Symmetry of approximate Hamiltonians generated in Birkhoff-Gustavson normal form

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Previous investigators have shown how the Birkhoff-Gustavson normal form (BGNF) procedure can be used to develop an integrable Hamiltonian which approximates a nonintegrable Hamiltonian system. We discuss the symmetry properties of these approximate Hamiltonians and how a certain flexibility inherent to the BGNF procedure can be used to affect this symmetry. Because the BGNF procedure involves a transformation of the coordinates, the symmetry issue has been somewhat neglected due to its complexity. The techniques we demonstrate here with the Hénon-Heiles Hamiltonian are easily generalized for use in molecular Hamiltonians of arbitrary symmetry. [S1050-2947(96)09307-9]

I. INTRODUCTION

The representation of physical systems as collections of classical coupled oscillators is ubiquitous in theoretical chemistry and physics, the vibrations of small molecules being an important example [1]. It is of considerable interest to find the quantum (vibrational) energy levels of such systems because knowledge of the vibrational ladder spacings would allow for the prediction of a molecule's spectroscopic transitions. It has long been recognized that semiclassical methods hold great promise for the determination of the quantum vibrational energy levels of coupled oscillator systems [2]. This promise is largely due to the fact that semiclassical methods are not encumbered by the need for the very large basis sets which make standard variational calculations intractable. One of the oldest and most common of the semiclassical methods, the EBK technique, due to Einstein, Brillouin and Keller [3], is really an extension of the Bohr-Sommerfeld quantization rule of the old quantum theory to multidimensional integrable Hamiltonian systems. Unfortunately, coupled oscillator systems are frequently *non*integrable and therefore considerable effort has been expended in an attempt to generalize semiclassical techniques to nonintegrable systems [3–7]. Another approach is to find an integrable system which closely approximates the nonintegrable system of interest, and then use the EBK procedure to find the quantum energy levels of the approximate integrable system. This technique was developed by Swimm and Delos [8] and Jaffé and Reinhardt [3]. These authors employed the Birkhoff-Gustavson normal form (BGNF) procedure to generate the approximating Hamiltonian [9]. Algebraic quantization has also been used to quantize these Hamiltonians from the BGNF procedure [10].

Our current interest, and the subject which is addressed in the present paper, lies in the determination of these approximating Hamiltonians. We have noted that these approximating Hamiltonians do not necessarily possess the same symmetry properties as the original nonintegrable system upon which they are based. This has important implications for quantization as the degeneracies of the quantum states are determined by the symmetries of the Hamiltonian. This issue of symmetry is more subtle than a cursory glance might suggest because the BGNF method of producing the approxi-

mating Hamiltonian involves transformation of not only the coordinates but also the momenta. In addition, we have also noted, as did several previous authors, that there is some flexibility in the BGNF transformation procedure. We will demonstrate that this flexibility can be utilized to maintain the symmetry properties of the full Hamiltonian in the approximating Hamiltonian expressed in the transformed coordinates. We believe this procedure will be useful in approximating molecular Hamiltonians describing vibrational motion as well as in other applications.

II. THE BIRKHOFF-GUSTAVSON NORMAL FORM PROCEDURE

Consider a Hamiltonian of the form

$$H(x_1,x_2,p_1,p_2) = H_2(x_1,x_2,p_1,p_2) + \epsilon H'(x_1,x_2,p_1,p_2),$$
(1)

where H_2 is an integrable, zeroth-order Hamiltonian, and H' is a perturbation which contains any nonintegrability. Here we represent a two-dimensional system with coordinate variables x_1,x_2 and conjugate momenta p_1,p_2. The generalization to more than two dimensions is straightforward. It is assumed that H_2 contains only quadratic terms in the four variables and can be written as a sum of uncoupled harmonic oscillators (e.g., normal modes). It is further assumed that H' can be written as a sum of polynomials of degree ≥ 3 in the four variables, each term having a constant real coefficient. In the Birkhoff normal form (BNF) procedure, a series of canonical transformations is found which transforms the Hamiltonian order-by-order into a "normal" form which is integrable. Each successive transformation normalizes the expression to one higher power in the perturbation parameter ϵ. For systems which are nonintegrable, this series cannot converge, but if it is truncated to some finite number of terms, the resulting expression is an integrable approximation to the original Hamiltonian. Gustavson's contribution to the Birkhoff-Gustavson normal form procedure was to generalize the Birkhoff normal form procedure to cases where the zeroth-order Hamiltonian H_2 is resonant [9].

