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Mixing quantum and classical mechanics

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Quantum-classical mixing is studied by a group-theoretical approach, and a quantum-classical equation of motion is derived. The quantum-classical bracket entering the equation preserves the Lie algebra structure of quantum and classical mechanics, and, therefore, leads to a natural description of interaction between quantum and classical degrees of freedom. The exact formalism is applied to coupled quantum and classical oscillators. Various approximations, such as the mean-field and the multiconfiguration mean-field approaches, which are of great utility in studying realistic multidimensional systems, are derived. Based on the formulation, a natural classification of the previously suggested quantum-classical equations of motion arises, and several problems from earlier works are resolved. [S1050-2947(97)03507-5]

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I. INTRODUCTION

Many phenomena in nature are described by quantum mechanics at a fundamental level and with high precision. Yet, there exist numerous situations where mixed quantum-classical models are needed. In some cases the phenomena are too complex to allow for a fully quantum approach, in others a consistent quantum theory is lacking. Classical mechanics often provides a more suggestive description and a clearer picture of physical events. Applications of various quantum-classical approaches range from biochemical and condensed-matter chemical reactions, where the large dimensionality of the systems of interest requires approximations, to the evolution of the Universe and cosmology, where no theory of quantum gravity has been established.

The issue of treating quantum and classical degrees of freedom within the same formalism has been discussed recently in a number of publications [1–10]. The interest was spurred [11] by the cosmological problem of defining the backreaction of quantum matter fields on the classical spacetime background, where classical variables should be independently correlated with each individual quantum state. The traditional quantum-classical mean-field approach fails the last requirement and was generalized in Ref. [12]. (For a fully quantum approach to cosmology see Ref. [13].) Earlier a similar situation was encountered in chemical physics, where quantum-classical trajectory methods were employed to simulate gas-phase scattering phenomena [14–21] and, later, chemical dynamics on surfaces [22] and in liquids [23–27]. It was noticed in these studies that asymptotically distinct quantum evolutions should correlate with different classical trajectories.

The first relationship between quantum and classical variables is due to Ehrenfest [28] who showed that the equation of motion for the average values of quantum observables coincides with the corresponding classical expression. (Surprisingly, the first mathematically rigorous treatment on the subject was not carried out until as late as 1974, see Ref. [29].) Ehrenfest's result leads to the mean-field approach, where classical dynamics is coupled to the evolution of the expectation values of quantum variables [30–33]. The mean-field equations of motion possess all of the properties of the purely classical equations and are rigorous insofaras the mean values of quantum operators are concerned. However, an expectation value does not provide information about the outcome of an individual process. The mean-field approach gives a satisfactory description of the classical subsystem as long as changes within the quantum part are fast compared to the characteristic classical time scale. If classical trajectories depend strongly on a particular realization of the quantum evolution, the mean-field approximation is inadequate. The problem can be corrected, for instance, by the introduction of stochastic quantum hops between preferred basis states, which define classical potential energy surfaces, with probabilities determined by the usual quantum-mechanical rules [34–36]. The decoherent histories interpretation of quantum mechanics [37,38] formulated on the level of individual histories [39,40] establishes a theoretical foundation of the surface hopping technique [41].

Similarity between the algebraic structures underlying quantum and classical mechanics provides a consistent way of improving upon the mean-field approximation, as explored in Refs. [2,12,15], which aim to derive a quantum-classical bracket that reduces to the quantum commutator and the Poisson bracket in the purely quantum and classical cases. In addition to the reduction property the bracket should satisfy other criteria so as to produce physically meaningful quantum-classical evolutions. For example, an antisymmetric bracket conserves the total energy.

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Recently, Kisil proposed a mathematical construction that naturally envelops classical and quantum mechanics and is named here p mechanics [42]. Formulated within the framework of operator algebras, the p-mechanical equation of motion reduces to the appropriate quantum or classical equations under suitable representations of the algebra of observables. In this paper we extend the ideas of p mechanics to incorporate mixed quantum-classical descriptions. We derive the quantum-classical bracket and explicitly show that it satisfies the properties common to quantum and classical mechanics. The technique described in this paper allows one to construct families of mixed quantum-classical approaches, each having a specific set of properties. Focusing on the simplest among such families we investigate the relationship between the quantum-classical equations of motions proposed by other authors [2,12,15].

The format of this paper is as follows: In Sec. II we summarize p mechanics and introduce the essential mathematical definitions. In Sec. III we construct the simplest p-mechanical model that adopts two distinct sets of variables associated with quantum and classical degrees of freedom. By taking an appropriate representation for the model we derive the quantum-classical bracket, show that it is antisymmetric, and discuss the Jacobi identity. The procedure presented in Sec. III allows us to discriminate between the very similar brackets of Refs. [12] and [15], and to obtain the antisymmetric analog of the bracket of Ref. [2]. In Sec. IV we consider coupled classical and quantum harmonic oscillators and illustrate how the dynamics of a mixed quantum-classical system can be studied by p mechanics without explicit reference to an equation of motion. In Sec. V we investigate various approximations to the general quantum-classical description, including the mean-field and the multiconfiguration mean-field approaches. In the concluding section we classify the quantum-classical equations of motion from earlier papers according to the present formalism and summarize our results.

II. *P* MECHANICS

A. The elements of *P* mechanics

We recall the constructions from Refs. [42,43] and introduce appropriate modifications.

Definition 1. An operator algebra $\mathfrak{P}$ gives a p-mechanical description [42] of a system if the following conditions hold.

(1.1) The set $\hat{\mathfrak{P}}$ of all irreducible representations π_h of $\mathfrak{P}$ is a disjoint union of subsets $\hat{\mathfrak{P}}=\cup_{p\in P}\hat{\mathfrak{P}}_p$ parametrized by the elements of a set P. The elements of the set P are associated with different values for the Planck constant. We refer to this set as the *set of Planck constants*. If for p_0 the set $\hat{\mathfrak{P}}_{p_0}$ consists of only commutative (and, therefore, one-dimensional) representations, then $\hat{\mathfrak{P}}_{p_0}$ gives a *classical* description. If $\hat{\mathfrak{P}}_{p_0}=\{\pi_{p_0}\}$ consists of a single noncommutative representation π_{p_0}, then $\hat{\mathfrak{P}}_{p_0}$ gives a purely *quantum* model. Sets $\hat{\mathfrak{P}}_p$ of other types provide *mixed* (quantum-classical) descriptions.

(1.2) Let $\hat{\mathfrak{P}}$ be equipped with a natural operator topology (for example, it may be the Jacobson topology [44] or the $*$-bundle topology [45,46]). Then P has a natural *factor topology* induced by the partition $\hat{\mathfrak{P}}=\cup_{p\in P}$.

(1.3) (Dynamics): the algebra $\mathfrak{P}$ is equipped with the one-parameter semigroup of transformations $G(t)$: $\mathfrak{P}\to\mathfrak{P}, t\in\mathbb{R}^+$. All sets $\hat{\mathfrak{P}}_p$, $p\in P$ are *preserved* by $G(t)$. Namely, for any $\pi\in\hat{\mathfrak{P}}_p$ all new representations $\pi_t=\pi\circ G(t)$ again belong to $\hat{\mathfrak{P}}_p$.

(1.4) (The correspondence principle): let S: $p\mapsto S(p)\in\mathfrak{P}_p$ be an operator-valued section, which is continuous in the $*$-bundle topology [45,46] over P. Then for any t, i.e., at any moment of time, the image $S_t(p)=G(t)S(p)$ is also a section due to statement (3). In the $*$-bundle topology the sections $S_t(p)$ are *continuous for all t*.

The above conditions are general. Next, we describe an important particular case of *group quantization* [43]. All components of p mechanics (operator algebra, partition of representations, topology) readily arise there.

Construction 2. Group quantization comprises the following steps.

(2.1) Let $\Omega=\{x_j\}$, $1\leqslant j\leqslant N$ be a set of physical variables defining the state of a classical system. Classical observables are real-valued functions on the states. The best known and the most important case is the set $\{x_j=q_j,\ x_{j+n}=p_j\}$, $1\leqslant j\leqslant n$, $N=2n$ of coordinates and momenta of n classical particles. The observables are real-valued functions on $\mathbb{R}^{2n}$. We will use this example throughout this section.

(2.2) We complete the set Ω with additional variables x_j, $N<j\leqslant\bar{N}$, such that the new set $\bar{\Omega}$ forms the smallest algebra, which contains Ω and is closed under the Poisson bracket

$$\{x_i,x_j\}\in\bar{\Omega},\quad \text{for all } x_i,x_j\in\bar{\Omega}.$$

In the above example we add the unit function $x_{2n+1}=1$. The complete set contains $\bar{N}=2n+1$ elements satisfying the famous relations

$$\{x_j,x_{j+n}\}=-\{x_{j+n},x_j\}=x_{2n+1}. \tag{1}$$

All other Poisson brackets are zero.

(2.3) We form an $\bar{N}$-dimensional Lie algebra $\mathfrak{p}$ with the frame $\{\hat{x}_j\}$, $1\leqslant j\leqslant\bar{N}$ defined by the formal mapping $\hat{}$: $x_j\mapsto\hat{x}_j$. The commutators of the frame vectors are formally defined by the formula

$$[\hat{x}_i,\hat{x}_j]=\widehat{\{x_i,x_j\}}. \tag{2}$$

We extend the commutator onto the whole algebra by linearity.

