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VIEW & PERSPECTIVE

# Geometric phase opens new frontiers in nonlinear frequency conversion of light

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Nonlinear frequency conversion of light involves interaction of photons through the nonlinear dielectric response of materials to generate light at new frequencies. The first demonstration of nonlinear optical frequency conversion was that of second-harmonic generation (SHG) [1]. In this process, two input photons at frequency $\omega$ combine in a material to coherently emit a single photon at frequency $2\omega$. Since then, many other nonlinear frequency conversion processes have been demonstrated, including different schemes of three and four-wave-mixing as well as higher order interactions [2].

The nonlinear optical response of materials can be illustrated by a simple model in which the induced polarization of a material is expanded in a power series of the applied light field

$$P\left(t\right)=\varepsilon_0\left[\chi^{(1)}E\left(t\right)+\chi^{(2)}E^2\left(t\right)+\chi^{(3)}E^3\left(t\right)+\dots\right]. \tag{1}$$

Nonlinear terms in this expression enable wave mixing and generation of light at new frequencies. In reality, various factors such as losses, dispersion, material anisotropy and light polarization must be accounted for. Nevertheless, when these are taken into consideration, efficient frequency conversion of light can be achieved.

Since its demonstration and theoretical formulation [3], the concept of nonlinear wave mixing has been driving technological progress and exploration of novel physical phenomena. In addition to the ability to generate coherent and intense light over the entire optical regime, from deep ultraviolet down to terahertz frequencies [4], it has opened the door for new schemes to control light and continues to stand at the focus of highly active research. Current frontiers of the field include for example, nonlinear holography [5, 6], investigations of nonreciprocal systems [7], as well as generation and manipulation of entangled states of light for development of modern quantum technologies [8, 9].

Within this broad field of research, a new and promising sub-field has emerged, studying expressions of geometric phase in nonlinear wave mixing and frequency conversion processes. In a recent review by Karnieli, Li and Arie [10], a comprehensive introduction and description of the development of this field is presented. From a few initial pioneering studies starting 30 years ago [11, 12], the concept attracts now an ever growing amount of attention, inspiring new insights on the fundamentals of frequency conversion processes, and unlocking new techniques to control and utilize nonlinear wave-mixing dynamics.

The concept of *geometric phase* (GP) goes back to the work by Berry [13], where he analyzed the state of a quantum mechanical system which varies in time in a cyclic, adiabatic and unitary manner. He found that apart from the usual dynamical phase accompanying the temporal evolution, the state also acquires a phase given by the expression

$$\gamma=\mathrm{i}\int_{\boldsymbol{R}(0)}^{\boldsymbol{R}(T)}\left\langle\psi\left(\boldsymbol{R}\right)\middle|\nabla_{\boldsymbol{R}}\middle|\psi\left(\boldsymbol{R}\right)\right\rangle\cdot\mathrm{d}\boldsymbol{R}, \tag{2}$$

where $\boldsymbol{R}(t)$ are the adiabatically varying parameters of the Hamiltonian and $\psi(\boldsymbol{R})$ is the wave function [13]. This phase can be interpreted as the rotation of a vector being subject to parallel transport on a curved surface [14]. Later, through the works of Aharonov & Anandan [15] and Samuel & Bhandari [16], the notion of a GP acquired by a quantum state was extended also to the cases of nonadiabatic, noncyclic and nonunitary evolution.