The BGNF procedure has been discussed in detail several times before [3,8,9,11], so we will restrict our presentation

here to a brief outline. The procedure is based on the equivalence of the Liouville operator to the time derivative operator [12]. For a dynamical quantity, α,

$$\frac{d\alpha}{dt}=\{\alpha,H\}=\sum_i\left(\frac{\partial H}{\partial p_i}\frac{\partial \alpha}{\partial q_i}-\frac{\partial H}{\partial q_i}\frac{\partial \alpha}{\partial p_i}\right). \qquad (2)$$

We recognize this as the Poisson bracket of α with H. The normal operator is the Poisson bracket with H_2, the zeroth-order Hamiltonian,

$$\mathbf{D}\alpha=\{\alpha,H_2\}=\sum_i\left(p_i\frac{\partial \alpha}{\partial q_i}-q_i\frac{\partial \alpha}{\partial p_i}\right). \qquad (3)$$

Functions which are in the null space of the normal operator are then constants of the motion of H_2.

The goal of the BNF procedure is to find a transformed Hamiltonian, $\widetilde{H}$, which can be written

$$\widetilde{H}=\sum_i a_iv_i, \qquad (4)$$

where

$$\mathbf{D}v_i=0 \qquad (5)$$

for all v_i. That is, the new Hamiltonian, $\widetilde{H}$, is to be a linear combination of the null-space vectors of $\mathbf{D}$. The v_i can be shown to all commute with each other (such a set of vectors is said to be ''in involution'') and will therefore be constants of the motion for the approximate Hamiltonian, $\widetilde{H}$. In a two-dimensional system, there will be one other *independent* constant of the motion in addition to the energy.

Gustavson [9] showed that in the resonant case the dimension of the set of vectors which span the null space of $\mathbf{D}$ is larger than in the nonresonant case, but this expanded set is no longer in involution. Nevertheless, it is still possible to construct a second independent constant of the motion. This constant of the motion $\widetilde{I}$ can be taken to be

$$\widetilde{I}=\widetilde{H}-\widetilde{H}_2. \qquad (6)$$

It follows from $\{v_i,H_2\}=0$, that $\{\widetilde{H},H_2\}=0$. Since H_2 is by definition already in normal form, $\{\widetilde{H}-\widetilde{H}_2,\widetilde{H}\}=0$, and therefore $\widetilde{I}$ is a constant of the motion of the approximate Hamiltonian, $\widetilde{H}$.

The jth transformation to the new Hamiltonian is given by the generating function [13],

$$S_j=\sum_i q_iP_i+\epsilon^jW_{j+2}(q_iP_i) \qquad (7)$$

where the lower case indicates old ''untransformed'' variables, and upper case denotes new ''transformed'' variables. Since there is a new transformation at each order j, the Q and P from the jth transformation are the q and p for the $(j+1)$st transformation. The following transformation rules follow from this generating function:

$$p_i=P_i+\epsilon^j\frac{\partial W}{\partial x_i};$$

$$x_i=X_i-\epsilon^j\frac{\partial W}{\partial P_i}. \qquad (8)$$

W is given by the solution of

$$\mathbf{D}W=\widetilde{H}-H. \qquad (9)$$

At first glance, this would appear to be problematic because both W and $\widetilde{H}$ are unknown. However, we can find $\widetilde{H}$ by using the condition from linear algebra that in order for a solution of the above equation to exist, $\widetilde{H}-H$ must be orthogonal to the null space of $\mathbf{D}^\dagger$ [9,11]. With $\widetilde{H}$ known, the solution for W is straightforward.

The solution, W, however, is not unique since W plus any multiple of a v_i satisfying Eq. (5) would result in a solution to Eq. (9). Typically, in order to specify a unique solution, the extra condition is added that W contain no component in the null space of $\mathbf{D}$ [3,8,9,11]. This is equivalent to requiring W to have the smallest possible norm.

