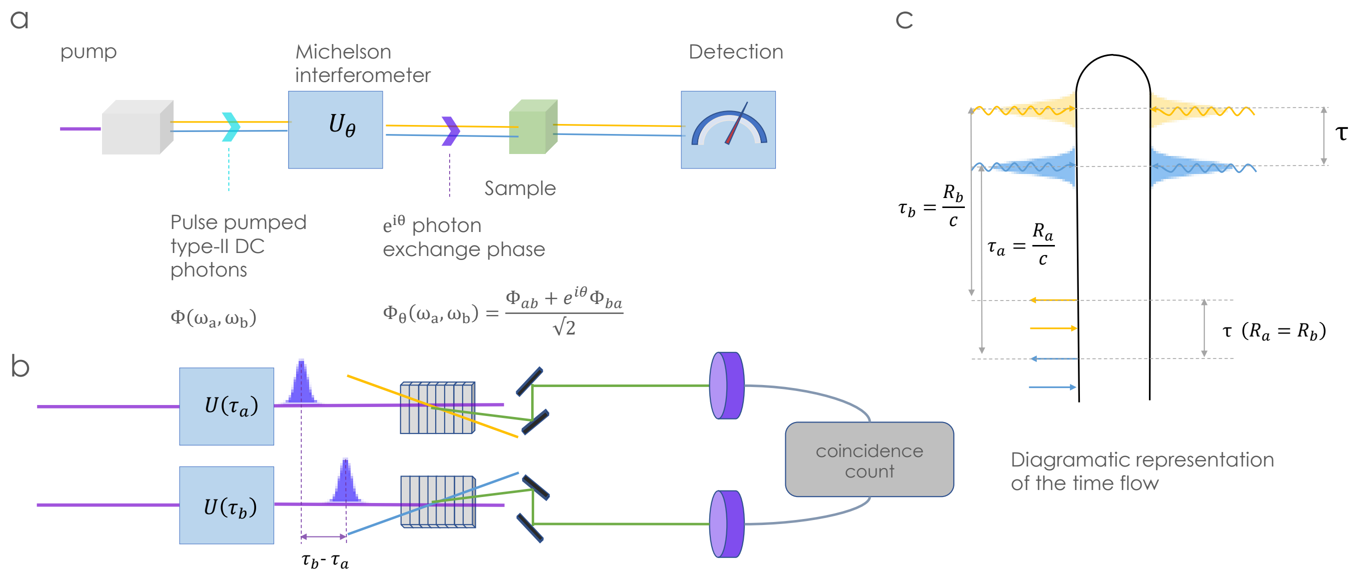

Figure 5. Time-domain switching platform. (a) A θ–symmetrized entangled pair is generated and directed and coupled to a sample. (b) The incident photons (yellow and blue) are separated using a polarization beam splitter (not shown), then scanned in time domain using an up conversion with the same (generating) a delayed pump beam ($U(T_a)$ and $U(T_b)$). The up-converted photons are detected in coincidence. (c) Time-domain diagrammatic description. Emitted light from matter de-excitation undergoes free (retarded) propagation to the detector. Ultrafast time-domain detection scheme with controlled time delay ($\tau = \tau_b - \tau_a$), used as a switch between pathways.

a controlled manner using the Michelson interferometer setup, producing a θ–symmetrized amplitude [70],

$$|\Psi_{\{2\}}\rangle = \int d\omega_a d\omega_b \Phi_\theta(\omega_a, \omega_b)\, a^\dagger(\omega_a)\, b^\dagger(\omega_b)\, |\text{vac}\rangle, \quad (8)$$

where $\Phi_\theta(\omega_a, \omega_b) = \left[\Phi(\omega_a, \omega_b) + e^{i\theta}\Phi(\omega_a, \omega_b)\right]/\sqrt{2}$. The exchange phase can thus be used to manipulate photonic pathways [71]. The pathway in which photon a is coupled to the matter at time t_1 preceding its entangled counterpart $(\omega_a; t_1) \to (\omega_b; t_2)$, and the opposite trajectory $(\omega_b; t_1) \to (\omega_a; t_2)$, carry a valuable phase difference. Repeating the measurement with different values of θ renders a set of signals from which single light-matter interaction pathways can be isolated (pathway selectivity). Generally, preparation interferometric procedures can extend this notion to an N photons amplitudes. N photons exchange phase cycling procedure introduces $\binom{N}{2}$ independent phases using the amplitude

$$\Phi_{\boldsymbol{\Theta}}(\omega_1, ..., \omega_N) = \mathcal{N}^{-1} \sum_{\{i,j\}} e^{i\theta_{ij}} \hat{\mathcal{P}}_{ij} \Phi(\omega_1, ..., \omega_N), \quad (9)$$

summing over all pair permutations $\{i, j\}$ with the normalization $\mathcal{N} = \sqrt{\binom{N}{2} + 1}$. $\hat{\mathcal{P}}_{ij}$ is the exchange operator between the i and j photons $\hat{\mathcal{P}}_{ij}\Phi_{\boldsymbol{\Theta}}(\omega_1, ..., \omega_i, ..., \omega_j, ..., \omega_N) = \Phi_{\boldsymbol{\Theta}}(\omega_1, ..., \omega_j, ..., \omega_i, ..., \omega_N)$. Generating a set of signals with independent exchange phases, one may independently access different pathways.

