

Perturbation Theory for Kerr Nonlinear Leaky Cavities

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In emerging open photonic resonators that support quasinormal eigenmodes, fundamental physical quantities and methods have to be carefully redefined. Here, we develop a perturbation theory framework for nonlinear material perturbations in leaky optical cavities. The ambiguity in specifying the stored energy due to the exponential growth of the quasinormal mode field profile is lifted by implicitly specifying it via the accompanying resistive loss. The capabilities of the framework are demonstrated by considering a third-order nonlinear ring resonator and verified by comparing against full-wave nonlinear finite element simulations. The developed theory allows for efficiently modeling nonlinear phenomena in contemporary photonic resonators with radiation and resistive loss. © 2020 Optical Society of America

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Optical cavities and electromagnetic resonant systems in general are ubiquitous in classical and quantum optics, finding use in a broad range of filtering, switching/routing, sensing, lasing, and nonlinear applications [1]. Photonic resonators of emerging technologies are open and typically become considerably leaky as physical dimensions are shrunk in order to achieve miniaturization and small mode volumes to harness the accompanying field enhancement and boost physical processes such as emission and nonlinearity. In addition, plasmonic materials such as noble metals and graphene further lower the total quality factor due to the accompanying high resistive loss. As a result, contemporary photonic/plasmonic cavities support quasinormal modes (QNMs) with complex eigenfrequencies whose eigenmode profiles feature a pronounced exponential increase away from the resonator (Fig. 1). In this context, fundamental physical and mathematical quantities, such as the mode volume and Purcell factor, have to be carefully (re)defined [2–4]. In addition, the well-known and practically-useful perturbation theory approach for efficiently assessing the effect of (small) material and structural modifications should be revised, as has been shown in the literature for linear systems [5–7].

However, the use of perturbation theory for *nonlinear* material modifications in leaky optical cavities has not yet been discussed and is of both fundamental and applied scientific interest. In this Letter, we develop a perturbation theory framework for nonlinear leaky resonators. The main obstacle we have to address concerns the ambiguity in defining the stored energy in leaky cavities, due to the eigenmode field divergence (Fig. 1). In classical perturbation theory of closed resonators, the stored energy is uniquely defined and used as the normalization parameter of the modes. We manage to lift this ambiguity by indirectly defining the stored energy via the definition of the resistive quality factor, which entails integration strictly in the resonator body and, thus, does not depend on the computational domain size and the mode's exponential divergence. The capabilities and potential of the derived framework are exemplified by considering a third-order nonlinear resonator and rigorously verified by comparing against full-wave nonlinear finite-element (FEM) simulations.

As has been discussed extensively in the literature, the classical, first order perturbation theory for resonant systems [8] is valid as long as light leakage (radiation damping) remains low [6]. Recently, a modification of the classical perturbation theory using the unconjugated, rather than the conjugated, form of the Lorentz reciprocity theorem has enabled dealing with leaky, open cavities as well [6, 9, 10]. This alternative form indicates that the complex resonance frequency $\tilde{\omega} = \omega_r + j\omega_i$ of a cavity ($\exp\{+j\omega t\}$ time-harmonic convention) is modified under a perturbation \mathbf{P}_{pert} (inside a volume V_p enclosing the resonator) by the complex quantity $\Delta\tilde{\omega}$, calculated through (see Supple-

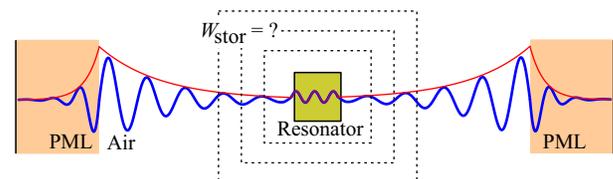


Fig. 1. Generic leaky resonator, demonstrating the exponential growth of the quasinormal mode field profile away from the resonator. Using perfectly matched layers (PMLs) the fields at the boundary of the computational domain can be zeroed out. Due to the this exponential growth, the stored energy cannot be unambiguously defined by integrating the energy density, as customarily exercised.