The BGNF procedure is most easily carried out in vector form. The direct product space of quadratic polynomials composed of the monomials x_1,p_1,x_2,p_2 is

$$(x_1^2,p_1^2,x_1p_1,x_2^2,p_2^2,x_2p_2,x_1x_2,p_1p_2,x_1p_2,x_2p_1)$$

and has dimension 10. The space of cubic polynomials is,

$$(x_1^3,p_1^3,x_1^2p_1,x_1p_1^2,x_2^3,p_2^3,x_2^2p_2,x_2p_2^2,x_1^2x_2,$$

$$x_1^2p_2,x_1x_2p_1,x_1p_1p_2,x_2p_1^2,p_1^2p_2,$$

$$x_1x_2^2,x_2^2p_1,x_1x_2p_2,x_2p_1p_2,x_1p_2^2,p_1p_2^2)$$

and has dimension 20. Functions which span the space of fourth, fifth, and higher degree polynomials can also be arranged into similar vectors. The general dimension of the vector space of nth degree polynomials in the four variables is

$$\frac{(n+1)(n+2)(n+3)}{6}, \qquad (10)$$

or

$$\frac{(j+3)(j+4)(j+5)}{6}, \qquad (11)$$

for order ϵ^j. The space of eighth degree polynomials, for example, has dimension 165.

The normal operator can also be written in matrix form. This matrix will have the same effect on the vector representation of a polynomial that the derivative form of the operator will have on the algebraic form of the polynomial. The matrix form of $\mathbf{D}$ in the space of quadratic polynomials is

$$\mathbf{D} = \begin{pmatrix} 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 2 & -2 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 2 & -2 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 & -1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 1 & -1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 1 & -1 & 0 & 0 \end{pmatrix}. \tag{12}$$

Similar matricies can be developed for $\mathbf{D}$ in the space of third, fourth, fifth, and higher degree polynomials. Note that the $\mathbf{D}$ operator is block diagonal. These blocks correspond to subspaces of the vectors of polynomials and can be divided up according to the sum of the degrees of the monomials with like subscripts [11]. Consider a polynomial of the form

$$x_1^a x_2^b p_1^c p_2^d. \tag{13}$$

In the matrix representation of the $\mathbf{D}$ operator in the space of second degree polynomials shown above, the first block (upper left) involves polynomials where $a+c=2$ and $b+d=0$. The second (middle) block involves polynomials where $a+c=0$ and $b+d=2$. The last block (lower right) involves polynomials where $a+c=b+d=1$. This block diagonal property is also present in the $\mathbf{D}$ operator in spaces of higher degree and can be used to improve computational efficiency in the practical application of the BGNF procedure. The maximum dimension of a block of nth degree polynomials is $(n+2)^2/4$ for n even or $(n+3)(n+5)/4$ for n odd [11]. Other block diagonalizations are also possible. One based on symmetry may prove particularly fruitful.

We used the MATHEMATICA [14] algebraic manipulation software in this investigation. The full procedure for the transformation is as follows.

(i) Construct $\mathbf{D}$ and $\mathbf{D}^\dagger$ in the space of polynomials of degree $j+2$ and find their respective null spaces v_i and w_i. ($\mathbf{D}v_i=0$ and $\mathbf{D}^\dagger w_i=0$).

(ii) Solve for $\widetilde{H}$ using the conditions that $\widetilde{H}-H$ must be orthogonal to the null space of $\mathbf{D}^\dagger$ [i.e., $(\widetilde{H}-H)\cdot w_i=0 \forall i$], and that $\widetilde{H}$ must be a linear combination of the v_i. This gives a set of linear equations for the coefficients of the v_i.

(iii) Solve $\mathbf{D}W=\widetilde{H}-H$ for W making $W\cdot v_i=0$ to minimize the norm of W.

(iv) Use the generating function S to develop the transformation rules.

(v) Apply the transformation rules to the untransformed Hamiltonian (or to the Hamiltonian resulting from the previous transformation). Since the right-hand sides of Eqs. (8) involve old variables as well as new variables, this must be done recursively.

(vi) Repeat the previous steps for the next j.

