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Abstract

Population harmonics distinguish ideal detuning and drive-amplitude modulation, but a small admixture of the two controls can change that comparison. We study single-tone, in-phase modulation of a dissipative two-level system with fixed dissipator and population readout. The standard bilinear response chain gives an extra inverse-frequency power for longitudinal even harmonics at their lowest modulation order. Near the longitudinal axis, the resonant second/fourth-harmonic ratio crosses from one to two through a scaling function of $\chi = 2\Gamma_2\nu\sin^2\theta/\Omega^2$, in a joint small-angle and high-frequency limit of the weak-modulation coefficients. The corresponding control-admixture tolerance decreases as $\nu^{-1/2}$ at fixed rates. A detuned stationary-state cancellation instead reproduces the longitudinal exponent pair with a ratio approaching 305/113. Thus the exponents alone do not identify a structural modulation axis. We derive an a posteriori full-periodic residual bound for positive longitudinal relaxation, separately from the asymptotic chain remainder. Independently represented periodic-state calculations test the crossover, counterexample and finite-frequency corrections. These results quantify the conditions under which population harmonics can compare modulation directions in this model.

Keywords: open quantum systems; population harmonics; bilinear response; modulation geometry; high-frequency expansion.

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1 Physical question and prior results

Population harmonics of a driven two-level system are an established spectroscopic signal. Phase-modulated atomic-candle experiments studied the second-harmonic Rabi resonance [1], and Tretiakov and LeBlanc calculated and observed higher-harmonic population response; their weak-modulation simulations give a quartic dependence of the fourth-harmonic resonance peak on phase depth [2]. Amplitude-modulated spin resonance likewise produces steady-state magnetization harmonics [3]. A related transverse-microwave/longitudinal-radiofrequency model has analytical responses at all modulation harmonics, evaluated through phase-sensitive electron paramagnetic resonance (EPR) absorption [4]. Neither the existence of higher harmonics nor their dependence on drive geometry is introduced here.

The longitudinal Bloch equations below map to those of Ref. [2], up to coherence sign and time origin, with modulation frequency $\omega_m = \nu$ and phase depth $m = 2\lambda/\nu$. A frequency scan at fixed m therefore differs from the fixed- λ scan studied here. The longitudinal second-harmonic high-frequency coefficient also follows from the published approximate small-signal line shape [5]. We calculate retained modulation-order coefficients and their finite-frequency corrections, rather than claim a separate discovery of that signal.

Shifting an affine stationary state gives a bilinear input-output system, whose single-tone chain is a specialization of established Volterra/generalized frequency-response theory [6, 7]. Observable-projected dissipative response [8] and cancellation of a nominal micromotion order [9] are likewise prior context. The distinction between modulation order and the first surviving inverse-frequency power is therefore a tool of the analysis, rather than a new general framework.

Can high-frequency population harmonics identify an intended detuning modulation when a small, in-phase drive-amplitude component is also present? We address this through a matched comparison, holding the unmodulated generator, dissipator and measured population fixed while changing the direction of the single-tone perturbation. The quantitative question is how the interpretation changes across the resulting crossover and how its error can be checked against the full periodic equation. Structural cancellation means that a projected operator vanishes for every input state. State-dependent cancellation means only its contraction with the chosen stationary state vanishes. An exact symmetry zero is a third, distinct mechanism. Keeping these mechanisms separate is necessary to interpret the exponents and the validity tests.

2 Model, readout and order of limits

Set $\hbar = 1$ and write

$$H_\theta(t) = \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}\sigma_x + \lambda \sin(\nu t)(\cos \theta \sigma_z + \sin \theta \sigma_x). \quad (1)$$

The endpoints $\theta = 0$ and $\pi/2$ are longitudinal and transverse modulation. Both perturbations have the same frequency and phase. The angle θ specifies an admixture in the Hamiltonian control plane spanned by detuning and drive amplitude; it need not equal a spatial laboratory-field angle. Independent relative phases, modulated dissipative rates and time-dependent optical pumping are outside this model. The jump operators are $\sqrt{\gamma_1}\sigma_-$ and $\sqrt{\gamma_\phi/2}\sigma_z$ [10, 11, 12]. We take $\Omega > 0$, $\nu > 0$, $\gamma_1, \gamma_\phi \geq 0$, excluding $\gamma_1 = \gamma_\phi = 0$. Results with nonzero harmonic ratios require $\gamma_1 > 0$ and nonzero denominators. The undamped system need not possess a unique attracting periodic state, so its limits are not covered by the stationary formulas.

With $\rho = (I + u\sigma_x + v\sigma_y + w\sigma_z)/2$, the Bloch vector obeys

$$\dot{\mathbf{r}} = [M_0 + \lambda \sin(\nu t) M_\theta] \mathbf{r} + \mathbf{c}, \quad M_\theta = \cos \theta M_z + \sin \theta M_x, \quad (2)$$

where $\Gamma_2 = \gamma_1/2 + \gamma_\phi$ and

$$\begin{aligned} M_0 &= \begin{pmatrix} -\Gamma_2 & -\Delta & 0 \\ \Delta & -\Gamma_2 & -\Omega \\ 0 & \Omega & -\gamma_1 \end{pmatrix}, & \mathbf{c} &= \begin{pmatrix} 0 \\ 0 \\ -\gamma_1 \end{pmatrix}, \\ M_z &= \begin{pmatrix} 0 & -2 & 0 \\ 2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, & M_x &= \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -2 \\ 0 & 2 & 0 \end{pmatrix}. \end{aligned} \quad (3)$$

All rates and frequencies are angular frequencies. Numerical dimensionless values use $\Omega = 1$; physical scans specify values divided by 2π consistently.