ment 1, section S1 for details)

$$\begin{aligned} \frac{\Delta\tilde{\omega}}{\tilde{\omega}_0} &= -\frac{\iiint_{V_p} \mathbf{P}_{\text{pert}} \cdot \mathbf{E}_0 \, dV}{\iiint_V \varepsilon_0 \frac{\partial\{\omega\varepsilon_r(\omega)\}}{\partial\omega} \mathbf{E}_0 \cdot \mathbf{E}_0 \, dV - \iiint_V \mu_0 \mathbf{H}_0 \cdot \mathbf{H}_0 \, dV} \\ &= -\frac{\iiint_{V_p} \mathbf{P}_{\text{pert}} \cdot \mathbf{E}_0 \, dV}{Q_{\text{QNM}}}. \end{aligned} \quad (1)$$

In Eq. (1), lossy nonmagnetic materials with arbitrary dispersion have been considered; the spectral derivative of the dispersive permittivity is calculated at $\omega = \tilde{\omega}_0$ (notation omitted for brevity). Extension to dispersive magnetic materials is trivial. Spatial dependence of the quantities in Eq. (1) has been suppressed for brevity. In studies so far, \mathbf{P}_{pert} typically refers to *linear* material perturbation [6, 9] in a volume V_p (inside or in the vicinity of the resonator), taking the form $\mathbf{P}_{\text{pert}} = \Delta\varepsilon\mathbf{E} \simeq \Delta\varepsilon\mathbf{E}_0$. Shape deformations are also possible but require different treatment [7]. The main advantage of the unconjugated formulation is that the end result can be complex, in contrast with the conjugated form where \mathbf{E}_0^* and \mathbf{H}_0^* appear in the numerator and the denominator, producing norms for the complex quantities and leading to solely real frequency shifts. Physically, the unconjugated approach allows to model not only resonance frequency shifts but also linewidth modifications, a capability that ultimately originates from the validity of the unconjugated Lorentz reciprocity theorem in lossy materials and radiation modes [11]. To reach Eq. (1) we have zeroed-out the boundary integral that arises; although this step is straightforward in cavities with negligible radiation [12], for leaky cavities it requires specific treatment. In this work, it is achieved by enclosing the computational space with perfectly matched layers (PMLs) and performing the integration *inside* the complex-valued PML stretched coordinates as well, as also exercised in Ref. [3] (care should be taken to accommodate the evanescent tails of the mode). Alternatively, the surface term that emerges should be appropriately treated [9, 13]. The two approaches have been proven equivalent [14].

The validity of Eq. (1) has been verified in the literature under *linear* perturbations [6, 7, 9]. Nevertheless, perturbation theory for leaky cavities should be also applicable to *nonlinear* perturbations. The key point when Eq. (1) is to be applied in nonlinear systems, is the ability to unambiguously define the stored energy W_{stor} (this is not required for linear perturbation theory) and cast Eq. (1) in the form

$$\Delta\tilde{\omega}(W_{\text{stor}}) = -\tilde{\gamma}W_{\text{stor}}^k, \quad (2)$$

where the power k is related to the nonlinear effect order/type ($k = 1$ for the Kerr effect, $k = 2$ for carrier effects, etc). However, due to the exponential growth of the quasinormal mode away from the resonator, the stored energy cannot be defined by simply integrating the energy density, since the result will depend on the integration domain size (Fig. 1). Here, we assume third-order nonlinearity and self-phase modulation due to the instantaneous Kerr effect; generalization to other nonlinearity types and phenomena should follow a similar approach. The Kerr effect induces a change in the refractive index of a material proportional to the illuminating light intensity ($\propto n_2I$) [15], or, in terms of the electric field, a nonlinear polarization of the form $\mathbf{P}_{\text{NL}} = (1/3)\varepsilon_0^2\text{Re}\{\varepsilon_r\}c_0n_2[2(\mathbf{E} \cdot \mathbf{E}^*)\mathbf{E} + (\mathbf{E} \cdot \mathbf{E})\mathbf{E}^*]$, with n_2 (in m^2/W) denoting the nonlinear index (isotropic nonlinearity has been assumed). For the Kerr effect, $\mathbf{P}_{\text{pert}} = \mathbf{P}_{\text{NL}}$