Each iteration will transform the Hamiltonian to the desired ''normal'' form ($\widetilde{H}=\Sigma_i a_i v_i$) to one higher power in ϵ. Once can use accelerated convergence techniques to normalize to order ϵ^{2n} in the nth transformation [15], or use Lie perturbation theory to obtain transformation equations which avoid the recursive substitutions [16–18], but the issues discussed below in Sec. III appear in these variations also.

The Hénon-Heiles Hamiltonian system has been the subject of considerable study over the past three decades, predominantly because it is one of the simplest Hamiltonian systems to exhibit regular *and* irregular motions and is a prototype for nonintegrable coupled oscillator systems [4,9–11,19–22]. The form of the Hamiltonian is

$$H = \left(\frac{p_x^2}{2} + \frac{x^2}{2} \right) + \left(\frac{p_y^2}{2} + \frac{y^2}{2} \right) + \epsilon \left(x^2 y - \frac{y^3}{3} \right). \tag{14}$$

While the Hénon-Heiles Hamiltonian is well known to be nonintegrable, extensive numerical experiments have shown that a considerable fraction of phase space is dense with regular trajectories, particularly at low energy [3,7,9,11]. The results of these numerical experiments then suggest that the Hénon-Heiles system might be well approximated by an integrable Hamiltonian, particularly at low energies. This property makes the Hénon-Heiles Hamiltonian system ideal for studying approximate Hamiltonians. The BGNF procedure has been used to develop an approximate Hamiltonian for the Hénon-Heiles system several times previously [3,8,9,11]. The form of the transformed BGNF Hamiltonian through order ϵ^6 is given in Refs. [3,10,15] and is available from the authors through order ϵ^{10}.

III. DISCUSSION OF SYMMETRY

Note that the zeroth-order Hamiltonian, H_2, has spherical symmetry. The cubic terms in the Hénon-Heiles Hamiltonian introduce a threefold rotational symmetry like that of an equilateral triangle. This threefold symmetry is easily seen when the cubic term is written in polar coordinates [4] where it takes the form

$$H'(r,\theta) = \left[\frac{r^3}{3} \sin(3\theta) \right]. \tag{15}$$

This means that the Hamiltonian is invariant under certain symmetry operations, specifically, reflection through the y axis, time reversal, rotation by $2\pi/3$, and their products [23]. We will represent these operations with the operators, $\mathbf{R}$, $\mathbf{T}$, and $\mathbf{O}$, respectively.

$$\mathbf{R} \Rightarrow (x \rightarrow -x, p_x \rightarrow -p_x), \tag{16}$$

$$\mathbf{T} \Rightarrow (p_x \rightarrow -p_x, p_y \rightarrow -p_y), \tag{17}$$

$$\mathbf{O} \Rightarrow \left(x \rightarrow \frac{-x}{2} - \frac{3^{1/2}y}{2}, y \rightarrow \frac{3^{1/2}x}{2} - \frac{y}{2}, p_x \rightarrow \frac{-p_x}{2} - \frac{3^{1/2}p_y}{2}, p_y \rightarrow \frac{3^{1/2}p_x}{2} - \frac{p_y}{2} \right). \tag{18}$$

In addition to those above, there are the following: rotation by $(4\pi/3)$ ($\mathbf{O}^2$), reflection through the line $y = (x/2)$ ($\mathbf{R}' = \mathbf{O}\mathbf{R}$), reflection through the line $y = -(x/2)$ ($\mathbf{R}'' = \mathbf{O}^2\mathbf{R}$), $\mathbf{TO}$, $\mathbf{TO}^2$, $\mathbf{TR}$, $\mathbf{TR}'$, and $\mathbf{TR}''$. Of course there is also the trivial identity operation $\mathbf{E}$. These 12 operations form a group which is the direct product of the C_{3v} point group with $\mathbf{T}$ and is isomorphic to D_{3d} or D_{3h}. Note that $\mathbf{R}$, $\mathbf{T}$, and $\mathbf{O}$ generate all group members. It is easily verified that

$$\mathbf{R}H = H \tag{19}$$

and that similar equalities hold for the other operators.