C. Time-domain QED – ultrafast pathway switching

Remarkable progress in time domain detection techniques, allows the observation of quantum electrodynamic (QED) processes such as electric-field vacuum fluctuations in subcycle scale [72, 73] and bunching at the femtosecond timescale [74]. This offers novel experimental possibilities, such as unraveling light-matter (spontaneous) pathways and sorting between relaxation mechanisms.

A Liouville pathway represents a distinct time ordering of events. Thus, coincidence measurements with a controlled delay, may prove useful for discriminating light-matter absorption-emission sequences. It is possible to recover the temporal profile of each photon of an entangled pair using setups such as the one reported in [75, 76]. Similar to the exchange-phase-cycling protocols, it relies on distinguishability to sort photonic degrees of freedom at the detection process. In . 5 we demonstrate this principle using polarization sorting of unidirectional entangled photon-pair. Controlled distinguishability can be employed by θ–symmetrization, or by applying more sophisticated single photon phase shaping techniques using electrooptic modulation [77].

All possible pathways contribute to the quantum state of the light-matter system. Elimination of multiple pathways is possible by applying a Fock state with fixed number of photons in conjunction with ultrafast time-domain coincidence detection. To demonstrate this, we consider two photons $N = 2$ as shown in Fig. 5 that undergo a coupling to matter and then detected in coincidence. Time domain scanning technique is employed based on up-conversion process [75, 76]. By fixing the detection time, only events in which both photons arrive simultaneously are counted. We define the temporally gated coincidence count [78]

$$\mathcal{C}\left(\theta,\bar{t}_a,\bar{t}_b\right) = \int dt_a dt_b \mathcal{D}\left(\bar{t}_a,t_a\right)\mathcal{D}\left(\bar{t}_b,t_b\right) W_B^{(2)}\left(t_a,t_b\right), \quad (10a)$$

$$W_B^{(2)} = \left\langle \mathcal{T}\mathcal{E}_{a,R}^{\dagger}\left(t_a\right)\mathcal{E}_{b,R}^{\dagger}\left(t_b\right)\mathcal{E}_{b,L}\left(t_b\right)\mathcal{E}_{a,L}\left(t_a\right) \times \exp\left\{-\frac{i}{\hbar}\int_{-\infty}^{t^*} du\, H_{int,-}\left(u\right)\right\}\right\rangle. \quad (10b)$$

Here $\mathcal{E}_R\left(\mathcal{E}_L\right)$ denotes the electromagnetic field superoperator that acts on a Hilbert space operator A from the right (left), $\mathcal{E}_R A \equiv A\mathcal{E}$ $\left(\mathcal{E}_L A \equiv \mathcal{E}A\right)$. Eq. 10a describes the the time-domain gated coincidence, given by integration over the bare signal given in Eq. 10b weighted by the temporal gating functions $\mathcal{D}_i\left(\bar{t}_i,t_i\right) = \left|F_t\left(t_i,\bar{t}_i\right)\right|^2$ which are determined by the pump temporal envelope. Since both photon emission events are spontaneous, no special meaning is attributed to the arrival time of a single photon, the time difference between detection events reveals the time ordering of emission events. This suggests an additional signal, that can be obtained experimentally by scanning and summing all coincidence events in which the relative pump delay $\tau = \tau_b - \tau_a$ is fixed. This corresponds to integration over the last interaction time of a temporally gated coincidence count, defining the signal

$$\mathcal{S}\left(\tau\right) = \int d\bar{t}_a \mathcal{C}\left(\theta,\bar{t}_a,\bar{t}_a+\tau\right). \quad (11)$$

When the pump is shorter than the measured photons wavepackets as depicted in Fig. 5c,each emission event is associated with a single detection, eliminating reversed order processes contributions in the coincidence count. Strikingly, when applied together with the $\theta-$symmetrized amplitude as in Fig. 5b, the signal is sensitive to the exchange in the order of the interactions (exchanging intermediate blue and yellow arrows). Thus, potentially sorting between pathways in addition to reversed emission discrimination.