is the perturbation term and for its introduction in Eq. (1), we allow $\mathbf{E} \simeq \mathbf{E}_0$ (first order perturbation theory). The crucial step is the next one: to reach the form of Eq. (2), we use the definition of the resistive quality factor Q_{res} to indirectly define the stored energy and multiply the right-hand side of Eq. (1) by $W_{\text{stor}}/W_{\text{stor}} = W_{\text{stor}}/(Q_{\text{res}}P_{\text{res}}/\omega_0)$, where $P_{\text{res}} = -(1/2)\iiint_{V_{\text{cav}}} \omega_0\varepsilon_0\text{Im}\{\varepsilon_r\}|\mathbf{E}_0|^2dV$. This action transforms Eq. (1) in the form of Eq. (2), rendering the nonlinear parameter $\tilde{\gamma}$ independent of the stored energy in the cavity. In addition, since P_{res} is specified by integrating strictly inside the cavity volume V_{cav} , its calculation is unambiguous and independent of the computational domain size. Note that the application of the developed framework requires a non-vanishing linear material loss. This is not a limiting factor whatsoever, since all physical structures suffer from some, even small, material loss or are attributed a non-vanishing phenomenological loss due to e.g. surface roughness. Even in lossless systems, a small level of material loss can always be appointed without affecting the response.

Eventually, Eq. (1) is transformed to (see Supplement 1, section S1 for a step-by-step extraction)

$$\Delta\tilde{\omega}(W_{\text{stor}}) = -\tilde{\gamma}_{\text{ucj}}W_{\text{stor}} = -4\left(\frac{\omega_0}{c_0}\right)^3 c_0\tilde{\omega}_0\tilde{\kappa}_{\text{ucj}}n_2^{\text{max}}W_{\text{stor}}, \quad (3)$$

where the quantities marked with tilde are in general complex. A complex $\tilde{\gamma}_{\text{ucj}}$ originating from a purely real n_2 means that radiation and ohmic lifetime (quality factor) modifications are induced by refractive index changes, due to resonant mode redistributions. The *nonlinear feedback parameter* $\tilde{\kappa}_{\text{ucj}}$ introduced in Eq. (3) constitutes a dimensionless, intensity-independent metric of the mode/nonlinear material overlap and is defined as

$$\tilde{\kappa}_{\text{ucj}} = \left(\frac{c_0}{\omega_0}\right)^3 \frac{\iiint_{V_p} n_2\text{Re}\{\varepsilon_r(\omega_0)\}|\mathbf{E}_0|^2(\mathbf{E}_0 \cdot \mathbf{E}_0) \, dV}{\frac{1}{\varepsilon_0^2}Q_{\text{QNM}}n_2^{\text{max}}} \frac{\omega_0}{4Q_{\text{res}}P_{\text{res}}}. \quad (4)$$

The expressions in Eq. (3) and Eq. (4) are the main result of this paper, generalizing previous works based on the conjugated form of the Lorentz reciprocity theorem to build a nonlinear perturbation theory framework, in analogy with Refs. [8, 16, 17]. For comparison, the conventional equations are

$$\Delta\omega(W_{\text{stor}}) = -\gamma_{\text{cj}}W_{\text{stor}} = -4\left(\frac{\omega_0}{c_0}\right)^3 c_0\omega_0\kappa_{\text{cj}}n_2^{\text{max}}W_{\text{stor}}, \quad (5)$$

and

$$\kappa_{\text{cj}} = \left(\frac{c_0}{\omega_0}\right)^3 \frac{\frac{1}{3}\iiint_{V_p} n_2\text{Re}\{\varepsilon_r(\omega_0)\}[2|\mathbf{E}_0|^4 + |\mathbf{E}_0 \cdot \mathbf{E}_0|^2] \, dV}{\frac{16}{\varepsilon_0^2}W_{\text{stor}}^2n_2^{\text{max}}}, \quad (6)$$

where $W_{\text{stor}} = (1/4)\iiint_V \varepsilon_0\text{Re}\{\partial(\omega\varepsilon_r)/\partial\omega\}|\mathbf{E}_0|^2dV + (1/4)\iiint_V \mu_0|\mathbf{H}_0|^2dV$ can be unambiguously specified in systems with very low leakage by integrating the energy density. Note that $\Delta\omega$ and κ_{cj} in Eq. (5) and Eq. (6) are strictly real, describing only resonance frequency shifts and not linewidth modifications. Importantly, if we were to apply Eq. (5) in leaky systems, the result would depend on the computational domain dimensions (see Fig. 4), as the stored energy term in the denominator diverges with increasing integration domain and $\kappa_{\text{cj}} \rightarrow 0$. Note that here we can also use the indirect definition