It is easily demonstrated, however, that $\mathbf{O}\widetilde{H} \neq \widetilde{H}$. This means that the approximate Hamiltonian $\widetilde{H}$ does not possess the same symmetry properties as the Hénon-Heiles system when the standard BGNF procedure is applied. This is readily seen in Eq. (20) of Ref. [3]. If the threefold rotational symmetry was maintained, the terms proportional to $\cos 2\theta_2$ and $\cos 4\theta_2$ would vanish.

We have noticed [7] that in the BGNF procedure, there is some flexibility in the choice of W. Note that in the solution of $\mathbf{D}W = \widetilde{H} - H$ for W, we can add any linear combination of the null-space vectors of $\mathbf{D}$ to W and still have a valid solution to the equation. This flexibility only exists for j even. For j odd, there are no null-space vectors of $\mathbf{D}$, and W is unique. Note that for polynomial degree $j + 2$ (order j in ϵ) there are M null-space vectors of $\mathbf{D}$, where $M = (j+4)^2/4$ for j even, and $M = 0$ for j odd. Since $\mathbf{D}$ only involves H_2, it conserves only the full spherical symmetry. In order to satisfy the reduced symmetry which comes from the higher-order terms in H, additional symmetry constraints must be imposed in the BGNF procedure artificially. We have therefore defined an extended W which we will denote

$$\mathcal{W} = W + \sum_i c_i v_i. \tag{20}$$

$\mathcal{W}$ is used in place of W in the generating function S from which the transformation rules are derived. After the transformation rules are applied, the transformed Hamiltonian contains the unknown coefficients c_i. We then determine the unknown coefficients c_i in such a way that the symmetry of the Hénon-Heiles system is preserved in the approximate Hamiltonian which we will now denote $\widetilde{H}_s$. This is accomplished by choosing the c_i coefficients so that the following equalities hold at order ϵ^{j+2}:

$$\mathbf{R}\widetilde{H}_s - \widetilde{H}_s = 0, \tag{21}$$

$$\mathbf{T}\widetilde{H}_s - \widetilde{H}_s = 0, \tag{22}$$

$$\mathbf{O}\widetilde{H}_s - \widetilde{H}_s = 0. \tag{23}$$

The other nine operators do not introduce any additional constraints since they can be expressed as products of these three. Since $\widetilde{H}_s$ is actually determined *before* $\mathcal{W}$ at a given order, the undetermined c_i coefficients which were introduced by the transformation vector $\mathcal{W}$ at order j are set at order $j + 2$, making this a "mixed-order" procedure.

There are no linear terms in the Hénon-Heiles Hamiltonian, and so no $\mathcal{W}_1$ transformation is required. The second-degree part of the Hénon-Heiles Hamiltonian is already in normal form, and therefore the $\mathcal{W}_2$ transformation is also unnecessary. The $\mathcal{W}_3$ transformation introduces no undetermined coefficients because $\mathbf{D}_3$ has no null-space vectors. The $\mathcal{W}_4$ transformation vector introduces nine undetermined coefficients because the dimension of the null space of $\mathbf{D}_4$ is nine. These undetermined coefficients first appear in terms of the transformed Hamiltonian having degree six. The $\mathcal{W}_5$ transformation does not affect the terms of degree six and so the undetermined c_i coefficients from the $\mathcal{W}_4$ transformation remain unspecified until it is necessary to find $\widetilde{H}_s$ for the sixth degree terms. The above symmetry equations uniquely determine eight of the nine coefficients. The remaining coefficient does not appear in $\widetilde{H}_s$ but does appear in the untransformed terms of degree six. (These are the terms of degree six following the first three iterations of the transformation procedure, using $\mathcal{W}_3$, $\mathcal{W}_4$, and $\mathcal{W}_5$.) In order to proceed with step (iii) in the transformation procedure in the space of sixth degree terms, we have found it necessary to give this remaining coefficient a numerical value [24] and we chose zero for simplicity since its value is irrelevant to the symmetry of the transformed Hamiltonian. It is then possible to solve for $\mathcal{W}_6$, which introduces 16 new undetermined coefficients. (The dimension of the null space of $\mathbf{D}_6$ is 16.) These coefficients first appear in terms of the transformed Hamiltonian of degree eight. The $\mathcal{W}_7$ transformation does not affect the terms of degree eight. We found that 15 of the 16 undetermined coefficients are determined uniquely by the symmetry equations (21)–(23). The remaining coefficient

does not appear in $\widetilde{H}_s$ but does appear in the untransformed terms of degree eight. Since we did not proceed on to higher order, it was unnecessary to set this coefficient. In order to find $\mathcal{W}_8$, however [step (iii) of the transformation proce-

dure], we would find it necessary to give this 16th coefficient a numerical value.

