Supplementary information:
Selection rules for breaking selection rules

Matan Even Tzur[1],* Ofer Neufeld[1], Avner Fleischer[2] and Oren Cohen[1]

[1]Solid State Institute and Physics Department, Technion - Israel Institute of Technology, 3200003, Haifa, Israel

[2]Raymond and Beverly Sackler Faculty of Exact Science, School of Chemistry and Center for Light Matter Interaction, Tel Aviv University, 6997801, Tel Aviv, Israel

This supplementary material file contains the derivations of the analytical results presented in the main text, as well as details on the methodology used in numerical calculations and complementary numerical results. In Section S.I we derive Eqs. (6,7) from the main text. In section S.II, we derive the selection rules for breaking selection rules presented in Table 1 of the main text. In section S.III we drive an analogous table for circularly polarized perturbations. In section S.IV we discuss the generalization of our results for fractional harmonic perturbations. In section V we discuss the applicability of main table 1 to multi-chromatic perturbations and present a complementary numerical example. In section S.VI we show that for the numerical examples we studied in the main text, and the numerical example we studied in section S.V, the same results are obtained when a spectral width around each harmonic is considered.

Throughout this file we follow the notation of reference [28] in the main text for Floquet perturbation theory (FPT) and reference [15] in the main text for dynamical symmetries.

S.I DERIVATION OF EQUATIONS (6,7) IN THE MAIN TEXT

In this section, we derive Eqs. (4-7) in the main text. Let $\left|\phi_{\alpha(0)}(t)\right\rangle$ be a Floquet state of the unperturbed system. When the system is perturbed by the term $\lambda\widehat{W}(t)$ this state may be corrected by FPT. First, one needs to lift the state $\left\{\left|\phi_{\alpha(0)}(t)\right\rangle\right\}_{t=0}^{t=T}$ from the Hilbert space $\mathcal{H}$ to the Floquet Hilbert space $\mathcal{F}=\mathcal{H}\otimes\mathcal{L}$ where $\mathcal{L}$ is the space of bounded periodic functions (i.e. loop functions) over $[0,T)$. The space $\mathcal{L}$ is spanned by the orthonormal basis $\{|t)\}, 0\leq t<T$, where the orthonormality condition is $(t|t')\equiv T\delta(t-t')$. The brackets $|\cdot\gg, |\cdot\rangle, |\cdot)$ describe states that live in $\mathcal{F},\mathcal{H}$ and $\mathcal{L}$, respectively.

The state $\left|\phi_{\alpha(0)}(t)\right\rangle$ is lifted to a loop in $\mathcal{F}$ by $|\phi_{\alpha t}\gg=\left|\phi_{\alpha(0)}(t)\right\rangle|t)$. The center of the loop is defined as

$$|\overline{\phi_{\alpha(0)}}\gg\equiv\int_0^T\left|\phi_{\alpha(0)}(t)\right\rangle|t)\frac{dt}{T} \tag{S 1}$$

To 1st order, $|\overline{\phi_{\alpha(0)}}\gg$ and is corrected by

$$|\overline{\phi_\alpha}\gg=|\overline{\phi_{\alpha(0)}}\gg+\lambda|\overline{\phi_{\alpha(1)}}\gg \tag{S 2}$$

where

(S 3)

$$|\overline{\phi_{\alpha(1)}} \gg = \sum_{(\beta,n)\neq(\alpha,0)} \frac{\ll \phi_{\beta n(0)} \left|\hat{\hat{W}}\right| \overline{\phi_{\alpha(0)}} \gg}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - n\omega} |\phi_{\beta n(0)} \gg$$

, and

$$|\phi_{\beta n(0)} \gg \equiv \int_0^T \frac{dt}{T} e^{in\omega t} \left|\phi_{\beta(0)}(t)\right\rangle |t) \tag{S 4}$$

$$\hat{\hat{W}} \equiv \int_0^T \frac{dt}{T} |t)\hat{W}(t| \tag{S 5}$$

$$\ll \phi_{\beta n(0)} \left|\hat{\hat{W}}\right| \overline{\phi_{\alpha(0)}} \gg = \int_0^T \frac{dt}{T} \left\langle \phi_{\beta(0)} \right| e^{-in\omega t} \hat{W}(t) \left| \phi_{\alpha(0)} \right\rangle \tag{S 6}$$

Finally, to obtain the corrected Floquet state $|\phi_\alpha(t,\lambda)\rangle$ we project $|\overline{\phi_\alpha} \gg$ back to the Hilbert space:

$$|\phi_\alpha(t,\lambda)\rangle = (t|\overline{\phi_\alpha} \gg \tag{S 7}$$

The Ω frequency component of the dipole moment expectation value $\hat{\boldsymbol{\mu}}(\lambda)$ is given by

$$\tilde{\mathbf{E}}(\Omega,\lambda) = \int_0^T \frac{dt}{T} \langle \phi_\alpha(t,\lambda)|\hat{\boldsymbol{\mu}} e^{i\Omega t}|\phi_\alpha(t,\lambda)\rangle = \ll \overline{\phi_\alpha}|\hat{\hat{\boldsymbol{\mu}}}_\Omega|\overline{\phi_\alpha} \gg \tag{S 8}$$

where

$$\hat{\hat{\boldsymbol{\mu}}}_\Omega \equiv \int_0^T \frac{dt}{T} |t)\hat{\boldsymbol{\mu}} e^{i\Omega t}(t| \tag{S 9}$$

Plugging Eq. (S 2) into Eq. (S 8) we obtain Eq.(4) in the main text:

$$\tilde{\mathbf{E}}(\Omega,\lambda) = \tilde{\mathbf{E}}_0(\Omega) + \lambda\tilde{\mathbf{E}}_1(\Omega) + \lambda^2\tilde{\mathbf{E}}_2(\Omega) \tag{S 10}$$

where

$$\tilde{\mathbf{E}}_0(\Omega) \equiv \ll \overline{\phi_{\alpha(0)}}|\hat{\hat{\boldsymbol{\mu}}}_\Omega|\overline{\phi_{\alpha(0)}} \gg \tag{S 11}$$

$$\tilde{\mathbf{E}}_1(\Omega) \equiv \ll \overline{\phi_{\alpha(0)}}|\hat{\hat{\boldsymbol{\mu}}}_\Omega|\overline{\phi_{\alpha(1)}} \gg + \ll \overline{\phi_{\alpha(1)}}|\hat{\hat{\boldsymbol{\mu}}}_\Omega|\overline{\phi_{\alpha(0)}} \gg \tag{S 12}$$

$$\tilde{\mathbf{E}}_2(\Omega) \equiv \ll \overline{\phi_{\alpha(1)}}|\hat{\hat{\boldsymbol{\mu}}}_\Omega|\overline{\phi_{\alpha(1)}} \gg \tag{S 13}$$

For the derivations of Eqs.(6,7) in the main text, we use the following mathematical identities:

$$\ll \overline{\phi_\alpha} \left|\hat{\hat{\boldsymbol{O}}}_\Omega\right| \overline{\phi_\beta} \gg = \int_0^T \frac{dt}{T} \langle \phi_\alpha(t)|\, \hat{\boldsymbol{O}} e^{i\Omega t} |\phi_\beta(t)\rangle \tag{S 14}$$

$$\begin{aligned} \ll \overline{\phi_{\alpha(0)}} \left|\hat{\hat{\boldsymbol{O}}}_{n\omega}\right| \phi_{\beta m(0)} \gg &= \ll \overline{\phi_{\alpha(0)}} \left|\hat{\hat{\boldsymbol{O}}}_{(n+m)\omega}\right| \overline{\phi_{\beta(0)}} \gg \\ &= \int_0^T \frac{dt}{T} \left\langle \phi_{\alpha(0)}(t) \right| \hat{\boldsymbol{O}} e^{i(n+m)\omega} \left|\phi_{\beta(0)}(t)\right\rangle \end{aligned} \tag{S 15}$$

$$\begin{aligned} \ll \phi_{\beta m(0)} \left|\hat{\hat{\boldsymbol{O}}}_{n\omega}\right| \overline{\phi_{\alpha(0)}} \gg &= \ll \overline{\phi_{\beta(0)}} \left|\hat{\hat{\boldsymbol{O}}}_{(n-m)\omega}\right| \overline{\phi_{\alpha(0)}} \gg \\ &= \int_0^T \frac{dt}{T} \left\langle \phi_{\alpha(0)}(t) \right| \hat{\boldsymbol{O}} e^{i(n-m)\omega} \left|\phi_{\beta(0)}(t)\right\rangle \end{aligned} \tag{S 16}$$

where $\hat{\boldsymbol{O}}$ is a vector operator. These identities can be verified by using definitions of $|\overline{\phi_{\alpha(0)}}\rangle$ (Eq.(S 1)) , $|\phi_{\beta n(0)} \gg$ (Eq.(S 4)), $\hat{\hat{\boldsymbol{O}}}_{n\omega}$ (Eq. (S **9**)), and the orthonormality condition $(t|t') \equiv T\delta(t-t')$.

The term $\widetilde{\boldsymbol{E}}_{\mathbf{1}}(\boldsymbol{\Omega})$ can also be written as

$$\widetilde{\boldsymbol{E}}_1(\Omega) = \ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{\Omega}\big|\overline{\phi_{\alpha(1)}} \gg + \left(\ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{-\Omega}\big|\overline{\phi_{\alpha(1)}} \gg\right)^{\dagger} \tag{S 17}$$

We plug Eq.(S 3) into the first term of Eq.(S 12)

$$\ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{n\omega}\big|\overline{\phi_{\alpha(1)}} \gg = \sum_{(\beta,m)\neq(\alpha,0)} \frac{\ll \phi_{\beta m}\left|\widehat{\widehat{W}}\right|\overline{\phi_{\alpha(0)}} \gg\ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{n\omega}\big|\phi_{\beta m} \gg}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - m\omega} = \tag{S 18}$$

$$= \sum_{(\beta,m)\neq(\alpha,0)} \frac{\ll \overline{\phi_{\beta(0)}}\left|\widehat{\widehat{W}}_{-m\omega}\right|\overline{\phi_{\alpha(0)}} \gg\ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{n\omega+m\omega}\big|\overline{\phi_{\beta(0)}} \gg}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - m\omega}$$

We plug

$$\lambda\widehat{\mathrm{W}}(\mathrm{t}) = \lambda\Re\{\mathbf{p}\cdot\widehat{\boldsymbol{\mu}}e^{is\omega t}\} \tag{S 19}$$

into (S 18) since the system is perturbed by a linearly polarized electric field of amplitude λ, polarization $\boldsymbol{p}\in\mathbb{R}^2$, and frequency $s\omega$, where $\omega = 2\pi/T$ and s may be any integer.

$$\ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{n\omega}\big|\overline{\phi_{\alpha(1)}} \gg = \sum_{\substack{(\beta,m)\neq(\alpha,0)\\ g=\pm1}} \frac{\ll \overline{\phi_{\beta(0)}}\big|\boldsymbol{p}_{\boldsymbol{g}}\cdot\widehat{\boldsymbol{\mu}}_{(gs-m)\omega}\big|\overline{\phi_{\alpha(0)}} \gg\ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{(n+m)\omega}\big|\overline{\phi_{\beta(0)}} \gg}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - m\omega} \tag{S 20}$$

where $\boldsymbol{p}_g \equiv \mathfrak{R}\{\boldsymbol{p}\} + ig\mathfrak{I}\{\boldsymbol{p}\}$ and the time-independent matrix element $\mathbf{F}_{\mathrm{n}}^{\beta\alpha}$

$$\mathbf{F}_{\mathrm{n}}^{\beta\alpha} \equiv \ll \overline{\phi_{\beta(0)}}\big|\widehat{\boldsymbol{\mu}}_{\mathrm{n}\omega}\big|\overline{\phi_{\alpha(0)}} \gg \tag{S 21}$$

represents the $n\omega$ frequency component of the time-dependent matrix element:

$$\langle\phi_{\beta(0)}|\widehat{\boldsymbol{\mu}}|\phi_{\alpha(0)}\rangle = \sum_n \boldsymbol{F}_n^{\beta\alpha}e^{in\omega t} \tag{S 22}$$