The unmodulated state is $M_0 \mathbf{r}_0 + \mathbf{c} = 0$. Defining $D = \Gamma_2 \Omega^2 + \gamma_1(\Delta^2 + \Gamma_2^2)$ gives

$$v_0 = \frac{\gamma_1 \Gamma_2 \Omega}{D}, \quad u_0 = -\frac{\Delta}{\Gamma_2} v_0, \quad w_0 = -\frac{\gamma_1(\Delta^2 + \Gamma_2^2)}{D}. \quad (4)$$

For $\Delta = \gamma_\phi = 0$, $\gamma_1 = \gamma$, one has $v_0 = 2\Omega\gamma/(\gamma^2 + 2\Omega^2)$ and $w_0 = -\gamma^2/(\gamma^2 + 2\Omega^2)$.

Expand the real periodic vector as $\mathbf{r}(t) = \sum_n \mathbf{r}_n \exp(in\nu t)$, $\mathbf{r}_{-n} = \mathbf{r}_n^*$. The population $P_e = (1 + w)/2$ has Fourier coefficient $p_n = w_n/2$ for $n \neq 0$. Its peak sinusoidal amplitude is

$$A_{n\nu} = 2|p_n| = |w_n|, \quad w_n = \mathbf{q}^T \mathbf{r}_n, \quad \mathbf{q} = (0, 0, 1)^T. \quad (5)$$

Thus the factor $1/2$ in the population and the factor two from a conjugate Fourier pair cancel. This readout differs from the coherence quadrature of Ref. [4] and from two-time resonance-fluorescence correlations [13].

For each fixed harmonic h , $w_h^{[h]}$ denotes the term proportional to λ^h in the Taylor expansion about $\lambda = 0$. Its large- ν expansion is taken with $M_0, \mathbf{r}_0, \theta$ fixed. These are sequential weak-modulation and high-frequency statements, except for the explicitly specified joint angle-frequency limit below. A full-response ratio means first taking its nonzero leading weak-modulation coefficients and then their high-frequency limit. Fixed phase depth ($\lambda \propto \nu$), simultaneous vanishing damping, and limits near a zero seed require separate analysis. The full periodic computations below test specified finite parameter sets; they do not prove uniformity of these limits.

3 Projected coefficients and matched ratios

The Fourier hierarchy is

$$(in\nu I - M_0) \mathbf{r}_n = \frac{\lambda}{2i} M_\theta (\mathbf{r}_{n-1} - \mathbf{r}_{n+1}) + \mathbf{c} \delta_{n0}. \quad (6)$$

Let $R_j = (ij\nu I - M_0)^{-1}$. At minimal order, only the monotone positive-frequency chain reaches harmonic h :

$$w_h^{[h]} = \left(\frac{\lambda}{2i} \right)^h \mathbf{q}^T R_h M_\theta R_{h-1} M_\theta \cdots R_1 M_\theta \mathbf{r}_0. \quad (7)$$

The next modulation orders are $h+2, h+4, \dots$. Expanding the resolvents for $\|M_0\|_2/\nu < 1$ yields, at $h=2$,

$$w_2^{[2]} = \frac{\lambda^2}{8\nu^2} \mathbf{q}^T M_\theta^2 \mathbf{r}_0 - \frac{i\lambda^2}{16\nu^3} \mathbf{q}^T (M_0 M_\theta^2 + 2M_\theta M_0 M_\theta) \mathbf{r}_0 + \mathcal{O}(\lambda^2 \nu^{-4}). \quad (8)$$

For longitudinal population readout $\mathbf{q}^T M_z = 0$, $M_z^2 = \text{diag}(-4, -4, 0)$ and $\mathbf{q}^T M_0 M_z^2 \mathbf{r}_0 = -4\Omega v_0$. The static drift rotates the modulation-induced transverse change back into population; it introduces the additional resolvent power. Transverse modulation acts directly on population, with $\mathbf{q}^T M_x^2 \mathbf{r}_0 = -4w_0$. The second- and fourth-harmonic leading terms are

$$\begin{aligned} w_2^{(z),[2]} &= \frac{i\lambda^2 \Omega v_0}{4\nu^3} + \mathcal{O}(\lambda^2 \nu^{-4}), & w_2^{(x),[2]} &= -\frac{\lambda^2 w_0}{2\nu^2} + \mathcal{O}(\lambda^2 \nu^{-3}), \\ w_4^{(z),[4]} &= -\frac{i\lambda^4 \Omega v_0}{96\nu^5} + \mathcal{O}(\lambda^4 \nu^{-6}), & w_4^{(x),[4]} &= \frac{\lambda^4 w_0}{24\nu^4} + \mathcal{O}(\lambda^4 \nu^{-5}). \end{aligned} \quad (9)$$

The z cancellations are operator identities, whereas the nonzero prefactors still require the stated stationary seeds. At $\gamma = \Omega$, $A_{2\nu}^{(x)}/A_{2\nu}^{(z)} \simeq \nu/\Omega$.