For our example, $\mathfrak{p}$ is the Lie algebra corresponding to the Heisenberg group (see the next subsection for details).

(2.4) We introduce the algebra $\mathfrak{P}$ of convolutions induced by $\mathfrak{p}$. The convolution operators are *observables* in the group quantization, and by analogy with the classical case they can be treated as functions of $\hat{x}_j$. Particular representations of the convolution algebra in spaces $\mathbb{L}^2(S)$ give different descriptions of a physical system. The family of all one-dimensional representations of $\mathfrak{P}$ corresponds to *classical* mechanics; various noncommutative representations lead to *quantum* and *quantum-classical* descriptions with different *Planck constants*.

For our example the following possibilities exist. (a) $S=\mathbb{R}^n$, $\hat{x}_j=X_j=M_{q_j}$, $\hat{x}_{j+n}=-i\hbar\partial/\partial q_j$, the convolutions are represented by pseudodifferential operators (PDO), and we obtain the *Dirac-Heisenberg-Schrödinger-Weyl quantization* by PDO. (b) $S=\mathbb{R}^{2n}$, $\hat{x}_j=X_j=M_{q_j}$, $\hat{x}_{j+n}=M_{p_j}$, the convolutions are represented by (operators of multiplication by) functions, and we obtain the classical description that we started from.

It is an empirical observation that the steps above lead to a nilpotent Lie group, with the dual $\hat{Z}$ of the center Z of the group interpreted as the set of Planck constants. Now we illustrate this fact by a well-known example of quantization, and later in Sec. III by constructing a quantum-classical model.

B. The Heisenberg group generates quantum and classical mechanics

In the previous subsection we claimed that the nth-order Heisenberg group $\mathbb{H}^n$ describes a set of n quantum particles. Here we show how this description is achieved.

$\mathbb{H}^n$ is generated by the n-dimensional translation and multiplication operators $e^{ip\cdot D}$, $e^{iq\cdot X}$, p, $q\in\mathbb{R}^n$ satisfying the Weyl commutator relations

$$e^{2\pi ip\cdot D}e^{2\pi iq\cdot X}=e^{2\pi ip\cdot q}e^{2\pi iq\cdot X}e^{2\pi ip\cdot D}. \tag{3}$$

An element of the Heisenberg group $g\in\mathbb{H}^n$ is defined by $2n+1$ real numbers (p,q,s), p, $q\in\mathbb{R}^n$, $s\in\mathbb{R}$. The composition of two elements g and g' is given by

$$(p,q,s)(p',q',s')=[p+p',q+q',s+s'+\tfrac{1}{2}(pq'-p'q)].$$

D_j, X_j, and I form a $(2n+1)$-dimensional basis of the Heisenberg algebra $\mathfrak{h}^n$ with a one-dimensional center $\mathbb{Z}=\{sI;s\in\mathbb{R}\}$. Since all second- and higher-order commutators of the basis elements vanish, $\mathbb{H}^n$ and $\mathfrak{h}^n$ are step-two nilpotent Lie group and algebra, respectively.

The unitary irreducible representations of the Heisenberg group are classified by the Stone–von Neumann theorem [47]. They are parametrized by a real number h, the character of the one-dimensional center $\mathbb{Z}$. A nonzero h gives noncommutative unitary representations acting on the Hilbert space $L^2(\mathbb{R}^n)$:

$$\rho_{h\neq 0}(p,q,s)=e^{2\pi i(p\cdot hD+q\cdot X+s\cdot hI)}. \tag{4}$$

The n components of X and hD are the usual quantum-mechanical position X_j (multiplication by x_j) and momentum hD_j ($h/2\pi i$ times differentiation with respect to x_j) operators characterized by the Heisenberg commutator relation

$$[hD_j,X_k]=\delta_{jk}\frac{h}{2\pi i}I. \tag{5}$$

In the limit of zero h the center $\mathbb{Z}$ of the Heisenberg group vanishes, and $\mathbb{H}^n$ becomes isomorphic to $\mathbb{R}^{2n}$. The irreducible representations of the latter are homomorphisms from $\mathbb{R}^{2n}$ into the circle group acting on $\mathbb{C}$

$$\rho_{h=0}(p,q)=e^{2\pi i(pk+qx)}. \tag{6}$$

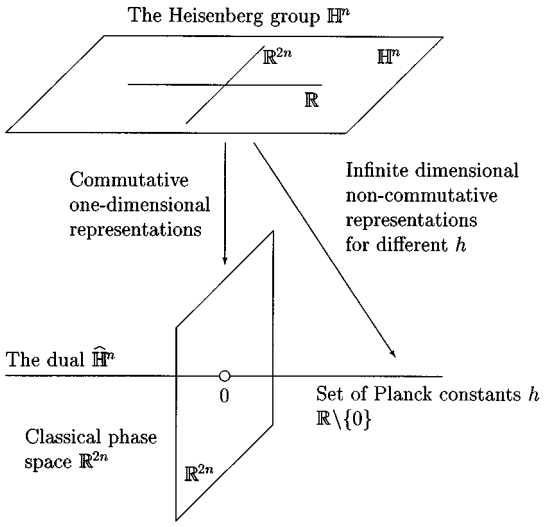

FIG. 1. The Heisenberg group and its dual.

The dual $\hat{\mathbb{H}}^n$ as a set is equal to $\{\mathbb{R}\backslash 0\}\cup\mathbb{R}^{2n}$ (see Fig. 1). It has the natural topology coinciding on $\{\mathbb{R}\backslash 0\}$ with the Euclidean topology. Any sequence of representations $\{\rho_{h_j}\}$, $h_j\to 0$, $h_j\neq 0$ is dense in whole $\mathbb{R}^{2n}$. The last property is fundamental for the correspondence principle.

The unitary representations of $\mathbb{H}^n$ can be extended to the convolution algebra $L^1(\mathbb{H}^n)$. Namely, if $A\in L^1(\mathbb{H}^n)$, then it defines a convolution on the Heisenberg group:

$$Ab(g)=\int_{\mathbb{H}^n}A(g')b(g\circ g')dg'.$$

The representation ρ_h maps the convolution to the operator

$$\rho_h(A)=\int_{\mathbb{H}^n}A(g)\rho_h(g)dg$$
$$=\int\int\int A(p,q,s)\rho_h(p,q,s)dp\,dq\,ds. \tag{7}$$

The p-mechanical equation of motion (see [42] for details) for an element $A(g)$ ($g\equiv\{p,q\}$) of the convolution algebra is defined by

$$\frac{\partial A(g)}{\partial t}=2\pi i[H,A](g) \tag{8}$$

with

$$[H,A](g)=\int_{\mathbb{H}^n}[H(g')A(g'\circ g)-A(g')H(g'\circ g)]dg',$$

where $H(g)$ is the Hamiltonian. The noncommutative unitary representations of Eq. (4) reduce this equation to the Heisenberg equation of motion for operators acting on the Hilbert space $L^2(\mathbb{R}^n)$. Under the commutative representations of Eq. (6) the p-mechanical equation of motion becomes the Hamilton equation for functions on the phase space $\mathbb{R}^{2n}$.

We consider the last statement in more detail by means of the pseudodifferential calculus, which is directly related to

the above group-theoretical developments and the problem of quantization. The noncommutative unitary representations of the Heisenberg group allow one to define integral operators corresponding to functions on phase space. Given a function $\sigma(k,x)$ on $\mathbb{R}^{2n}$ one obtains the operator $\sigma(D,X)$ on $L^2(\mathbb{R}^n)$ by the formula

$$\begin{aligned}\sigma(hD,X) &= \int_{\mathbb{H}^n} \mathcal{F}^{-1}[\sigma](g)\rho_{h\neq 0}(g)dg \\ &= \int\int \mathcal{F}^{-1}[\sigma](p,q)e^{2\pi i(p\cdot hD+q\cdot X)}dp\ dq,\end{aligned} \tag{9}$$

where $\mathcal{F}^{-1}[\ ]$ denotes the inverse Fourier transform, and the trivial integration over s has been carried out. The action of the operator $\sigma(D,X)$ on a function $f(x)\in L^2(\mathbb{R}^n)$ follows from the definition of hD and X [see Eq. (4) and the related paragraph], and is given by

$$\begin{aligned}\sigma(hD,X)f(x) &= \int\int \mathcal{F}^{-1}[\sigma](p,q)e^{\pi ihpq+2\pi iqx} \\ &\quad\times f(x+hp)dp\ dq \\ &= h^{-n}\int\int \mathcal{F}^{-1}[\sigma]\left(\frac{y-x}{h},q\right) \\ &\quad\times e^{\pi iq(x+y)}f(y)dy\ dq \\ &= h^{-n}\int\int \sigma\left(k,\frac{x+y}{2}\right)e^{2\pi i(x-y)k/h}f(y)dydk\end{aligned} \tag{10}$$

or

$$\begin{aligned}\sigma(hD,X)f(x) &= \int K_\sigma(x,y)f(y)dy, \\ K_\sigma(x,y) &= h^{-n}\int \sigma\left(k,\frac{x+y}{2}\right)e^{2\pi i(x-y)k/h}dk,\end{aligned} \tag{11}$$

where K_σ is the kernel of the integral operator $\sigma(hD,X)$. In the language of the pseudodifferential calculus the function $\sigma(k,x)$ is called the symbol of the operator $\sigma(hD,X)$. If instead of $\rho_{h\neq 0}$ one uses a commutative representation $\rho_{h=0}$, the transformation of Eq. (9) reduces to identity and we recover the classical observable $\sigma(k,x)$. Equations (9)–(11) are known as the Weyl correspondence principle.