Rewriting Eq.(S 20) using $\mathbf{F}_{\mathrm{n}}^{\beta\alpha}$, we obtain

$$\ll \overline{\phi_{\alpha(0)}}\big|\widehat{\boldsymbol{\mu}}_{n\omega}\big|\overline{\phi_{\alpha(1)}} \gg = \sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0)\\ \mathrm{g}=\pm1}} \frac{\left(\mathbf{p}_{\boldsymbol{g}}\cdot\mathbf{F}_{\mathrm{gs-m}}^{\beta\alpha}\right)\mathbf{F}_{\mathrm{n+m}}^{\alpha\beta}}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - \mathrm{m}\omega} \tag{S 23}$$

Plugging Eq.(S 23) into Eq.(S 17), we have

$$\widetilde{\mathbf{E}}_1(\Omega) = \sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0)\\ \mathrm{g}=\pm1}} \frac{\left(\mathbf{p}_{\boldsymbol{g}}\cdot\mathbf{F}_{\mathrm{gs-m}}^{\beta\alpha}\right)\mathbf{F}_{\mathrm{n+m}}^{\alpha\beta} + \left(\mathbf{p}_{\boldsymbol{g}}\cdot\mathbf{F}_{\mathrm{m-gs}}^{\alpha\beta}\right)\mathbf{F}_{\mathrm{n-m}}^{\beta\alpha}}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - \mathrm{m}\omega} \tag{S 24}$$

This is exactly Eq.(6) of the main text. Similarly, the term $\widetilde{\boldsymbol{E}}_2(\Omega)$ is given by

$$\widetilde{\boldsymbol{E}}_2(n\omega) = \ll \overline{\phi_{\alpha(1)}}\big|\widehat{\boldsymbol{\mu}}_{n\omega}\big|\overline{\phi_{\alpha(1)}} \gg = \tag{S 25}$$

$$= \frac{1}{4}\sum_{\substack{(\beta,m)\neq(\alpha,0)\\ (\kappa,l)\neq(\alpha,0)\\ g_1,g_2=\pm1}} \frac{\ll \overline{\phi_{\alpha(0)}}\big|\boldsymbol{p}_{\boldsymbol{g_1}}\cdot\widehat{\boldsymbol{\mu}}_{(l-g_1s)\omega}\big|\overline{\phi_{\kappa(0)}} \gg\ll \overline{\phi_{\beta(0)}}\ \big|\boldsymbol{p}_{\boldsymbol{g_2}}\cdot\widehat{\boldsymbol{\mu}}_{(g_2s-m)\omega}\big|\overline{\phi_{\alpha(0)}} \gg\ll \overline{\phi_{\kappa(0)}}\big|}{\left(\epsilon_{\alpha(0)} - \epsilon_{\kappa(0)} - l\omega\right)\left(\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - m\omega\right)}$$

Plugging in Eq.(S 21) into (S 25) we obtain

$$\tilde{\mathbf{E}}_2(\mathrm{n}\omega) = \frac{1}{4}\sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0)\\(\kappa,\mathrm{l})\neq(\alpha,0)\\ \mathrm{g}_1,\mathrm{g}_2=\pm1}} \frac{\left(\mathbf{p}_{g_1}\cdot\mathbf{F}^{\alpha\kappa}_{\mathrm{l}-\mathrm{g}_1\mathrm{s}}\right)\left(\mathbf{p}_{g_2}\cdot\mathbf{F}^{\beta\alpha}_{\mathrm{g}_2\mathrm{s}-\mathrm{m}}\right)\mathbf{F}^{\kappa\beta}_{\mathrm{n}+\mathrm{m}-\mathrm{l}}}{\left(\epsilon_{\alpha(0)}-\epsilon_{\kappa(0)}-\mathrm{l}\omega\right)\left(\epsilon_{\alpha(0)}-\epsilon_{\beta(0)}-\mathrm{m}\omega\right)} \tag{S26}$$

This is Eq.(7) of the main text.

S.II DERIVATION OF TABLE 1 IN THE MAIN TEXT

In this section, we derive the selection rules for breaking selection rules presented in Table 1 of the main text.

$\hat{C}_{N,M}$ and $\hat{e}_{N,M}$ symmetry breaking

The $\hat{C}_{N,M}$ operation is defined as $\hat{C}_{N,M} = \hat{R}_{N,M}\cdot\hat{\tau}_N$ where $\hat{R}_{N,M}$ is a $2\pi M/N$ rotation and $\hat{\tau}_N$ is a T/N time translation. The eigenvalues of the $\hat{R}_{N,M}$ operation are $exp(\pm i\,2\pi M/N)$ and the eigenvalues of $\hat{\tau}_N$ are $\{exp(-i\,2\pi k/N)\mid k=0,\dots,N-1\}$. Hence, the eigenvalues of $\hat{C}_{N,M}$ are $\left\{exp\left(-i\frac{2\pi k}{N}\pm i\frac{2\pi M}{N}\right)\mid k=0,\dots,N-1\right\} = \left\{exp\left(-i\frac{2\pi k}{N}\right)\mid k=0,\dots,N-1\right\}$. If the unperturbed system is $\hat{C}_{N,M}$ symmetric, the unperturbed Floquet states $|\phi_{\alpha(0)}\rangle, |\phi_{\beta(0)}\rangle$ are eigenfunctions of $\hat{C}_{N,M}$ with eigenvalues $e^{-i2\pi k_\alpha}, e^{-i2\pi k_\beta}$ respectively.

To obtain the selection rules for $\boldsymbol{F^{\beta\alpha}_n}$, we operate with $\boldsymbol{\hat{C}_{N,M}}$ on Eq.(S 22) to find

$$\begin{aligned}\hat{C}_{N,M}\langle\phi_{\beta(0)}|\hat{\boldsymbol{\mu}}|\phi_{\alpha(0)}\rangle &= e^{\frac{i2\pi(k_\beta-k_\alpha)}{N}}\sum_n \boldsymbol{F^{\beta\alpha}_n} e^{in\omega t}\\ &= \sum_n e^{\frac{i2\pi n}{N}}\hat{R}_{N,M}\cdot\boldsymbol{F^{\beta\alpha}_n} e^{in\omega t}\end{aligned} \tag{S 27}$$

, and

$$e^{\frac{i2\pi(k_\beta-k_\alpha-n)}{N}}\boldsymbol{F^{\beta\alpha}_n} = \hat{R}_{N,M}\cdot\boldsymbol{F^{\beta\alpha}_n} \tag{S 28}$$

Since the eigenvalues of $\boldsymbol{\hat{R}_{N,M}}$ are $\boldsymbol{e^{\pm i\frac{2\pi M}{N}}}$, Eq.(S 28) has a nontrivial solution (where $\boldsymbol{F^{\beta\alpha}_n}$ is nonzero) only if there exists an integer $\boldsymbol{z}\in\mathbb{Z}$ such that

$$n = N\times z + k_\beta - k_\alpha \pm M \tag{S 29}$$

We consider a specific $\boldsymbol{\beta, m, g}$ contribution to $\boldsymbol{\tilde{E}_1(n\omega)}$ (Eq.(S 24)). By Eq.(S 29), this contribution is nonzero only if there are two integers $\boldsymbol{z_1, z_2}$ that simultaneously fulfill

$$gs - m = N\times z_1 + k_\beta - k_\alpha \pm M \tag{S 30}$$

$$n + m = N\times z_2 + k_\alpha - k_\beta \pm M \tag{S 31}$$

By summing Eqs.(S 30)(S 31), we find that $\tilde{\mathbf{E}}_\mathbf{1}(\mathrm{n}\omega)$ is only nonzero for values of n for which there exists an integer $z\in\mathbb{Z}$ that fulfills one of the conditions

$$\begin{aligned}\pm s \pm n &= N\times z \pm 2M\\ \pm s\pm n &= N\times z\end{aligned} \tag{S 32}$$

Next, we consider a specific $(\boldsymbol{\beta}, \boldsymbol{m}), (\boldsymbol{\kappa}, \boldsymbol{l}), \boldsymbol{g_1}, \boldsymbol{g_2}$ contribution to $\boldsymbol{\tilde{E}_2(n\omega)}$ (Eq.(S26)). We denote the eigenvalues of $|\boldsymbol{\phi_{\alpha(0)}}\rangle, |\boldsymbol{\phi_{\beta(0)}}\rangle, |\boldsymbol{\phi_{\kappa(0)}}\rangle$ by $\boldsymbol{e^{-\frac{i2\pi k_\alpha}{N}}}, \boldsymbol{e^{-\frac{i2\pi k_\beta}{N}}}, \boldsymbol{e^{-\frac{i2\pi k_\kappa}{N}}}$ respectively. By Eq.(S 29) , there is a nonzero contribution only if there exists three integers $\boldsymbol{z_1, z_2, z_3}\in\mathbb{Z}$ such that

$$l - g_1 s = N z_1 + k_\alpha - k_\kappa \pm M \qquad \text{(S 33)}$$
$$g_2 s - m = N z_2 + k_\beta - k_\alpha \pm M$$
$$n + m - l = N z_3 + k_\kappa - k_\beta \pm M$$

By summing the three lines of Eq.(S 34), we find that $\widetilde{\boldsymbol{E}}_{\boldsymbol{2}}(n\omega)$ is only nonzero for values of n for which there exists three integers $z_1, z_2, z_3 \in \mathbb{Z}$ that fulfill

$$n + (g_2 - g_1)s = N(z_1 + z_2 + z_3) \pm M \pm M \pm M \qquad \text{(S 35)}$$

More compactly, $\widetilde{\boldsymbol{E}}_{\boldsymbol{2}}(n\omega)$ is only nonzero for values of n for which there exists an integer $z \in \mathbb{Z}$ that fulfills

$$n \pm (1 \pm 1)s = N \times z \pm (2 \pm 1)M \qquad \text{(S 36)}$$

We note that the same conditions also apply to discrete elliptical symmetries, denoted $\hat{e}_{N,M}$, where the operation $\hat{e}_{N,M}$ is defined as $\hat{e}_{N,M} = \hat{\tau}_N \cdot \hat{L}_b \cdot \hat{R}_{N,M} \cdot \hat{L}_{1/b}$ where:

$$\hat{L}_b = \begin{pmatrix} 1 & 0 \\ 0 & b \end{pmatrix} \qquad \text{(S 37)}$$

The derivation and selection rules remain the same, because the eigenvalues of $\hat{\boldsymbol{e}}_{\boldsymbol{N,M}}$ are identical to the eigenvalues of $\widehat{\boldsymbol{C}}_{\boldsymbol{N,M}}$, and the eigenvalues of $\hat{\boldsymbol{L}}_{\boldsymbol{b}} \cdot \widehat{\boldsymbol{R}}_{\boldsymbol{N,M}} \cdot \hat{\boldsymbol{L}}_{\boldsymbol{1/b}}$ are identical to the eigenvalues of $\widehat{\boldsymbol{R}}_{\boldsymbol{N,M}}$.

In the case where $\boldsymbol{s = r/q}$ is a rational number, the same selection rules may be used, since a $\widehat{\boldsymbol{C}}_{\boldsymbol{N,M}}$ symmetric Floquet system whose frequency is $\boldsymbol{\omega}$, perturbed by an $\boldsymbol{r\omega/q}$ perturbation can be treated as a $\widehat{\boldsymbol{C}}_{\boldsymbol{qN,qM}}$ symmetric Floquet system whose frequency is $\boldsymbol{\omega' = \omega/q}$, perturbed by an $\boldsymbol{r\omega'}$ perturbation.

