Supplementary information: New photonic conservation laws in parametric nonlinear optics

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The supplementary information file contains the following sections: in section 1 we derive the photonic spin conservation law (row 4 of table 1 in the main text). Section 2 discusses the generalization of the photonic spin conservation law to conservation of ellipticity, and explains why ellipticity is in fact not generally conserved. In section 3 we derive the photonic parity conservation law. In section 4 we discuss general symmetries and conservation laws in anisotropic media with discrete symmetries. Section 5 describe basic concepts of superspace symmetries which we used in the main text.

1) <u>Conservation of spin</u>

In order to obtain the photonic spin conservation law (row 4 of table in the main text), we consider a superposition of circularly polarized EM fields:

$$\vec{E}(t, X_1, X_2, \dots, X_M) = \hat{e}_+ \sum_{\substack{n_+=1\\N_-}}^{N_+} a_{n_+} e^{i(\omega_{n_+}t - \sum_{m=1}^M k_{n_+,m} \cdot X_m)}$$

$$+ \hat{e}_- \sum_{\substack{n_-=1\\n_-=1}}^{N_-} a_{n_-} e^{i(\omega_{n_-}t - \sum_{m=1}^M k_{n_-,m} \cdot X_m)}$$

$$(1)$$

where \hat{e}_+ and \hat{e}_- are the normalized Jones vectors of the circularly polarized field with lefthelicity and right-helicity, respectively, and X_m are M different orthogonal space dimensions, ω_n and $k_{n,m}$ are the angular frequency and wavevector of \vec{E}_n , respectively. Similarly, to the derivations of conservation laws in the main text, we represent the field in super space by:

$$\vec{E}(t_{1}, t_{2}, ..., t_{N}, X_{1}, X_{2}, ..., X_{M}) = \hat{e}_{+} \sum_{\substack{n_{+}=1\\N_{-}}}^{N_{+}} a_{n_{+}} e^{i(\omega_{n_{+}} t_{n_{+}} - \sum_{m=1}^{M} k_{n_{+},m} \cdot X_{m})} + \hat{e}_{-} \sum_{\substack{n_{-}=1\\n_{-}=1}}^{N_{-}} a_{n_{-}} e^{i(\omega_{n_{-}} t_{n_{-}} - \sum_{m=1}^{M} k_{n_{-},m} \cdot X_{m})}$$
(2)

This field, in the super space representation, has the symmetry, $t_{n_+} \rightarrow t_{n_+} + \frac{\delta}{\omega_{n_+}}$, $t_{n^-} \rightarrow \frac{\delta}{\omega_{n_+}}$

 $t_{n_{-}}-rac{\delta}{\omega_{n_{-}}}$, \hat{r}_{δ} .

The selection rules for such a continuous symmetry, according to eq. (5) in the main text is:

$$\sum_{n_{+}=1}^{N_{+}} q_{n_{+}} - \sum_{n_{-}=1}^{N_{-}} q_{n_{-}} = \pm 1$$
(3)

where the + (-) sign corresponds to emission of \hat{e}_+ (\hat{e}_-) circularly polarized harmonics, and $q_{n_+}(q_{n_-})$ is the number of annihilated photons of each n_+ (n_-) driver component. This corresponds to the conservation of spin in the fourth row of table 1 in the main text.

2) <u>Conservation of ellipticity</u>

In this section we first derive conservation of ellipticity and then discuss the reason it is usually irrelevant in nonlinear optics. To this end, we replace in Eqs. (1) and (2) \hat{e}_+ and \hat{e}_- by $\hat{e}_{\epsilon+}$ and $\hat{e}_{\epsilon-}$ that correspond to orthogonal elliptical polarization normalized Jones vectors, both with ellipticity ϵ and with the same ellipse major axes, yet opposite helicity.