The symbol $\sigma\sharp_h\tau(k,x)$ of the product of two operators $\sigma\sharp_h\tau(hD,X)=\sigma(hD,X)\tau(hD,X)$ can be obtained by application of a noncommutative representation to the convolution on the Heisenberg group [see Eq. (7)] or directly from the Weyl rule. It is given in terms of the symbols of individual operators by

$$\begin{aligned}&\sigma\sharp_h\tau(k,x) \\ &\quad= \left(\frac{2}{h}\right)^{2n}\int\int\int\int \sigma(\zeta,u)\tau(\eta,v) \\ &\qquad\times e^{4\pi i[(x-u)(k-\eta)-(x-v)(k-\zeta)]/h}du\ dv\ d\eta\ d\zeta.\end{aligned} \tag{12}$$

It follows that the noncommutative representations of the Heisenberg group transform the p-mechanical equation of motion (8) into the equation for operators on $L^2(\mathbb{R}^n)$:

$$\frac{\partial}{\partial t}A(hD,X) = \frac{2\pi i}{h}[H,A]_{\sharp_h}(hD,X), \tag{13}$$

where $[H,A]_{\sharp_h}\equiv[H\sharp_hA-A\sharp_hH]$, the operation of taking the product of two symbols $\sharp_h$ is defined by Eq. (12), and the operators $A(hD,X)$ and $[H,A]_{\sharp_h}(hD,X)$ are recovered from their symbols $A(k,x)$ and $[H,A]_{\sharp_h}(k,x)$ by the application of the Weyl transform Eqs. (9)–(11). This is the quantum-mechanical law of motion in the Heisenberg form.

In order to obtain the corresponding classical expression it is useful to cast the product rule of Eq. (12) in the form of an asymptotic expansion in powers of h. The integration over η and ζ and the change of variables $(u-x)/h\to u$, $(v-x)/h\to v$ converts Eq. (12) to

$$\begin{aligned}\sigma\sharp_h\tau(k,x) &= h^{-2n}\int\int \mathcal{F}_1^{-1}[\sigma](v,x+uh) \\ &\quad\times\mathcal{F}_1[\tau](u,x+vh)e^{4\pi i(v-u)k}du\ dv,\end{aligned}$$

where $\mathcal{F}_1$ and $\mathcal{F}_1^{-1}$ denote the Fourier transform and its inverse with respect to the first variable only. Expanding σ and τ in the second variable around x and applying the Fourier inversion formula to each term in the Taylor series we obtain

$$\begin{aligned}\sigma\sharp_h\tau(k,x) &= \sum_{\alpha+\beta\leq\gamma}\frac{(i\pi h)^{\alpha+\beta}(-1)^\alpha}{\alpha!\beta!}D_k^\beta D_x^\alpha\sigma(k,x)D_k^\alpha D_x^\beta\tau(k,x)+O(h^\gamma) \\ &= \sum_{j=0}^{\gamma}\frac{(i\pi h)^j}{j!}[D_{k,\sigma}D_{x,\tau}-D_{k,\tau}D_{x,\sigma}]^j\sigma(k,x)\tau(k,x)+O(h^\gamma),\end{aligned} \tag{14}$$

where the second subscripts σ and τ of D indicate the symbol to be acted upon. The asymptotic expression for the symbol of the commutator of two operators follows from Eq. (14). The even-order terms in the sum cancel out to produce

$$\begin{aligned}[\sigma\sharp_h\tau-\tau\sharp_h\sigma](k,x) &= 2i\sum_{j=0}^{\gamma}\frac{(-1)^j(\pi h)^{2j+1}}{(2j+1)!} \\ &\quad\times[D_{k,\sigma}D_{x,\tau}-D_{k,\tau}D_{x,\sigma}]^{2j+1} \\ &\quad\times\sigma(k,x)\tau(k,x)+O(h^{2\gamma+1}).\end{aligned} \tag{15}$$

The series expansion of the symbol of the commutator Eq. (15) allows one to derive the Poisson bracket as the classical limit of the symbol of the Heisenberg commutator of two quantum operators:

$$\lim_{h\to 0}\frac{2\pi i}{h}[\sigma\sharp_h\tau-\tau\sharp_h\sigma](k,x)=\{\sigma(k,x),\tau(k,x)\}. \tag{16}$$

Since the commutative representations of the Heisenberg group leave symbols of operators unchanged, i.e.,

$$\begin{aligned}\int_{\mathbb{H}^n}\mathcal{F}^{-1}[\sigma](g)\rho_{h=0}(g)dg&=\int\int\mathcal{F}^{-1}[\sigma]\\&\quad\times(p,q)e^{2\pi i(pk+qx)}dp\ dq\\&=\sigma(k,x),\end{aligned}$$

we deduce that under the commutative representations the p-mechanical equation of motion (8) reduces to the Hamilton equation

$$\frac{\partial}{\partial t}A(k,x)=\{H(k,x),A(k,x)\}. \tag{17}$$

In summary, the Heisenberg group contains the exact quantum and classical descriptions of a system of particles and provides the correspondence principle between the descriptions. We refer the reader to Chapters 1 and 2 of Ref. [47] for further information on the subject.

III. THE QUANTUM-CLASSICAL EQUATION OF MOTION

We proceed to derive an equation of motion for a mixed quantum-classical system by considering an appropriate Lie group that will play the role of the Heisenberg group of the standard quantization. The desired group can be constructed based on the following observations. Two distinct sets of variables $\{D,X\}$ and $\{D',X'\}$ should correspond to quantum and classical parts, accordingly. An operator from the first set may or may not commute with an operator from the second set. Each set should have a Planck constant of its own. Then the Planck constant of the second set can approach zero leading to the classical limit for the second subsystem and leaving the first subsystem quantum. ''Planck constants'' arise as characters of the center of a Lie group, therefore, the Lie group should possess a two-dimensional center.

The ''quantum-classical'' group is generated by two sets of variables $\{hD,X\}$ and $\{h'D',X'\}$ satisfying the commutator relations

$$[hD_j,X_k]=-ih\delta_{jk}I,\quad [h'D'_{j'},X'_{k'}]=-ih'\delta_{jk}I',$$
$$1\leqslant j,k\leqslant n;\quad 1\leqslant j',k'\leqslant n'. \tag{18}$$

Other commutators are zero. The group has a two-dimensional center $\mathbb{Z}=\{sI+s'I';s,s'\in\mathbb{R}\}$. The irreducible representations of a nilpotent Lie group are induced by the characters of the center [48]. For the quantum-classical group the characters are

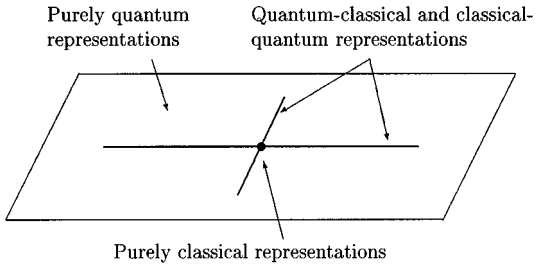

FIG. 2. Representations of the step-two nilpotent Lie group with a two-dimensional center.

$$\mu:(z,z')\mapsto\exp[i(hz+h'z')].$$

It is clear that for $hh'\neq 0$ the induced representation coincides with the irreducible representation of $\mathbb{H}^{n+n'}$ on $L_2(\mathbb{R}^{n+n'})$. This corresponds to purely quantum behavior of both sets of variables (see definition 1.1). The trivial character $h=h'=0$ gives the family of one-dimensional representations parametrized by $\mathbb{R}^{2(n+n')}$ and a purely classical description. These situations were studied in detail in the previous section. A new situation appears where $h\neq 0$ and $h'=0$ producing quantum behavior for the first set and classical behavior for the second set. (The choice $h=0$, $h'\neq 0$ just permutes the quantum and classical parts.) Figure 2 illustrates these facts. In the topology on the dual to the quantum-classical group the quantum descriptions are dense in the quantum-classical and classical descriptions, and the quantum-classical descriptions are dense in the classical ones.