The symmetry-preserved transformed Hamiltonian through terms of degree eight is

$$\widetilde{H}_s = \frac{p_1^2}{2} + \frac{p_2^2}{2} + \frac{x_1^2}{2} + \frac{x_2^2}{2} + \frac{\epsilon^2}{48}(-5p_1^4 - 10p_1^2p_2^2 - 5p_2^4 - 10p_1^2x_1^2 + 18p_2^2x_1^2 - 5x_1^4 - 56p_1p_2x_1x_2 + 18p_1^2x_2^2 - 10p_2^2x_2^2 - 10x_1^2x_2^2$$

$$- 5x_2^4) + \frac{\epsilon^4}{3456}(101p_1^6 - 2721p_1^4p_2^2 + 2319p_1^2p_2^4 - 235p_2^6 + 303p_1^4x_1^2 - 3510p_1^2p_2^2x_1^2 + 219p_2^4x_1^2 + 303p_1^2x_1^4 - 789p_2^2x_1^4$$

$$+ 101x_1^6 - 3864p_1^3p_2x_1x_2 + 4200p_1p_2^3x_1x_2 - 3864p_1p_2x_1^3x_2 - 789p_1^4x_2^2 + 2538p_1^2p_2^2x_2^2 - 705p_2^4x_2^2 - 3510p_1^2x_1^2x_2^2$$

$$+ 2538p_2^2x_1^2x_2^2 - 2721x_1^4x_2^2 + 4200p_1p_2x_1x_2^3 + 219p_1^2x_2^4 - 705p_2^2x_2^4 + 2319x_1^2x_2^4 - 235x_2^6) + \frac{\epsilon^6}{2\,488\,320}(108\,467p_1^8$$

$$- 2\,278\,660p_1^6p_2^2 - 253\,374p_1^4p_2^4 + 1\,940\,828p_1^2p_2^6 - 192\,925p_2^8 + 433\,868p_1^6x_1^2 - 6\,862\,356p_1^4p_2^2x_1^2 - 4\,176\,204p_1^2p_2^4x_1^2$$

$$- 1\,702\,252p_2^6x_1^2 + 650\,802p_1^4x_1^4 - 6\,888\,732p_1^2p_2^2x_1^4 + 5\,290\,850p_2^4x_1^4 + 433\,868p_1^2x_1^6 - 2\,305\,036p_2^2x_1^6 + 108\,467x_1^8$$

$$+ 52\,752p_1^5p_2x_1x_2 + 7\,338\,912p_1^3p_2^3x_1x_2 + 7\,286\,160p_1p_2^5x_1x_2 + 105\,504p_1^3p_2x_1^3x_2 - 29\,515\,808p_1p_2^3x_1^3x_2$$

$$+ 52\,752p_1p_2x_1^5x_2 - 2\,305\,036p_1^6x_2^2 - 4\,176\,204p_1^4p_2^2x_2^2 + 2\,179\,404p_1^2p_2^4x_2^2 - 771\,700p_2^6x_2^2 - 6\,888\,732p_1^4x_1^2x_2^2$$

$$+ 46\,929\,672p_1^2p_2^2x_1^2x_2^2 - 1\,463\,676p_2^4x_1^2x_2^2 - 6\,862\,356p_1^2x_1^4x_2^2 - 4\,176\,204p_2^2x_1^4x_2^2 - 2\,278\,660x_1^6x_2^2$$

$$- 29\,515\,808p_1^3p_2x_1x_2^3 + 14\,572\,320p_1p_2^3x_1x_2^3 + 7\,338\,912p_1p_2x_1^3x_2^3 + 5\,290\,850p_1^4x_2^4 - 1\,463\,676p_1^2p_2^2x_2^4$$