A field with $\hat{e}_{\epsilon+}$ and $\hat{e}_{\epsilon-}$ in eq. (1) have the symmetry, $t_{n_+} \rightarrow t_{n_+} + \frac{\delta}{\omega_{n_+}}$, $t_{n^-} \rightarrow t_{n_-} - \frac{\delta}{\omega_{n_-}}$, \hat{e}_{δ} . Here, $\hat{e}_{\delta} = \begin{pmatrix} 1 & 0 \\ 0 & \epsilon \end{pmatrix} \hat{r}_{\delta} \begin{pmatrix} 1 & 0 \\ 0 & 1/\epsilon \end{pmatrix}$ is an elliptical rotation operation. This DS is a continuous super-space version of the discrete elliptical DS [1]. The resulted selection rule from this DS is written in eq. (3), but here the + (-) signs correspond to emission of $\hat{e}_{\epsilon+}$ ($\hat{e}_{\epsilon-}$) elliptical polarized harmonics. This selection rule indicates, for example, that if all the pump photons have the same ellipticity then this ellipticity is conserved in the generation processes, e.g. all harmonics will exhibit the same ellipticity [2,3]. However, there are two important issues which prevent this selection rule from becoming a general conservation law: I) Only very specific fields can be easily and compactly represented by the { \hat{e}_+ , \hat{e}_- } basis, and only for this kind of fields the selection rules will be useful. II) Natural materials are not symmetrical under \hat{e}_{δ} . Even an isotropic medium does not exhibit elliptical symmetry. Therefore, even if the field exhibits this symmetry, most materials will break it. Hence, this fundamental symmetry does not correspond to a general elliptical photonic conservation law, but might only be realized in specialized systems.

3) <u>Conservation of pairity</u>

The general superspace EM field in eq.(7) in the main text also exhibit another symmetry in super-space, $t_n \rightarrow t_n + \frac{\pi}{\omega_n}$ for all $n = 1 \dots N$ simultaneously, \hat{i} , where \hat{i} is the microscopic inversion operator (i.e., $\hat{i}\vec{E} = -\vec{E}$). According to eq. (4, 9) in the main text, this symmetry produces the constraint $\sum_{n=1}^{N} q_n \omega_n \hat{t}_n \cdot \frac{\pi}{\omega_n} = 2\pi Q$, hence the sum $\sum_n q_n$ most be odd. This constrain is the parity conservation law which appears in row 5 in table 1 in the main text.

4) Conservation laws in anisotropic media with discrete symmetries

Until now we derived conservations laws in isotropic media. For a medium that lacks continuous symmetries, but still exhibits discrete symmetries, the general superspace symmetries will also become discrete. This discretization will modify the conservations laws. One well-known example of such modification is the conservation of crystal momentum which is a modified linear momentum. In this section we derive the selection rules in such systems.

First, for the case of a medium that has an *S* fold microscopic rotational symmetry, we should use the circularly polarized basis described in eq. (1) of the SI. In this case the systems lacs the continues symmetry, but still has the discrete symmetry, $t_{n_+} \rightarrow t_{n_+} + \frac{2\pi}{S\omega_{n_+}}$, $t_{n^-} \rightarrow t_{n_-} - t_{n_-}$

 $\frac{2\pi}{\omega_{n_{-}}}$, \hat{r}_{S} , which leads to the photonic equation:

$$\sum_{n_{+}=1}^{N_{+}} q_{n_{+}} - \sum_{n_{-}=1}^{N_{-}} q_{n_{-}} = QS \pm 1$$
(4)

where Q is an integer. This equation is similar to eq. (3) with an additional QS term. Here, the photonic spin equation in row 3 of table 1 becomes $s_f = QS + \sum_n q_n s_n$, with the same constraint that all the spins are $s = \pm 1$. For example, a monochromatic circularly polarized electric field with a helicity of s and a frequency of ω_1 interacting with a medium that has an S fold microscopic rotational symmetry can generate only $QS \pm 1$ harmonic orders with spin of $\pm s$.

A similar derivation can be performed for other discrete symmetries of the medium. When the medium is time periodic with a period of $2\pi/\Omega$, the energy conservation in row 1 of table 1 becomes $\omega_f = Q\Omega + \sum_n q_n \omega_n$. Likewise, for a medium with space periodicity, the momentum equation in row 2 of table 1 becomes $\vec{k}_f = Q\vec{K} + \sum_n q_n\vec{k}_n$, where \vec{K} is the wavevector of the medium periodicity. When the medium has an *L* fold macroscopic rotational symmetry, the OAM equation in row 3 of table 1 becomes $l_f = QL + \sum_n q_n l_n$.