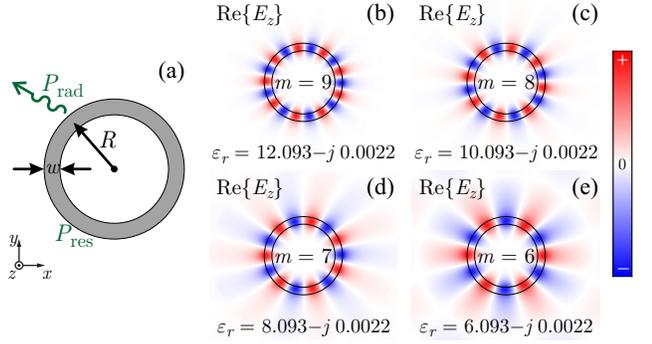


Fig. 2. (a) Nonlinear ring resonator considered in this work. $R = 0.79 \mu\text{m}$ and $w = 200 \text{ nm}$ throughout. (b-e) Field plots (E_z -components) of resonance modes (of azimuthal order m) with gradually increasing radiation damping, owing to the decrease in $\text{Re}\{\epsilon_r\}$ that weakens field confinement. The permittivity values are 12.093, 10.093, 8.093, 6.093 for the four cases respectively and the imaginary part equals -0.0022 throughout.

of the stored energy with the resistive quality factor. This solves the divergence problem; however, Eq. (5) and Eq. (6) fail to predict an imaginary part for the frequency shift and, thus, information for linewidth modifications cannot be attained (see the respective figure in Supplement 1, section S4).

To support the above discussion, we examine a two-dimensional ring resonator of radius $R = 0.79 \mu\text{m}$ and width $w = 200 \text{ nm}$, as shown in Fig. 2(a). The relative permittivity of the ring is varied from 12.093 (ϵ_r of silicon) to 6.093 in steps of 2 RIU in order to access modes with higher radiation loss. The surrounding medium is air ($n = 1$). An imaginary part encompassing ohmic and possibly other fabrication-related losses equal to -0.0022 is assumed in all cases. In Fig. 2(b-e), we plot the E_z -component of the four modes considered, revealing the increasing radiation/bending loss as $\text{Re}\{\epsilon_r\}$ decreases. The depicted results were produced with the eigenvalue solver of COMSOL Multiphysics[®]. In all four cases considered, the resonance wavelength is around $1.52 \mu\text{m}$ and the respective radiation quality factors are approximately 550 000, 27 000, 1 700, and 140 (see Supplement 1, section S5 for a detailed report). Material dispersion is not considered but it could have been easily introduced; in this case, extra care should be exercised for the correct calculation of the quality factors [14, 18].

To confirm our claims, we plot in Fig. 3 the nonlinear feedback parameter κ with respect to the computational domain size (a circular domain of diameter d , enclosed by PMLs) for the four cases considered. We use both Eq. (4) and Eq. (6) for the calculations to highlight the significance of the newly-developed framework (integration order and the normalization term power are reduced by one to fit the 2D geometry chosen). For the first two resonances ($m = 9$ and $m = 8$) for which radiation leakage is weak, the two formulations give identical results for $\text{Re}\{\kappa\}$ [Fig. 3(a)], regardless of the computational domain size. However, it is evident that for the more leaky sixth and seventh order modes, κ_{cj} unphysically depends on the computational domain size, while $\tilde{\kappa}_{\text{ucj}}$ remains constant, as required. Importantly, the imaginary part $\text{Im}\{\kappa\}$ can be calculated only through Eq. (4) [Fig. 3(b)] and is also constant with the computational domain size. As anticipated, leakier resonances exhibit a higher value of $\text{Im}\{\kappa\}$, becoming significant for the sixth order mode ($Q_{\text{rad}} \simeq 140$) as $\text{Im}\{\tilde{\kappa}_{\text{ucj}}\}/\text{Re}\{\tilde{\kappa}_{\text{ucj}}\} = 0.035$. Thus,