$$- 1\,157\,550p_2^4x_2^4 - 4\,176\,204p_1^2x_1^2x_2^4 + 2\,179\,404p_2^2x_1^2x_2^4 - 253\,374x_1^4x_2^4 + 7\,286\,160p_1p_2x_1x_2^5 - 1\,702\,252p_1^2x_2^6$$

$$- 771\,700p_2^2x_2^6 + 1\,940\,828x_1^2x_2^6 - 192\,925x_2^8). \tag{24}$$

Interestingly, we have found that the same result for $\widetilde{H}_{s8}$ can be found by taking symmetry-adapted linear combinations of the terms of degree eight produced by Jaffé and Reinhardt [3]. (Through $\widetilde{H}_6$ Jaffé's transformed Hamiltonian is identical to ours.) with the standard BGNF procedure. The appropriate linear combination is given by

$$\widetilde{H}_{s8} = \frac{1}{12}\sum_{N=1}^{12} \boldsymbol{\sigma}_N \widetilde{H}_8. \tag{25}$$

Here the index on the summation N runs over the 12 operators in the symmetry group, the $\boldsymbol{\sigma}_N$ being the 12 operators.

It is straightforward to demonstrate that the equalities (21)–(23) hold for our $\widetilde{H}_s$ at all orders but not for the $\widetilde{H}$ of Jaffé and Reinhardt. [see Eq. (20) of Ref. [3].] We conclude that the flexibility in W can be used to impose certain symmetry restrictions on the transformed Hamiltonian $\widetilde{H}$ *in the transformed coordinate system.*

While it may be desirable (for quantization, for example) to constrain the transformed Hamiltonian to have the same symmetry properties as the original Hamiltonian, there are other constraints which could lead to a different choice for

the undetermined coefficients c_i in $\mathcal{W}$ which might be required for purposes different than those considered here.

Consider that the variables x_1, x_2, p_1, p_2 upon which the original Hamiltonian depends define the axes of an orthogonal rectilinear coordinate system in a four-dimensional space. The new transformed variables, X_1, X_2, P_1, P_2 define the axes of a new coordinate system which is curvilinear with respect to the untransformed coordinates (although the two coordinate systems become identical near the phase space origin). The exact nature of this distortion is contained in the transformation rules (8). We have demonstrated how to constrain the transformed Hamiltonian in the transformed coordinates to obey the same mathematical symmetry operators that the original Hamiltonian does in the untransformed coordinates.

In related work, Finkler, Jones, and Sowell [23] have sought to find an approximate constant of the motion, K, independent of the energy for the Hénon-Heiles system. Their procedure employs a power series with unknown coefficients for the desired conserved quantity, K. To determine the coefficients, they require the Poisson bracket of K with the Hamiltonian to be zero, $\{K,H\}=0$. Some arbitrariness in the determination of the coefficients is removed by using the symmetries of the Hénon-Heiles Hamiltonian. Finkler, Jones, and Sowell [25] note that if the nonuniqueness is

removed in some arbitrary manner at a given order, the equations which specify the expansion coefficients of K at higher order will in general have no solution. This raises some concern about our procedure. Recall that the nine unspecified constants introduced by $\mathcal{W}_4$ are set by applying the symmetry conditions to $\widetilde{H}_6$. These symmetry constraints only specify eight of the nine coefficients, however. Since the ninth coefficient does not appear in $\widetilde{H}_6$, we set it to zero for simplicity. We do so because it is practically (though not mathematically) necessary to pick a particular numerical value for this coefficient in order to utilize $\mathcal{W}_6$. It is possible that in making this somewhat arbitrary choice for the remaining unspecified coefficient, we may be preventing the preservation of symmetry at some higher order, although no such problem was encountered through terms of degree eight. We assume that if this occurs, some value of the coefficient can be found to satisfy the latter condition. We always found adequate flexibility in the transformed Hamiltonian to preserve the desired symmetry properties to the order that we have extended our work.

The Finkler, Jones, and Sowell [23,25] procedure does not involve any transformation of the coordinate system and therefore there is no question of the symmetry properties in a transformed representation. The resulting constant of the motion K is expressed in the same variables as the original Hamiltonian. Unfortunately, their procedure does not yield an integrable approximate Hamiltonian and so no advantage is gained for quantization. It is also interesting to note that the Finkler, Jones, and Sowell [23,25] procedure is a mixed-order procedure like ours.

