# Third harmonic generation as a third-order quasi-exactly solvable system

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## Abstract
The two degrees of freedom in the usual third harmonic generation effective Hamiltonian can be separated, both classically and quantum mechanically, into a harmonic oscillator and a nonlinear oscillator. In turn, the quantum Hamiltonian of the nonlinear oscillator can be written as a cubic polynomial in the generators of a quasi-exactly solvable Lie algebra, and the physically relevant eigenstates are precisely those that can be determined exactly (although, in general, not explicitly). Since the standard Jeffreys–Wentzel–Kramers–Brillouin methods are not easily applicable to general third-order differential equations, we use a Bohr–Sommerfeld quantization of the classical orbits to obtain approximate explicit formulas for the corresponding quantum eigenvalues.

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## 1. Introduction

Second harmonic generation and down conversion are among the most extensively studied nonlinear optical processes that can be described by the following effective Hamiltonian,

$$H_{\text{eff}} = a_1^\dagger a_1 + \tfrac{1}{2} + 2\left(a_2^\dagger a_2 + \tfrac{1}{2}\right) + g\left(a_2^\dagger a_1^2 + a_1^{\dagger 2} a_2\right) \tag{1}$$

which features two harmonic modes of angular frequencies one and two, respectively, coupled by a cubic interaction in the creation and destruction operators [1–13].

Although the quantum evolution generated by $H_{\text{eff}}$ can be reduced to the diagonalization of a finite matrix, explicit expressions for the eigenvalues and eigenvectors are not known. Alternatively, the exact solution can be framed within the algebraic Bethe ansatz [8], but again the resulting expressions are neither explicit nor suitable for subsequent analyses. This situation has prompted the development of numerous approximate methods that capitalize

on specific features of certain experimental situations, parameter ranges or time intervals. For example, the process of second harmonic generation, in which two quanta of angular frequency one (the fundamental or pump mode) yield a quantum of angular frequency two (the second harmonic mode), has been studied numerically [2, 5, 6, 12], classically [14, 15], treating the fundamental mode as a classical field (the parametric approximation) [1, 3, 4, 7], by semiclassical methods [11] and by algebraic methods [13, 16].

Apparently considered at the beginning as a mathematical curiosity, in 1987 Turbiner and Ushveridze [17] found a family of sextic polynomial potentials for which a finite number of (the infinite) bound states of the corresponding Schrödinger equation could be 'decoupled' from the rest and exactly calculated by diagonalization of a finite matrix. Although there are elementary ways of constructing nontrivial potentials with exactly calculable bound states, the Hamiltonians found by Turbiner and Ushveridze have an underlying algebraic structure that accounts for the phenomenon, which has been termed 'quasi-exact solvability' (QES).

Further research produced an increasing number of QES 'physical' systems, including electrons moving in an external oscillator potential [18–20], and two-dimensional Schrödinger [21–23], Klein–Gordon [24] and Dirac [25] equations for charged particles in Coulomb and magnetic fields. (Very recently, Chiang and Ho [26] have given a unified treatment of most of these systems.) Simultaneously, the algebraic structure of these Hamiltonians was being studied from a more mathematical point of view: first, the Hamiltonians were identified as *quadratic* polynomials in the generators of certain Lie algebras (the QES Lie algebras); and later, all such quadratic polynomials were classified with due care of the equivalence under changes of variables and the normalizability of the eigenfunctions[27–34].

In a previous paper [35], we identified the second harmonic generation Hamiltonian (1) as an instance of a QES system in nonlinear optics. More concretely, we have shown that the two degrees of freedom in $H_{\text{eff}}$ can be explicitly separated into a linear oscillator and a nonlinear oscillator, and that the latter is equivalent to a (second order) Schrödinger equation with precisely the family of sextic polynomial potentials originally found by Turbiner and Ushveridze. The leading role in the separation of variables is played by the phase difference between the fundamental mode and the second harmonic mode—in fact this phase difference becomes the natural coordinate to describe the evolution (for more details on this topic, see [12, 36, 37]). Finally, with the separated differential equations in Schrödinger form, we used standard Jeffreys–Wentzel–Kramers–Brillouin (JWKB) techniques to obtain explicit semiclassical formulas for the eigenvalues.

It seems natural to ask if a similar situation holds for higher order processes, and the purpose of this paper is to give a detailed account in the case of third harmonic generation [13, 16, 38, 39]. Third harmonic generation is physically interesting not only as a higher order effect, but as the dominant effect in centrosymmetric nonlinear materials (in which second harmonic generation is suppressed), and also as a two-mode restriction of a quantum field theory with quartic self-interaction. As we will see, third harmonic generation is an example of a QES physical system that escapes previous classifications, because it has to be written as a *cubic* polynomial in the generators of a QES Lie algebra. Furthermore, it sets the pattern for higher order processes.