5) <u>Super-space concepts</u>

This section overviews the basics of super-space representation that we used to derive the conservation laws.

Periodicity is a very important symmetry in many physical systems. Many crystals are classified according to their periodic symmetries. However, there are also aperiodic crystals, e.g. quasicrystals. The Fourier transforms of both periodic and quasicrystals consist of sharp peaks, and the positions of the peaks is spanned by a finite number (N) of vectors:

$$\vec{B} = q_1 \vec{b}_1 + q_2 \vec{b}_2 + \dots + q_N \vec{b}_N \tag{5}$$

where q_n are integers and \vec{b}_n are rationally independent vectors.

The collection of all \vec{B} vectors is called the Fourier module, \mathcal{M} , which has a rank of N. Thus, the Fourier decomposition of (periodic and aperiodic) crystal function (e.g. the density function of atoms in an aperiodic crystal) is:

$$f(\vec{X}) = \sum_{\vec{k} \in \mathcal{M}} F_{\vec{k}} \exp(i\vec{k} \cdot \vec{X})$$
(6)

where \vec{X} is the physical coordinates and $F_{\vec{k}}$ are the Fourier coefficients. For periodic crystals, N is the physical dimensionality (D) of the crystal, the Fourier module is the reciprocal lattice, and \vec{b}_n is the reciprocal primitive vectors. For aperiodic crystals, N is greater than the physical dimensionality, and the Fourier module can be considered as an N-dimensional reciprocal lattice that is projected onto the physical Fourier space. This N-dimensional reciprocal lattice has N reciprocal primitive vectors of $\vec{b}_n = (\vec{b}_n, \vec{b}_n^I)$, where the dimension of the \vec{b}_n^I vectors is N - D. The lattice periodic function in the N-dimensional space is defined by:

$$\tilde{f}(\vec{X}, \vec{X}^{I}) = \sum_{\vec{k} \in \mathcal{M}} F_{\vec{k}} \exp(i\vec{k} \cdot \vec{X} + i\vec{k}^{I} \cdot \vec{X}^{I})$$
(7)

By construction, the function, $\tilde{f}(\vec{X}, \vec{X}^{l})$, is periodic in the (\vec{X}, \vec{X}^{l}) space. The (\vec{X}, \vec{X}^{l}) space is called super-space. The $f(\vec{X})$ function is exactly $\tilde{f}(\vec{X}, \vec{X}^{l} = 0)$ when all the spanning \vec{b}_{n} are orthogonal or parallel to each other. For example, when the field

$$f(X_1, X_2) = \sum_{\substack{q_1, q_2, q_3, q_4 \\ + iq_4b_4X_2)}} F_{\vec{q}} \exp(iq_1b_1X_1 + iq_2b_2X_1 + iq_3b_3X_2$$
(8)

has two parallel pairs, such as $\vec{b}_1 \parallel \vec{b}_2$ and $\vec{b}_3 \parallel \vec{b}_4$, which are orthogonal to each other (e.g. $\vec{b}_2 \perp \vec{b}_3$), then we can choose a super-space function,

$$\tilde{f}(X'_{1}, X''_{1}, X'_{2}, X''_{2}) = \sum_{\substack{q_{1}, q_{2}, q_{3}, q_{4} \\ + iq_{3}b_{3}X'_{2}}} F_{\vec{q}} \exp(iq_{1}b_{1}X'_{1} + iq_{2}b_{2}X''_{1})$$
(9)

such that $\tilde{f}(X'_1, X''_1, X'_2, X''_2)$ is periodic along the main axis. Returning to the physical space is done by setting $X'_1 = X''_1 = X_1$ and $X'_2 = X''_2 = X_2$. Transferring to super symmetric space allows the theories of periodic structures to be applied to aperiodic structures.

Similarly to of the case of crystals, the superposition of monochromatic waves (including vectorial field waves, like EM waves) also exhibits sharp peaks in Fourier space; hence, transformation to super-space may be useful. In [4] we showed that symmetries of a vectorial

field lead to constrains in Fourier space. These symmetries can be in D-dimensional physical space or in N-dimensional super-space. Specifically, for optical quasicrystals, some of the DS will be in super-space. These super-space DSs also lead to selection rules. We utilized the super-symmetry transformation to derive photonic conservation laws, including two new laws.

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