D. Time-frequency coincidence of entangled photons

Joint properties of systems described by an entangled state are not necessarily constrained by uncertainty restrictions that apply to single systems. By exploiting some distinguishability handles (polarization, color, etc.), it is possible measure simultaneously conjugate properties in an apparent violation of uncertainty relations, e.g., joint position-momentum detection of EPR states (See Sec. 4 of the SM). Such phenomenon is easily demystified using the appropriate definitions for conjugate quantities in terms of the many-body wavefunction [79]. These type of nonlocal effects can be further employed to perform quantum microscopy and spectroscopy with unprecedented joint resolutions.

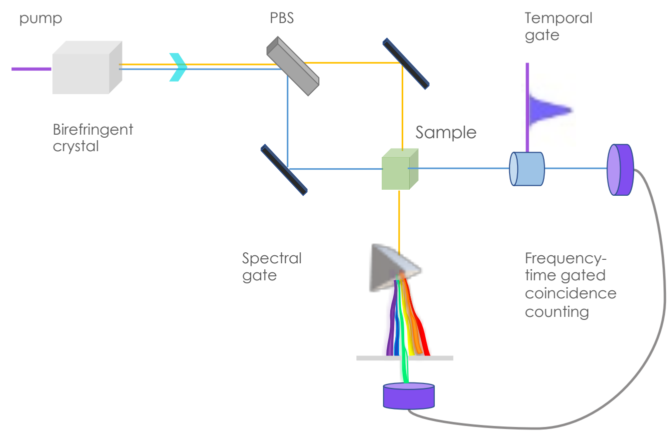

Figure 6. Frequency-time gated coincidence counting setup. Entangled photon pair separated using a polarization beam splitter (PBS). Then coupled to a sample and detected individually. One photon is characterized spectrally while the other in time domain. The photons are ultimately measured in coincidence, producing two dimensional spectra-temporal information with superior joint resolution compared with classical sources.

In the setup described in Fig. 6, time-frequency entangled photon pair is coupled to matter. One photon is detected in the time domain using ultrafast up-conversion detection technique, revealing its temporal profile. Its entangled counterpart is spectrally gated, thus recovering its frequency profile. The photons are measured in coincidence,

$$\mathcal{C}\left(\bar{t}_s,\bar{\omega}_i\right) = \left\langle E_{s,R}^{\dagger}\left(\bar{t}_s\right)E_{i,R}^{\dagger}\left(\bar{\omega}_i\right)E_{i,L}\left(\bar{\omega}_i\right)E_{s,L}\left(\bar{t}_s\right) \exp\left\{-\frac{i}{\hbar}\int_{-\infty}^{t^*} du H_{\mathrm{int},-}\left(u\right)\right\}\right\rangle. \quad (12)$$

Here $\bar{t}_s$ and $\bar{\omega}_i$ are the scanned time and frequency covered by the respective gates. From the initial frequency correlations of the photons, some knowledge can be obtained regarding the spectral spread of the temporally gated photon, beyond the minimal time-frequency uncertainty. This is possible thanks to the frequency gating of its counterpart, combined with the initial nonlocal correlations. Complementary information is obtained for the frequency gated photon. The two dimensional time-frequency map, potentially exhibits superior joint resolution. The main control parameters are then the gating functions and the initial state of the light (pump bandwidth and crystal length).

IV. SUMMARY

This perspective article surveys several schemes in which interferometry can be combined with quantum states of light in spectroscopy applications. We distinguish between linear and nonlinear interferometric techniques, and describe them using the formalism of linear transformations; relating the input to the output ports. Inclusion of matter degrees of freedom along the electromagnetic flux lines breaks the symmetries of these linear transformation and induces photocurrent that carries matter information. For simplicity, we have considered a colinear propagation direction after the light matter interaction. Phase matching conditions, can generate additional radiation directions induced by nonlinear spontaneous processes, that can be associated with altered transformations. Our algebraic-geometric description of interferometric building blocks, offers a simple explanation for the nonlocal light-matter state after the coupling in terms of vector rotations. These transformations can be employed either before, or after the coupling to matter. These affect the resulting observables, and may give access to different processes as shown above.

Quantum interferometry combined with sequences of light-matter interactions can single out noncausal contributions to nonlinear response functions. Usually such contributions are uniformly summed when classical light probes are involved. Quantum states of light offer sensitivity to the order of events and thus give weights to different pathways. We identify in this case the excess (quantum) information with pathway selectivity. Distinguishability between pathways can be used to separate decay channels in many-body systems, sorting them separately in experiments. OTOC contribute naturally to the interferometric-spectroscopy signals. Distinct OTOC can be extracted in full and measured by ultrafast pulse sequences. Ranging from a single molecule to many-body systems, OTOC tells a story regarding the quantum information scrambling; how a single perturbation that propagates through a quantum system affects different degrees of freedom. Characterizing such behavior becomes increasingly important for materials designed for novel quantum technologies. Also for detecting quantum coherent pathways in systems in which it is not clear whether there are any. From a theoretical point of view, such analysis poses an interesting inference-challenge for few-photon detection of distant atomic processes occurring in curved space-time (which are outside the scope of this perspective).