The layout of the paper is as follows. In section 2 we discuss the quantum mechanical aspects of the third harmonic generation effective Hamiltonian, including general properties of the spectrum, separation into two one-dimensional problems, and expression of the nontrivial separated equation as a cubic polynomial in the generators of a QES Lie algebra. Since JWKB methods for third-order differential equations are not fully developed [40] (except in some special cases [41, 42]), and for physical applications we are also interested in explicit formulas for the eigenvalues, we have resorted to a Bohr–Sommerfeld quantization of the

classical orbits. Therefore, in section 3 we discuss in some detail the phase space of the corresponding classical system and in section 4 we rewrite the classical Hamiltonian in terms of action variables, quantize it and present numerical examples of the accuracy of this Bohr–Sommerfeld quantization. The paper ends with a brief summary and a few final comments.

## 2. Third harmonic generation as a QES system

The usual third harmonic generation effective Hamiltonian in second-quantization formalism is

$$H = H_0 + gH_1 \tag{2}$$

where the unperturbed part is the free Hamiltonian for two modes of angular frequencies one and three, respectively,

$$H_0 = a_1^\dagger a_1 + \tfrac{1}{2} + 3\left(a_2^\dagger a_2 + \tfrac{1}{2}\right) \tag{3}$$

and the perturbation, proportional to the coupling constant $g$, is

$$H_1 = a_2^\dagger a_1^3 + a_1^{\dagger 3} a_2. \tag{4}$$

We first transform the Hamiltonians (2)–(4) to the Bargmann representation, where the creation and destruction operators are represented by multiplication and derivation operators with respect to a complex variable $z$ according to the rules

$$a_i^\dagger \to z_i \tag{5}$$

$$a_i \to \frac{\mathrm{d}}{\mathrm{d}z_i}. \tag{6}$$

The associated scalar product is

$$\langle f, g \rangle = \frac{1}{\pi^2} \int_{\mathbf{R}^2 \times \mathbf{R}^2} \mathrm{d}^2 z_1 \, \mathrm{d}^2 z_2 \, \mathrm{e}^{-|z_1|^2 - |z_2|^2} \, \overline{f(z_1, z_2)} g(z_1, z_2) \tag{7}$$

where $\mathrm{d}^2 z_i = \mathrm{d}x_i \, \mathrm{d}y_i$, the normalized harmonic oscillator eigenfunctions are

$$f_{n_1, n_2}(z_1, z_2) = \frac{z_1^{n_1} z_2^{n_2}}{\sqrt{n_1! \, n_2!}} \quad (n_1, n_2 = 0, 1, \ldots) \tag{8}$$

and the transformed Hamiltonian is

$$H = z_1 \frac{\partial}{\partial z_1} + \frac{1}{2} + 3\left(z_2 \frac{\partial}{\partial z_2} + \frac{1}{2}\right) + g\left(z_2 \frac{\partial^3}{\partial z_1^3} + z_1^3 \frac{\partial}{\partial z_2}\right). \tag{9}$$

Since the free part $H_0$ and the perturbation $H_1$ commute, the eigenvalues of $H$ are linear functions of the coupling constant $g$, and the perturbation $H_1$ has to be diagonalized in the subspaces of constant unperturbed energy

$$E_0 = n_1 + \tfrac{1}{2} + 3\left(n_2 + \tfrac{1}{2}\right). \tag{10}$$

In particular, any non-degenerate eigenvector of $H_0$ is an eigenvector of the total Hamiltonian, as is the case for the ground state $(n_1, n_2) = (0, 0)$ and the two lowest excited states $(n_1, n_2) = (1, 0)$ and $(n_1, n_2) = (2, 0)$, with respective eigenfunctions $f_{0,0}(z_1, z_2) = 1$, $f_{1,0}(z_1, z_2) = z_1$ and $f_{2,0}(z_1, z_2) = z_1^2/\sqrt{2}$.

Equations (8) and (10) show that to stay within the subspace of constant unperturbed energy $E_0$, the eigenvectors of $H_1$ have to be homogeneous polynomials of degree

$$k = \frac{n_1}{3} + n_2 \tag{11}$$

in $z_1^3$ and $z_2$, which we write as

$$p(z_1, z_2) = c_1 z_1^{3k} + c_2 z_1^{3k-3} z_2 + \cdots + c_{k+1} z_2^k. \tag{12}$$

Therefore, as we anticipated in the introduction, the action of the perturbation within this subspace is given by a $(k+1) \times (k+1)$ self-adjoint matrix whose nonvanishing elements are

$$[H_1]_{i,i+1} = [H_1]_{i+1,i} = \sqrt{i(3k-3i+3)(3k-3i+2)(3k-3i+1)} \quad (i = 1, \ldots, k). \tag{13}$$