For any fixed even $h = 2m \geq 2$, the same chain gives

$$\begin{aligned} w_h^{(z),[h]} &= -i(-1)^m \frac{\lambda^h \Omega v_0}{h h! \nu^{h+1}} + \mathcal{O}(\lambda^h \nu^{-h-2}), \\ w_h^{(x),[h]} &= (-1)^m \frac{\lambda^h w_0}{h! \nu^h} + \mathcal{O}(\lambda^h \nu^{-h-1}). \end{aligned} \quad (10)$$

This is a fixed-minimal-order family for this Bloch model, not a sum over all modulation orders or a classification of general Lindblad dynamics. The derivation and certified chain remainder are given in Online Resource 1 (the technical companion). A ratio removes the common seed only where both relevant harmonics are nonzero. In particular,

$$R_{42} = \frac{24\nu^2 A_{4\nu}}{\lambda^2 A_{2\nu}}, \quad \lim_{\nu \rightarrow \infty} \lim_{\lambda \rightarrow 0^+} R_{42} = \begin{cases} 1, & \theta = 0, \\ 2, & \theta = \pi/2. \end{cases} \quad (11)$$

For general even h , $\nu^2 A_{(h+2)\nu}/(\lambda^2 A_{h\nu})$ approaches $h/[(h+1)(h+2)^2]$ longitudinally and $1/[(h+1)(h+2)]$ transversely, with the same order-of-limits qualification.

4 Axis tilt, exact zeros and validity

For the unit tilted axis in Eq. (1),

$$s_\theta = \mathbf{q}^T M_\theta^2 \mathbf{r}_0 = 4 \sin \theta \cos \theta u_0 - 4 \sin^2 \theta w_0, \quad M_\theta^4 = -4M_\theta^2. \quad (12)$$

If $s_\theta \neq 0$, the leading powers are therefore $\lambda^2 \nu^{-2}$ and $\lambda^4 \nu^{-4}$, and the minimal-order ratio in Eq. (11) tends to two. At resonance $u_0 = 0$, every nonzero small tilt with $\gamma_1 > 0$ gives a nonzero seed. The exact-axis suppression is robust to detuning and damping changes within the longitudinal model but is fragile to a transverse modulation component. The $\theta \rightarrow 0$ and $\nu \rightarrow \infty$ limits of the minimal-order ratio need not commute. At nonzero detuning an isolated state-dependent zero can also occur; it should not be called a structural axis symmetry.

4.1 A quantitative resonant crossover

For $\Delta = 0$, fixed $\Omega, \gamma_1 > 0$ and $\gamma_\phi \geq 0$, define

$$\chi = \frac{2\nu|w_0|\sin^2\theta}{\Omega v_0} = \frac{2\Gamma_2\nu\sin^2\theta}{\Omega^2}. \quad (13)$$

Take $\nu \rightarrow \infty, \theta \rightarrow 0$ with χ fixed, after extracting the lowest weak-modulation coefficients. Uniformly on bounded χ intervals, their scaled limits are

$$\frac{4\nu^3 w_2^{[2]}}{\lambda^2 \Omega v_0} \rightarrow \chi + i, \quad \frac{96\nu^5 w_4^{[4]}}{\lambda^4 \Omega v_0} \rightarrow -(2\chi + i). \quad (14)$$

The amplitude ratio therefore follows the crossover law

$$F(\chi) = \sqrt{\frac{1 + 4\chi^2}{1 + \chi^2}}, \quad R_{42}^{\text{minimal}} \rightarrow F(\chi). \quad (15)$$

This joint limit resolves the noncommuting ideal-axis and fixed-tilt limits; it is not an all-modulation-order theorem. With the sine phase in Eq. (1) as reference, the complex ratio is

$$\mathcal{R}_{42} = \frac{24\nu^2 w_4^{[4]}}{\lambda^2 w_2^{[2]}} \rightarrow -\frac{2\chi + i}{\chi + i}. \quad (16)$$

It traces the upper semicircle $(\text{Re } \mathcal{R}_{42} + 3/2)^2 + (\text{Im } \mathcal{R}_{42})^2 = 1/4$. Changing the common drive phase by φ multiplies the unreferenced complex ratio by $\exp(2i\varphi)$; this circle requires a phase reference. The magnitude is independent of that common phase.

For a prescribed leading-ratio deviation $0 < \epsilon < 1$, $F(\chi) - 1 \leq \epsilon$ gives

$$\chi \leq \chi_\epsilon = \sqrt{\frac{(1 + \epsilon)^2 - 1}{4 - (1 + \epsilon)^2}}, \quad \sin^2\theta \leq \frac{\Omega^2 \chi_\epsilon}{2\Gamma_2\nu}. \quad (17)$$

Thus increasing frequency narrows the admissible in-phase control admixture on the scale $|\theta| \propto \nu^{-1/2}$ at fixed rates. This is an asymptotic interpretation tolerance, not a certified finite-frequency or detector-noise guarantee. The companion supplies a uniform retained-order remainder, and full-response calculations test finite points separately.