$\hat{\mathbf{T}}$ symmetry breaking

The time reversal operation is denoted by $\hat{T}$. Its eigenvalues are ± 1. To obtain the selection rules for $\boldsymbol{F}_n^{\beta\alpha}$, we operate with $\hat{T}$ on Eq.(S 22):

$$\hat{T}\langle \phi_{\beta(0)} | \hat{\boldsymbol{\mu}} | \phi_{\alpha(0)} \rangle = \pm \sum_n \boldsymbol{F}_n^{\beta\alpha} e^{i\omega n t} = \sum_n \boldsymbol{F}_{-n}^{\beta\alpha} e^{i\omega n t} \qquad \text{(S 38)}$$

and

$$\pm \boldsymbol{F}_n^{\beta\alpha} = \boldsymbol{F}_{-n}^{\beta\alpha} \qquad \text{(S 39)}$$

where a plus (minus) sign is used when $|\boldsymbol{\phi}_{\boldsymbol{\alpha}(\mathbf{0})}\rangle, |\boldsymbol{\phi}_{\boldsymbol{\beta}(\mathbf{0})}\rangle$ have the same (different) eigenvalue. Therefore, if $\boldsymbol{\alpha}, \boldsymbol{\beta}$ have the same (different) eigenvalue $\boldsymbol{F}_{\boldsymbol{n}}^{\boldsymbol{\beta\alpha}}$ is real (imaginary). We consider a specific $\boldsymbol{\beta}, \boldsymbol{m}, \boldsymbol{g}$ contribution to $\widetilde{\boldsymbol{E}}_{\boldsymbol{1}}(\boldsymbol{n\omega})$ (Eq.(S 24)). The contribution is comprised of a multiplication of either two real entities, or two imaginary entities, therefore it is real. Since all contributions to Eq. (S 24) are real, $\tilde{\mathbf{E}}_{\mathbf{1}}(\boldsymbol{n\omega})$ is a real vector, i.e. it is linearly polarized and is in an equal or opposite phase with the perturbation. Next, we consider a specific $(\boldsymbol{\beta}, \boldsymbol{m}), (\boldsymbol{\kappa}, \boldsymbol{l}), \boldsymbol{g}_{\boldsymbol{1}}, \boldsymbol{g}_{\boldsymbol{2}}$ contribution to $\widetilde{\boldsymbol{E}}_{\boldsymbol{2}}(\boldsymbol{n\omega})$ (Eq.(S 25)). If $|\boldsymbol{\phi}_{\boldsymbol{\alpha,\beta,\kappa}(\mathbf{0})}\rangle$ all have the same eigenvalue, the contribution is comprised of a multiplication of three real entities and is therefore real. If one of $|\boldsymbol{\phi}_{\boldsymbol{\alpha,\beta,\kappa}(\mathbf{0})}\rangle$ has a different eigenvalue from the other two, the contribution is comprised of a multiplication of one real entity and two imaginary entities, hence the contribution is real. Therefore, $\tilde{\mathbf{E}}_{\mathbf{2}}(\boldsymbol{n\omega})$ is a real vector as well.

$\hat{\mathbf{Q}}$ symmetry breaking

The operation $\hat{Q}$ is defined by $\hat{Q} = \hat{T} \cdot \hat{R}_2$ where $\hat{T}$ is the time reversal operation and $\hat{R}_2$ is a π rotation. Its eigenvalues are ± 1. To obtain the selection rules for $\boldsymbol{F}_n^{\beta\alpha}$, we operate with $\hat{Q}$ on Eq.(S 22):

$$\hat{Q}\langle\phi_{\beta(0)}|\hat{\boldsymbol{\mu}}|\phi_{\alpha(0)}\rangle = \pm\sum_n \boldsymbol{F}_n^{\beta\alpha} e^{i\omega nt} = -\sum_n \boldsymbol{F}_{-n}^{\beta\alpha} e^{i\omega nt} \quad \text{(S 40)}$$

and

$$\mp\boldsymbol{F}_n^{\beta\alpha} = \boldsymbol{F}_{-n}^{\beta\alpha} \quad \text{(S 41)}$$

where a minus (plus) sign is used when $|\phi_{\alpha(0)}\rangle, |\phi_{\beta(0)}\rangle$ have the same (different) eigenvalue. Therefore, if α, β have the same (different) eigenvalue $\boldsymbol{F}_n^{\beta\alpha}$ is imaginary (real). From the same considerations as above, $\tilde{\boldsymbol{E}}_1(n\omega)$ is real and $\tilde{\boldsymbol{E}}_2(n\omega)$ is imaginary.

$\hat{\text{G}}$ symmetry breaking

The operation $\hat{G}$ is defined by $\hat{G} = \hat{T} \cdot \hat{\tau}_2 \cdot \hat{R}_2$ where $\hat{T}$ is the time reversal operation, $\hat{R}_2$ is a π rotation and $\hat{\tau}_2$ is a $T/2$ time translation. Its eigenvalues are ± 1. To obtain the selection rules for $\boldsymbol{F}_n^{\beta\alpha}$, we operate with $\hat{G}$ on Eq.(S 22):

$$\hat{G}\langle\phi_{\beta(0)}|\hat{\boldsymbol{\mu}}|\phi_{\alpha(0)}\rangle = \pm\sum_n \boldsymbol{F}_n^{\beta\alpha} e^{i\omega nt} = \sum_n (-1)^{n+1} \boldsymbol{F}_{-n}^{\beta\alpha} e^{i\omega nt} \quad \text{(S 42)}$$

and

$$\mp\boldsymbol{F}_n^{\beta\alpha} = (-1)^n \boldsymbol{F}_{-n}^{\beta\alpha} \quad \text{(S 43)}$$

	$\boldsymbol{\alpha, \beta}$ **have the same eigenvalue**	$\boldsymbol{\alpha, \beta}$ **have different eigenvalues**
n is even	$\boldsymbol{F}_n^{\beta\alpha} \in i\mathbb{R}$	$\boldsymbol{F}_n^{\beta\alpha} \in \mathbb{R}$
n is odd	$\boldsymbol{F}_n^{\beta\alpha} \in \mathbb{R}$	$\boldsymbol{F}_n^{\beta\alpha} \in i\mathbb{R}$

Table S1.
selection rules for $\hat{\text{G}}$ symmetric matrix elements.

We consider a specific $\boldsymbol{\beta}, \boldsymbol{m}, \boldsymbol{g}$ contribution to $\tilde{\boldsymbol{E}}_1(\boldsymbol{n\omega})$ (Eq.(S 24)). If s and n are of the same parity, $\boldsymbol{gs} - \boldsymbol{m}$ and $\boldsymbol{n} + \boldsymbol{m}$ are of the same parity as well, and the contribution is comprised of either a multiplication of real or two imaginary entities. Therefore, if s and n are of the same (different) parity $\tilde{\mathbf{E}}_1(\boldsymbol{n\omega})$ is real (imaginary). By a similar analysis, we find that $\tilde{\boldsymbol{E}}_2(\boldsymbol{2m\omega})$ is imaginary and $\tilde{\boldsymbol{E}}\big((\boldsymbol{2m}+\boldsymbol{1})\boldsymbol{\omega}\big)$ is real (Table S2).

$\tilde{\boldsymbol{E}}_1$	$\tilde{\boldsymbol{E}}_1\big((\boldsymbol{2m} \pm \boldsymbol{s})\boldsymbol{\omega}\big) \in \mathbb{R}^2$	$\tilde{\boldsymbol{E}}_1\big((\boldsymbol{2m} \pm \boldsymbol{s} + \boldsymbol{1})\boldsymbol{\omega}\big) \in \boldsymbol{i}\mathbb{R}^2$
$\tilde{\boldsymbol{E}}_2$	$\tilde{\boldsymbol{E}}_2(2m\omega) \in i\mathbb{R}^2$	$\tilde{\boldsymbol{E}}_2\big((2m+1)\omega\big) \in \mathbb{R}^2$

Table S2.
selection rules for $\tilde{\boldsymbol{E}}_1(\boldsymbol{n\omega}), \tilde{\boldsymbol{E}}_2(\boldsymbol{n\omega})$, given that the unperturbed system is $\hat{\boldsymbol{G}}$ symmetric.

$\widehat{D}_y$ symmetry breaking

The operation $\widehat{D}_y$ is defined by $\widehat{D}_y = \hat{T} \cdot \hat{\sigma}_y$ where $\hat{T}$ is the time reversal operation and $\hat{\sigma}_y$ is a reflection relative to $\hat{y}$ (i.e $x \rightarrow -x$). Its eigenvalues are ± 1. To obtain the selection rules for $\boldsymbol{F}_n^{\beta\alpha}$, we operate with $\widehat{D}_y$ on Eq.(S 22):

$$\widehat{D}_y \langle \phi_{\beta(0)} | \widehat{\boldsymbol{\mu}} | \phi_{\alpha(0)} \rangle = \pm \sum_n \begin{pmatrix} F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} = \sum_n \begin{pmatrix} -F_{-nx}^{\beta\alpha} \\ F_{-ny}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} \tag{S 44}$$

and

$$\pm \begin{pmatrix} F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} = \begin{pmatrix} -F_{-nx}^{\beta\alpha} \\ F_{-ny}^{\beta\alpha} \end{pmatrix} \tag{S 45}$$

where a plus (minus) sign is used when $|\boldsymbol{\phi}_{\alpha(0)}\rangle, |\boldsymbol{\phi}_{\beta(0)}\rangle$ have the same (different) eigenvalue. Table S3 is obtained from Eq.(S 45).

	α, β have the same eigenvalue	α, β have different eigenvalues
$\boldsymbol{F}_n^{\beta\alpha} \cdot \hat{\boldsymbol{x}}$	$\boldsymbol{F}_n^{\beta\alpha} \cdot \hat{\boldsymbol{x}} \in i\mathbb{R}$	$\boldsymbol{F}_n^{\beta\alpha} \cdot \hat{\boldsymbol{x}} \in \mathbb{R}$
$\boldsymbol{F}_n^{\beta\alpha} \cdot \hat{\boldsymbol{y}}$	$\boldsymbol{F}_n^{\beta\alpha} \cdot \hat{\boldsymbol{y}} \in \mathbb{R}$	$\boldsymbol{F}_n^{\beta\alpha} \cdot \hat{\boldsymbol{y}} \in i\mathbb{R}$

Table S3.

Selection rules for $\widehat{\mathrm{D}}_\mathrm{y}$ symmetric matrix elements.

By Eqs.(S 24)(S26) and Table S3,

$$\widetilde{\boldsymbol{E}}_{\boldsymbol{1}}(n\omega) = p_x \begin{pmatrix} a \\ ib \end{pmatrix} + p_y \begin{pmatrix} ic \\ d \end{pmatrix} \; ; a,b,c,d \in \mathbb{R} \tag{S 46}$$

$$\widetilde{\boldsymbol{E}}_{\boldsymbol{2}}(n\omega) = \left(p_x^2 a + p_y^2 b + i p_x p_y c\right) \begin{pmatrix} id \\ e \end{pmatrix} \; ; a,b,c,d,e \in \mathbb{R} \tag{S 47}$$

$\widehat{\mathrm{H}}_\mathrm{y}$ symmetry breaking

The operation $\widehat{H}_y$ is defined by $\widehat{H}_y = \hat{T} \cdot \hat{\tau}_2 \cdot \hat{\sigma}_y$ where $\hat{T}$ is the time reversal operation , $\hat{\sigma}_y$ is a reflection relative to the y axis (i.e $x \rightarrow -x$) and $\hat{\tau}_2$ is a $T/2$ time translation. Its eigenvalues are ± 1. To obtain the selection rules for $\boldsymbol{F}_n^{\beta\alpha}$, we operate with $\widehat{H}_y$ on Eq.(S 22):

$$\widehat{H}_y \langle \phi_{\beta(0)} | \widehat{\boldsymbol{\mu}} | \phi_{\alpha(0)} \rangle = \pm \sum_n \begin{pmatrix} F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} = \sum_n (-1)^n \begin{pmatrix} -F_{-nx}^{\beta\alpha} \\ F_{-ny}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} \tag{S 48}$$

and

$$\pm \begin{pmatrix} F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} = (-1)^n \begin{pmatrix} -F_{-nx}^{\beta\alpha} \\ F_{-ny}^{\beta\alpha} \end{pmatrix} \tag{S 49}$$

where a plus (minus) sign is used when $|\boldsymbol{\phi}_{\alpha(0)}\rangle, |\boldsymbol{\phi}_{\beta(0)}\rangle$ have the same (different) eigenvalue. Table S4 is obtained from Eq.(S 49).