Consider the quantum-classical case in more detail. The quantum-classical representation is given by

$$\rho_h(p,q,s,p',s')=e^{2\pi i(s\cdot hI+p\cdot hD+q\cdot X+p'\cdot k'+q'\cdot x')}. \tag{19}$$

where $k',x'\in\mathbb{R}^{n'}$ and $h\in\mathbb{R}\setminus\{0\}$. In this representation an element of the convolution algebra on the quantum-classical group is identified with a quantum-classical operator acting on $L_2(\mathbb{R}^n)\otimes\mathbb{R}^{2n'}$. The operator can be computed in terms of the Weyl transform of its symbol taken with respect to the quantum (unprimed) coordinates

$$\sigma(hD,X,k',x')f(x)=\int K_\sigma(x,y,k',x')f(y)dy \tag{20}$$

$$K_\sigma(x,y,k',x')=h^{-n}\int\sigma\left(k,\frac{x+y}{2},k',x'\right)e^{2\pi i(x-y)k/h}dk. \tag{21}$$

The quantum-classical analog of the commutator is determined by the limit $h\neq 0$, $h'\to 0$, which is similar to that used to derive the Poisson bracket from the quantum commutator. We proceed as follows. First we need to obtain the expression for the symbol of the product of two operators. We start with the expression analogous to Eq. (12), but having two, rather than one, sets of variables. Focusing on the primed variables, we carry out the transformations identical to those performed in deriving Eq. (16):

$$
\begin{aligned}
&\sigma \sharp_h \tau(k,x,k',x') \\
&= \left(\frac{2}{h}\right)^{2n} \int\int\int\int du\ dv\ d\eta\ d\zeta\ e^{4\pi i[(x-u)(k-\eta)-(x-v)(k-\zeta)]/h} \left(\frac{2}{h'}\right)^{2n'} \int\int\int\int du'\ dv'\ d\eta'\ d\zeta' \\
&\quad \times e^{4\pi i[(x'-u')(k'-\eta')-(x'-v')(k'-\zeta')]/h'} \sigma(\zeta,u,\zeta',u')\,\tau(\eta,v,\eta',v') \\
&= \left(\frac{2}{h}\right)^{2n} \int\int\int\int du\ dv\ d\eta\ d\zeta\ e^{4\pi i[(x-u)(k-\eta)-(x-v)(k-\zeta)]/h} \left(\frac{1}{h'}\right)^{2n'} \int\int du'\ dv' e^{4\pi i(v'-u')k'} \\
&\quad \times \mathcal{F}_3^{-1}[\sigma](\zeta,u,v',x'+u'h')\,\mathcal{F}_3[\tau](\eta,v,u',x'+v'h') \\
&= \left(\frac{2}{h}\right)^{2n} \int\int\int\int du\ dv\ d\eta\ d\zeta\ e^{4\pi i[(x-u)(k-\eta)-(x-v)(k-\zeta)]/h} \sum_{j=0}^{\gamma} \frac{(i\pi h')^j}{j!} \\
&\quad \times [D_{k',\sigma}D_{x',\tau} - D_{k',\tau}D_{x',\sigma}]^j [\sigma(\zeta,u,k',x')\,\tau(\eta,v,k',x')] + O(h'^{\gamma}).
\end{aligned} \tag{22}
$$

At this point we drop explicit dependence of the symbols on the primed variables, since the unprimed variables alone account for the noncommuting nature of the symbols. Applying the limit $\lim_{h'\to 0} h/h'$ behind the zero-order terms, which are kept intact to preserve the purely quantum part, we obtain the expression for the symbol of the quantum-classical commutator:

$$
\begin{aligned}
[\sigma \sharp_h \tau - \tau \sharp_h \sigma](k,x) = \left(\frac{2}{h}\right)^{2n} \int\int\int\int du\ dv\ d\eta\ d\zeta e^{4\pi i[(x-u)(k-\eta)-(x-v)(k-\zeta)]/h} \Bigg\{ & \sigma(\zeta,u)\,\tau(\eta,v) - \tau(\zeta,u)\,\sigma(\eta,v) \\
+ \lim_{h'\to 0} \frac{h}{h'} \sum_{j=0}^{\infty} \frac{(-1)^j(\pi h')^{2j+1}}{(2j+1)!} & [D_{k',\sigma}D_{x',\tau} - D_{k',\tau}D_{x',\sigma}]^{(2j+1)} [\sigma(\zeta,u)\,\tau(\eta,v) - \tau(\zeta,u)\,\sigma(\eta,v)] \Bigg\}
\end{aligned} \tag{23}
$$

or

$$
\begin{aligned}
[\sigma,\tau]_{\sharp h}(k,x) = \left(\frac{2}{h}\right)^{2n} \int\int\int\int du\ dv\ d\eta\ d\zeta e^{4\pi i[(x-u)(k-\eta)-(x-v)(k-\zeta)]/h} \Bigg[& \sigma(\zeta,u)\,\tau(\eta,v) - \tau(\zeta,u)\,\sigma(\eta,v) \\
+ \frac{1}{2}\frac{h}{2\pi i}\left(\frac{\partial\sigma(\zeta,u)}{\partial k'}\frac{\partial\tau(\eta,v)}{\partial x'} - \frac{\partial\sigma(\zeta,u)}{\partial x'}\frac{\partial\tau(\eta,v)}{\partial k'}\right) & - \frac{1}{2}\frac{h}{2\pi i}\left(\frac{\partial\tau(\zeta,u)}{\partial k'}\frac{\partial\sigma(\eta,v)}{\partial x'} - \frac{\partial\tau(\zeta,u)}{\partial x'}\frac{\partial\sigma(\eta,v)}{\partial k'}\right)\Bigg].
\end{aligned} \tag{24}
$$

Equation (24) for the symbol of the commutator, together with the rule for calculating operators from their symbols given by Eqs. (20) and (21), leads to the following equation of motion for a mixed quantum-classical system

$$
\frac{\partial}{\partial t} A(hD,X,k',x') = \frac{2\pi i}{h} [H,A]_{\sharp_h}(hD,X,k',x'). \tag{25}
$$

This formula determines the evolution of an operator A, which depends on quantum and classical position and momentum variables and acts on $L^2(\mathbb{R}^n)\otimes\mathbb{R}^{2n}$. The kernel of the operator with respect to the $L^2(\mathbb{R}^n)$ subspace is given by Eq. (21).

The limiting procedure that leads from the quantum-quantum commutator [Eq. (22)] to the quantum-classical one [Eq. (24)] constitutes an essential part of the current formalism. The procedure can be summarized as follows. First, compute the quantum-quantum commutator. Second, discard terms of second and higher order in h'. Third, substitute h for h' in the first-order terms.

Example 3. Consider the quantum-classical bracket for the following functions:

$$
\sigma(k,x,k',x') = x^2 k'^2, \quad \tau(k,x,k',x') = k^2 x'^2. \tag{26}
$$

With this choice of σ and τ the integral in Eq. (22) factorizes, and the symbol of the quantum-quantum commutator becomes

$$
\begin{aligned}
[x^2k'^2, k^2x'^2]_{\sharp_{h,h'}} &= (x^2 \sharp_h k^2)(k'^2 \sharp_{h'} x'^2) - (k^2 \sharp_h x^2) \\
&\quad \times (x'^2 \sharp_{h'} k'^2) \qquad (27) \\
&= -4i\hbar x'k' \left(x^2k^2 - \frac{\hbar^2}{2}\right) \\
&\quad + 4i\hbar xk \left(x'^2k'^2 - \frac{\hbar'^2}{2}\right), \qquad (28)
\end{aligned}
$$

where $\hbar = h/2\pi$ is used for notational convenience. The details of evaluation of individual compositions in Eq. (27) are given in the Appendix. The limiting procedure applied to Eq. (28) leads to

$$-4i\hbar x'k'\left(x^2k^2-\frac{\hbar^2}{2}\right)+4i\hbar xkx'^2k'^2. \tag{29}$$

This result can be obtained directly from Eq. (24), which for the current choice of σ and τ factorizes as

$$[x^2k'^2,k^2x'^2]_{\sharp_{h,h'\to 0}} = -4i\hbar x'k'\,\frac{x^2\sharp_h k^2 + k^2\sharp_h x^2}{2} + x'^2k'^2[x^2,k^2]_{\sharp_h}. \tag{30}$$

The quantum-classical correspondence of Sec. II and the quantum-classical bracket of Eq. (24) are based on the Weyl correspondence principle. It is well known, however, that the Weyl correspondence is not unique in mapping phase space functions to Hilbert space operators. In fact, there exist arbitrarily many such mappings, differing in the order assigned to products of position and momentum operators. For example, the Weyl rule maps kx to the symmetrized product $\frac{1}{2}(hD\cdot X + X\cdot hD)$. Another variant of the correspondence principle, which is widely used in the mathematics community because of its simpler form, is due to Kohn and Nirenberg (KN). It keeps momentum operators on the right, mapping kx to $X\cdot hD$. It is straightforward to obtain a mixed quantum-classical equation of motion using the Kohn-Nirenberg calculus. One starts with $\rho_h = e^{2\pi i q\cdot X}e^{2\pi i p\cdot hD}$ instead of $\rho_h = e^{2\pi i(p\cdot hD + q\cdot X)}$ (see [47]) and follows the same steps. The result is

$$\frac{\partial}{\partial t}A(hD,X,k',x') = \frac{2\pi i}{h}[H,A]^{\mathrm{KN}}_{\sharp_h}(hD,X,k',x') \tag{31}$$

with the correspondence rule

$$\sigma(hD,X,k',x')^{\mathrm{KN}}f(x) = \int K^{\mathrm{KN}}_\sigma(x,y,k',x')f(y)dy,$$

$$K^{\mathrm{KN}}_\sigma(x,y,k',x') = h^{-n}\int \sigma(k,x,k',x')e^{2\pi i(x-y)k/h}dk, \tag{32}$$

and the following formula for the quantum-classical bracket of two symbols

$$[\sigma,\tau]^{\mathrm{KN}}_{\sharp_h}(k,x,k',x') = \left(\frac{2}{h}\right)^n\int\int du\,dv\,e^{4\pi i(x-u)(v-k)/h}\left\{\sigma(v,x)\tau(k,u)-\tau(v,x)\sigma(k,u) + \frac{h}{2\pi i}\left[\frac{\partial\sigma(v,x)}{\partial k'}\frac{\partial\tau(k,u)}{\partial x'}-\frac{\partial\tau(v,x)}{\partial k'}\frac{\partial\sigma(k,u)}{\partial x'}\right]\right\}. \tag{33}$$

These expressions are somewhat simpler than Eqs. (20), (21) and (24), (25), which were obtained by the Weyl correspondence. The Weyl rule results are preferable, though, since they preserve the simplectic invariance of the phase space variables [47] and lead via the Wigner transform from the density matrix to the quantum quasiprobability function that is closest to the classical probability density [49].