4.2 State-dependent cancellation and exact zeros

Take $\Delta = \Omega = \gamma_1 = 1$, $\gamma_\phi = 0$ and $\tan\theta = 4/5$. The unmodulated stationary state is $\mathbf{r}_0 = (-4/7, 2/7, -5/7)^T$. It gives $s_\theta = 0$ although $\mathbf{q}^T M_\theta \neq 0$. The first surviving weighted drift sums yield

$$w_2^{[2]} = i\frac{113\lambda^2}{574\nu^3} + \mathcal{O}(\lambda^2\nu^{-4}), \quad w_4^{[4]} = -i\frac{305\lambda^4}{13776\nu^5} + \mathcal{O}(\lambda^4\nu^{-6}). \quad (18)$$

Thus $\lim_{\nu \rightarrow \infty} \lim_{\lambda \rightarrow 0^+} R_{42} = 305/113 \simeq 2.699115$, despite the same exponent pair as the longitudinal model. The exponent pair or leading seed zero alone does not identify a structural modulation axis. This exact counterexample was checked by independent rational matrix calculations and a separately frozen three-point full-solver campaign.

At longitudinal resonance, the half-period transformation $S = \text{diag}(-1, 1, 1)$ satisfies $SM_0S = M_0$, $SM_zS = -M_z$ and $S\mathbf{c} = \mathbf{c}$. Uniqueness of the attracting state implies

$\mathbf{r}(t) = S\mathbf{r}(t + T/2)$. Consequently every odd population harmonic is exactly zero, at any modulation strength. This symmetry result is distinct from the inverse-frequency cancellation in even harmonics. For longitudinal modulation off resonance, $p_n(-\Delta) = (-1)^n p_n(\Delta)$ follows by the corresponding transformed equations. The technical companion gives the proofs and separates exact zeros from numerical-floor values.

For pure dephasing with $\gamma_1 = 0$, $\gamma_\phi > 0$, the source vanishes and $\mathbf{r} = 0$ is an exact solution. The Lyapunov derivative is $d\|\mathbf{r}\|^2/dt = -2\gamma_\phi(u^2 + v^2)$. In the stated model a nonzero w cannot remain in the zero-derivative set because the static transverse drive is nonzero. The attracting periodic population is therefore constant and all harmonics vanish. No ratio is defined in this zero-signal case.

Validity has three separate requirements: convergence/truncation of the fixed-order resolvent expansion, negligibility of higher modulation orders, and accuracy of the full periodic numerical solution. A norm bound for the first does not prove the other two. The earlier amplitude-change diagnostic $\eta_h = |A_h^{[2]}/A_h^{(0)} - 1|$, where $A_h^{[2]}$ retains two additional inverse-frequency orders, is retained as a heuristic. It is not a bound on the omitted tail. Nor is it well-conditioned at a zero seed. Its correlations with actual amplitude errors describe the fixed tested tuples, without statistical generalization or a universal threshold.

For example, with the induced Euclidean norm, take $\mu \geq \|M_0\|_2$, $t = \mu/\nu < 1$, $b = \|M_1\|_2$ and $G_h(t) = \prod_{j=1}^h (1 - t/j)^{-1}$. When $\mathbf{q}^T M_1 = 0$, let $W_h^{[1]}$ retain the first potentially nonzero inverse-frequency order of the chain. An absolute bound is

$$|w_h^{[h]} - W_h^{[1]}| \leq K_h[G_h(t) - 1], \quad K_h = \frac{|\lambda|^h \|\mathbf{q}^T M_0\|_2 \|\mathbf{r}_0\|_2 b^h}{2^h h! h \nu^{h+1}}. \quad (19)$$

The resolvent identity $\mathbf{q}^T R_h M_1 = (i h \nu)^{-1} \mathbf{q}^T M_0 R_h M_1$ exposes the extra drift exactly; submultiplicativity and the nonnegative coefficients of G_h bound its remaining tail. The companion proves the corrected-chain bound as well. These bounds exclude omitted powers of λ . A relative certificate additionally requires a nonzero denominator, and the conservative norm estimate becomes loose near a small stationary seed.

4.3 A full-periodic residual certificate

The fixed-order bound above does not control the complete response. For $\gamma_1 > 0$, set $\kappa = \min(\Gamma_2, \gamma_1) > 0$. Let $\tilde{\mathbf{r}}(t)$ be any differentiable T -periodic approximation, with residual

$$\mathbf{r}_d = \dot{\tilde{\mathbf{r}}} - [M_0 + \lambda \sin(\nu t) M_\theta] \tilde{\mathbf{r}} - \mathbf{c}, \quad \|f\|_{T,2}^2 = \frac{1}{T} \int_0^T \|f(t)\|_2^2 dt. \quad (20)$$

The symmetric part of the Bloch generator is bounded above by $-\kappa I$. The periodic energy identity and Cauchy–Schwarz inequality give

$$\|\tilde{\mathbf{r}} - \mathbf{r}_*\|_{T,2} \leq E, \quad E = \frac{\|\mathbf{r}_d\|_{T,2}}{\kappa}, \quad |\tilde{p}_h - p_h| \leq \frac{E}{2}, \quad |\tilde{A}_{h\nu} - A_{h\nu}| \leq E. \quad (21)$$

For a truncated Fourier polynomial, the residual includes the interior algebraic residuals and the two exterior sectors $n = \pm(N + 1)$. Parseval’s identity therefore evaluates the full residual without assuming that the unknown harmonic tail vanishes. This certificate applies to the full finite- λ periodic solution, at any frequency, under the stated positive-relaxation condition. It does not apply at $\kappa = 0$. Outward interval evaluation must include coefficient serialization and intended parameter values; agreement between solvers alone is a separate consistency check. A ratio bound additionally requires that its denominator enclosure exclude zero; the companion gives explicit quotient-error propagation.