	α, β have the same eigenvalue	α, β have different eigenvalues
$\boldsymbol{F}_{2n}^{\beta\alpha} \cdot \hat{\boldsymbol{x}}$	$\boldsymbol{F}_{2n}^{\beta\alpha} \cdot \hat{\boldsymbol{x}} \in i\mathbb{R}$	$\boldsymbol{F}_{2n}^{\beta\alpha} \cdot \hat{\boldsymbol{x}} \in \mathbb{R}$
$\boldsymbol{F}_{2n}^{\beta\alpha} \cdot \hat{\boldsymbol{y}}$	$\boldsymbol{F}_{2n}^{\beta\alpha} \cdot \hat{\boldsymbol{y}} \in \mathbb{R}$	$\boldsymbol{F}_{2n}^{\beta\alpha} \cdot \hat{\boldsymbol{y}} \in i\mathbb{R}$

$\boldsymbol{F}_{\boldsymbol{2n+1}}^{\boldsymbol{\beta\alpha}} \cdot \hat{\boldsymbol{x}}$	$\boldsymbol{F}_{\boldsymbol{2n+1}}^{\boldsymbol{\beta\alpha}} \cdot \hat{\boldsymbol{x}} \in \mathbb{R}$	$\boldsymbol{F}_{\boldsymbol{2n+1}}^{\boldsymbol{\beta\alpha}} \cdot \hat{\boldsymbol{x}} \in i\mathbb{R}$
$\boldsymbol{F}_{\boldsymbol{2n+1}}^{\boldsymbol{\beta\alpha}} \cdot \hat{\boldsymbol{y}}$	$\boldsymbol{F}_{\boldsymbol{2n+1}}^{\boldsymbol{\beta\alpha}} \cdot \hat{\boldsymbol{y}} \in i\mathbb{R}$	$\boldsymbol{F}_{\boldsymbol{2n+1}}^{\boldsymbol{\beta\alpha}} \cdot \hat{\boldsymbol{y}} \in \mathbb{R}$

Table S4.
Selection rules for $\widehat{\mathrm{H}}_{\mathrm{y}}$ symmetric matrix elements

By Eqs. (S 24)(S26) and Table S4, $\tilde{\mathbf{E}}_{\mathbf{1}}(\mathbf{n\omega})$ scales as:

$$\widetilde{\boldsymbol{E}}_{\boldsymbol{1}}((2m \pm s)\omega) = p_x \begin{pmatrix} a \\ ib \end{pmatrix} + p_y \begin{pmatrix} ic \\ d \end{pmatrix} \; ; a, b, c, d \in \mathbb{R} \qquad \text{(S 50)}$$

$$\widetilde{\boldsymbol{E}}_{\boldsymbol{1}}\big((2m \pm s + 1)\omega\big) = p_x \begin{pmatrix} ia \\ b \end{pmatrix} + p_y \begin{pmatrix} c \\ id \end{pmatrix} \; ; a, b, c, d \in \mathbb{R} \qquad \text{(S 51)}$$

and $\widetilde{\boldsymbol{E}}_{\boldsymbol{2}}(n\omega)$ scales as

$$\widetilde{\boldsymbol{E}}_{\boldsymbol{2}}(2m\omega) = \left(ip_x^2 a + ip_y^2 b + p_x p_y c\right) \begin{pmatrix} d \\ ie \end{pmatrix} \; ; a, b, c, d, e \in \mathbb{R} \qquad \text{(S 52)}$$

$$\widetilde{\boldsymbol{E}}_{\boldsymbol{2}}\big((2m+1)\omega\big) = \left(ip_x^2 a + ip_y^2 b + p_x p_y c\right) \begin{pmatrix} id \\ e \end{pmatrix} \; ; a, b, c, d, e \in \mathbb{R} \qquad \text{(S 53)}$$

$\hat{\mathbf{Z}}_{\mathbf{y}}$ symmetry breaking

The operation $\hat{\mathrm{Z}}_{\mathrm{y}}$ is defined by $\hat{\mathrm{Z}}_{\mathrm{y}} = \hat{\tau}_2 \cdot \hat{\sigma}_y$ where $\hat{\sigma}_y$ is a reflection relative to the y axis (i.e. $x \to -x$) and $\hat{\tau}_2$ is a $T/2$ time translation. Its eigenvalues are ± 1. To obtain the selection rules for $F_n^{\beta\alpha}$, we operate with $\hat{Z}_y$ on Eq.(S 22):

$$\hat{Z}_y \langle \phi_{\beta(0)} | \hat{\mu} | \phi_{\alpha(0)} \rangle = \pm \sum_n \begin{pmatrix} F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} e^{i\omega n t} = \sum_n (-1)^n \begin{pmatrix} -F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} e^{i\omega n t} \qquad \text{(S 54)}$$

and

$$\pm \begin{pmatrix} F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} = (-1)^n \begin{pmatrix} -F_{nx}^{\beta\alpha} \\ F_{ny}^{\beta\alpha} \end{pmatrix} \qquad \text{(S 55)}$$

where a plus (minus) sign is used when $|\phi_{\alpha(0)}\rangle, |\phi_{\beta(0)}\rangle$ have the same (different) eigenvalue. Table S5 is obtained from Eq.(S 55).

	α, β have the same eigenvalue	**α, β have different eigenvalues**
$\boldsymbol{F}_{\boldsymbol{2n}} \cdot \hat{\boldsymbol{x}}$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{x}}_{(2n)\omega} \big| \overline{\phi_{\alpha(0)}} \gg = 0$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{x}}_{(2n)\omega} \big| \overline{\phi_{\alpha(0)}} \gg \neq 0$
$\boldsymbol{F}_{\boldsymbol{2n}} \cdot \hat{\boldsymbol{y}}$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{y}}_{(2n)\omega} \big| \overline{\phi_{\alpha(0)}} \gg \neq 0$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{y}}_{(2n)\omega} \big| \overline{\phi_{\alpha(0)}} \gg = 0$
$\boldsymbol{F}_{\boldsymbol{2n+1}} \cdot \hat{\boldsymbol{x}}$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{x}}_{(2n+1)\omega} \big| \overline{\phi_{\alpha(0)}} \gg \neq 0$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{x}}_{(2n+1)\omega} \big| \overline{\phi_{\alpha(0)}} \gg = 0$
$\boldsymbol{F}_{\boldsymbol{2n+1}} \cdot \hat{\boldsymbol{y}}$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{y}}_{(2n+1)\omega} \big| \overline{\phi_{\alpha(0)}} \gg = 0$	$\ll \overline{\phi_{\beta(0)}} \big| \hat{\hat{y}}_{(2n+1)\omega} \big| \overline{\phi_{\alpha(0)}} \gg \neq 0$

Table S5.
Selection rules for $\hat{\mathrm{Z}}_{\mathrm{y}}$ symmetric matrix elements

By Eqs. (S 24)(S26) and Table S5, $\tilde{\mathbf{E}}_{\mathbf{1}}$ scales as:

$$\tilde{\mathbf{E}}_{\mathbf{1}}\big((2m \pm s)\omega\big) = \begin{pmatrix} p_x a \\ p_y b \end{pmatrix} \;; a, b \in \mathbb{C} \tag{S 56}$$

$$\tilde{\mathbf{E}}_{\mathbf{1}}\big((2m \pm s + 1)\omega\big) = \begin{pmatrix} p_y a \\ p_x b \end{pmatrix} \;; a, b \in \mathbb{C} \tag{S 57}$$

and $\tilde{\mathbf{E}}_{\mathbf{2}}$ scales as

$$\tilde{\mathbf{E}}_{\mathbf{2}}(2m\omega) = \begin{pmatrix} p_x p_y a \\ p_x^2 b + p_y^2 c \end{pmatrix} \;; a, b, c \in \mathbb{C} \tag{S 58}$$

$$\tilde{\mathbf{E}}_{\mathbf{2}}\big((2m \pm 1)\omega\big) = \begin{pmatrix} p_x^2 a + p_y^2 b \\ p_x p_y a \end{pmatrix} \;; a, b, c \in \mathbb{C} \tag{S 59}$$

S.III DERIVATION OF TABLE 1 IN CIRCULARLY POLARIZED BASIS

In this section we derive selection rules for breaking selection rules induced by circularly polarized perturbations. That is, we consider perturbations whose polarization vector can be written in a cartesian basis as

$$\boldsymbol{p}^{(cartesian)} = \frac{p_R}{\sqrt{2}}(1, -i) + \frac{p_L}{\sqrt{2}}(1, i) \tag{S 60}$$

where $p_R, p_L \in \mathbb{R}$ are the amplitudes of the right-handed and left-handed circular polarization components of the perturbation. In circularly polarized basis (CP), p is written as

$$\boldsymbol{p}^{(CP)} = (p_R, p_L) \tag{S 61}$$

Notably, Eqs. (S 24)(S26) are basis independent. Therefore, in this section, we write vector quantities in the CP basis, for example $\tilde{\mathbf{E}}_1(\mathrm{n}\omega) = \big(\tilde{\mathrm{E}}_{1\mathrm{R}}(\mathrm{n}\omega), \tilde{\mathrm{E}}_{1\mathrm{L}}(\mathrm{n}\omega)\big)$, $\tilde{\mathbf{E}}_2(\mathrm{n}\omega) = \big(\tilde{\mathrm{E}}_{2\mathrm{R}}(\mathrm{n}\omega), \tilde{\mathrm{E}}_{2\mathrm{L}}(\mathrm{n}\omega)\big)$, $\mathbf{F}_{\mathrm{n}}^{\beta\alpha} = \big(\mathbf{F}_{\mathrm{R,n}}^{\beta\alpha}, \mathbf{F}_{\mathrm{L,n}}^{\beta\alpha}\big)$ etc. The results of this section are summarized in Table S6.