The advantage of the BGNF procedure is that it yields not only an approximate Hamiltonian but also an approximate constant of the motion. In the transformed coordinates, this quantity is typically chosen using Eq. (6) [9,11],

$$\widetilde{I} = \widetilde{H} - \widetilde{H}_2. \tag{26}$$

Other choices are possible. The constant of motion, I, in original untransformed variables can be generated from $\widetilde{I}$ by applying to $\widetilde{I}$ the reverse of the transformations used to find $\widetilde{H}$ from H. To determine the reverse transformation rules, Eqs. (8) are simply reversed to give the new transformed variables in terms of the old untransformed variables instead of vice versa. We have made such an inversion on

$$\widetilde{I}_s = \widetilde{H}_s - \widetilde{H}_2. \tag{27}$$

Interestingly, the approximate constant of the motion in untransformed variables, I_s, does not satisfy the symmetry operations of the Hénon-Heiles system (21)–(23), however.

Through terms of degree six, our transformed Hamiltonian, $\widetilde{H}_s$, and approximate constant of motion in untransformed variables, I_s, are identical to those of Jaffé and Reinhardt [3]. A comparison of our I_s in untransformed coordinates to the constant K found by Finkler, Jones, and

Sowell can therefore be found in Table I of Ref. [25], where they make a comparison with the I of Gustavson [9] through terms of degree six [26]. (Note that Gustavson's Hamiltonian and constant of motion are the same as those found by Jaffé and Reinhardt [3], the only difference being that the coefficients were found numerically in Gustavson's work and algebraically in the work of Jaffé and Reinhardt [3].) Apparently, constraining the symmetry of the approximate Hamiltonian in the transformed coordinates does *not* lead to the same approximate constant of the motion as the procedure of Finkler, Jones, and Sowell [23,25]. While one coefficient in $\mathcal{W}_4$ was set in a somewhat arbitrary way, this can only affect terms of degree $\geqslant 7$. Since our $\widetilde{H}_s$ is unique through terms of degree six, we see that determining a symmetry correct approximate constant of the motion in the untransformed variables and generating a symmetry correct approximate Hamiltonian in the transformed variables are incompatible goals.

It might be possible to invert the transformation procedure before specifying the undetermined coefficients c_i in $\widetilde{H}$. These unspecified constants could then be chosen *after* the reverse transformation in such a way that I possessed the same symmetry properties as the Hamiltonian. In this case, I would presumably have to be identical to the expression for I found by Finkler, Jones, and Sowell. While conceptually reasonable, we have not been able to perform the reverse transformation on $\widetilde{H}$ before specifying the undetermined coefficients.

One can argue that the ''correct'' transformed Hamiltonian is the one which produces an untransformed approximate constant of the motion I which possesses the same symmetry properties as the original Hamiltonian. Certainly a ''true'' constant of the motion must possess the same symmetry properties as the Hamiltonian system to which it belongs. It is important to note, however, that such an approximate Hamiltonian, when quantized, may not give the proper degeneracies for the eigenvalues of the system which it describes because it may not have the correct symmetries. In the case considered here, the Hénon-Heiles system, the approximate Hamiltonian in transformed variables $\widetilde{H}_s$ which possesses the same symmetry properties as the Hénon-Heiles system does not have an associated untransformed approximate constant of the motion which is symmetry correct.

The procedures demonstrated here for the two-dimensional Hénon-Heiles Hamiltonian can be used on the more interesting Hamiltonians describing molecular vibrations. For example, ammonia has a vibrational potential in which two oscillators are degenerate, methane has a vibrational potential with twofold and threefold degeneracies, and Buckminsterfullerene (C_{60}) has a vibrational potential with threefold, fourfold, and fivefold degeneracies. In all of these cases, the molecular Hamiltonian will therefore have a high degree of symmetry. Thus, a symmetry-adapted BGNF for the Hamiltonian should provide a description of the system which can be quantized semiclassically to produce information about the high energy spectrum of molecules which are not accessible by quantum basis set methods.

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