5 Computational tests and results

Two independently transcribed methods solve the full periodic state. The time method propagates the affine monodromy map over one period and solves its fixed point, then integrates the complex Fourier coefficient directly. Harmonic balance (HB) solves the two-sided truncated Fourier system. Arbitrary-precision refinement, conjugacy, residuals, periodic closure, positivity and operational coefficient floors are checked separately. The methods share physical tuples and normalization, rather than extraction or equation-transcription code. A finite retained-order resolvent calculation supplies a third reference, which is not itself a full periodic solver.

OpenAI Codex assisted with literature comparison, editorial preparation, numerical implementation, analytic proposals and checks, and audit coordination. Validation uses separately transcribed time-domain and harmonic-balance equations, exact projected identities, retained-order references and precision/truncation refinement; AI agreement is not scientific evidence. Parameters, endpoints, acceptance gates and the diagnostic outputs to be retained were specified before each physical campaign. Historical model/version identifiers were not retained in the supplied records. The author is the sole scientific decision-maker and retains responsibility for the final selection, interpretation and verification of the original sources. The detailed methods and implementation records accompany the numerical results in Online Resources 1–3.

The deposited benchmark provides additional checks of the coefficients. Its 74 primary fourth-harmonic points receive both full periodic methods and the exact order- λ^4 chain; the full methods agree at relative complex discrepancy below 10^{-20} . Finite-domain frequency slopes are -5.000189 longitudinally and -4.006594 transversely. Their descriptive residual intervals are not measurement uncertainties. Leading amplitude errors are 0.0263% and 0.921% , while transverse complex phase corrections reach 18.6% ; amplitude agreement must not be substituted for complex-response agreement. The technical companion retains the complete grids, fits, old figures, coefficient floors and failure records.

The deposited parameter-diverse study uses 683 fixed tuples from 695 supplied rows, with independent 50/70-digit harmonic-balance checks and 16 time-domain anchors. Its historical random seed and sampler were not documented. Accordingly, distributions and correlations describe these supplied inputs, without population generalization. The earlier clean full reconstruction remains evidence for that dataset; it is not claimed as a new 683-point campaign here.

The initial extension campaign tests the fixed-order family and axis sensitivity separately from those deposited data. Its 40 points were fixed before physical calculation: 12 primary tuples (two stationary seeds, two endpoint geometries, $\nu/\Omega = 40, 80, 160$, $\lambda/\Omega = 0.1$); six low-frequency crossover tuples; six fixed- $\lambda/\nu = 0.2$ diagnostics; and 16 tilt tuples ($\theta = 0, 0.05, 0.2, \pi/2$ and $\nu/\Omega = 20, 80, 320, 1280$, $\lambda/\Omega = 0.2$). With $\Omega = \gamma_1 = 1$, the primary seeds use $(\Delta, \gamma_\phi) = (0, 0)$ and $(1, 0.5)$. Every point receives both full methods at two declared precision/truncation settings. The primary points additionally test $h = 6, 8$, giving 104 retained harmonic rows in total; none failed the numerical gates or was excluded.

The maximum cross-method relative complex discrepancy is 4.40×10^{-48} , and every coefficient exceeds its operational floor by at least 7.63×10^{26} . These values reflect the arbitrary-precision mathematical benchmark, rather than laboratory detectability. All 16 primary harmonic endpoint comparisons and all four ratio endpoint gates pass the predeclared criteria at $\nu/\Omega = 160$. The maximum retained-order versus leading complex error at these endpoints is below 4.58% , while its amplitude error is below 0.059% ; finite-frequency phase effects remain visible. Across the complete primary grid, the maximum

full-response versus exact minimal-order complex discrepancy is below 1.05×10^{-5} ; this is the finite-grid check of omitted modulation orders, separate from the leading-term error. At the resonant endpoints $R_{42} = 1.00000531$ longitudinally and 2.00035999 transversely; for the detuned/dephased seed they are 1.00008102 and 2.00010407 .