Dynamical Symmetry	$\tilde{\mathbf{E}}_1(\mathbf{n\omega})$ and $\tilde{\mathbf{E}}_2(\mathbf{n\omega})$ selection rules	
$\hat{\mathbf{C}}_{\mathbf{N,M}}$ $\hat{\mathbf{e}}_{\mathbf{N,M}}$	RCP perturbation: $\tilde{E}_{1R}(n_R\omega) \rightarrow n_R = Nq \pm s - 2M$ $\tilde{E}_{1L}(n_L\omega) \rightarrow n_L = Nq \pm s$ $\tilde{E}_{2R}(n_R\omega) \rightarrow n_R = Nq \pm (1 \pm 1)s - 3M$ $\tilde{E}_{2L}(n_R\omega) \rightarrow n_L = Nq \pm (1 \pm 1)s - M$	LCP perturbation: $\tilde{E}_{1R}(n_R\omega) \rightarrow n_R = Nq \pm s$ $\tilde{E}_{1L}(n_L\omega) \rightarrow n_L = Nq \pm s + 2M$ $\tilde{E}_{2R}(n_R\omega) \rightarrow n_R = Nq \pm (1 \pm 1)s + M$ $\tilde{E}_{2L}(n_R\omega) \rightarrow n_L = Nq \pm (1 \pm 1)s + 3M$
$\hat{\mathbf{T}}$	$\tilde{\mathbf{E}}_1(\mathrm{n\omega})$ and $\tilde{\mathbf{E}}_2(\mathrm{n\omega})$ are real vectors in $\mathbb{R}^2$ when represented in the circularly polarized basis	
$\hat{\mathbf{Q}}$	$\tilde{\mathbf{E}}_1(\mathrm{n\omega}) \in \mathbb{R}^2$; $\tilde{\mathbf{E}}_2(\mathrm{n\omega}) \in i\mathbb{R}^2$ (when represented in the circularly polarized basis)	
$\hat{\mathbf{D}}_{\mathbf{y}}$	$\tilde{\boldsymbol{E}}_1(n\omega) = \begin{pmatrix} p_R a + p_L \bar{b} \\ p_R b + p_L \bar{a} \end{pmatrix}$; $a, b \in \mathbb{C}$ $\tilde{\boldsymbol{E}}_2(n\omega) = \begin{pmatrix} p_R^2 a + p_R p_L b + p_L^2 c \\ p_R^2 \bar{c} + p_R p_L \bar{b} + p_L^2 \bar{a} \end{pmatrix}$; $a, b, c \in \mathbb{C}$	
$\hat{\mathbf{H}}_{\mathbf{y}}$	$\tilde{\boldsymbol{E}}_1(n\omega) = \begin{pmatrix} p_R a + p_L b \\ p_L \bar{b}(-1)^{n+s} + p_L \bar{a}(-1)^{n+s} \end{pmatrix}$; $a, b \in \mathbb{C}$ $\tilde{\boldsymbol{E}}_2(n\omega) = \begin{pmatrix} p_R^2 a + p_R p_L b + p_L^2 c \\ (-1)^{n+1}(p_R^2 \bar{c} + p_R p_L \bar{b} + p_L^2 \bar{a}) \end{pmatrix}$; $a, b, c \in \mathbb{C}$	
$\hat{\mathbf{G}}$	$\tilde{\mathbf{E}}_1\big((2\mathrm{m} \pm \mathrm{s})\omega\big) \in \mathbb{R}^2$; $\tilde{\mathbf{E}}_1\big((2\mathrm{m} + 1 \pm \mathrm{s})\omega\big) \in i\mathbb{R}^2$ when represented in the circularly polarized basis. $\tilde{\mathbf{E}}_2(2\mathrm{m}\omega) \in i\mathbb{R}^2$; $\tilde{\mathbf{E}}_2\big((2\mathrm{m}+1)\omega\big) \in \mathbb{R}^2$ when represented in the circularly polarized basis	
$\hat{\mathbf{Z}}_{\mathbf{y}}$	$\tilde{\boldsymbol{E}}_1(n\omega) = \begin{pmatrix} p_R a + p_L b \\ p_L b(-1)^{n+s} + p_L a(-1)^{n+s} \end{pmatrix}$; $a, b \in \mathbb{C}$ $\tilde{\boldsymbol{E}}_2(n\omega) = \begin{pmatrix} p_R^2 a + p_R p_L b + p_L^2 c \\ (-1)^{n+1}(p_R^2 c + p_R p_L b + p_L^2 a) \end{pmatrix}$; $a, b, c \in \mathbb{C}$	

Table S6.
Selection rules for $\tilde{E}_1(\mathrm{n\omega})$ and $\tilde{E}_2(\mathrm{n\omega})$ written in circularly polarized basis.

$\hat{\mathbf{C}}_{\mathbf{N,M}}$ and $\hat{\mathbf{e}}_{\mathbf{N,M}}$ symmetry breaking

RCP perturbation: We assume that the perturbation is right-handed circularly polarized, i.e. $\boldsymbol{p}$ can be written in a circularly polarized basis as $\boldsymbol{p} = (p_R, 0)$ where $p_R \in \mathbb{R}$. Plugging in $\boldsymbol{p} = (p_R, 0)$, Eqs.(S 24)(S26) read in the CP basis:

$$\tilde{\mathrm{E}}_1(n\omega) = \sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0) \\ \mathrm{g}=\pm1}} \frac{\left(p_R \mathrm{F}_{\mathrm{R,gs-m}}^{\beta\alpha}\right)\mathbf{F}_{\mathrm{n+m}}^{\alpha\beta} + \left(\mathrm{p_R} \mathrm{F}_{\mathrm{R,m-gs}}^{\alpha\beta}\right)\mathbf{F}_{\mathrm{n-m}}^{\beta\alpha}}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - \mathrm{m}\omega} \qquad \text{(S 62)}$$

$$\tilde{\mathbf{E}}_2(n\omega) = \frac{1}{4} \sum_{\substack{(\beta,m)\neq(\alpha,0)\\(\kappa,l)\neq(\alpha,0)\\g_1,g_2=\pm1}} \frac{\left(p_R \cdot F^{\alpha\kappa}_{R,l-g_1 s}\right)\left(p_R \cdot F^{\beta\alpha}_{R,g_2 s-m}\right)\mathbf{F}^{\kappa\beta}_{n+m-l}}{\left(\epsilon_{\alpha(0)} - \epsilon_{\kappa(0)} - l\omega\right)\left(\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - m\omega\right)} \quad \text{(S 63)}$$

To obtain the selection rules for $\tilde{\mathbf{E}}_1(n\omega), \tilde{\mathbf{E}}_2(n\omega)$, we first need to obtain the selection rules for $\boldsymbol{F}^{\beta\alpha}_{R,n}, \boldsymbol{F}^{\beta\alpha}_{L,n}$. They can be derived from Eq. (S 28),

$$e^{\frac{i2\pi(k_\beta - k_\alpha - n)}{N}} \begin{pmatrix} F^{\beta\alpha}_{Rn} \\ F^{\beta\alpha}_{Ln} \end{pmatrix} = \hat{R}_{N,M} \cdot \begin{pmatrix} F^{\beta\alpha}_{Rn} \\ F^{\beta\alpha}_{Ln} \end{pmatrix} = \begin{pmatrix} e^{\frac{i2\pi M}{N}} F^{\beta\alpha}_{Rn} \\ e^{-\frac{i2\pi}{N}M} F^{\beta\alpha}_{Ln} \end{pmatrix} \quad \text{(S 64)}$$

$\boldsymbol{F}^{\beta\alpha}_{R\backslash L, n_{R\backslash L}}$ is only nonzero for

$$n_{R\backslash L} = Nq + k_\beta - k_\alpha \mp M \quad \text{(S 65)}$$

where a minus (plus) sign is used for $\boldsymbol{F}^{\boldsymbol{\beta\alpha}}_{\boldsymbol{R,n_R}}\left(\boldsymbol{F}^{\boldsymbol{\beta\alpha}}_{\boldsymbol{L,n_L}}\right)$. We consider a specific $\boldsymbol{\beta}, \boldsymbol{m}, \boldsymbol{g}$ contribution to $\widetilde{\boldsymbol{E}}_{\boldsymbol{1R}}(\boldsymbol{n\omega})$ (Eq.(S 62)). This contribution is nonzero only if there are two integers $\boldsymbol{z_1}, \boldsymbol{z_2}$ that simultaneously fulfill

$$gs - m = N \times z_1 + k_\beta - k_\alpha - M \quad \text{(S 66)}$$
$$n_R + m = N \times z_2 + k_\alpha - k_\beta - M$$

That is, for an RCP perturbation, $\tilde{\mathbf{E}}_{1\mathrm{R}}(n_R\omega)$ is nonzero only for $n_R = N \times z \pm s - 2M$. Similarly, $\tilde{\mathbf{E}}_{1\mathrm{L}}(n_L\omega)$ is only nonzero for $n_L = Nz \pm s$.
Next, we consider a specific $(\beta, m), (\kappa, l), g_1, g_2$ contribution to $\widetilde{\boldsymbol{E}}_2(n\omega)$ (Eq.(S 63)). We denote the eigenvalues of $|\phi_{\alpha(0)}\rangle, |\phi_{\beta(0)}\rangle, |\phi_{\kappa(0)}\rangle$ by $e^{-\frac{i2\pi k_\alpha}{N}}, e^{-\frac{i2\pi k_\beta}{N}}, e^{-\frac{i2\pi k_\kappa}{N}}$ respectively. By Eq.(S 65) , there is a nonzero contribution only if there exists three integers $z_1, z_2, z_3 \in \mathbb{Z}$ such that

$$l - g_1 s = Nz_1 + k_\alpha - k_\kappa - M \quad \text{(S 67)}$$
$$g_2 s - m = Nz_2 + k_\beta - k_\alpha - M$$
$$n_{R/L} + m - l = Nz_3 + k_\kappa - k_\beta \mp M$$

For an RCP perturbation, $\tilde{E}_{2R}(n_R\omega), \tilde{E}_{2L}(n_L\omega)$ are nonzero only for values of $n_{R\backslash L}$ for which there exists an integer $z \in \mathbb{Z}$ that fulfills

$$n_R = N \times z \pm (1 \pm 1)s - 3M \quad \text{(S 68)}$$
$$n_L = N \times z \pm (1 \pm 1)s - M$$

LCP perturbation: By similar arguments, when the perturbation is LCP, $\tilde{\mathbf{E}}_{1\mathrm{R}}(n_R\omega)$ is nonzero only for $n_R = N \times z \pm s$ and $\tilde{\mathbf{E}}_{1\mathrm{L}}(n_L\omega)$ is only nonzero for $n_L = Nz \pm s + 2M.$, $\widetilde{\boldsymbol{E}}_{\boldsymbol{2R}}(n_R\omega), \widetilde{\boldsymbol{E}}_{\boldsymbol{2L}}(n_L\omega)$ are only nonzero for

$$n_R = N \times z \pm (1 \pm 1)s + M \quad \text{(S 69)}$$
$$n_L = N \times z \pm (1 \pm 1)s + 3M$$

$\hat{\boldsymbol{T}}, \hat{\boldsymbol{Q}}, \hat{\boldsymbol{G}}$ symmetry breaking

For $\hat{T}, \hat{Q}$ and $\hat{G}$ symmetries, the same derivations as Table 1 of the main text hold, with the only difference being that they are carried out in the CP basis. Therefore, the structure of the results is unchanged. For example, if we obtained that for a linearly polarized perturbation $\widetilde{\boldsymbol{E}}_1(n\omega)$ is a real vector in the cartesian basis, for a CP perturbation, $\widetilde{\boldsymbol{E}}_1(n\omega)$ is a real vector in the CP basis.

$\widehat{\boldsymbol{T}}$ symmetry breaking

In the CP basis, $\tilde{\mathrm{E}}_1(\mathrm{n}\omega), \tilde{\mathrm{E}}_2(\mathrm{n}\omega)$ are real vectors.

$\widehat{\mathbf{Q}}$ symmetry breaking

In the CP basis, $\tilde{\mathrm{E}}_1(\mathrm{n}\omega)$ is real and $\tilde{\mathrm{E}}_2(\mathrm{n}\omega)$ is imaginary.

$\widehat{\mathbf{G}}$ symmetry breaking

In the CP basis, $\tilde{\mathrm{E}}_1\big((2\mathrm{m} \pm \mathrm{s})\omega\big) \in \mathbb{R}^2$; $\tilde{\mathrm{E}}_1\big((2\mathrm{m}+1 \pm \mathrm{s})\omega\big) \in i\mathbb{R}^2$ and $\tilde{\mathrm{E}}_2(2\mathrm{m}\omega) \in i\mathbb{R}^2$; $\tilde{\mathrm{E}}_2\big((2\mathrm{m}+1)\omega\big) \in \mathbb{R}^2$.

$\widehat{\boldsymbol{D}}_{\boldsymbol{y}}$ symmetry breaking

To obtain the selection rules for $F_{nR}^{\beta\alpha}, F_{nL}^{\beta\alpha}$, we operate with $\widehat{D}_y$ on Eq. (S 22):

$$\widehat{D}_y\langle\phi_{\beta(0)}|\widehat{\boldsymbol{\mu}}|\phi_{\alpha(0)}\rangle = \pm\sum_n \begin{pmatrix} F_{nR}^{\beta\alpha} \\ F_{nL}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} = \sum_n \begin{pmatrix} -F_{-nL}^{\beta\alpha} \\ -F_{-nR}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} \tag{S 70}$$

Thus

$$F_{nR}^{\beta\alpha} = \mp F_{-nL}^{\beta\alpha} = \mp\overline{F_{nL}^{\beta\alpha}} \tag{S 71}$$

where a minus (plus) sign is used when $|\boldsymbol{\phi}_{\boldsymbol{\alpha}(\mathbf{0})}\rangle, |\boldsymbol{\phi}_{\boldsymbol{\beta}(\mathbf{0})}\rangle$ have the same (different) eigenvalue.