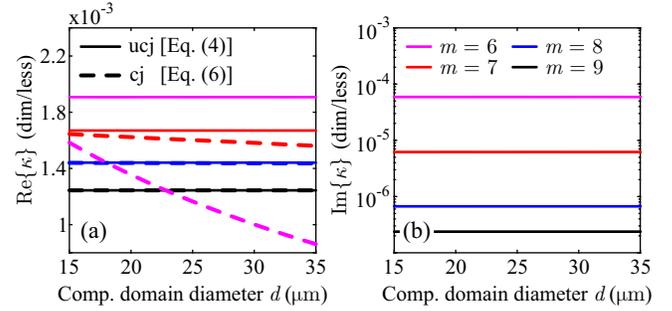


Fig. 3. Nonlinear feedback parameter calculations, $\tilde{\kappa}_{\text{ucj}}$ and κ_{cj} , versus computational domain diameter d for the four resonant modes in Fig. 2. (a) Real part ($\text{Re}\{\kappa\}$): as radiation damping increases, the conjugated formulation breaks down but the unconjugated one remains unaffected. (b) Imaginary part ($\text{Im}\{\kappa\}$): Linewidth modifications can only be predicted by the unconjugated framework.

it is safe to assume that when Q_{rad} falls under 10 000, calculations using the traditional conjugated framework becomes questionable and break down completely when Q_{rad} falls under 1 000. Although these limits are vaguely defined and refer to this system, the observation of κ dependence on d highlights the necessity of resorting to the proposed unconjugated framework and can pinpoint the corresponding limits when working with highly leaky resonators such as compact integrated (rings, disks, Bragg filters, photonic crystal slabs) or free-space resonant systems (nanoparticles/nanoantennas, diffraction gratings, subwavelength metasurfaces).

For validating the proposed framework, we side-couple the ring resonator to a straight bus waveguide [Fig. 4(a)]. We take care to approximately fulfill the critical coupling condition for each case by correctly choosing the coupling gap g . This is not generally necessary for the demonstration but we choose it since it stretches the system most by forcing it to pass through an almost vanishing transmission point (see Fig. 4). Then, we solve the nonlinear time-harmonic problem in CW conditions using the nonlinear solver of COMSOL Multiphysics[®] and compare the results with those obtained using the proposed framework and temporal coupled-mode theory (CMT) [17]. Specifically, we monitor the transmission of the ring resonator which, in the CMT context, is calculated through (see Supplement 1, section S2 for a detailed extraction)

$$\frac{p_{\text{out}}}{p_{\text{in}}} = \frac{(\delta + p_i)^2 + (1 - r_Q + r_{\tilde{\gamma}} p_i)^2}{(\delta + p_i)^2 + (1 + r_Q + r_{\tilde{\gamma}} p_i)^2}, \quad (7a)$$

$$p_{\text{in}} - p_{\text{out}} - p_i = r_{\tilde{\gamma}} p_i^2. \quad (7b)$$

The above polynomial system can exhibit at most three real and positive solutions, pointing to the phenomenon of optical bistability [16, 17]. In Eq. (7), we have defined the normalized frequency detuning $\delta = 2Q_i(\omega - \omega_0)/\omega_0$ (ω is the operating frequency of the feeding wave), the intrinsic over external quality factor ratio $r_Q = Q_i/Q_e$, and the complex nonlinear parameter ratio $r_{\tilde{\gamma}} = -\text{Im}\{\tilde{\gamma}_{\text{ucj}}\}/\text{Re}\{\tilde{\gamma}_{\text{ucj}}\}$, quantifying the contribution of the imaginary part of $\tilde{\gamma}$. All power quantities in Eq. (7) are normalized with respect to the characteristic power of the system, $P_0 = \omega_0^2/2Q_i^2\text{Re}\{\tilde{\gamma}\}$; an incorrect estimation of $\text{Re}\{\tilde{\gamma}\}$ will result in a different normalization and thus erroneous results. Finally, p_i is the power loss due to the initial intrinsic losses (ohmic and radiation), before the possible modifications