It is immediate to check that if a polynomial $p(z_1, z_2)$ is an eigenvector of $H_1$ with eigenvalue $E$, then $p(z_1, -z_2)$ is also an eigenvector but with eigenvalue $-E$, that is the eigenvalues are symmetrically distributed around zero. For example, the $k = 1$ results are:

$$E = -\sqrt{6} \qquad p_-(z_1, z_2) = \frac{1}{\sqrt{2}} \left( \frac{z_1^3}{\sqrt{6}} - z_2 \right) \tag{14}$$

$$E = +\sqrt{6} \qquad p_+(z_1, z_2) = \frac{1}{\sqrt{2}} \left( \frac{z_1^3}{\sqrt{6}} + z_2 \right) \tag{15}$$

where the polynomial eigenfunctions have been normalized with respect to the Bargmann scalar product (7).

Furthermore, since the diagonalization of the perturbation $H_1$ is equivalent to finding polynomial solutions of the differential equation

$$\left( z_2 \frac{\partial^3}{\partial z_1^3} + z_1^3 \frac{\partial}{\partial z_2} \right) p(z_1, z_2) = E p(z_1, z_2) \tag{16}$$

and, as we have said, the polynomials in equation (12) are homogeneous in $z_1^3$ and $z_2$, we can reduce the problem to an ordinary differential equation by the following change of dependent and independent variables:

$$z = z_1^3 / z_2 \tag{17}$$
$$p(z_1, z_2) = z_2^k P(z) \tag{18}$$

where $P(z)$ is again a polynomial of degree $k$ in $z$. Substitution of equations (17) and (18) into equation (16) gives the following equation for $P(z)$:

$$27 z^2 P'''(z) + 54 z P''(z) + (6 - z^2) P'(z) + k z P(z) = E P(z). \tag{19}$$

The left-hand side of equation (19) is a third-order differential operator which admits polynomial solutions for certain values of the parameter $E$. Therefore [32], it can be written as a polynomial in the following operators, which satisfy the commutation relations of the Lie algebra $\mathrm{sl}_2$:

$$J_k^+ = z^2 \partial_z - k z \tag{20}$$

$$J_k^0 = z \partial_z - \frac{1}{2} k \tag{21}$$

$$J_k^- = \partial_z. \tag{22}$$

The privileged role of these three operators is intuitively clear: $J_k^-$ lowers by one the degree of any monomial $z^n$; $J_k^0$ keeps the degree, and $J_k^+$ raises the degree by one *except* for the monomial $z^k$, which is cancelled; hence, the polynomials in $z$ of degree less than or equal to $k$ are a finite-dimensional invariant subspace for any differential operator that can be written as a polynomial (with constant coefficients) in the above generators.

In our case, it is simple to check that the differential operator in the left-hand side of equation (19) can be written as

$$H_1 = 27 \left[ J_k^- J_k^+ J_k^- + k J_k^0 J_k^- + \tfrac{1}{2} k(k+2) J_k^- \right] + 6 J_k^- - J_k^+. \tag{23}$$

Moreover, all the terms but the last in equation (23) lower the degree of a monomial by one and, consequently, any polynomial eigenvector of $H_1$ has to be exactly of degree $k$.

At this point we have been able to separate the third harmonic generation Hamiltonian into two one-dimensional systems: a trivial harmonic part represented by the $z_2^k$ factor in equation (18), and a nontrivial, QES system represented by the differential equation (19) or, equivalently, by the cubic polynomial in the QES Lie algebra generators given in equation (23). Nevertheless, the QES character of equation (19) does not help much directly to obtain (approximate) explicit formulas for the eigenvalues. In our previous paper on second harmonic generation [35] we took advantage of the second order of the analogous QES equation: we transformed it into Schrödinger form, and used standard JWKB techniques to obtain the desired formulas.

Although formal Hamiltonians with third-order derivatives are not new (they appear, for example, in the application of the inverse scattering method to the Korteweg–de Vries equation), JWKB methods for third-order differential equations like (19) are not easily applicable [40] except in some particular situations [41, 42], usually when both the second-order and first-order derivatives are missing. To circumvent this problem, we resort to a Bohr-Sommerfeld quantization of the corresponding classical system. As we have seen, the change of variables (17)–(18) is very convenient to understand the QES structure of the Hamiltonian, but not so to carry on the semiclassical quantization program. For reasons that will be clarified later, we use new variables