Consider the RCP and LCP components of $\tilde{\boldsymbol{E}}_1(n\omega)$:

$$\tilde{\mathbf{E}}_{1\mathrm{R}}(n\omega) = \sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0)\\ \mathrm{g}=\pm1}} \frac{\left(p_R\mathrm{F}_{\mathrm{R,gs-m}}^{\beta\alpha} + p_L\mathrm{F}_{\mathrm{L,gs-m}}^{\beta\alpha}\right)\mathrm{F}_{\mathrm{R,n+m}}^{\alpha\beta} + \left(\mathrm{p_R}\mathrm{F}_{\mathrm{R,m-gs}}^{\alpha\beta} + \mathrm{p_L}\mathrm{F}_{\mathrm{L,m-gs}}^{\alpha\beta}\right)\mathrm{F}_{\mathrm{R,n-m}}^{\beta\alpha}}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - \mathrm{m}\omega} \tag{S72}$$

$$\tilde{\mathbf{E}}_{1\mathrm{L}}(n\omega) = \sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0)\\ \mathrm{g}=\pm1}} \frac{\left(p_R\mathrm{F}_{\mathrm{R,gs-m}}^{\beta\alpha} + p_L\mathrm{F}_{\mathrm{L,gs-m}}^{\beta\alpha}\right)\mathrm{F}_{\mathrm{R,n+m}}^{\alpha\beta} + \left(\mathrm{p_R}\mathrm{F}_{\mathrm{R,m-gs}}^{\alpha\beta} + \mathrm{p_L}\mathrm{F}_{\mathrm{L,m-gs}}^{\alpha\beta}\right)\mathrm{F}_{\mathrm{R,n-m}}^{\beta\alpha}}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - \mathrm{m}\omega} \tag{S73}$$

By Eqs.((S **71**(S72(S73), $\tilde{E}_1(n\omega)$ can be written as

$$\tilde{\boldsymbol{E}}_{\mathbf{1}}(n\omega) = \begin{pmatrix} p_R a + p_L\bar{b} \\ p_R b + p_L\bar{a} \end{pmatrix} ; a, b \in \mathbb{C} \tag{S74}$$

Consider the RCP and LCP components of $\tilde{\boldsymbol{E}}_2(n\omega)$:

$$\tilde{E}_{2\mathrm{R}}(\mathrm{n}\omega) = \frac{1}{4}\sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0)\\(\kappa,\mathrm{l})\neq(\alpha,0)\\ \mathrm{g}_1,\mathrm{g}_2=\pm1}} \frac{\begin{pmatrix} \mathrm{p}_R^2\mathrm{F}_{\mathrm{R,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{R,\mathrm{g_2s-m}}^{\beta\alpha} + p_Rp_L\mathrm{F}_{\mathrm{R,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{L,\mathrm{g_2s-m}}^{\beta\alpha} + \\ +p_Lp_R\mathrm{F}_{\mathrm{L,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{R,\mathrm{g_2s-m}}^{\beta\alpha} + \mathrm{p}_L^2\mathrm{F}_{\mathrm{L,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{L,\mathrm{g_2s-m}}^{\beta\alpha} \end{pmatrix}\mathrm{F}_{\mathrm{R,n+m-l}}^{\kappa\beta}}{\big(\epsilon_{\alpha(0)} - \epsilon_{\kappa(0)} - \mathrm{l}\omega\big)\big(\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - \mathrm{m}\omega\big)}$$

(S 75)

$$\tilde{E}_{2\mathrm{L}}(\mathrm{n}\omega) = \frac{1}{4}\sum_{\substack{(\beta,\mathrm{m})\neq(\alpha,0)\\(\kappa,\mathrm{l})\neq(\alpha,0)\\ \mathrm{g}_1,\mathrm{g}_2=\pm1}} \frac{\begin{pmatrix} \mathrm{p}_R^2\mathrm{F}_{\mathrm{R,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{R,\mathrm{g_2s-m}}^{\beta\alpha} + p_Rp_L\mathrm{F}_{\mathrm{R,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{L,\mathrm{g_2s-m}}^{\beta\alpha} + \\ +p_Lp_R\mathrm{F}_{\mathrm{L,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{R,\mathrm{g_2s-m}}^{\beta\alpha} + \mathrm{p}_L^2\mathrm{F}_{\mathrm{L,l-g_1s}}^{\alpha\kappa}\mathrm{F}_{L,\mathrm{g_2s-m}}^{\beta\alpha} \end{pmatrix}\mathrm{F}_{\mathrm{L,n+m-l}}^{\kappa\beta}}{\big(\epsilon_{\alpha(0)} - \epsilon_{\kappa(0)} - \mathrm{l}\omega\big)\big(\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - \mathrm{m}\omega\big)}$$

(S 76)

By Eq.((S73), we can write $\tilde{\boldsymbol{E}}_{\mathbf{2}}(n\omega)$ as

$$\tilde{E}_2(n\omega) = \begin{pmatrix} p_R^2 a + p_R p_L b + p_L^2 c \\ p_R^2 \bar{c} + p_R p_L \bar{b} + p_L^2 \bar{a} \end{pmatrix}; a, b, c \in \mathbb{C} \tag{S77}$$

$\widehat{\mathbf{H}}_{\mathbf{y}}$ symmetry breaking

To obtain the selection rules for $F_{nR}^{\beta\alpha}, F_{nL}^{\beta\alpha}$, we operate with $\widehat{H}_y$ on Eq. (S 22):

$$\widehat{H}_y \langle \phi_{\beta(0)} | \widehat{\boldsymbol{\mu}} | \phi_{\alpha(0)} \rangle = \pm \sum_n \begin{pmatrix} F_{nL}^{\beta\alpha} \\ F_{nR}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} = \sum_n (-1)^n \begin{pmatrix} -F_{-nL}^{\beta\alpha} \\ -F_{-nR}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} \tag{S78}$$

and

$$\pm \begin{pmatrix} F_{nL}^{\beta\alpha} \\ F_{nR}^{\beta\alpha} \end{pmatrix} = (-1)^{n+1} \overline{\begin{pmatrix} F_{nL}^{\beta\alpha} \\ F_{nR}^{\beta\alpha} \end{pmatrix}} \tag{S79}$$

where a plus (minus) sign is used when $|\boldsymbol{\phi}_{\boldsymbol{\alpha}(\mathbf{0})}\rangle, |\boldsymbol{\phi}_{\boldsymbol{\beta}(\mathbf{0})}\rangle$ have the same (different) eigenvalue. By Eqs.(S72(S73(S79)

$$\tilde{E}_1(n\omega) = \begin{pmatrix} p_R a + p_L b \\ p_L \bar{b}(-1)^{n+s} + p_L \bar{a}(-1)^{n+s} \end{pmatrix} \tag{S80}$$

Similarly, by Eqs.((S 75(S 76(S79)

$$\tilde{E}_2(n\omega) = \begin{pmatrix} p_R^2 a + p_R p_L b + p_L^2 c \\ (-1)^{n+1}(p_R^2 \bar{c} + p_R p_L \bar{b} + p_L^2 \bar{a}) \end{pmatrix} \tag{S81}$$

$\widehat{\mathbf{Z}}_{\mathbf{y}}$ symmetry breaking

To obtain the selection rules for $F_{nR}^{\beta\alpha}, F_{nL}^{\beta\alpha}$, we operate with $\hat{Z}_y$ on Eq.(S 22):

$$\begin{aligned} \pm \sum_n \begin{pmatrix} F_{nR}^{\beta\alpha} \\ F_{nL}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} &= \sum_n (-1)^n \hat{\sigma}_y \begin{pmatrix} F_{nR}^{\beta\alpha} \\ F_{nL}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} \\ &= \sum_n (-1)^{n+1} \begin{pmatrix} F_{nL}^{\beta\alpha} \\ F_{nR}^{\beta\alpha} \end{pmatrix} e^{i\omega nt} \end{aligned} \tag{S82}$$

That is

$$\pm F_{nR}^{\beta\alpha} = (-1)^{n+1} F_{nL}^{\beta\alpha} \tag{S83}$$

By Eqs.((S72(S73,(S83)

$$\tilde{E}_1(n\omega) = \begin{pmatrix} p_R a + p_L b \\ p_L b(-1)^{n+s} + p_L a(-1)^{n+s} \end{pmatrix} \tag{S84}$$

By Eqs.((S **75**(S 76,(S83)

$$\tilde{E}_2(n\omega) = \begin{pmatrix} p_R^2 a + p_R p_L b + p_L^2 c \\ (-1)^{n+1}(p_R^2 c + p_R p_L b + p_L^2 a) \end{pmatrix} \tag{S85}$$

S.IV GENERALIZATION FOR FRACTIONAL HARMONIC PERTURBATIONS

In this section, we discuss the generalization of previous section derivations to account for fractional Floquet harmonic perturbations. Generally, the selection rules for all DS can be applied in their present form for rational values of s, by reformulating the problem in terms of an appropriate fundamental Floquet frequency. Alternatively, they may be explicitly derived, using the guidelines presented here.

To illustrate the first point, we show in the next paragraph how the predictions for $\hat{C}_{N,M}$ symmetry breaking can be applied for fractional harmonic perturbations of frequency $s = r/q$ (where r, q are integers) without an additional derivation. A T-periodic, $\hat{C}_{N,M}$ symmetric, Floquet system, perturbed by an $s\omega = r\omega/q$ perturbation, is equivalent to qT periodic, $\hat{C}_{qN,qM}$ symmetric Floquet system perturbed by an $r\omega' \equiv r\omega/q$ perturbation. Therefore, the selection rules for $\widetilde{\boldsymbol{E}}_1(n'\omega'), \widetilde{\mathbf{E}}_\mathbf{2}(n'\omega')$ are obtained by substituting $N \to qN, M \to qM, n \to n' = qn, s \to qs = r$ into the selection rules that have been derived in the previous section. Notably, the selection rules are invariant under this substitution. A similar reformulation may be carried out for other DSs.

Alternatively, the selection rules may be explicitly derived using the following perturbation term :

$$\lambda\widehat{W}(t) = \lambda\Re\left\{\boldsymbol{p}\cdot\widehat{\boldsymbol{\mu}}e^{ir\omega t/q}\right\} \tag{S 86}$$

and modified versions of Eqs. (S 24)(S26). We denote the fundamental frequency of the perturbed system by $\omega' \equiv \omega/q$, and the frequency of the perturbation by $r\omega'$ where r is an integer.

The $n'\omega'$ frequency component of the linear contribution $\widetilde{\boldsymbol{E}}_1(n'\omega')$ is calculated by a modified version of Eq.(S 20):

$$\ll \overline{\phi_{\alpha(0)}}\left|\widehat{\boldsymbol{\mu}}_{n'\omega/q}\right|\overline{\phi_{\alpha(1)}} \gg = \tag{S 87}$$

$$= \sum_{\substack{(\beta,m)\neq(\alpha,0)\\ g=\pm1}} \frac{\ll \overline{\phi_{\beta(0)}}\left|\boldsymbol{p}\cdot\widehat{\boldsymbol{\mu}}_{(g\times r-m)\omega/q}\right|\overline{\phi_{\alpha(0)}} \gg\ll \overline{\phi_{\alpha(0)}}\left|\widehat{\boldsymbol{\mu}}_{(n'+m)\omega/q}\right|\overline{\phi_{\beta(0)}} \gg}{\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - m\omega}$$

The $n'\omega' = n'\omega/q$ frequency component of the quadratic contribution $\widetilde{\boldsymbol{E}}_2(n'\omega')$ is calculated by a modified version of Eq.(S 25):

$$\widetilde{\boldsymbol{E}}_2(n'\omega') = \ll \overline{\phi_{\alpha(1)}}\left|\widehat{\boldsymbol{\mu}}_{n'\omega/q}\right|\overline{\phi_{\alpha(1)}} \gg = \tag{S 88}$$

$$= \frac{1}{4}\sum_{\substack{(\beta,m)\neq(\alpha,0)\\ (\kappa,l)\neq(\alpha,0)\\ g_1,g_2=\pm1}} \frac{\ll \overline{\phi_{\alpha(0)}}\left|\widehat{\boldsymbol{p}\widehat{\boldsymbol{\mu}}}_{\frac{(l-g_1 r)\omega}{q}}\right|\overline{\phi_{\kappa(0)}} \gg\ll \overline{\phi_{\beta(0)}}\left|\widehat{\boldsymbol{p}\widehat{\boldsymbol{\mu}}}_{\frac{(g_2 r-m)\omega}{q}}\right|\overline{\phi_{\alpha(0)}} \gg\ll \overline{\phi_{\kappa(0)}}\left|\widehat{\boldsymbol{\mu}}_{\frac{(n'+m-l)\omega}{q}}\right|\overline{\phi_{\beta(0)}} \gg}{\left(\epsilon_{\alpha(0)} - \epsilon_{\kappa(0)} - l\omega\right)\left(\epsilon_{\alpha(0)} - \epsilon_{\beta(0)} - m\omega\right)}$$