It is instructive to consider Example 3 within the KN correspondence. The quantum-quantum commutator is evaluated (see Appendix) as

$$[x^2k'^2,k^2x'^2]^{\mathrm{KN}}_{\sharp_{h,h'}} = (x^2\sharp^{\mathrm{KN}}_h k^2)(k'^2\sharp^{\mathrm{KN}}_{h'}x'^2)-(k^2\sharp^{\mathrm{KN}}_h x^2)(x'^2\sharp^{\mathrm{KN}}_{h'}k'^2) = -x^2k^2\left(4i\hbar x'k'+\frac{\hbar'^2}{2}\right)+\left(4i\hbar xk+\frac{\hbar^2}{2}\right)x'^2k'^2. \tag{34}$$

Multiplication of each term containing $\hbar'$ by $\hbar/\hbar'$ and the limit $\hbar'\to 0$ produce

$$-x^2k^2(4i\hbar x'k')+\left(4i\hbar xk+\frac{\hbar^2}{2}\right)x'^2k'^2 = -4i\hbar x'k'(x^2\sharp^{\mathrm{KN}}_h k^2)+x'^2k'^2[x^2,k^2]^{\mathrm{KN}}_{\sharp_h}, \tag{35}$$

which also directly follows from Eq. (33). The Weyl and KN expressions for the quantum-classical commutator in the example [Eqs. (29) and (35)] disagree in the second-order term in $\hbar$.

The quantum-classical equations of motion [Eq. (25) and (31)] exhibit many desired features. If A and H depend solely on quantum or classical variables, they reduce to the purely quantum and classical equations, Eqs. (13) and (17), respectively. Since the quantum-classical brackets of the right-hand side of Eqs. (25) and (31) were obtained by selecting a representation for the Lie bracket of a Lie group, they possess the general properties of Lie brackets. In particular, both quantum-classical brackets are antisymmetric, since their symbols are antisymmetric: the integrands of Eqs. (24) and (33) change sign under the permutation $\sigma\leftrightarrow\tau$. The Jacobi identity is a more subtle issue. If the limiting procedure introduced in the previous paragraphs is applied to derive the quantum-classical Jacobi identity from the quantum-quantum one, no problems are encountered. Indeed, by successive application of Eq. (22) the symbol of the product of three operators

$$\sigma(hD,X,k',x')\tau(hD,X,k',x')\phi(hD,X,k',x')$$

takes the form

$$
\begin{aligned}
&\sigma \sharp_h \tau \sharp_h \phi(k,x) \\
&= \left(\frac{2}{h}\right)^{2n} \int\int\int\int du_1 dv_1 d\eta_1 d\zeta_1 e^{4\pi i[(x-u_1)(k-\eta_1)-(x-v_1)(k-\zeta_1)]/h} \left(\frac{2}{h'}\right)^{2n} \int\int\int\int du\ dv\ d\eta\ d\zeta [\sigma(\zeta,u)\tau(\eta,v) \\
&- \tau(\zeta,u)\sigma(\eta,v)]\{[e^{4\pi i[(\zeta_1-u)(u_1-\eta)-(\zeta_1-v)(u_1-\zeta)]/h}\phi(\eta_1,v_1) - e^{4\pi i[(\eta_1-u)(v_1-\eta)-(\eta_1-v)(v_1-\zeta)]/h}\phi(\zeta_1,u_1)] \\
&+ i\pi h'\{[D_{k'}\sigma(\zeta,u)\tau(\eta,v) + \sigma(\zeta,u)D_{k'}\tau(\eta,v) - D_{k'}\tau(\zeta,u)\sigma(\eta,v) - \tau(\zeta,u)D_{k'}\sigma(\eta,v)] \\
&\times e^{4\pi i[(\zeta_1-u)(u_1-\eta)-(\zeta_1-v)(u_1-\zeta)]/h} D_{xD'}\phi(\eta_1,v_1) - e^{4\pi i[(\eta_1-u)(u_1-\eta)-(\eta_1-v)(v_1-\zeta)]/h} D_{k'}\phi(\zeta_1,u_1) \\
&\times [D_{x'}\sigma(\zeta,u)\tau(\eta,v) + \sigma(\zeta,u)D_{x'}\tau(\eta,v) - D_{x'}\tau(\zeta,u)\sigma(\eta,v) - \tau(\zeta,u)D_{x'}\sigma(\eta,v)]\} + O(h'^2)\}. \qquad (36)
\end{aligned}
$$

The limit $\lim_{h'\to 0} h/h'$, applied after the zero-order term, discards second- and higher-order terms. Subsequent integrations lead to an expression that can be rewritten in a compact symbolic form as

$$
\sigma \sharp \tau \sharp \phi = \sigma\tau\phi - \phi\tau\sigma + i(\sigma_{k'}\tau\phi_{x'} + \sigma\tau_{k'}\phi_{x'} - \tau_{k'}\sigma\phi_{x'} - \tau\sigma_{k'}\phi_{x'} - \phi_{k'}\sigma_{x'}\tau - \phi_{k'}\sigma\tau_{x'} + \phi_{k'}\tau_{x'}\sigma + \phi_{k'}\tau\sigma_{x'}), \qquad (37)
$$

where the subscripts k' and x' indicate differentiation with respect to these variables, and the ordering of the symbols is to be kept track of. Given Eq. (37) it is straightforward to check that the Jacobi identity holds for the symbols of operators: $[[\sigma,\tau]_{\sharp h},\phi]_{\sharp h}(k,x,k',x')$ and, therefore, for the operators themselves.

Evaluation of the double quantum-quantum commutators followed by removal of the h'^2 terms leads to the same result. However, when an attempt is made to check the Jacobi identity based only on the quantum-classical bracket, the outcome is negative: repeated application of Eq. (24) or Eq. (33) leaves some of the second-order terms as observed, for instance, in Ref. [7]. Close examination of the quantum-*classical* Jacobi identity shows that the second-order terms that do not cancel out are associated in expansion (36) with h'^2 [see also Eq. (22)], i.e., with the Planck constant for the classical set of variables. If one takes the view that transition from quantum to classical leaves only the first-order contributions from the classical part, then the second-order terms in the Jacobi identity must disappear. Examination of the quantum-classical bracket alone does not show the origin of the extra terms. The subtlety with the Jacobi identity is due to the nontrivial topological structure of the unitary dual to the quantum-classical Lie group.

The quantum-classical Lie bracket itself does not define the dynamics of a mixed quantum-classical system. The ''naive'' proposition of Eq. (25) for the time derivative of a quantum-classical operator A meets the problem that the second time derivative given by a double bracket must contain the h'^2 terms, so as to reduce to the double Poisson bracket in the purely classical limit. Formulation of a consistent dynamical equation for the evolution of a quantum-classical system remains incomplete. It is stressed, however, that quantum-classical dynamics can be analyzed by p mechanics even without an explicit form for the equation of motion. An example of such an analysis is presented in the next section.

IV. QUANTUM-CLASSICAL COUPLING FOR HARMONIC OSCILLATORS

Quantization of a classical system is particularly suitable for a description of harmonic oscillators. The relevant results are briefly summarized below and are applied to coupled quantum and classical oscillators. The reader is referred to Refs. [50–56] for further information on the quantization procedure used below.

Let $L_2(\mathbb{C}^n, d\mu_n)$ be a space of functions on $\mathbb{C}^n$ that are square integrable with respect to the Gaussian measure

$$
d\mu_n(z) = \pi^{-n} e^{-z\cdot\bar{z}} dv(z),
$$

where $dv(z) = dx\ dy$ is the Euclidean volume measure on $\mathbb{C}^n = \mathbb{R}^{2n}$. The Segal-Bargmann [57,58], or, equivalently, Fock [59] space $F_2(\mathbb{C}^n)$ is the subspace of $L_2(\mathbb{C}^n, d\mu_n)$ consisting of all entire functions, i.e., functions $f(z)$ that satisfy

$$
\frac{\partial f}{\partial \bar{z}_j} = 0, \quad 1 \leqslant j \leqslant n.
$$

Denote by P_Q the orthogonal Bargmann projection [57] of $L_2(\mathbb{C}^n, d\mu_n)$ onto the Fock space $F_2(\mathbb{C}^n)$. Then

$$
k(q,p) \to T_{k(q+ip)} = P_Q k(q+ip) I \qquad (38)
$$

defines the Berezin-Toeplitz (anti-Wick) quantization, which maps a function $k(q,p) = k(q+ip)$ on $\mathbb{R}^{2n} = \mathbb{C}^n$ to the Toeplitz operator T_k. There exists an identification between the Berezin and Weyl quantizations [50,53,56]. The identification has an especially transparent form for the observables depending only on p and q.