SUPPLEMENTARY MATERIAL

The supplementary material includes detailed and self-contained derivations of the mathematical expressions used in the main text.

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DATA AVAILABILITY

All the information used in this manuscript is available in the supplementary material files.

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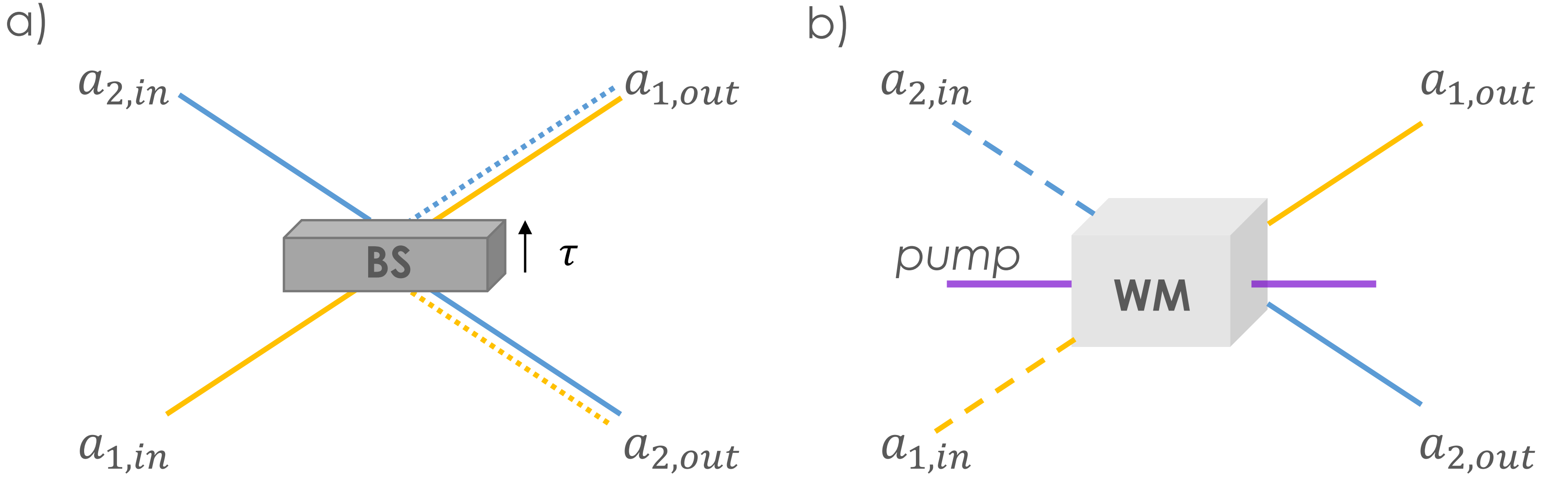
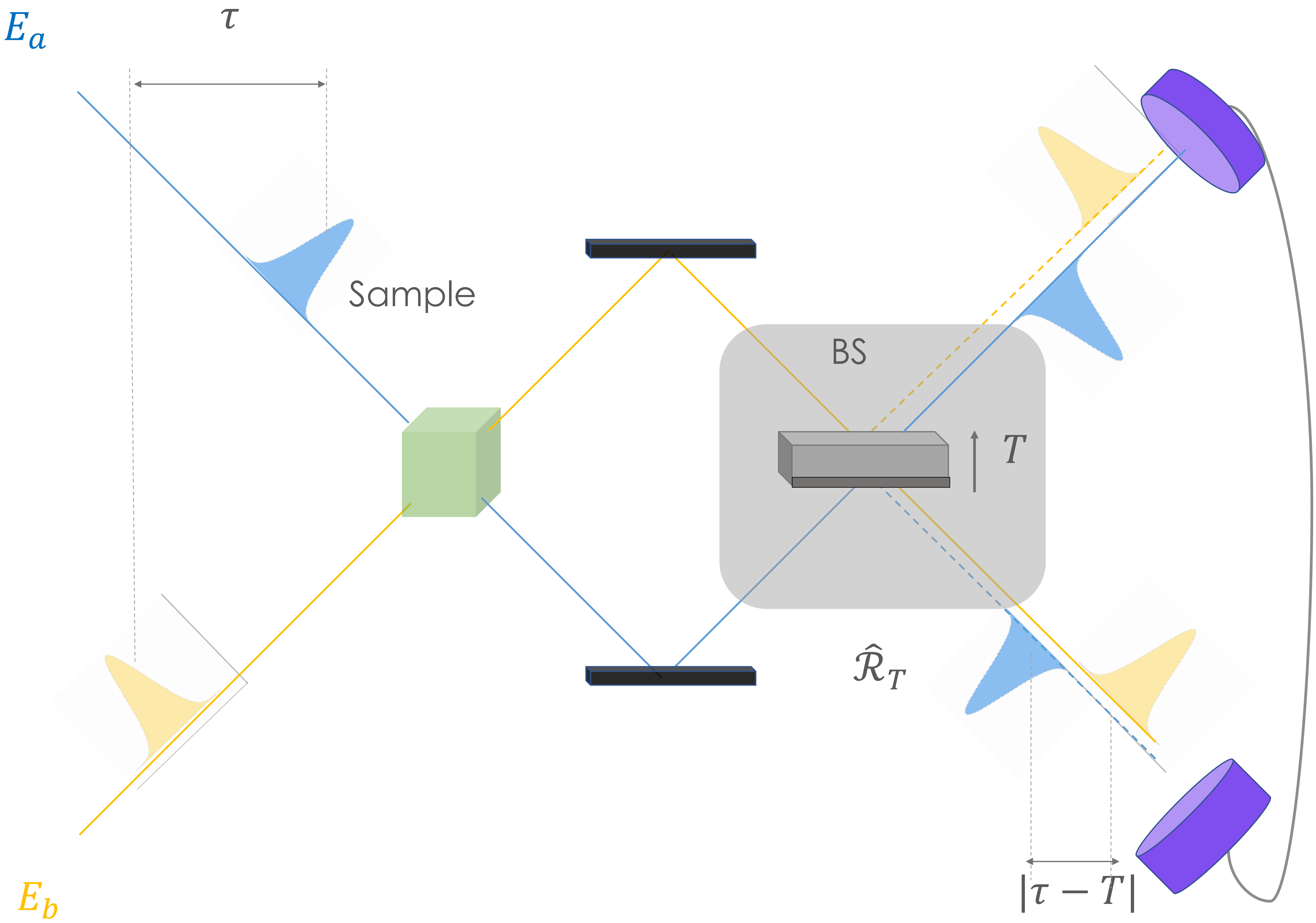
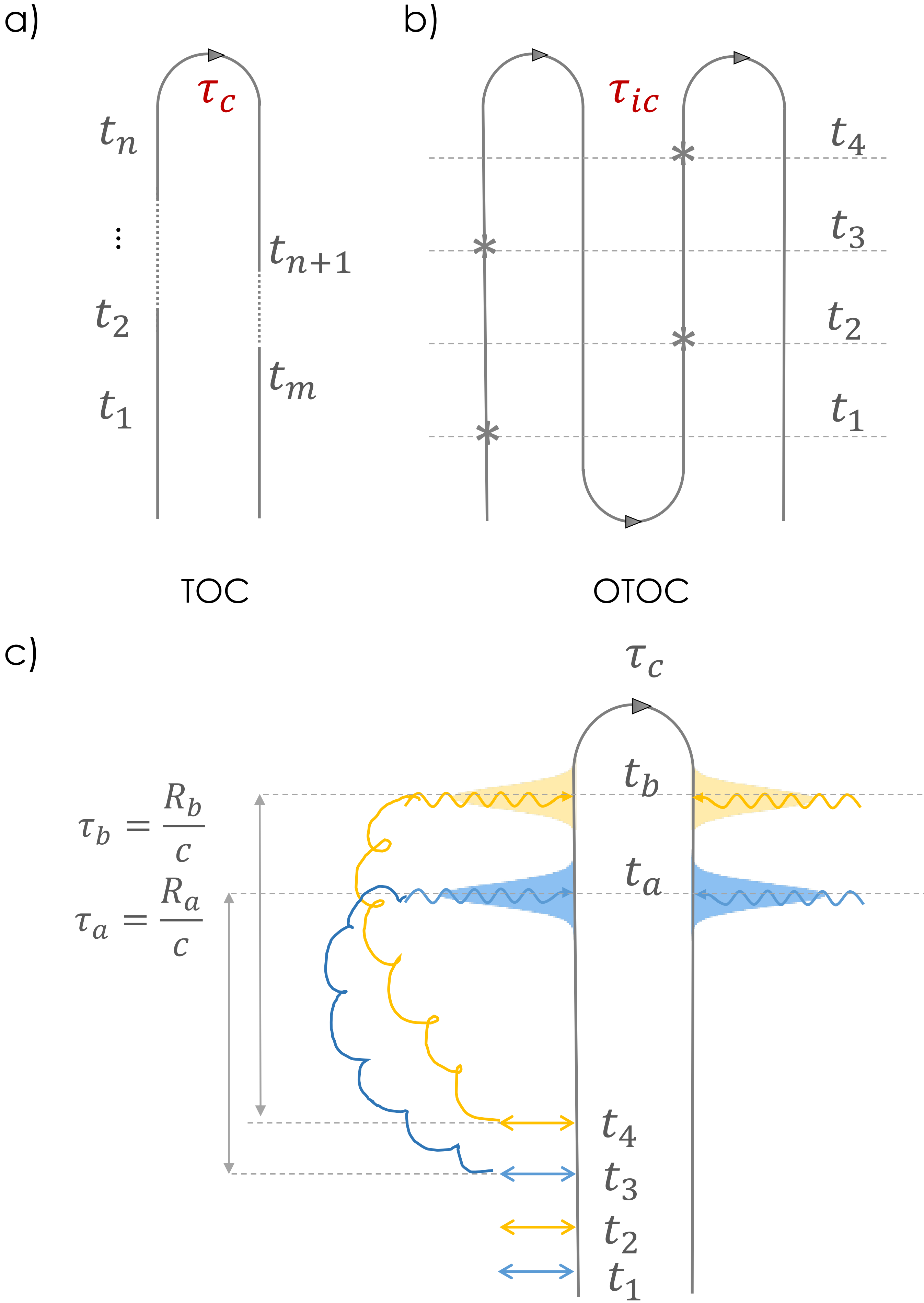
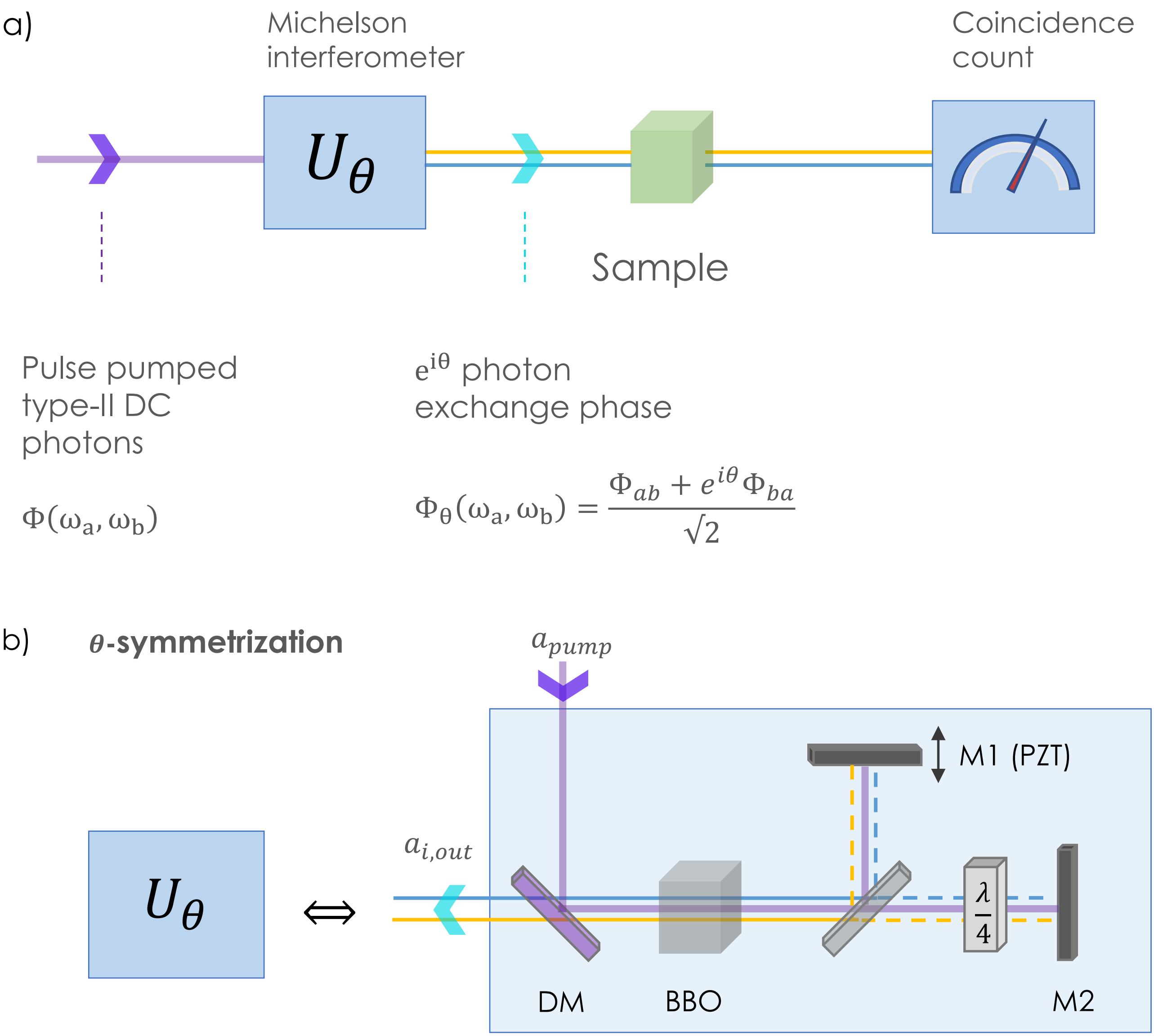