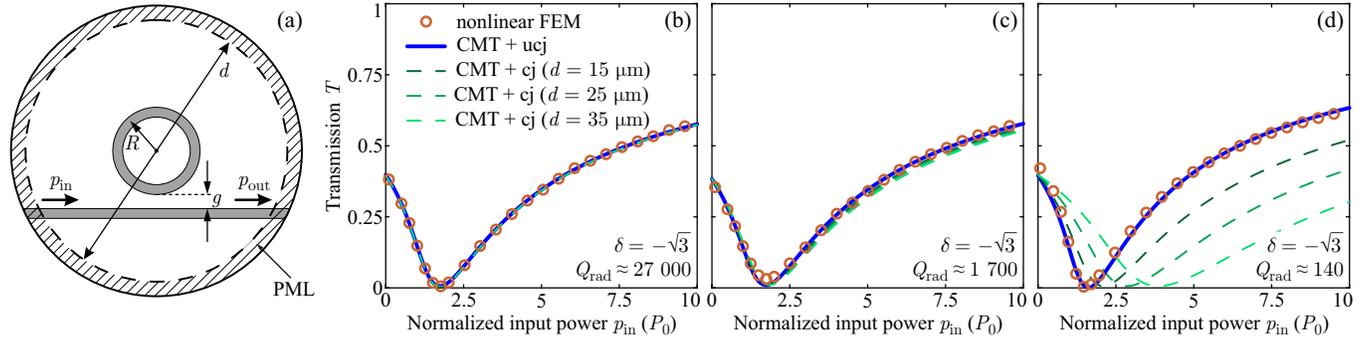


Fig. 4. Verification of the developed, unconjugated (ucj) framework against full-wave nonlinear FEM simulations and comparison with the conventional conjugated (cj) framework. (a) Schematic of the ring structure and the integration domain used. (b-d) Transmission versus input power using CMT and nonlinear FEM for $Q_{rad} \approx 27\,000$, $1\,700$, and 140 , respectively. Excellent agreement is found when the proposed framework is applied, even for highly leaky systems where modifications of the linewidth with input power are pronounced. The conjugated alternative fails and unphysically depends on the chosen domain size d .

induced by the nonlinearity. Note that when the conjugated framework is applied, $r_{\tilde{\gamma}} = 0$.

In Fig. 4, we plot the comparison between the proposed nonlinear framework and the full-wave nonlinear FEM simulations. A normalized detuning parameter equal to $\delta = -\sqrt{3}$ is chosen for all demonstrations. Having approximately fulfilled the critical coupling conditions means that $r_Q \simeq 1$ and, therefore, as the input power increases the transmission curve begins from a high state ($\delta \neq 0$), reaches a minimum which is equal to zero, and then starts to rise again for higher input power levels. This expected behaviour is well captured by the full-wave simulations and perfectly reproduced by perturbation theory/CMT when the unconjugated framework is utilized. Calculations using the conjugated form are also included [for various computational domain sizes, cf. Fig. 4(a)]. For the first two cases [Figs. 4(b,c)] they show good agreement, but start revealing a (weak) dependence on the choice of d , as anticipated [Fig. 4(c)]. This is because the $r_{\tilde{\gamma}}$ parameter acquires small values equal to -0.00060 and -0.0042 , respectively, meaning that linewidth modifications are not yet significant. The most interesting result is obtained for the most leaky case [Fig. 4(d)], for which $r_{\tilde{\gamma}} = -0.039$. In this case, the linewidth changes are notable but can still be captured by the proposed framework. As already mentioned, the effect of linewidth modification is attributed to the mode redistribution induced by the nonlinearity which in turn affects ohmic and radiation losses in the resonator. The proposed unconjugated approach is capable of correctly capturing these modifications as the excellent agreement between the blue solid line and the dot markers (nonlinear FEM) reveals. On the contrary, the conjugated framework (green dashed lines) fails to reproduce the full wave results exhibiting a strong unphysical dependence on the selected integration domain. The framework is expected to hold for even lower Q -factors, in the order of ten or below. Similar conclusions are reached for linear perturbations as well (see Supplement 1, section S3).

In conclusion, we have developed a perturbation theory framework for studying nonlinear material modifications in leaky optical cavities, by implicitly specifying the stored energy in the cavity via its impact on resistive losses in the resonator material. The theory is verified for the case study of a third-order nonlinear ring resonator which undergoes self-phase modulation resulting in frequency and linewidth changes. Apart from integrated resonant structures, the theory can be applied to free-space resonant systems, which are inher-

ently leaky. This work opens the way to applying nonlinear perturbation theory to a broad range of nonlinearity types and phenomena in the blooming field of leaky resonant systems.

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