The Berezin-Toeplitz quantization is related to the Heisenberg group more intuitively than the representation of Eq. (4). On a geometrical level, consider the group of Euclidean shifts $a: z \mapsto z + a$ of $\mathbb{C}^n$. To obtain unitary operators on $L_2(\mathbb{C}^n, d\mu)$ the shifts are multiplied by the weight function

$$
a: f(z) \mapsto f(z+a) e^{-z\bar{a} - a\bar{a}/2}. \qquad (39)
$$

This mapping determines [60] a unitary representation of the $(2n+1)$-dimensional Heisenberg group acting on $L_2(\mathbb{C}^n, d\mu)$. The mapping preserves the Fock space $F_2(\mathbb{C}^n)$, and, hence, all operators of the form of Eq. (39) commute with P_Q. The operators are generated by infinitesimal displacements

$$i\sum_{k=1}^{n}\left[a_j'\left(\frac{\partial}{\partial z_j'}-z_j'-iz_j''\right)+a_j''\left(\frac{\partial}{\partial z_j''}-z_j''+iz_j'\right)\right],$$

where $a=(a_1,\ldots,a_n)$, $z=(z_1,\ldots,z_n)\in\mathbb{C}^n$, and $a_j=(a_j',a_j'')$, $z=(z_j',z_j'')\in\mathbb{R}^2$. The generators form a linear space with the basis

$$A_j^{f\prime}=\frac{1}{i}\left(\frac{\partial}{\partial z_j'}-z_j'-iz_j''\right),\quad A_j^{f\prime\prime}=\frac{1}{i}\left(\frac{\partial}{\partial z_j''}-z_j''+iz_j'\right). \tag{40}$$

The basis vectors commute with the Bargmann projector P_Q. The operators

$$X_j^{f\prime}=\frac{1}{i}\left(\frac{\partial}{\partial z_j'}-z_j'+iz_j''\right),\quad X_j^{f\prime\prime}=\frac{1}{i}\left(\frac{\partial}{\partial z_j''}-z_j''-iz_j'\right) \tag{41}$$

commute with the basis vectors, and we anticipate that P_Q produces a self-adjoint representation of convolutions with respect to $X_j^{f\prime}$, $X_j^{f\prime\prime}$, and unit operators.

Proposition 4 [43]. The Bargmann projector P_Q defines a representation of convolutions induced by the Weyl-Heisenberg Lie algebra $\mathfrak{h}_n$ operating on $\mathbb{C}^n$ by Eqs. (41). The kernel $b(t,\zeta)$, $t\in\mathbb{R}$, $\zeta\in\mathbb{C}^n$ of the representation is given by the formula

$$\hat{b}(t,\zeta)=2^{n+1/2}e^{-1}e^{-(t^2+\zeta\bar{\zeta}/2)}.$$

We move on to apply the Bargmann projection technique to the quantum-classical coupling of harmonic oscillators.

Example 5 [52]. In the Segal-Bargmann representation the creation and annihilation operators are $a_j^{\dagger}=z_jI$ and $a_j^{-}=\partial/\partial z_j$, respectively. Consider an n degree of freedom harmonic oscillator with the classical Hamilton function

$$H(q,p)=\frac{1}{2}\sum_{j=1}^{n}(q_j^2+p_j^2).$$

The corresponding quantum Hamiltonian is obtained by the Bargmann projection

$$T_{H(q,p)}=\frac{1}{2}P_Q\sum_{j=1}^{n}(q_j^2+p_j^2)I=\frac{1}{2}\left(nI+\sum_{j=1}^{n}z_j\frac{\partial}{\partial z_j}\right). \tag{42}$$

The right side of Eq. (42) is the celebrated Euler operator. It generates the well-known dynamical group [[61], Chap. 1, Eq. (6.35)]

$$e^{itT_{H(p,q)}}f(z)=e^{int/2}f(e^{it}z),\quad f(z)\in F_2, \tag{43}$$

which induces rotation of the $\mathbb{C}^n$ space. The evolution of the classical oscillator is also given by a rotation, that of the phase space $\mathbb{R}^{2n}$:

$$z(t)=G_tz_0=e^{it}z_0,\quad z(t)=p(t)+iq(t),\quad z_0=p_0+iq_0. \tag{44}$$

The projection P_Q leads to the Segal-Bargmann representation, providing a very straightforward correspondence between quantum and classical mechanics of oscillators, in contrast to the rather complicated case of the Heisenberg representation [[61], Prop. 7.1 of Chap. 1]. The powers of z are the eigenfunctions $\phi_n(z)=z^n$ of the Hamiltonian (42), and the integers n are the eigenvalues. Either pure or mixed, any initial state of the oscillator remains unchanged during the Eq. (43) evolutions and no transitions between states are observed.

Now consider classical and quantum oscillators coupled by a quadratic term

$$H(p,q;p',q')=\tfrac{1}{2}[p'^2+p^2+x'^2+x^2+\alpha(x'-x)^2]. \tag{45}$$

Applying the canonical transformation (see [62], Sec. 23 D)

$$q'=\frac{q_1+q_2}{\sqrt{2}},\quad q=\frac{q_1-q_2}{\sqrt{2}},\quad p'=\frac{p_1+p_2}{\sqrt{2}},\quad p=\frac{p_1-p_2}{\sqrt{2}}, \tag{46}$$

or, equivalently, introducing complex variables $z=q+ip$, $z'=q'+ip'$, $z_1=q_1+ip_1$, $z_2=q_2+ip_2$,

$$z'=\frac{z_1+z_2}{\sqrt{2}},\quad z=\frac{z_1-z_2}{\sqrt{2}},$$

$$z_1=\frac{z'+z}{\sqrt{2}},\quad z_2=\frac{z'-z}{\sqrt{2}}, \tag{47}$$

we get rid of the coupling term

$$H(p_1,q_1;p_2,q_2)=\tfrac{1}{2}(p_1^2+p_2^2+\omega_1^2q_1^2+\omega_2^2q_2^2), \tag{48}$$

where $\omega_1=1$, $\omega_2=\sqrt{1+2\alpha}$. The two uncoupled oscillators evolve independently:

$$z_1(t)=e^{2i\omega_1t}z_1(t_0),\quad z_2(t)=e^{2i\omega_2t}z_2(t_0).$$

The dynamics in the original coordinates, however, is not trivial. The primed and unprimed (classical and quantum) variables mix:

$$z'(t)=\frac{(e^{2i\omega_1t}+e^{2i\omega_2t})z'(t_0)+(e^{2i\omega_1t}-e^{2i\omega_2t})z(t_0)}{2}, \tag{49}$$

$$z(t)=\frac{(e^{2i\omega_1t}-e^{2i\omega_2t})z'(t_0)+(e^{2i\omega_1t}+e^{2i\omega_2t})z(t_0)}{2}. \tag{50}$$

Suppose that the classical subsystem is initially localized at a point z_0' of the phase space and the quantum subsystem is in its nth pure state: $\phi(z',z;t_0)=\delta(z_0'-z')\otimes z^n$. Then, the dynamics of the combined system is given by

$$\phi(z',z;t)=\delta\left(z_0'-\frac{(e^{2i\omega_1t}+e^{2i\omega_2t})z'+(e^{2i\omega_1t}-e^{2i\omega_2t})z}{2}\right) \tag{51}$$

$$\otimes\left(\frac{(e^{2i\omega_1t}-e^{2i\omega_2t})z'+(e^{2i\omega_1t}+e^{2i\omega_2t})z}{2}\right)^n. \tag{52}$$

During the evolution, the classical subsystem [Eq. (51)] is always sharply supported, i.e., represented by the delta function, while the quantum subsystem [Eq. (52)] evolves into a mixed state. The binomial (52) contains all powers of z less than or equal to n (z^k, $k \leqslant n$). Therefore, there exists a nonzero probability for the quantum subsystem to make a transition from the initial pure state z^n into a lower-energy state z^k, $k<n$. It is remarkable that in this particular case the interaction with the classical subsystem can only decrease the initial energy of the quantum one. If the quantum subsystem is originally in the ground state ($n=0$), expression (52) identically equals one, and energy exchange is not observed. The overall dynamics is (quasi)periodic with the recurrence time determined by the frequencies ω_1 and ω_2.