In the decades since Berry's formulation, the concept of GP has been applied throughout condensed matter physics and optics [14]. However, even before Berry's general description of GP, there were a few precursors to this concept [17]. In the field of optics in 1956, Pancharatnam published his work on the interference between monochromatic polarized states of light [18]. To obtain insight into the general properties of the interference between different polarization states, Pancharatnam used the geometrical representation of the Poincaré sphere. On this sphere, the north and south poles correspond to right and left handed circularly polarized light, the equator describes states of linear polarization and the rest of the sphere contains elliptically polarized light. Actions performed by polariza-

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tion affecting elements are marked as the geodesic paths between different points on the sphere. It was shown by Pancharatnam that transformations of polarization may involve an accumulated phase that depends on the path made on the Poincaré sphere. For example, in a cyclic set of transformations, the accumulated phase is equal to half of the solid angle enclosed by the path. A later analysis by Berry showed that the Pancharatnam phase is analogous to the GP observed in time varying quantum mechanical systems [19, 20]. This equivalence led to the well-known term *Pancharatnam–Berry* (PB) *phase* commonly used to describe the GP acquired in polarization transformations. PB phase has since played a key role in various developments in the field of linear optical manipulations of light. Specifically in the field of metasurfaces, it has been used in the development of various new schemes to control light beams by flat optical elements [21].

The manifestation of GP in nonlinear wave mixing processes has so far been studied in two areas defined by the type of state transformation. The first deals with GP acquired in spectral domain transformations that occur during propagation of the interacting waves in bulk materials. The second has to do with GP accompanying polarization transformations, most commonly in relation to frequency conversion on metasurfaces.

As an example of the manifestation of GP in the spectral domain, it is instructive to look at the parametric process of sum-frequency-generation (SFG). In SFG a pump wave at frequency $\omega_p$, an idler wave at frequency $\omega_i$ and a signal wave at frequency $\omega_s = \omega_p + \omega_i$ interact in a quadratic nonlinear material. Under the undepleted pump approximation, the amplitude of the pump wave, $A_p$, remains constant and energy is exchanged between the signal and idler waves. In this case the complex slowly-varying envelopes, $A_i$ and $A_s$ of the idler and signal waves respectively, are governed by two coupled linear equations [2]:

$$\begin{aligned}\frac{\partial A_i}{\partial z} &= \mathrm{i}\frac{2d(z)\omega_i^2}{k_i c^2} A_p^* A_s \mathrm{e}^{-\mathrm{i}\Delta k_0 z},\\ \frac{\partial A_s}{\partial z} &= \mathrm{i}\frac{2d(z)\omega_s^2}{k_s c^2} A_p A_i \mathrm{e}^{\mathrm{i}\Delta k_0 z},\end{aligned} \tag{3}$$

where $z$ is the propagation axis in the nonlinear crystal, $d(z)$ the location dependent quadratic nonlinear coefficient, $k_{i,s,p}$ represent the corresponding wavenumbers in the material, $\Delta k_0 = k_p + k_i - k_s$ is proportional to the momentum mismatch in the wave mixing process, and $c$ is the speed of light. One of the ways to control the rate of energy flow between the two frequency components is by momentum mismatch compensation. This can be done by spatially varying $d(z)$ through engineered poling of the nonlinear coefficient, as commonly done to achieve quasi-phase-matching (QPM) [2] [see illustration in Fig. 1(a)].