$$z = z_1/z_2^{1/3} \tag{24}$$
$$p(z_1, z_2) = z_2^k Q(z) \tag{25}$$

(in fact $Q(z) = P(z^3)$, i.e. $Q(z)$ is a polynomial of degree $k$ in $z^3$) which substituted into equation (16) yield the following equation for the polynomial $Q(z)$:

$$Q'''(z) - \tfrac{1}{3} z^4 Q'(z) + Q(z)(kz^3 - E) = 0. \tag{26}$$

There is an interesting point concerning the scalar product associated with this differential equation. If we put equations (24) and (25) into the Bargmann scalar product (7) and integrate the variable $z_2$, we arrive at an *apparently* general expression of the scalar product between any two sufficiently well-behaved functions $Q_i(z)$,

$$\langle Q_2, Q_1 \rangle = \frac{1}{\pi} \int_{\mathbf{R}^2} d^2 z \, \Lambda(z) \overline{Q_2(z)} Q_1(z) \tag{27}$$

where

$$\Lambda(z) = 2\pi \int_0^\infty dt \, t^{2k+5/3} e^{-|z|^2 t^{2/3} - t^2}. \tag{28}$$

Equation (27), however, may be divergent even for polynomial $Q_i(z)$ if the degree of each factor is greater than $3k$ in $z$. The reason is that in the derivation of equation (26) it is assumed that $Q(z)$ is a polynomial of degree $k$ in $z^3$ (otherwise, $p(z_1, z_2)$ would not be entire in $z_2$). With this restriction, the integrations in equations (27) and (28) can be trivially done with the expected result,

$$\langle z^{3n}, z^{3m} \rangle = \delta_{n,m} (3n)! \quad (n, m = 0, \ldots, k). \tag{29}$$

A completely equivalent situation happens with the scalar product associated with equation (19).

## 3. The classical Hamiltonian

The classical third harmonic generation Hamiltonian in cartesian coordinates is

$$H = \tfrac{1}{2}\left(p_1^2 + x_1^2\right) + \tfrac{3}{2}\left(p_2^2 + x_2^2\right) + \tfrac{g}{2}\left[x_1^3 x_2 - p_1^3 p_2\right.$$
$$\left. + \tfrac{3}{2}\left(p_1 x_1^2 + x_1^2 p_1\right) p_2 - \tfrac{3}{2}\left(p_1^2 x_1 + x_1 p_1^2\right) x_2\right]. \tag{30}$$

To remove ambiguities, we have written the preceding Hamiltonian in symmetrized form, so that direct quantization of the cartesian coordinates and momenta with the standard rules

$$x_i \to \frac{1}{\sqrt{2}}\left(a_i^+ + a_i\right) \tag{31}$$

$$p_i \to \frac{\mathrm{i}}{\sqrt{2}}\left(a_i^+ - a_i\right) \tag{32}$$

lead to the second-quantized operators (3) and (4). The classical counterpart of the commutation between the unperturbed Hamiltonian and the perturbation is the vanishing of the Poisson bracket:

$$[H_0, H_1]_{\mathrm{PB}} = 0. \tag{33}$$

We begin the classical study by switching to the action-angle variables of the harmonic oscillator,

$$x_i = \sqrt{2\mathcal{J}_i}\cos\vartheta_i \tag{34}$$
$$p_i = -\sqrt{2\mathcal{J}_i}\sin\vartheta_i \tag{35}$$

in which the Hamiltonian (30) has the standard form

$$H = \mathcal{J}_1 + 3\mathcal{J}_2 + 2g\mathcal{J}_1^{3/2}\mathcal{J}_2^{1/2}\cos(3\vartheta_1 - \vartheta_2) \tag{36}$$

which displays the distinctive feature of this system, the three-to-one resonance that makes it a suitable model for third harmonic generation. This resonance can be removed by a linear transformation to new canonical variables $(\theta_1, j_1)$ and $(\theta_2, j_2)$ with generating function

$$F_2(\vartheta_1, \vartheta_2, j_1, j_2) = (3\vartheta_1 - \vartheta_2)j_1 + \vartheta_2 j_2. \tag{37}$$

The old and new variables are related by

$$\mathcal{J}_1 = 3j_1 \tag{38}$$
$$\mathcal{J}_2 = j_2 - j_1 \tag{39}$$
$$\theta_1 = 3\vartheta_1 - \vartheta_2 \tag{40}$$
$$\theta_2 = \vartheta_2 \tag{41}$$

and the transformed Hamiltonian is

$$H = 3j_2 + 6\sqrt{3}g j_1^{3/2}\sqrt{j_2 - j_1}\cos\theta_1. \tag{42}$$

The coordinate $\theta_2$ is cyclic, and therefore the conjugate action $j_2$ is a constant of the motion (namely, one third of the unperturbed energy), while the nontrivial dynamical variables are the phase difference between the two modes $\theta_1$, and its conjugate action $j_1$. Their equations of motion are,