a)
$a_{2,in}$
$a_{1,out}$
BS
τ
$a_{1,in}$
$a_{2,out}$
b)
$a_{2,in}$
$a_{1,out}$
pump
WM
$a_{1,in}$
$a_{2,out}$

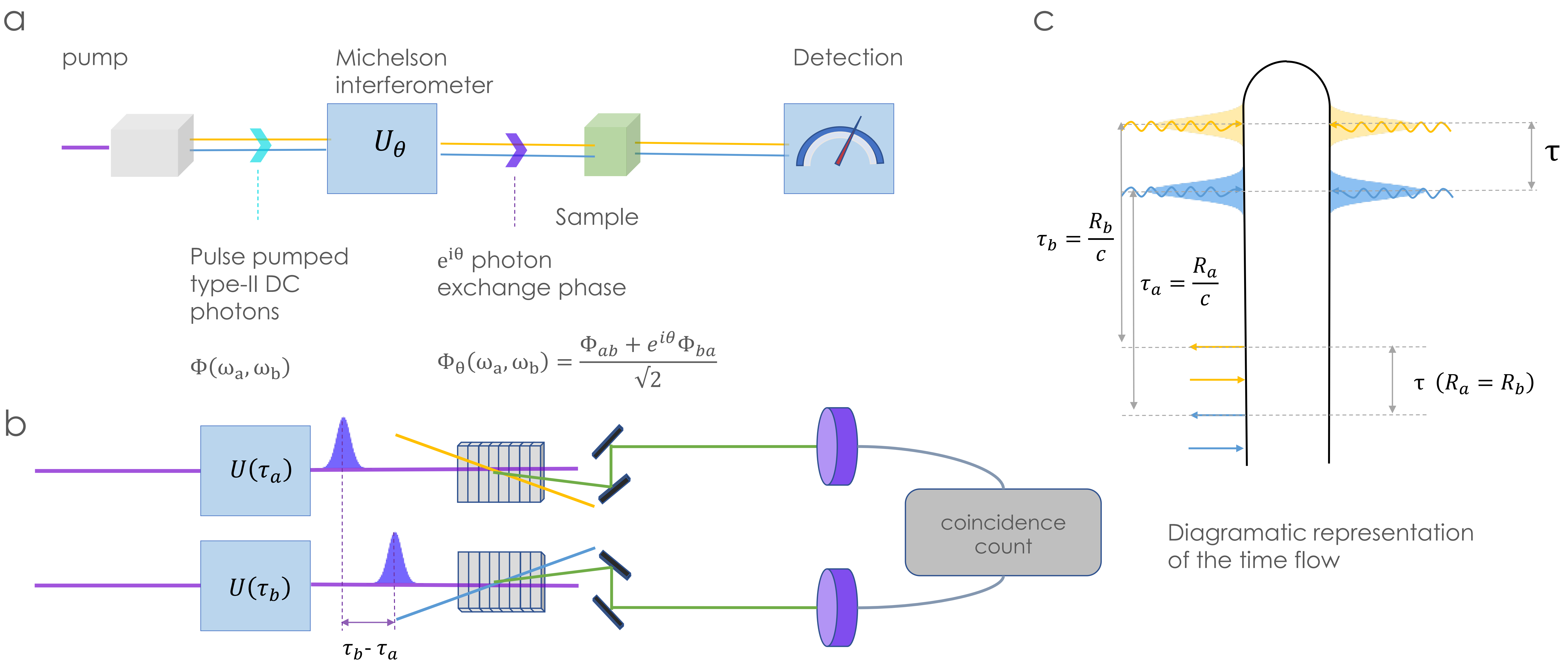


E_a
τ
Sample
BS
T
$\hat{\mathcal{R}}_T$
$|\tau - T|$
E_b

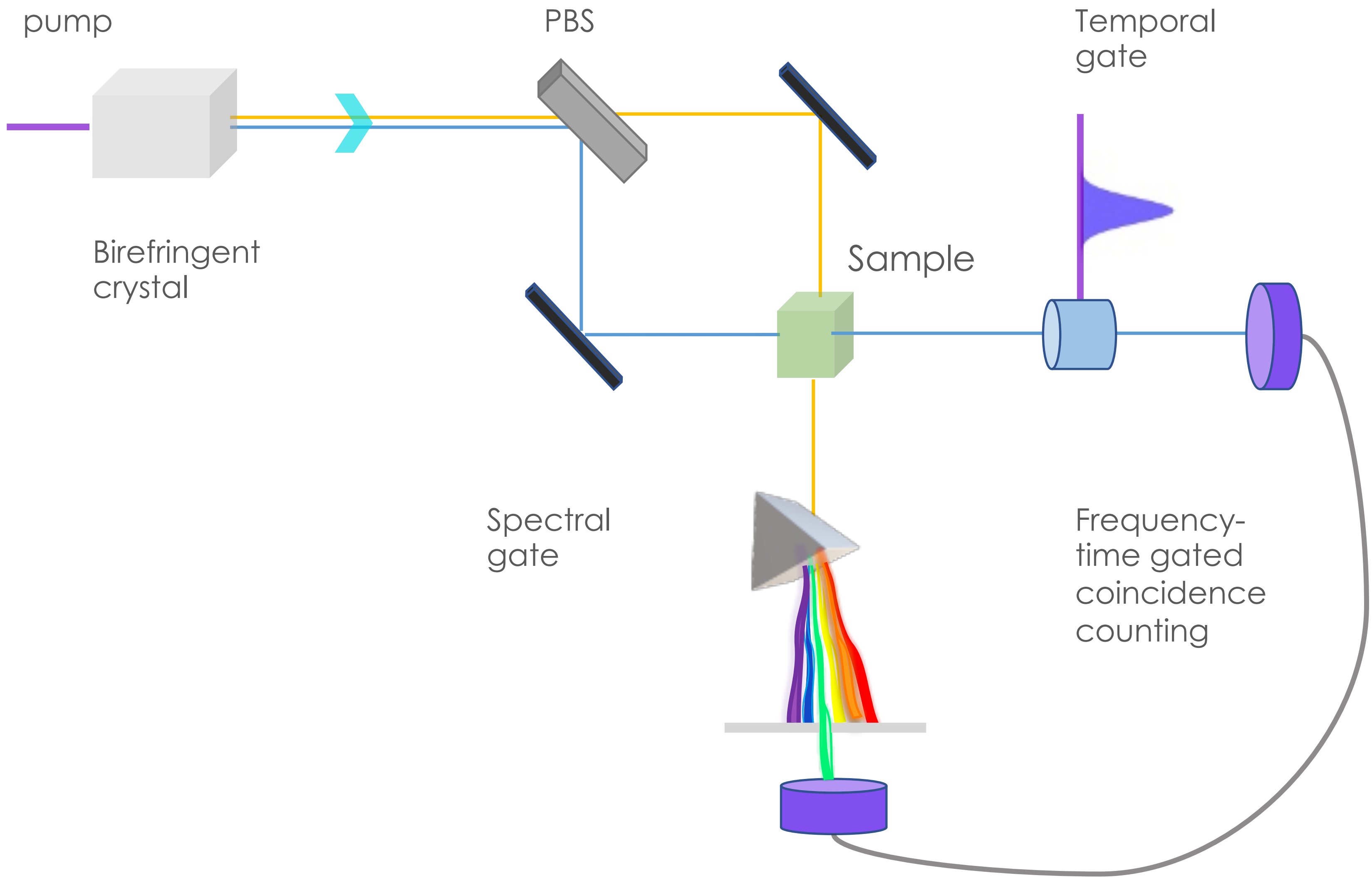

a)
b)
τ_c
t_n
⋮
t_2
t_1
t_{n+1}
t_m
τ_{ic}
t_4
t_3
t_2
t_1
TOC
OTOC
c)
τ_c
t_b
t_a
$\tau_b = \frac{R_b}{c}$
$\tau_a = \frac{R_a}{c}$
t_4
t_3
t_2
t_1



a)
Michelson interferometer
U_θ
Coincidence count
Sample
Pulse pumped type-II DC photons
$\Phi(\omega_a, \omega_b)$
$e^{i\theta}$ photon exchange phase
$\Phi_\theta(\omega_a, \omega_b) = \frac{\Phi_{ab} + e^{i\theta}\Phi_{ba}}{\sqrt{2}}$
b)
θ-symmetrization
U_θ
$\Leftrightarrow$
a_{pump}
$a_{i,out}$
M1 (PZT)
$\frac{\lambda}{4}$
DM
BBO
M2



a
pump
Michelson interferometer
Detection
U_θ
Sample
Pulse pumped type-II DC photons
$e^{i\theta}$ photon exchange phase
$\Phi(\omega_a, \omega_b)$
$\Phi_\theta(\omega_a, \omega_b) = \frac{\Phi_{ab} + e^{i\theta}\Phi_{ba}}{\sqrt{2}}$
b
$U(\tau_a)$
$U(\tau_b)$
τ_b- τ_a
coincidence count
c
τ
$\tau_b = \frac{R_b}{c}$
$\tau_a = \frac{R_a}{c}$
$\tau\ (R_a = R_b)$
Diagramatic representation of the time flow


pump
PBS
Temporal gate
Birefringent crystal
Sample
Spectral gate
Frequency-time gated coincidence counting