We note that since $|\overline{\phi_{\alpha,\beta,\kappa(0)}}\rangle\rangle$ are all eigenfunctions of an $\omega = q\omega'$ periodic Floquet system, the numerators of Eq.(S 87) can only be nonzero if

$$\exists z_1 \in \mathbb{Z}: g \times r - m = q \times z_1 \quad \text{(S 89)}$$

$$\exists z_2 \in \mathbb{Z}\, n' + m = q \times z_2 \quad \text{(S 90)}$$

, and the numerators of Eq.(S 88) can only be nonzero if

$$\exists z_1 \in \mathbb{Z}: l - g_1 \times r = q \times z_1 \quad \text{(S 91)}$$

$$\exists z_2 \in \mathbb{Z}: g_2 \times r - m = q \times z_2 \quad \text{(S 92)}$$

$$\exists z_3 \in \mathbb{Z}: n' + m - l = q \times z_3 \quad \text{(S 93)}$$

, regardless of the DS of the unperturbed system. Therefore $\tilde{\mathbf{E}}_{\mathbf{1}}(n'\omega')$ can only be nonzero if

$$\exists z \in \mathbb{Z}: n' \pm r = qz \quad \text{(S94)}$$

That is, $\tilde{\mathbf{E}}_{\mathbf{1}}(n\omega)$ can only be nonzero if

$$\exists z \in \mathbb{Z}: n \pm s = z \quad \text{(S95)}$$

$\tilde{\mathbf{E}}_{\mathbf{2}}(n'\omega')$ can only be nonzero if

$$\exists z \in \mathbb{Z}: n' \pm (1 \pm 1)r = qz \quad \text{(S96)}$$

That is, $\tilde{\mathbf{E}}_{\mathbf{2}}(n\omega)$ can only be nonzero if

$$\exists z \in \mathbb{Z}: n \pm (1 \pm 1)s = z \quad \text{(S97)}$$

One immediate conclusion is that to 1st order, only the $\pm s = \pm r/q$ harmonic sidebands are affected by the perturbation. The selection rules for $\tilde{\boldsymbol{E}}_{\mathbf{1}}\big((n \pm r/q)\omega\big), \tilde{\boldsymbol{E}}_{\mathbf{2}}\big((n \pm (1 \pm 1)\, r/q)\omega\big)$ (where n is an integer) may be derived using Eqs. (S 87)(S 88), similarly to the derivations for integer values of s.

S.V APPLICATION OF MAIN TABLE 1 TO BI-CHROMATIC PERTURBATIONS

In this section, we discuss the applicability of Table 1 in the main text to symmetry breaking induced by multi-chromatic perturbing electric fields with a general polarization. Such a field can always be divided to a coherent sum of linearly polarized, monochromatic components. Thus, the multichromatic field induces a perturbation term $\widehat{\mathrm{W}}$ in the Hamiltonian that can be written as a sum of perturbations described in table 1, i.e. $\lambda_1\widehat{\mathrm{W}}_1 + \lambda_2\widehat{\mathrm{W}}_2 + \cdots$, where λ_{i} is the amplitude of the i'th component. Hence, $\tilde{\mathbf{E}}(\Omega, \lambda)$ is of the form

$$\tilde{\mathbf{E}}(\Omega, \lambda) = \tilde{\mathbf{E}}_0(\Omega) + \lambda_1\tilde{\mathbf{E}}_{1_1}(\Omega) + \lambda_1^2\tilde{\mathbf{E}}_{2_1}(\Omega) + \lambda_2\tilde{\mathbf{E}}_{1_2}(\Omega) + \lambda_2^2\tilde{\mathbf{E}}_{2_2}(\Omega) + \lambda_1\lambda_2\tilde{\mathbf{E}}_{12}(\Omega) + \cdots \quad \text{(S 98)}$$

Where $\tilde{\mathbf{E}}_{1_1}, \tilde{\mathbf{E}}_{1_2}$ are the linear contributions induced by $\widehat{\mathrm{W}}_1, \widehat{\mathrm{W}}_2$, respectively, $\tilde{\mathbf{E}}_{2_1}, \tilde{\mathbf{E}}_{2_2}$ are the quadratic contributions induced by $\widehat{\mathrm{W}}_1, \widehat{\mathrm{W}}_2$, respectively, and $\lambda_1\lambda_2\tilde{\mathbf{E}}_{12}(\Omega)$ is a contribution that is induced by the interference of $\widehat{\mathrm{W}}_1, \widehat{\mathrm{W}}_2$. Note that the selection rules for $\tilde{\mathbf{E}}_{1_1}(\Omega), \tilde{\mathbf{E}}_{2_1}(\Omega), \tilde{\mathbf{E}}_{1_2}(\Omega), \tilde{\mathbf{E}}_{2_2}(\Omega)$ are still given in Table 1 in the main text, since they arise purely from a linearly polarized monochromatic field, whereas the selection rules for the cross contribution $\tilde{\mathbf{E}}_{\mathbf{12}}(\Omega)$ are not covered by Table 1 in the main text. Thus, Table 1 in the main text can be used to completely describe the linear-orders selection rules, and partially the quadratic orders, without additional derivations. With this in mind, we consider a numerical example of a bi-chromatic perturbative field. Consider the $\omega - 2\omega$ cross-linear driving field, $\boldsymbol{E}(t) = \sin(2\omega t)\,\hat{\boldsymbol{x}} + \cos(\omega t)\,\hat{\boldsymbol{y}}$. This field exhibits $\hat{\mathrm{Z}}_{\mathrm{x}} = \hat{\tau}_2 \cdot \hat{\sigma}_{\mathrm{x}}$ symmetry and therefore only $\hat{y}$ ($\hat{x}$) polarized $2\mathrm{m} \pm 1$ ($2m$) harmonics are allowed. The ellipticity of the cross linear drivers can be modified from zero to some finite value λ, by adding an additional bi-chromatic cross linear field, that breaks the $\hat{Z}_x$ symmetry and its associated polarization restrictions. The perturbed field, known as a bi-elliptical field, is given by $\boldsymbol{E}(t) = (\lambda \sin(\omega t) + \sin(2\omega t))\hat{\boldsymbol{x}} + (\cos(\omega t) + \lambda \cos(2\omega t))\hat{\boldsymbol{y}}$ **(**Lissajous curves in Figure S1). Since the perturbing field is not linearly polarized or monochromatic, we split the perturbation into two separate terms - $\widehat{W}_1 = \lambda x \sin(\omega t)$ and $\widehat{W}_2 = \lambda y \cos(2\omega t)$. By plugging ($\widehat{W}_1$: $p_x = 1$, $p_y = 0, s = 1$) and ($\widehat{W}_2$: $p_x = 0$, $p_y = 1, s = 2$) into the $\hat{Z}$ prediction of table 1 in the main text (and keeping in mind that the symmetry of the unperturbed system is $\hat{\mathbf{Z}}_{\mathbf{x}}$ and not $\hat{Z}_y$), we find that

$$\tilde{\mathbf{E}}_{2_1}(2m\omega) \cdot \hat{\mathbf{y}} = \tilde{\mathbf{E}}_{1_1}(2m\omega) \cdot \hat{\mathbf{x}} = 0 \quad \text{(S 99)}$$

$$\tilde{\mathbf{E}}_{2_1}\big((2m+1)\omega\big) \cdot \hat{\mathbf{x}} = \tilde{\mathbf{E}}_{1_1}\big((2m+1)\omega\big) \cdot \hat{\mathbf{y}} = 0 \quad \text{(S 100)}$$

Hence, for spectral regions where $\tilde{\mathbf{E}}_{\mathbf{12}}(\Omega)$ may be neglected, $\hat{x}$ ($\hat{y}$) polarized odd (even) harmonics are predicted to scale linearly in λ, whereas $\hat{x}$ ($\hat{y}$) polarized even (odd) harmonics are predicted to scale quadratically in λ. All these selection rules are numerically observed in the TDSE calculation, in the spectral region between harmonics 28 to 43 with an average $R^2>0.95$.

S.VI. VALIDITY OF THE THEORY FOR INTEGRATED HARMONIC INTENSITIES

When an ultrashort driver is used for HHG, the spectrum may display spectral shifts such that the peaks are not centered around an integer harmonic $n\omega$. In those scenarios, it is experimentally useful to integrate the intensity of the emitted radiation around each integer harmonic, and to use the result as a measure for the harmonic intensity.

In this section, we numerically demonstrate that our analytical results also hold when the spectral width around each harmonic is considered. Instead of considering the scaling of $\left|\widetilde{\boldsymbol{E}}(\lambda,\Omega) - \widetilde{\boldsymbol{E}}(0,\Omega)\right|$ with λ, we consider the scaling of the integrated harmonics amplitudes (i.e., square root of the integrated harmonic intensities) with λ. Explicitly, we show that for the numerical examples we studied in the main text, the following quantity consistently follows the predictions of Table 1 in the main text

$$\sqrt{\int_{\Delta=-\omega/2}^{\Delta=+\omega/2} d\Delta |E(\lambda,\Omega+\Delta)|^2} - \sqrt{\int_{\Delta=-\omega/2}^{\Delta=+\omega/2} d\Delta |E(0,\Omega+\Delta)|^2} \quad \textbf{(S 101)}$$

$\widehat{\boldsymbol{C}}_{\mathbf{5,3}}$ Symmetry broken by a $5\boldsymbol{\omega}$ linearly polarized field

In Figure 1 of the main text, we numerically demonstrated that when a $\hat{C}_{5,3}$ DS is broken by a 5ω linearly polarized laser, $\left|\widetilde{\boldsymbol{E}}(\lambda,5m\omega) - \widetilde{\boldsymbol{E}}(0,5m\omega)\right| \propto \lambda$ and $\left|\widetilde{\boldsymbol{E}}(\lambda,(5m\pm 2)\omega) - \widetilde{\boldsymbol{E}}(0,(5m\pm 2)\omega)\right| \propto \lambda^2$, in agreement with the analytical prediction $\widetilde{\boldsymbol{E}}_{\mathbf{1}}\big((5m\pm 2)\omega\big) = \widetilde{\boldsymbol{E}}_{\mathbf{2}}(5m\omega) = 0$. In Figure S2, we show that the same predictions hold for the integrated harmonic amplitudes (Eq.(S 101)). For quadratically scaling harmonics, some of the harmonics display small deviations from the analytical prediction due to the inclusion of the linearly scaling background radiation in the computation.

$\hat{\boldsymbol{Z}}_{\boldsymbol{y}}$ DS broken by a $5\boldsymbol{\omega}$ linearly polarized field

In Figure 2 of the main text, we numerically demonstrated that when a $\hat{Z}_y$ DS is broken by a 5ω linearly polarized laser (polarized along $\hat{y}$), 2m harmonics scale linearly (quadratically) with λ if polarized along $\hat{x}$ ($\hat{y}$), and $2\mathrm{m} \pm 1$ harmonics scale linearly (quadtatically) with λ if polarized along $\hat{y}$ ($\hat{x}$). This is consistent with the analytical prediction $\widetilde{\mathrm{E}}_{2\mathrm{x}}(2\mathrm{m}\omega) = \widetilde{\mathrm{E}}_{1\mathrm{y}}(2\mathrm{m}\omega) = \widetilde{\mathrm{E}}_{2\mathrm{y}}\big((2\mathrm{m} \pm 1)\omega\big) = \widetilde{\mathrm{E}}_{1\mathrm{x}}\big((2\mathrm{m} \pm 1)\omega\big) = 0$. In Figure S3, we show the same predictions hold for the integrated harmonic amplitudes (Eq.(S 101)).