$$\frac{\mathrm{d}\theta_1}{\mathrm{d}t} = 3\sqrt{3}\cos\theta_1 (3j_2 - 4j_1)\sqrt{\frac{j_1}{j_2 - j_1}} \tag{43}$$

$$\frac{\mathrm{d}j_1}{\mathrm{d}t} = 6\sqrt{3}\sin\theta_1 j_1^{3/2}\sqrt{j_2 - j_1}. \tag{44}$$
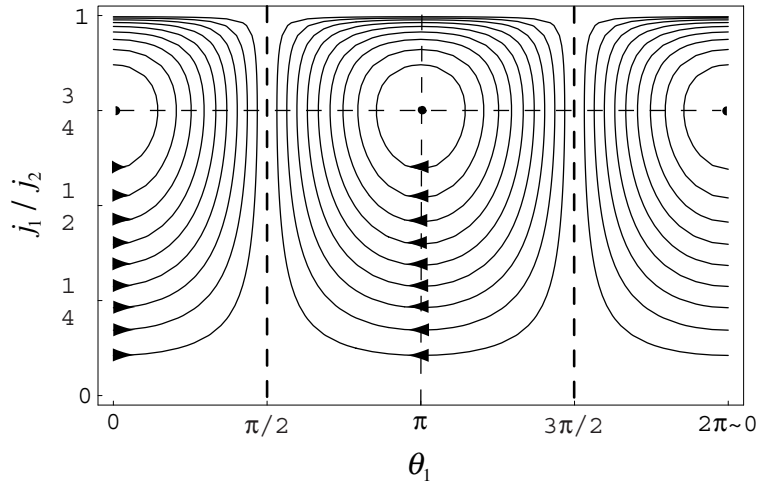
**Figure 1.** Phase map of the perturbation Hamiltonian $H_1(\theta_1, j_1) = 6\sqrt{3}\, j_1^{3/2}(j_2 - j_1)^{1/2} \cos \theta_1$ for a fixed value of $j_2 = 3 + 2/3$, which corresponds to $k = 3$ in the quantum case. The vertical dashed lines are the separatrices, and the dots at $(0, 3/4)$ and $(\pi, 3/4)$ mark the constant solutions of the Hamilton equations (44) and (43) with maximum and minimum energy, respectively.

In figure 1 we have plotted the phase map corresponding to these Hamilton equations for a fixed value of $j_2 = 11/3$ ($k = 3$ in the quantum case). The obvious symmetry of the map is easy to understand: if $(\theta_1(t), j_1(t))$ is a solution of the Hamilton equations corresponding to a perturbation energy $E$, then

$$\hat{\theta}_1(t) = \theta_1(-t) + \pi \tag{45}$$

$$\hat{j}_1(t) = j_1(-t) \tag{46}$$

is also a solution, but corresponding to a perturbation energy $\hat{E} = -E$. In other words, the phase map is symmetrical around $E = 0$.

For a fixed value of $j_2$, the perturbation energy is bounded by

$$-E_{\max} \leqslant E \leqslant E_{\max} = \frac{27}{8} j_2^2. \tag{47}$$

The maximum perturbation energy is reached by the constant trajectory $(\theta_1(t), j_1(t)) \equiv (0, 3j_2/4)$, and the minimum by $(\theta_1(t), j_1(t)) \equiv (\pi, 3j_2/4)$, in agreement with equations (45) and (46). In these constant solutions the cartesian variables $(x_1, p_1)$ and $(x_2, p_2)$ are oscillating harmonically with the non-natural frequencies $\omega_{\pm} = 1 \pm 9j_2/4$ and $3\omega_{\pm}$, respectively, the fundamental mode has $3/4$ of the unperturbed energy, the third harmonic mode has $1/4$ of the unperturbed energy, and there is not any energy exchange between the modes.

The trajectories with $0 < E < E_{\max}$ are oscillations whose phase curves in the $(\theta_1, j_1)$ plane are ovals with vertices $(0, j_1^-)$, $(\theta_{\max}, 3j_2/4)$, $(0, j_1^+)$, and $(-\theta_{\max}, 3j_2/4)$, where $\cos \theta_{\max} = E/E_{\max}$ and the equations for $j_1^{\pm}$ will be given in the next section. An analogous situation with the rotation sense reversed holds for the trajectories with $-E_{\max} < E < 0$ around the center at $(\pi, 3j_2/4)$. In these trajectories, the two modes exchange energy periodically.