V. MULTICONFIGURATION MEAN-FIELD APPROXIMATION

The quantum-classical equation of motion [Eq. (25)] can be applied in several ways depending on the description chosen for the quantum and classical subsystems. The great utility of quantum-classical models based on a trajectory description for the classical subsystem has found realization in numerous molecular-dynamics techniques [63,64], which are inexpensive computationally and predict dynamical characteristics of many degree of freedom systems by averaging over just a few sample trajectories. In the simplest case the classical subsystem can be represented by a point in the phase space $\{k_i', x_i'\}$ evolving according to the Hamilton equations with the quantum-classical bracket of Eq. (24) regarded as a modification of the Poisson bracket. If at the same time the Heisenberg equation of motion with the quantum-classical bracket in place of the commutator is used to describe the evolution of quantum operators, the mean-field approximation follows. Namely, the quantum-mechanical average of Eq. (25) with respect to the wave function Ψ is given by

$$\frac{\partial}{\partial t}\langle\Psi|A(k',x')|\Psi\rangle = \frac{2\pi i}{h}\langle\Psi|[H,A]_{\sharp_h}(k',x')|\Psi\rangle. \tag{53}$$

If A is a purely quantum mechanical observable independent of classical variables, the derivatives $\partial A/\partial k'$ and $\partial A/\partial x'$ in the quantum-classical bracket of Eq. (24) vanish, and we obtain

$$\frac{\partial}{\partial t}\langle\Psi|A|\Psi\rangle = \frac{2\pi i}{h}\langle\Psi|[H(k',x'),A]|\Psi\rangle \tag{54}$$

with the Hamiltonian H parametrically dependent on the classical phase space variables k' and x'. Substituting these variables in place of $A(k',x')$ in the quantum-classical bracket of Eq. (53), we compute the integrals in the definition of the bracket [Eq. (24)] and recover the classical equations of motion with the classical Hamiltonian being equal to the quantum mechanical average of the total Hamiltonian

$$\frac{\partial k'}{\partial t} = -\frac{\partial\langle\Psi|H(k',x')|\Psi\rangle}{\partial x'}, \qquad \frac{\partial x'}{\partial t} = \frac{\partial\langle\Psi|H(k',x')|\Psi\rangle}{\partial k'}. \tag{55}$$

Equations (54) and (55) constitute the traditional mean-field approximation: classical variables are coupled to the expectation values of quantum observables [30–33].

The quantum-classical equation of motion [Eq. (25)] can also be regarded as a Liouville–von Neumann equation for a mixed density matrix–phase space distribution function $\rho(hD,X,k',x')$. In a quantum basis the evolution of ρ is given by a set of coupled equations for classical phase space distribution functions $\rho_{ij}(k',x')$ corresponding to each pair of the quantum basis states i,j:

$$\frac{\partial\rho_{ij}}{\partial t} = \frac{2\pi i}{h}\sum_k\left[H_{ik}^*\rho_{kj} - \rho_{ik}^*H_{kj} + \frac{1}{2}\frac{h}{2\pi i}\left(\frac{\partial H_{ik}^*}{\partial k'}\frac{\partial\rho_{kj}}{\partial x'} - \frac{\partial H_{ik}^*}{\partial x'}\frac{\partial\rho_{kj}}{\partial k'}\right) - \frac{1}{2}\frac{h}{2\pi i}\left(\frac{\partial\rho_{ik}^*}{\partial k'}\frac{\partial H_{kj}}{\partial x'} - \frac{\partial\rho_{ik}^*}{\partial x'}\frac{\partial H_{kj}}{\partial k'}\right)\right]. \tag{56}$$

The above set of coupled Liouville equations is based on the Weyl correspondence.

In the purely quantum and classical limits the corollaries [Eqs. (54), (55), and Eq. (56)] of the quantum-classical equation of motion [Eq. (25)] are equivalent. In the mixed case they are not due to nonlocal correlations within the classical subsystem induced by interactions with the quantum one. Such correlations, inherent in the Liouville–von Neumann equation, do not appear in the mean-field approach. In particular, if Eqs. (56) are integrated with the initial conditions $\rho_{11}(k',x') = \delta(k'-k_0')\,\delta(x'-x_0')$, $\rho_{ij}=0$, $\forall\{ij\}\neq\{11\}$, at later times $\rho_{ij}\neq 0$ because of the nonzero couplings H_{ij}. Classical phase space distribution functions associated with different quantum states differ and mix. Mutual mixing enhances spreading of classical distributions. An initially localized phase space distribution ρ_{11} corresponding to the ground quantum state populates excited-state distributions. Since, in general, ground- and excited-state distributions undergo diverging evolutions, ρ_{11} delocalizes due to mixing with excited states. This, of course, cannot happen in the mean-field approach, where the classical subsystem is described by a single trajectory and the nonlocal correlations are averaged out. In the absence of coupling between quantum states, phase space distributions do not mix, and Eq. (56) splits into a set of uncoupled equations for classical distribution functions for individual quantum states. The evolution of each distribution function can be equivalently described by the classical trajectory mean-field approach [Eqs. (54) and (55)] with the corresponding basis state wave function in place of Ψ. Adiabatic molecular-dynamics methods [63] take

advantage of this situation by evolving classical particles on adiabatic quantum states and neglecting nonadiabatic coupling.

The Liouville–von Neumann equation for the evolution of the mixed quantum-classical distribution reduces to the coupled equations of the mean-field approximation when the distribution is restricted to have the following functional form

$$\rho = |\Psi\rangle\langle\Psi| \, \delta(k_0' - k') \, \delta(x_0' - x'), \tag{57}$$

where k' and x' are the n-dimensional classical momentum and position vectors. Under this constraint the quantum part of the mixed distribution remains a pure state at all times, and the classical part is always represented by a delta function. Substituting expression (57) for ρ in place of A in Eq. (25) and integrating out the phase space variables we obtain the von Neumann equation with the quantum Hamiltonian depending on classical coordinates $H(k',x')$. For a pure state the von Neumann equation is equivalent to the Schrödinger equation and to Eq. (54). In order to recover the mean-field equations for the classical positions and momenta [Eqs. (55)] we substitute Eq. (57) into Eq. (25), multiply both sides by either k' or x', and integrate over quantum and classical variables.

The mean-field approximation does not account for the nonlocal correlations that appear in the fully quantum description and are reproduced by the Liouville–von Neumann equation. Such correlations are important, for instance, when quantum motion involves tunneling between two distinct subspaces. Unfortunately, the quantum-classical Liouville–von Neumann equation does not provide a computational advantage over the fully quantum von Neumann equation, since both deal with delocalized distributions. At the same time, propagation of an individual classical trajectory via the Hamilton equations of motion is far less demanding than integration of the Liouville equation. In order to account for the quantally induced nonlocal correlations among the phase space variables, while retaining a trajectory description for the classical subsystem, we interpolate between the mean field and Liouville–von Neumann approaches by developing a multiconfiguration version of the mean-field method. Starting with the quantum-classical distribution of the following functional form,

$$\rho = \sum_i \sum_j \varrho_{ij} |\Psi_i\rangle\langle\Psi_j| \, \delta(k_{ij}' - k') \, \delta(x_{ij}' - x'), \tag{58}$$

where the sums run over configurations, we assume orthonormality of the wave functions $\langle\Psi_i|\Psi_j\rangle = \delta_{ij}$, substitute Eq. (58) into Eq. (25), integrate over the classical variables ($\int dk' \int dx'$), and obtain the von Neumann equation for the quantum density matrix

$$\frac{\partial \varrho_{ij}}{\partial t} = \frac{2\pi i}{h} \sum_k [H_{ik}^*(k_{ik}', x_{ik}') \varrho_{ik} - \varrho_{kj}^* H_{kj}(k_{kj}', x_{kj}')]. \tag{59}$$

Note that in contrast to $\rho_{ij}(k',x')$ of Eq. (56), ϱ_{ij} do not explicitly depend on classical variables and are complex numbers rather than phase space distribution functions. To get the equations of motion for the classical momenta and positions k_{ij}, x_{ij} we substitute Eq. (58) into Eq. (25), multiply both sides by the corresponding variable, and integrate over all degrees of freedom. The ''diagonal'' positions and momenta evolve according to

$$\frac{\partial k_{ii}'}{\partial t} = -\frac{1}{2} \sum_k \left(\frac{\partial H_{ik}^*(k_{ik}', x_{ik}')}{\partial x_{ik}'} + \frac{\partial H_{ki}(k_{ki}', x_{ki}')}{\partial x_{ki}'} \right),$$

$$\frac{\partial x_{ii}'}{\partial t} = \frac{1}{2} \sum_k \left(\frac{\partial H_{ik}^*(k_{ik}', x_{ik}')}{\partial k_{ik}'} + \frac{\partial H_{ki}(k_{ki}', x_{ki}')}{\partial k_{ki}'} \right). \tag{60}$$

The corresponding expressions for the ''nondiagonal'' variables are more complicated

$$\frac{\partial x_{ij}'}{\partial t} = \sum_k H_{ik}^*(k_{ik}, x_{ik}) x_{kj}' - x_{ik}' H_{kj}(k_{ik}, x_{ik}) + \frac{1}{2} \left(\frac{\partial H_{ik}^*(k_{ik}, x_{ik})}{\partial k_{ik}'} + \frac{\partial H_{kj}(k_{kj}', x_{kj}')}{\partial k_{kj}'} \right), \tag{61}$$

and similarly for momenta. In order to keep the Hamiltonian matrix Hermitian $H_{ij} = H_{ji}^*$ and to avoid situations where $H_{ij}(k_{ij}', x_{ij}')$ and $H_{ji}^*(k_{ji}', x_{ji}')$ are computed at different locations of classical particles, we stipulate that the position and momentum ''matrices'' are symmetric: $k_{ij}' = k_{ji}'$, $x_{ij}' = x_{ji}'$. With this constraint the ''nondiagonal'' evolutions simplify, the first two terms in formula (61) disappear, and the dynamics of the ''nondiagonal'' phase space variables coincide with the average dynamics of the ''diagonal'' variables