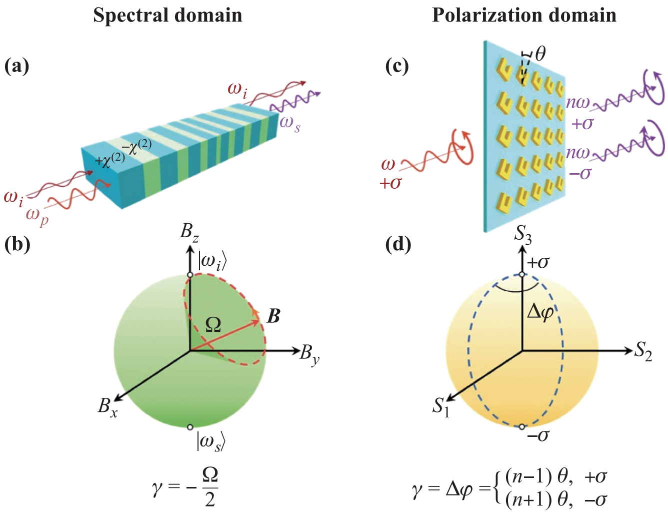

**Fig. 1** Manifestation of GP in nonlinear wave-mixing processes in the spectral (a), (b) and polarization (c), (d) domains. **(a)** SFG in an engineered nonlinear crystal with aperiodic alternating regions of opposite signs of the nonlinear coefficient $\chi^{(2)}$. **(b)** The system supports an analogous effective magnetic field $\boldsymbol{B}$ that varies adiabatically as light propagates in the material. Here $\boldsymbol{B}$ is represented on the Bloch sphere. The dashed line illustrates the path of this adiabatically varying effective field. The GP $\gamma$ accumulated in one cycle is equal to half of the solid angle enclosed by the path. Schemes for non-cyclic transitions and non-adiabatic variations of $\boldsymbol{B}$ can also be represented. **(c)** Harmonic generation in a nonlinear metasurface comprised of rotated split-ring resonators. **(d)** Geometric phase accompanying polarization conversion represented on the Poincaré sphere. The GP is the angle $\Delta\varphi$ between the two geodesic trajectories of polarization transformation. $\Delta\varphi$ is determined by the rotation angle of the meta-atom $\theta$ and by the final polarization state. In the nonlinear case the GP can be increased by the number of interacting photons. For harmonic generation the final GP is given by the presented expression for $\gamma$.

The complex amplitudes that are described by the set of equations in Eq. (3) can be mapped onto the Bloch sphere, a geometrical representation commonly used in the context of quantum two-level systems [22]. In the case of SFG, the Bloch sphere is constructed so that the north and south poles correspond with pure idler and signal waves respectively, the equator marks equal superposition of idler and signal waves, and the rest of the sphere contains all other combinations [22, 23]. The geometrical representation of wave mixing processes and their analogy to quantum mechanical systems, have proven to be highly advantageous for obtaining new fundamental insights, as well as developing new abilities to control nonlinear interactions [22, 24]. This geometrical representation has also advanced the formulation of GP accompanying frequency conversion processes. For example, the analogy between the SFG wave dynamics in modulated QPM crystals to the dynamics of a spin 1/2 particle in a magnetic field was used to calculate and measure the accumulated GP during SFG [23, 25]. The analogous effective magnetic field in this system depends on the magnitude and phase of the pump wave, the momentum mismatch and the modulated crystal design. By varying the modulation pattern, the effective magnetic field can be made to evolve adiabatically in closed paths on the Bloch sphere [Fig. 1(b)]. This results in a GP added to the signal and idler fields over one cycle, similar to the phase that Berry calculated for the spin 1/2 particle case [13, 23].

Apart from the above example, the geometric representation of nonlinear wave mixing and the GP acquired in the process inspired various interesting developments in the field. The geometric representation in general was leveraged to enhance the nonlinear conversion efficiency and to improve control over the wave mixing process. GP was used for nonlinear beam shaping, and its nonreciprocal manifestation in nonlinear wave mixing was studied and observed. GP was calculated for a variety of three- and four-wave-mixing processes in bulk materials and beyond the undepleted pump approximation. Additionally, GP accumulated during frequency conversion in birefringent crystals was studied. Moreover, GP acquired in the context of propagation in nonlinear artificial gauge fields attracted recent interest. Extensive elaboration on the development and the current state of research in the field appears in Ref. [10].

The second case of GP accompanies polarization transformations in frequency conversion processes. This type of GP can be seen as a nonlinear extension of the PB phase. The nonlinear PB phase was mostly studied and demonstrated to date in nonlinear optical metasurfaces. These are thin layers, built of subwavelength building blocks called meta-atoms, that exhibit a strong engineered nonlinear optical response. The prospects of fabricating ultrathin metasurface-based optical elements, capable of highly controlled frequency conversion, have drawn a lot of attention and research efforts to this field [6, 26].