$\hat{\boldsymbol{Z}}_{\boldsymbol{x}}$ DS broken by a bi-chromatic perturbation

In Figure S1, we numerically demonstrated that when an $\omega - 2\omega$ cross linear driver with a DS $\hat{Z}_x$ becomes a cross-elliptical driver with ellipticity λ, there exists a spectral region where $2\mathrm{m} \pm 1$ harmonics scale linearly (quadratically) with λ if polarized along $\hat{x}$ ($\hat{y}$), and $2\mathrm{m}$ harmonics scale linearly (quadtatically) with λ if polarized along $\hat{y}$ ($\hat{x}$). In Figure S4, we show the same predictions hold for the integrated harmonic amplitudes (Eq.(S 101)).

$$\boldsymbol{E}(t) = (\sin(2\omega t) + \lambda \sin(\omega t))\hat{\boldsymbol{x}} + \big(\cos(\omega t) + \lambda cos(2\omega t)\big)\hat{\boldsymbol{y}}$$

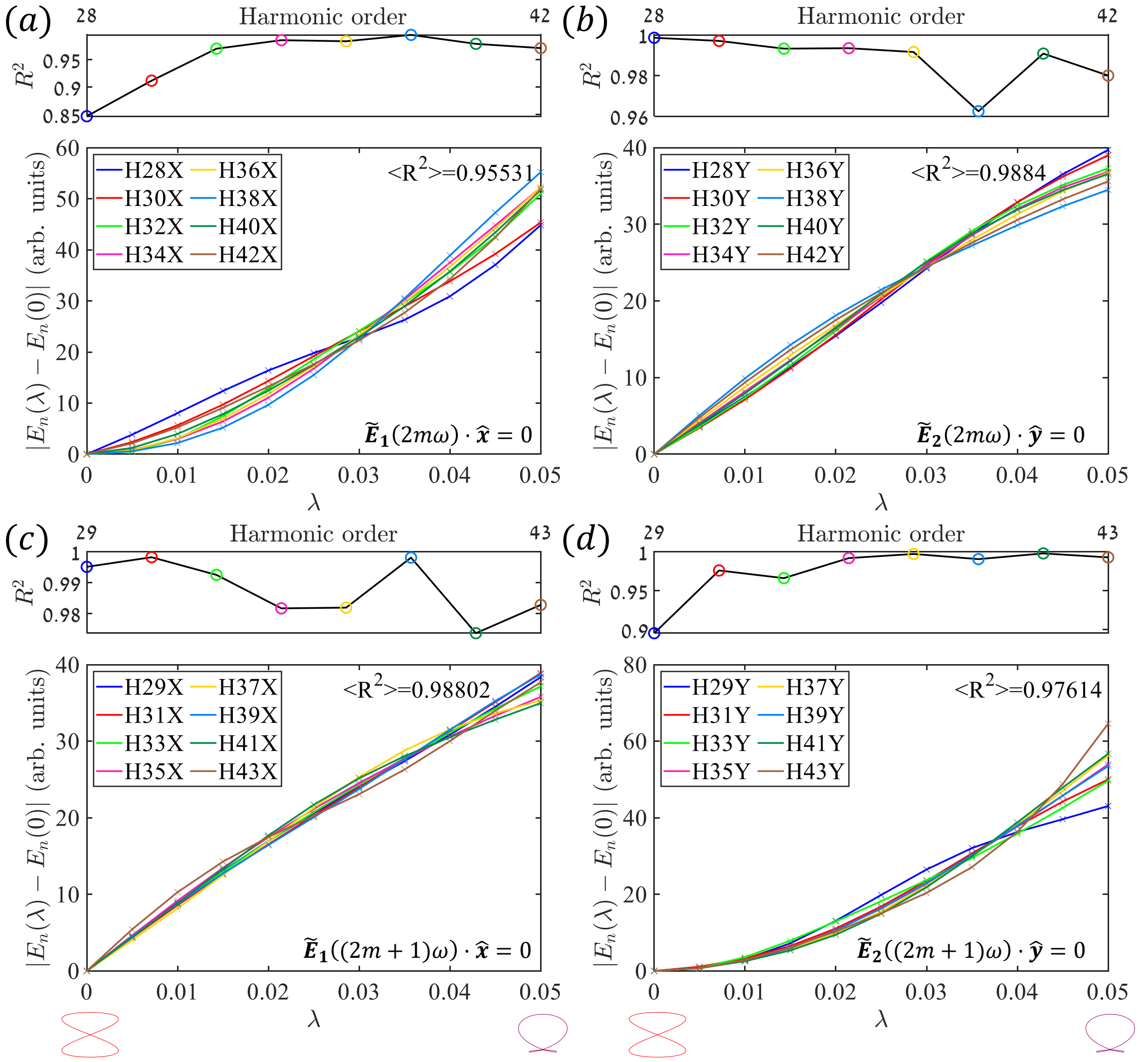

Fig. S1.

Numerical demonstration of selection rules for breaking $\hat{Z}_x = \hat{\tau}_2 \cdot \hat{\sigma}_x$ selection rules in HHG by multi-chromatic, cross linear perturbative field. For $\lambda = 0$, $\hat{y}$ polarized 2m harmonics and $\hat{x}$ polarized 2m+1 harmonics are forbidden. For $\lambda > 0$, the ellipticity breaks the DS and induces (a,d) quadratic scaling of $\hat{x}(\hat{y})$ polarized even (odd) harmonics (b,c) linear scaling of $\hat{y}(\hat{x})$ polarized even (odd) harmonics.The scaling of the harmonic amplitudes with perturbation strengths are presented in color, , and the individual R^2 values are marked with the corresponding color above each subfigure. The harmonic amplitudes are multiplied by a factor to appear on the same graph.

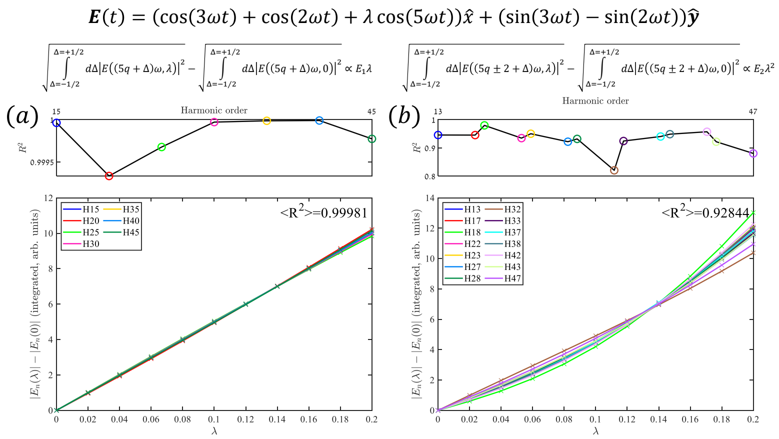

Fig. S2.

Numerical demonstration of selection rules for breaking $\hat{\boldsymbol{C}}_{5,3} = \hat{\boldsymbol{\tau}}_5 \cdot \hat{\boldsymbol{R}}_{5,3}$ selection rules in HHG by an $\hat{\boldsymbol{x}}$-polarized perturbative electric field (the same driving field and perturbation as Figure 1 in the main text). The scaling of integrated harmonic amplitudes (Eq.(S 101)) is linear for $5m$ harmonics and quadratic for $5m \pm 2$ harmonics, in accordance with the analytical predictions of Table 1 in the main text. The scaling of the harmonic amplitudes with perturbation strengths are presented in color, and the individual $\boldsymbol{R}^2$ values are marked with the corresponding color above each subfigure. Each harmonic amplitude is multiplied by a factor such that all the harmonics fit to the graph.

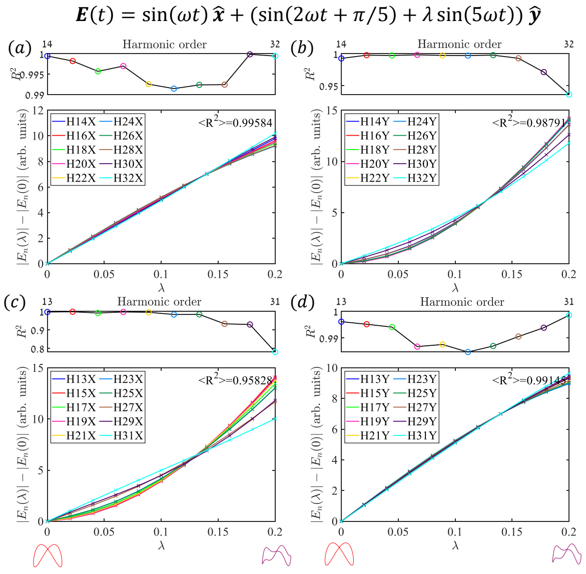

Fig. S3.

Numerical demonstration of selection rules for breaking $\hat{Z}_y = \hat{\tau}_2 \cdot \hat{\sigma}_y$ selection rules in HHG by an $\hat{y}$-polarized perturbative electric field (the same driving field and perturbation as Figure 2 in the main text). The scaling of integrated harmonic amplitudes (Eq.(S 101)) is linear for odd (even) harmonics polarized along the $\hat{y}(\hat{x})$ axis, and quadratic for even (odd) harmonics polarized along the $\hat{y}(\hat{x})$ axis. The scaling of the harmonic amplitudes with perturbation strengths are presented in color, and the individual R^2 values are marked with the corresponding color above each subfigure. Each harmonic amplitude is multiplied by a factor such that all the harmonics fit to the graph.

$$\boldsymbol{E}(t) = (\sin(2\omega t) + \lambda \sin(\omega t))\hat{\boldsymbol{x}} + (\cos(\omega t) + \lambda cos(2\omega t))\hat{\boldsymbol{y}}$$

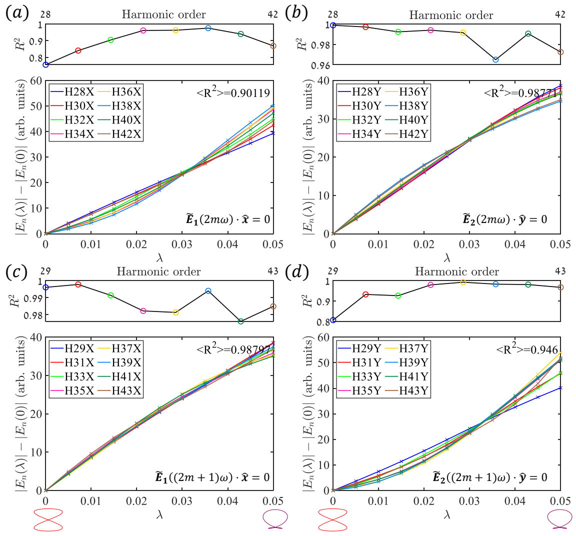

Fig. S4.

Numerical demonstration of selection rules for breaking $\widehat{\boldsymbol{Z}}_{\boldsymbol{x}} = \hat{\boldsymbol{\tau}}_{\boldsymbol{2}} \cdot \widehat{\boldsymbol{\sigma}}_{\boldsymbol{x}}$ selection rules in HHG by a bi-chromatic perturbation (the same driving field and perturbation as Figure 3 in the main text). The scaling of integrated harmonic amplitudes (Eq.(S 101)) is linear for even (odd) harmonics polarized along the $\widehat{\boldsymbol{y}}(\widehat{\boldsymbol{x}})$ axis, and quadratic for odd (even) harmonics polarized along the $\widehat{\boldsymbol{y}}(\widehat{\boldsymbol{x}})$ axis. The scaling of the harmonic amplitudes with perturbation strengths are presented in color, and the individual $\boldsymbol{R}^{\boldsymbol{2}}$ values are marked with the corresponding color above each subfigure. Each harmonic amplitude is multiplied by a factor such that all the harmonics fit to the graph.