The remaining trajectories (with $E = 0$) belong to the separatrices at $\theta_1 = \pi/2$ and $\theta_1 = 3\pi/2$. Note that for these values of $\theta_1$ the Hamilton equations (43) and (44) have non-unique solutions. This is an artifact of the change of variables, and the simplest way out is to study directly the initial conditions corresponding to $E = 0$ in cartesian variables. Except for the trivial solution with $(x_1(t), p_1(t)) \equiv (0, 0)$, in which all the unperturbed energy is always

in the higher mode, the $E = 0$ solutions correspond to situations in which all the unperturbed energy is in the higher mode at $t \to -\infty$, as $t$ increases the energy is first transferred to the fundamental mode, and then fully back to the higher mode, where it ends at $t \to +\infty$.

We finish this section by making contact with the separated quantum Hamiltonian. Consider the following transformation from the angle-action variables $(\theta_1, j_1)$ to new variables $(x, p)$:

$$x = \sqrt{3}e^{i\theta_1/3} j_1^{1/2}(j_2 - j_1)^{-1/6} \tag{48}$$

$$p = -i\sqrt{3}\, e^{-i\theta_1/3} j_1^{1/2}(j_2 - j_1)^{1/6}. \tag{49}$$

This transformation is complex canonical, since

$$[x, p]_{PB} = \frac{\partial x}{\partial \theta_1}\frac{\partial p}{\partial j_1} - \frac{\partial x}{\partial j_1}\frac{\partial p}{\partial \theta_1} = 1 \tag{50}$$

and maps the classical perturbation Hamiltonian

$$H_1 = 6\sqrt{3}j_1^{3/2}\sqrt{j_2 - j_1}\cos\theta_1 \tag{51}$$

to the formally complex Hamiltonian

$$H_1 = -ip^3 - \frac{i}{6}\left(px^4 + x^4p\right) + j_2 x^3 \tag{52}$$

which we have written again in symmetrized form. (Incidentally, we mention that Krichever [43] used a complexification of a classical Hamiltonian with a Weierstrass elliptic potential to prove its algebraically integrability, although in that case there is no known explicit construction of real action-angle variables.) If we formally apply to the Hamiltonian (52) the usual quantization rules,

$$x \to z \tag{53}$$

$$p \to -i\frac{d}{dz} \tag{54}$$

and substitute the classical unperturbed energy $E_0 = 3j_2$ in terms of the quantum unperturbed energy $E_0 = 3k + 2$, that is if we replace $j_2 \to k + 2/3$, we arrive precisely at equation (26). Again, the classical variable $x$ and the quantum variable $z$ carry in their arguments the difference of phases between the modes, $\theta_1 = 3\vartheta_1 - \vartheta_2$, which is a natural variable to describe the dynamics of the perturbation.

## 4. Bohr–Sommerfeld quantization

In this section, we will perform a Bohr–Sommerfeld quantization [44] of the classical perturbation Hamiltonian (51) to obtain explicit formulas for the eigenvalues of the third harmonic generation Hamiltonian. This quantization consists in writing the Hamiltonian as a function of the good action variable

$$J_1(E) = \frac{1}{2\pi}\oint_{H_1=E} j_1(\theta_1)\,d\theta_1 \tag{55}$$

(which, apart from the prefactor, is the area enclosed in phase space by the orbit $H_1 = E$), and then setting $J_1 \to n + 1/2$. Instead of integrating directly in equation (55), we have found a more efficient indirect route to write the Hamiltonian (51) as a function of the good action variables: we first obtain an analytic expression for the period of the oscillation $T(E)$, and then integrate the equation $\partial E/\partial J_1 = 2\pi/T(E)$ that relates the period to the good action variable $J_1$. Finally, we obtain the perturbation energy as a function of the action by inversion of an appropriate series expansion. Let us proceed now with the details of the calculation.

Eliminating $\cos \theta_1$ between the energy $E$ and the equation of motion for $j_1$ we can write the period of the oscillation as

$$T(E) = 2 \int_{j_1^-}^{j_1^+} \frac{\mathrm{d}j_1}{\left(-108 j_1^4 + 108 j_1^3 j_2 - E^2\right)^{1/2}} \tag{56}$$

where $j_1^{\pm}$ are the two real roots of the quartic polynomial under the square root. It is convenient to introduce non-dimensional parameters

$$\epsilon = 1 - \frac{E^2}{E_{\max}^2} \tag{57}$$

$$\tau = \frac{4 j_1}{3 j_2} - 1 \tag{58}$$

and rewrite the period as

$$T(E) = \frac{4}{9 j_2} \int_{\tau_-}^{\tau_+} \frac{\mathrm{d}\tau}{(\epsilon - 6\tau^2 - 8\tau^3 - 3\tau^4)^{1/2}} \tag{59}$$

where $\tau_{\mp}$ are the two real roots of $p(\tau) = \epsilon - 6\tau^2 - 8\tau^3 - 3\tau^4$ (we will denote by $\tau_{1,2}$ the two complex roots). Equation (59) can be conveniently expressed in terms of Gauss' hypergeometric function $F(a, b; c; z)$,

$$T(E) = \frac{4}{9 j_2} \frac{\pi}{[3(\tau_- - \tau_1)(\tau_+ - \tau_2)]^{1/2}} F\left(\frac{1}{2}, \frac{1}{2}; 1; \frac{(\tau_- - \tau_+)(\tau_1 - \tau_2)}{(\tau_- - \tau_1)(\tau_+ - \tau_2)}\right). \tag{60}$$