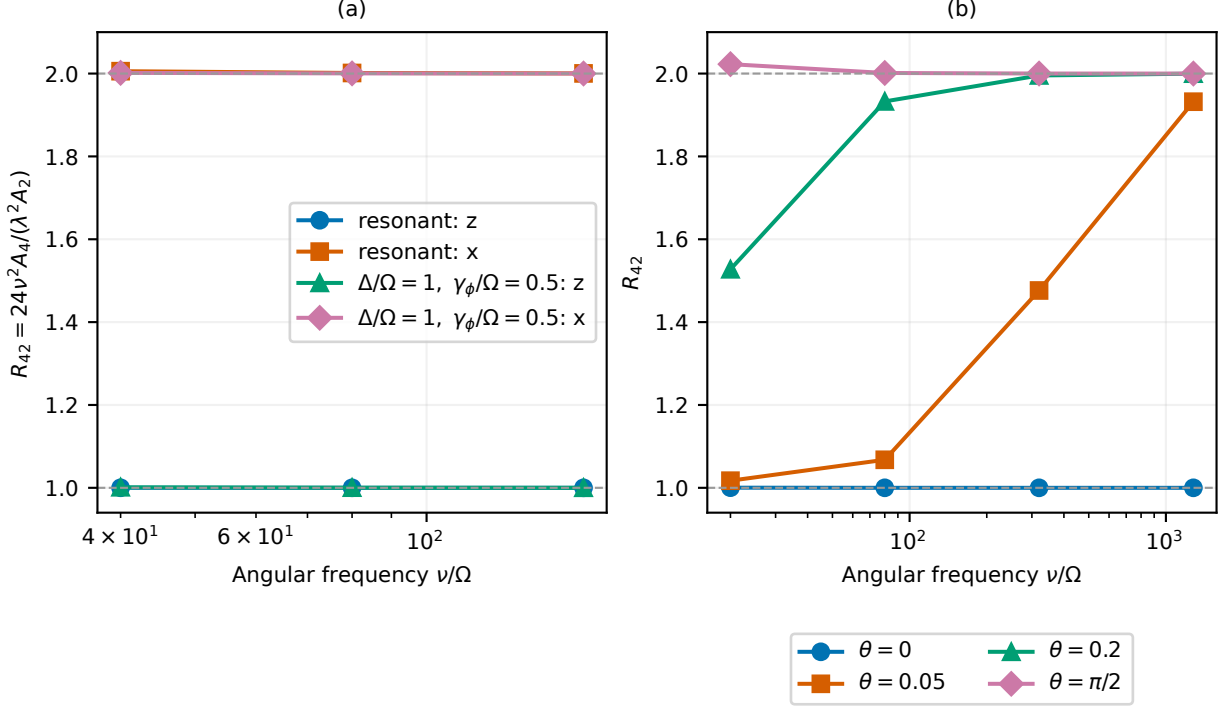


Fig. 1 Full periodic-response ratios on every declared plotted point, with $\Omega = \gamma_1 = 1$. Left: the matched endpoint geometries at $\lambda/\Omega = 0.1$, for the resonant seed and the second seed $\Delta/\Omega = 1$, $\gamma_\phi/\Omega = 0.5$. Right: resonant axis-tilt scan at $\lambda/\Omega = 0.2$; angles are in radians. Symbols are the full harmonic-balance ratios, independently checked by the time solver. Lines join discrete computed points; dashed levels are the minimal-order high-frequency predictions, rather than fitted curves. All 28 plotted points are retained

Figure 1 shows the matched and tilted-axis comparisons. The tilt scan makes the nonuniformity visible. At $\theta = 0.05$, R_{42} changes from 1.017215 at $\nu/\Omega = 20$ to 1.932086 at 1280; it remains approximately 3.4% below two at this last declared point. At $\theta = 0.2$ the corresponding values are 1.527488 and 1.999712. The exact longitudinal axis instead gives 1.000000085 at 1280. The fixed- $\lambda/\nu = 0.2$ diagnostics give $R_{42} = 1.005370$ and 2.011058 at $\nu/\Omega = 160$: these finite-grid deviations illustrate why that scan cannot be substituted for the sequential limit. We do not infer its infinite-frequency value from three points.

The separately frozen state-dependent-zero control of Eq. (18) uses $\lambda/\Omega = 0.1$, $\nu/\Omega = 40, 80, 160$. Its full ratios are 2.700402, 2.699437 and 2.699195, approaching the retained-order value 305/113; both full methods pass every numerical gate on these three points. Together, the positive, crossover and counterexample cases test the interpretation as well as the coefficients. A portable minimal reproducer regenerates the complete new campaign, while a separate launcher stages the full deposited S2 workflow without manual directory changes. Its four preselected diverse-anchor smoke checks pass, and its input and anchor registries are byte-identical. A subsequent complete reexecution on 8 October 2026 ran the frozen 683-point scalar and paired arbitrary-precision workflow, all 16 independent time-domain anchors and all 36 pure-dephasing controls. The separately deposited fourth-harmonic campaign was also reexecuted in full, including all 74 primary arbitrary-precision

points and 2682 registered configurations. Every declared numerical gate passed, and the canonical numerical tables reproduced the archived values exactly. The complete replay sources and original parameter memberships are supplied in Online Resource 2. This reproduction does not restore the undocumented historical sampler or add parameter-general claims.

A separately preregistered 12-point joint-limit campaign uses $\Omega = \gamma_1 = 1$, $\lambda/\Omega = 0.1$ and $\chi = 0.1, 0.5, 2$. For $\gamma_\phi = 0$, each χ receives $\nu/\Omega = 80, 320, 1280$, with $\theta = \arcsin \sqrt{\chi\Omega^2/(2\Gamma_2\nu)}$. Three further points use $\gamma_\phi/\Omega = 0.5$ at the declared endpoint 1280. Both full periodic methods evaluate every point. At that endpoint the maximum amplitude-ratio departure from Eq. (15) is 0.0837%, below the predeclared 2% gate. Lower-frequency deviations and the complex trajectory remain visible; no endpoint is selected after computing the results. Figure 2 compares all twelve new points with the curve and also maps all sixteen earlier tilt points retrospectively. Those older arbitrary-angle points are not presented as new joint-limit confirmation.