The nonlinearity of metasurfaces can originate from the materials that compose the meta-atoms, their surfaces, substrates, and also from their structural design. These material or structural nonlinearities can be greatly enhanced by designing the meta-atoms to confine the electromagnetic fields, through plasmonic and Mie resonances, as well as through collective effects on the metasurfaces [27, 28]. Therefore, design parameters such as the size, shape, symmetry, and position of the meta-atoms play a major role in the overall nonlinear response and functionality of the metasurface.

In the frequency conversion process on the metasurface, the interacting waves can accumulate a nonlinear phase. Since the metasurfaces are optically thin, this phase is usually related to the complex effective nonlinear susceptibility of the metasurface, rather than propagation. Independent of this dynamic type of phase, variations of the meta-atom orientation can add a GP to the interacting waves [29–31], as depicted in Fig. 1(c). This manifestation of the nonlinear PB phase provides an increasingly important tool to control the nonlinear response and functionality of metasurfaces.

To understand the origin of this type of GP, it is helpful to consider a harmonic generation process of circularly polarized interacting states on a meta-atom whose principal axis is tilted at an angle $\theta$, as shown in Fig. 1(c). By coordinate transformations between the lab frame and the meta-atom local frame, the nonlinear polarization can be calculated,

$$P^{(n)}_{\pm\sigma} = \overline{\boldsymbol{\alpha}} \mathrm{e}^{\mathrm{i}\sigma(n\mp1)\theta}(E_\sigma)^n, \tag{4}$$

where $\overline{\boldsymbol{\alpha}}$ is the polarizability tensor, $n$ is the order of nonlinearity, $\sigma = \pm1$ indicates the circular state of polarization (right or left) and $E_\sigma$ is the fundamental electric field at state of polarization $\sigma$. It can be seen that the GP depends on the harmonic order (number of interacting waves), input and output circular polarization states, and on the orientation of the particle. Importantly, the interaction and conversion between circularly polarized states through materials, necessitates the conservation of spin-angular moment. This condition sets strict selection rules on the allowed processes which relate to the GP and the material (or meta-atom) symmetry [26, 32, 33]. The selection rules for meta-atoms with $m$-fold rotational symmetry, states that the $n$-th harmonic generation is allowed for a circular polarization pump, if the following holds:

$$n = ml \pm 1, \quad l \text{ is an integer}, \tag{5}$$

where the sign ± stands for the same or opposite circular polarization of the generated wave compared to the pump. Keeping in mind these strict selection rules, the ability to continuously vary the orientation of the meta-atoms provides a useful tool to obtain continuous phase control over the nonlinear interaction.

This concept was used in various wave-mixing schemes to control generated waves mainly in the near infrared and visible regime [26], and very recently also for THz generation [34]. Various advanced nonlinear functionalities were demonstrated by nonlinear GP metasurfaces, including beam shaping, imaging, holography, image encoding and beam steering. It was also used to study various spin-angular momentum interactions in the nonlinear wave-mixing process and obtain control over nonlinear generation of vector fields. A detailed description of the most exciting developments in the field can be found in Ref. [10].

The lessons learned from Berry and Pancharatnam on the notion of GP in evolving physical systems, have deepened and expanded our understanding and control capabilities of nonlinear wave mixing processes. This new understanding has so far been applied in spectral and polarization domain transformations. In domains, progress has included a correlated advancement in theoretical understanding, as well as a strong dependence on advanced fabrication capabilities. In spectral domain transformations, the progress of engineered poling of quadratic nonlinear crystals played a major role in many of the recent works, and for polarization domain transformations, nanofabrication capabilities of metasurfaces were instrumental. With major improvements in the upcoming years in the techniques of 3D poling of nonlinear crystals [35] and fabrication of nonlinear metamaterials, an interesting future path involves the combination of GP in frequency and polarization domains together. Such advancement will open the door to create hybrid nonlinear metastructures, in which both the polarization and frequency domain GPs are tuned. This may pave the way for substantial conversion efficiency improvements in nonlinear wave mixing, as well as fundamental studies in new domains of nonlinear wave propagation and its control.

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