It is elementary to find as many terms as desired of the power series (in $\epsilon$) expansions of the four roots $\tau_{\pm,1,2}$, which substituted in equation (60) yield the expansion for the period. We show the lowest terms:

$$T(E) = \frac{4\pi}{9\sqrt{6} j_2} \left(1 + \frac{31}{144}\epsilon + \frac{10465}{82944}\epsilon^2 + \frac{9769375}{107495424}\epsilon^3 + \cdots\right). \tag{61}$$

The expansion for the new action variable $J_1$ is readily obtained by termwise integration:

$$J_1 = -\frac{1}{2\pi} \int_0^\epsilon T(\epsilon)\frac{\partial E}{\partial \epsilon} \, \mathrm{d}\epsilon \tag{62}$$

$$= \frac{\sqrt{3} j_2}{8\sqrt{2}} \left(\epsilon + \frac{103}{288}\epsilon^2 + \frac{50497}{248832}\epsilon^3 + \frac{58821031}{429981696}\epsilon^4 + \cdots\right). \tag{63}$$

We will rename $j_2 = J_2$, since then $(J_1, J_2)$ are good action variables for the full Hamiltonian. The actions $J_1$ and $J_2$ appear only in the combination

$$J = \sqrt{\frac{2}{3} \frac{J_1}{J_2}}. \tag{64}$$

Consequently, we will pull out the appropriate factors in equation (63) and obtain an expansion for $J = J(\varepsilon)$. This series, in turn, has to be reversed to obtain $\varepsilon = \varepsilon(J)$, and then plugged into equation (57) to obtain the perturbation energy $E$ as a function of the actions. Thus we obtain a series expansion of the full third harmonic generation Hamiltonian as a series in the good action variables, whose lowest terms are

$$H(J_1, J_2) = 3J_2 + g\frac{27 J_2^2}{8} \left[1 - 4\left(\sqrt{\frac{2}{3} \frac{J_1}{J_2}}\right) + \frac{31}{9}\left(\sqrt{\frac{2}{3} \frac{J_1}{J_2}}\right)^2 + \frac{235}{972}\left(\sqrt{\frac{2}{3} \frac{J_1}{J_2}}\right)^3 + \cdots\right]. \tag{65}$$

The Bohr–Sommerfeld quantization of this expansion is achieved by the replacements

$$J_1 \rightarrow n + \tfrac{1}{2} \tag{66}$$

$$J_2 \rightarrow k + \tfrac{2}{3}. \tag{67}$$

We remark that equation (65) has been obtained by a *convergent* expansion around $\epsilon = 0$ (i.e. $E = E_{\max}$), and that the radius of convergence $J_{1,\max}$ is the value $J_1(E = 0)$ corresponding to the separatrix, namely $(2\pi)^{-1}$ times the area of the rectangle $[-\pi/2, \pi/2] \times [0, J_2]$:

$$J_{1,\max} = \frac{J_2}{2}. \tag{68}$$

For the same reason, the Bohr–Sommerfeld quantized equation (65) gives only approximations to the $[k/2]$ positive eigenvalues (if $k$ is odd, there is an additional zero eigenvalue), that is the allowed values of $n$ in equation (66) are $n = 0, \ldots, [k/2]$, and since

$$\frac{k/2}{k + 2/3} < \frac{1}{2} \tag{69}$$

all the positive eigenvalues fall within the disk of convergence of the series.

Note that $J_1$ is an increasing function of the parameter $\epsilon$, that all the terms in equation (63) are positive and, consequently, that all the terms in equation (65) beyond $J_1/J_2$ are also positive. Therefore, we should consider at least terms up to $(J_1/J_2)^2$ in the Bohr–Sommerfeld quantization. As a first, very crude test, we recall that the exact positive eigenvalue for $k = 1$ ($n = 0$) is $E = \sqrt{6} \approx 2.45$, while equation (65) up to $(J_1/J_2)^2$ gives $E \approx 2.13$.