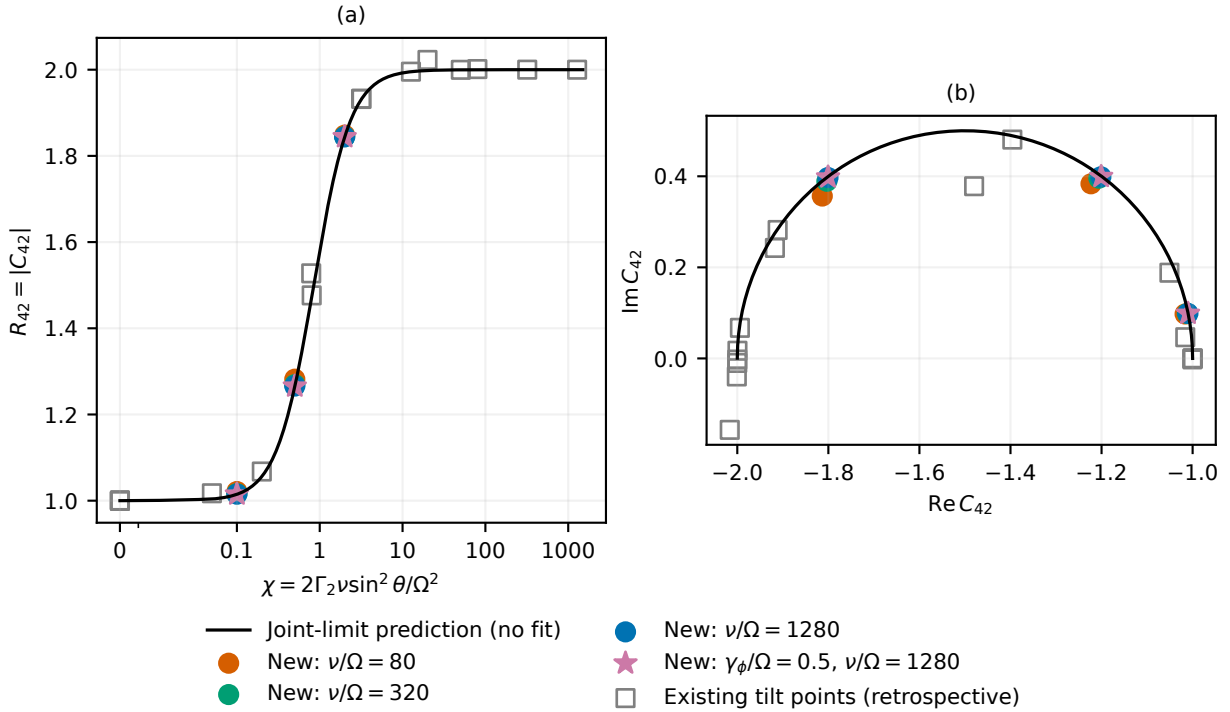


Fig. 2 Amplitude and phase in the resonant crossover. Colored symbols show all twelve preregistered full-response points at $\lambda/\Omega = 0.1$, with $\chi = 0.1, 0.5, 2$; the three starred points have $\gamma_\phi/\Omega = 0.5$. Black curves are the minimal-order joint-limit predictions, without fitting. Gray squares map all sixteen earlier tilt points at $\lambda/\Omega = 0.2$ retrospectively; arbitrary fixed angles and low frequencies need not satisfy the joint small-angle limit. The phase reference is the common sine in Eq. (1). In the right panel the plotted full-response complex ratio uses the same factor $24\nu^2/\lambda^2$ as Eq. (16); it is compared with a minimal-order prediction, rather than equated to it. Every plotted point and its source identity are retained

The density-matrix and joint-crossover solvers, frozen inputs and full-residual reduction records are supplied in Online Resource 3. An orthogonal implementation constructs the 2×2 density-matrix master equation directly from Hamiltonian commutators and jump operators, and extracts ρ_{ee} instead of importing Bloch equations or their population normalization. Nine predeclared controls cover weak positive relaxation, strong dephasing,

signed detuning, a common drive-phase shift, finite modulation depth and both sides of the state-cancellation angle. All 27 required $h = 1, 2, 4$ coefficient checks pass. This tests representation and normalization errors in addition to solver agreement; the implementations still share arbitrary-precision arithmetic and Taylor machinery.

For the 43 previously computed validation points, outward interval reductions at 100 and 140 decimal digits evaluate the complete Fourier-polynomial residual, including both exterior sectors. The 440 required harmonic enclosures all pass their predeclared gates. The largest absolute complex-population error bound, divided by the stored coefficient magnitude, is below 7.71×10^{-41} . This bounds the error of that polynomial against the full periodic model; it is not an error bound for the asymptotic formula. A true-relative error requires a positive lower bound on the unknown coefficient magnitude. Exact decimal parameters are distinguished from intended mathematical transverse and seed-cancellation axes, which are separately evaluated with their canonical matrices. The enclosure relies on outward interval arithmetic and its library implementation; it is not a formal verification of that software. Operational floors of the older campaigns retain their original meaning.

The same a posteriori procedure is evaluated separately on the 21 density-control and joint-crossover HB polynomials. Its 168 additional harmonic enclosures pass, with a true-relative complex-population error bound below 7.12×10^{-43} . Quotient-error propagation gives 128 ratio enclosures across the 43 earlier and 21 additional points, with serialized and intended parameter targets treated separately. Every denominator enclosure excludes zero; the largest relative ratio-interval width is below 1.55×10^{-40} . These posterior error bounds assess the computed full solutions. They are not fitted tolerances or evidence that an asymptotic prediction is exact at a finite point.