A more elaborate example corresponding to $k = 9$ is presented in table 1. In each column we list the partial sums up to $(J_1/J_2)^{10}$ of the Bohr–Sommerfeld quantized perturbation energy, the sum of the full series, and the exact quantum eigenvalue obtained by numerical diagonalization of the matrix (13). The zeroth order sum is the classical limit $E_{\max}$, independent of the eigenvalue. As we argued in the previous paragraph, the correct qualitative behaviour sets in at order $(J_1/J_2)^2$. Even for such a small value of $k$, four of the five eigenvalues have almost converged at order 10, and the differences between the converged results and the quantum mechanical results are due to higher quantum corrections. As is typical in semiclassical approximations, the accuracy decreases as the energy tends to the separatrix.

**Table 1.** Partial sums of the Bohr–Sommerfeld quantized perturbation energy for the third harmonic generation Hamiltonian with $k = 9$. The row labelled "$\infty$" is the converged value of the series, and the last row, labelled "QM", is the quantum mechanical result obtained by numerical diagonalization of the corresponding matrix (13). Only the positive eigenvalues are listed.

| Order in $J_1/J_2$ | $n = 0$ | $n = 1$ | $n = 2$ | $n = 3$ | $n = 4$ |
|---|---|---|---|---|---|
| 0 | 315.38 | 315.38 | 315.38 | 315.38 | 315.38 |
| 1 | 262.10 | 155.55 | 48.99 | −57.56 | −164.11 |
| 2 | 264.04 | 172.98 | 97.43 | 37.38 | −7.18 |
| 3 | 264.04 | 173.14 | 98.15 | 39.35 | −2.99 |
| 4 | 264.04 | 173.17 | 98.41 | 40.36 | −0.22 |
| 5 | 264.04 | 173.18 | 98.51 | 40.86 | 1.54 |
| 6 | 264.04 | 173.18 | 98.54 | 41.11 | 2.65 |
| 7 | 264.04 | 173.18 | 98.54 | 41.23 | 3.36 |
| 8 | 264.04 | 173.18 | 98.55 | 41.29 | 3.81 |
| 9 | 264.04 | 173.18 | 98.56 | 41.32 | 4.12 |
| 10 | 264.04 | 173.18 | 98.56 | 41.34 | 4.33 |
| $\infty$ | 264.04 | 173.18 | 98.56 | 41.36 | 5.02 |
| QM | 264.31 | 173.44 | 98.82 | 41.66 | 5.54 |

From a practical point of view, we can summarize the results of this section in the statement that, for moderate and large values of $k$, the behavior of the eigenvalues is captured by just the first three terms of the series in equation (65),

$$E(J_1, J_2) \approx \frac{27 J_2^2}{8} \left[ 1 - 4 \sqrt{\frac{2}{3}} \frac{J_1}{J_2} + \frac{62}{27} \left( \frac{J_1}{J_2} \right)^2 \right] \tag{70}$$

which gives an equation simple enough to be used in analytic work.

## 5. Summary and final comments

In this paper, we have seen that the two degrees of freedom in the usual third harmonic generation effective Hamiltonian can be explicitly separated, both classically and quantum mechanically, into a harmonic oscillator and a nonlinear oscillator. The quantum Hamiltonian of the nonlinear oscillator can be written as a cubic polynomial in the generators of a quasi-exactly solvable Lie algebra, and the physically relevant states for third harmonic generation are precisely the part of the spectrum that can be determined exactly in the quasi-exactly solvable system. This is, to our knowledge, the first example of a third-order QES physical system.

The statement that part of the spectrum of a QES system can be solved exactly, however, does not mean that it can be determined as an explicit function of suitable quantum numbers and the parameters in the Hamiltonian, but implicitly as the solutions of a finite set of algebraic equations which can be written in several equivalent ways (for example, as the eigenvalues of a matrix, or as the roots of a set of Bethe ansatz equations). For physical applications, however, the interest of explicit (albeit approximate) formulas is clear. In the case of second harmonic generation and, for that matter, for any quadratic polynomial in the generators of the algebra, we can put the second-order differential equation in Schrödinger form and use standard JWKB techniques to find approximate semiclassical equations for the eigenvalues. This is not the case for third-order and higher order differential equations, because the JWKB techniques are not easily applicable. We have circumvented this problem by performing a Bohr–Sommerfeld quantization of the classical orbits of the nonlinear oscillator. We emphasize the numerical consistency between the Bohr–Sommerfeld eigenvalues and the exact quantum results even for very moderate values of the unperturbed energy.

We finally point out that besides the intrinsic interest of the third harmonic generation Hamiltonian (for example, as the leading effect in centrosymmetric nonlinear materials), our study sets the pattern for the analysis of higher order processes as separable, QES physical systems for which a Bohr–Sommerfeld quantization of the classical orbits yields explicit semiclassical eigenvalues.

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