At $\nu/2\pi = 40$ MHz the deposited second-harmonic amplitudes are 3.25519×10^{-7} longitudinally and 2.60562×10^{-5} transversely: the ratio is $A_{2\nu}^{(x)}/A_{2\nu}^{(z)} = 80.0452$, compared with $\nu/\Omega = 80$. The fourth-harmonic compensated amplitude is dimensionless,

$$\frac{A_{4\nu}^{(z)}\nu^5}{\lambda^4\Omega} \longrightarrow \frac{x}{48(x^2 + 2)}, \quad x = \gamma_1/\Omega, \quad (22)$$

at resonance without additional pure dephasing. Its value $1/144$ at $x = 1$ holds for any $\Omega > 0$; no hidden choice of $\Omega = 1$ is required in this formula.

6 Interpretation and limitations

The physical mechanism is a readout projection followed by a static-drift conversion, as made explicit by Eq. (8). It does not imply weaker dissipation, a new dynamical symmetry, or absence of the even harmonics. The ratio distinguishes the two ideal matched endpoints at the retained order, but a small axis tilt can eventually produce the generic ratio. Finite-frequency deviations within a fixed geometry do not establish a new symmetry class. These facts qualify the use of the asymptotic ratio as a geometry diagnostic.

Detuning and dephasing affect the prepared seeds. Their cancellation from the longitudinal ratio makes it an internal consistency test, rather than an independent estimator of either parameter. The drive amplitude and frequency must be known. A measured ratio would also require correction for unequal readout transfer at 2ν and 4ν . We infer no detection-noise, calibration or sensitivity advantage. The phenomenological dissipator is held fixed throughout the comparison; a laboratory system in which modulation alters its bath coupling requires a different model. Upward thermal transitions, multilevel structure, colored noise and non-Markovian effects are not tested here.

The resonant crossover quantifies how the interpretation of suppression changes with an in-phase control admixture. The fixed-even-harmonic family and separate chain and full-periodic residual bounds support this comparison within the specified model. They do not prove a uniform high-frequency expansion of the complete population, especially when the seed vanishes or modulation strength scales with frequency. Existing Rabi-harmonic studies [2, 4, 3], the general bilinear hierarchy [6, 7], and open-system high-frequency theory [9, 14] supply the context. Multi-tone dissipative Rabi calculations [15] address related modulations with different approximations. The present scope is an explicit matched population comparison, a test of its fragility and finite-domain numerical validation.

7 Conclusion

Population-harmonic suppression can compare ideal modulation directions, but its interpretation changes under an in-phase admixture of detuning and drive-amplitude controls. At resonance the small-angle crossover is organized by $\chi = 2\Gamma_2\nu \sin^2 \theta / \Omega^2$, with the minimal-order ratio approaching $F(\chi)$. A specified leading-ratio tolerance consequently narrows the allowed control admixture on a $\nu^{-1/2}$ scale at fixed rates. The detuned state-cancellation example gives the same longitudinal exponent pair with ratio 305/113, demonstrating the limits of inference from those exponents. Fixed-order asymptotic remainders and full-periodic residual bounds address different errors. Independent representations and frozen finite grids make these scoped predictions and their deviations directly checkable.

Statements and Declarations

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Author contributions. Yigit Yardimci: conceptualization, methodology, formal analysis, investigation, validation, data curation, software, visualization, writing—original draft, and writing—review and editing. Computational assistance from OpenAI Codex is described in Section 5.

Data and code availability. Supporting code, frozen parameter tables, numerical result tables, figure sources and mathematical proof/check sources are publicly available in Zenodo v3.0.0 at <https://doi.org/10.5281/zenodo.23249084> [16]. This release preserves the original benchmark archive unchanged and includes the population-harmonic and density-matrix/residual reproducibility packages. Online Resource 1 provides the detailed derivations and numerical protocols. The frozen parameter tables, executable reproducers and result tables for the original benchmarks and the fixed-order, tilted-axis and state-cancellation validations are available as the repository supplementary archive Online Resource 2 (ESM_2.zip) within the published Zenodo v3.0.0 package cited above. The density-matrix and joint-crossover calculations, full-periodic residual and ratio enclosures, and proof sources are available as the repository supplementary archive Online Resource 3 (ESM_3.zip) within that package. The earlier v2.0.0 record at <https://doi.org/10.5281/zenodo.23112065>

retains its historical metadata, but its files now have restricted access; the current public supporting package is identified by the v3.0.0 DOI.

Supplementary Information

Online Resource 1 (ESM_1.pdf). Technical companion: derivations, domain restrictions, fixed-order remainder, full-periodic residual bound, crossover analysis, numerical methods and retained benchmark evidence.

Online Resource 2 (ESM_2.zip). Repository supplementary archive within the Zenodo v3.0.0 package cited in Data and code availability. Reproducible population-harmonic benchmarks: frozen parameter tables, independent solvers, canonical fourth-harmonic and full-ensemble evidence, control/counterexample data and figure/units reproduction.

Online Resource 3 (ESM_3.zip). Repository supplementary archive within the Zenodo v3.0.0 package cited in Data and code availability. Independent density-matrix validation and fixed- χ crossover: frozen numerical grids, full-periodic residual and ratio enclosures, portable replay, proof sources and archived results.

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