$$\begin{aligned} \frac{\partial x_{ij}'}{\partial t} &= \frac{1}{2} \left\{ \frac{\partial x_{ij}'}{\partial t} + \frac{\partial x_{ji}'}{\partial t} \right\} \\ &= \frac{1}{2} \sum_k \left\{ H_{ik}^* x_{kj}' - x_{ik}' H_{kj} + \frac{1}{2} \left(\frac{\partial H_{ik}^*}{\partial k_{ik}'} + \frac{\partial H_{kj}}{\partial k_{kj}'} \right) \right. \\ &\quad \left. + H_{jk}^* x_{ki}' - x_{jk}' H_{ki} + \frac{1}{2} \left(\frac{\partial H_{jk}^*}{\partial k_{jk}'} + \frac{\partial H_{ki}}{\partial k_{ki}'} \right) \right\} \\ &= \frac{1}{4} \left(\frac{\partial H_{ik}^*}{\partial k_{ik}'} + \frac{\partial H_{kj}}{\partial k_{kj}'} + \frac{\partial H_{jk}^*}{\partial k_{jk}'} + \frac{\partial H_{ki}}{\partial k_{ki}'} \right) = \frac{1}{2} \left(\frac{\partial x_{ii}'}{\partial t} + \frac{\partial x_{jj}'}{\partial t} \right), \end{aligned} \tag{62}$$

and similarly for momenta. Apart from assigning the ''nondiagonal'' coordinates and momenta unique values with a simple physical meaning: $x_{ij}' = (x_{ii}' + x_{jj}')/2$, $k_{ij}' = (k_{ii}' + k_{jj}')/2$, Eq. (62) reduces the number of independent classical trajectories in the n-dimensional multiconfiguration mean-field approximation from n^2 to n.

The multiple configuration technique is widely used in quantum chemistry as an improvement on the self-consistent field solution to the time-independent Schrödinger equation [65]. The time-dependent fully quantum multiconfiguration self-consistent field approach is discussed in Ref. [66]. Equations of motion similar to our version of the quantum-classical multiconfiguration approximation were first proposed in Ref. [14]. Our approach has a different derivation and is less computationally demanding, since it needs a factor of n fewer trajectories. Other versions of the quantum-

classical multiconfiguration mean-field approximation can be found in Refs. [67, 68]. We point out that the number of configurations in the multiconfiguration mean-field method does not have to equal and is usually less than the number of quantum basis states. The indices i,j,k in Eq. (59) denote configurations, which do not, in general, correspond to basis states. In the case when the overall space is separated into several weakly connected regions, the number of configurations is determined by the number of such regions. For example, the double well system [69] requires two configurations and two classical trajectories, each originating in its own well [70].

VI. DISCUSSION AND CONCLUSIONS

The technique presented in the earlier sections provides a unifying framework for the quantum-classical equations of motion proposed by other authors [2,12,15]. The equations of Refs. [12, 15] are very similar and correspond to Eq. (25) of this paper. Thus, Ref. [12] proposes

$$\dot{\rho}=\frac{i}{\hbar}[\rho,H]+\frac{1}{2}\{\rho,H\}-\frac{1}{2}\{H,\rho\}. \tag{63}$$

[See Eqs. (3.4) and (3.6) of Ref. [12].] The equation of motion presented in Ref. [15], Eq. (11), is slightly different:

$$\dot{\rho}=i[\rho,H]+\frac{\hbar}{2}\{\rho,H\}-\frac{\hbar}{2}\{H,\rho\}. \tag{64}$$

Our results based on the Weyl correspondence principle [cf. Eqs. (24) and (25)] give preference to Eq. (63).

The quantum-classical bracket of Ref. [2], Eq. (2),

$$[A,B]_{(q-c)}=[A,B]+i\{A,B\} \tag{65}$$

is not antisymmetric. It is very similar to Eq. (33) of the current work, which is derived by the Kohn-Nirenberg calculus and is antisymmetric as expected from the derivation. The example given in the paragraph following Eq. (2) in Ref. [2] considers a separable case with f,g being classical functions and U,V being quantum operators:

$$[fU,gV]_{(q-c)}=fg[U,V]+iUV\{f,g\}. \tag{66}$$

If U and V do not commute, the result is clearly not antisymmetric. For this example, Eq. (33) of this work gives the expression

$$fg[U,V]+i(UV\partial_p f\partial_x g-VU\partial_p g\partial_x f), \tag{67}$$

which does change sign under the permutation $f\leftrightarrow g$, $U\leftrightarrow V$. Thus, Eq. (33) can be regarded as the corrected version of the bracket of Ref. [2].

The brackets of Refs. [12, 15] and the bracket of Ref. [2] relate to each other via the choice of the correspondence principle, i.e., via the ordering of products of position and momentum operators used in constructing quantum operators from classical phase space functions. References [12,15] employ symmetrized products, while Ref. [2] uses expressions, which keeps momentum operators on the right. The relationship between the quantum-classical correspondence rules and pseudodifferential calculus explored, for instance, in Ref. [71] leads to a family of quantization rules parametrized by a continuous parameter $0\leqslant\lambda\leqslant 1$. The general form of pseudodifferential operators underlying this family of quantum-classical correspondences is given in Refs. [42, 72]. λ equal to 1/2 and 1 produces the Weyl and Kohn-Nirenberg correspondence rules, respectively. Arbitrarily many quantum-classical brackets can be generated for different choices of λ.

Our approach explains why the author of Ref. [8] came to the conclusion of the algebraic impossibility of quantum-classical coupling. The assumptions imposed in Ref. [8] on a possible quantum-classical theory restrict consideration to step-two nilpotent Lie groups with a one-dimensional center. Under this exceedingly stringent constraint any interaction between quantum and classical degrees of freedom is impossible, indeed. However, once the constraint is relaxed, and we see no reason to restrict our attention to groups with a one-dimensional center only, quantum-classical interactions naturally reappear. The step-two nilpotent Lie group with a two-dimensional center considered here in detail produces satisfactory results. Other groups in principle suitable to describe quantum-classical mixing do exist. For instance, one may consider a step-three nilpotent Lie group and the corresponding algebra, which, as a vector space, is decomposable into the three subspaces V_0, V_1, and V_2 having the following properties. The elements of V_0 commute with all elements and form the center. Commutators of vectors from V_1 belong to V_0. Commutators of vectors from V_2 belong to V_1. By taking a representation of this group that maps the center V_0 to zero, we would obtain another model for a mixed quantum-classical system, where vectors from V_1 would correspond to classical degrees of freedom, since their commutators vanish, while vectors from V_2 would describe quantum variables. It is likely, though, that this scheme will exhibit properties atypical for quantum and classical mechanics, since step-three nilpotent Lie groups differ from step-two groups and from the Heisenberg group in particular. There exist, however, some advantages in dealing with general nilpotent Lie groups. For example, the relativistic quantization of Ref. [73] is based on a representation of the simplest step-three nilpotent Lie group (meta-Heisenberg group [74]) spanned by the Schrödinger representation of the Heisenberg group and the operators of multiplication by functions. Application of the quantization rules to the appropriate Lie algebras leads to quantum-classical constructions for string theory, conformal field theory, and Yang-Mills theories [75].

In summary, we presented a systematic approach to coupling quantum and classical degrees of freedom based on a generalization of the unified description of quantum and classical mechanics given in terms of convolutions on the Heisenberg group. Considering the simplest extension of the Heisenberg group that allows for two distinct sets of variables, we derived a quantum-classical equation of motion. The quantum-classical bracket entering the equation is a Lie bracket and, therefore, possesses the two most important properties common to the quantum commutator and the Poisson bracket: it is antisymmetric and satisfies the Jacobi identity. The quantum-classical Jacobi identity meets with subtleties that we associate with the dynamical interpretation of the bracket. Further work is needed in order to clarify this issue,

and the simplistic equation $-i\hbar dA/dt=[H,A]_{q-c}$ might have to be abandoned. We constructed the quantum-classical dynamics of coupled harmonic oscillators without explicitly appealing to such an equation. Starting from a general group-theoretical formulation we derived the mean-field and the multiconfiguration mean-field approximations, which are trajectory-based approaches of great utility in studying realistic multidimensional physical and chemical systems. The proposed technique allowed us to classify the quantum-classical equations of motion and to resolve some problems from earlier works.

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APPENDIX

We present details of computation of the $\sharp$ compositions encountered in Example 3 of Sec. III. The transformations below use the standard properties of the delta function. Within the Weyl correspondence [Eq. (12)] a typical composition in Example 3 is calculated as follows:

$$\begin{aligned}
k^2\sharp_h x^2&=\left(\frac{2}{h}\right)^2\int\int\int\int \zeta^2 v^2 e^{4\pi i[(x-u)(k-\eta)-(x-v)(k-\zeta)]/h}du\ dv\ d\eta\ d\zeta\\
&=\left(\frac{2}{h}\right)^2\int e^{4\pi ix(k-\eta)/h}d\eta\int e^{-4\pi iu(k-\eta)/h}du\int\int \zeta^2 v^2 e^{-4\pi i(x-v)(k-\zeta)/h}dv\ d\zeta\\
&=\left(\frac{2}{h}\right)^2\int e^{4\pi ix(k-\eta)/h}d\eta\,\delta\left[\frac{2(\eta-k)}{h}\right]\int\int \zeta^2 v^2 e^{-4\pi i(x-v)(k-\zeta)/h}dv\ d\zeta\\
&=\frac{2}{h}\int \zeta^2 e^{-4\pi ix(k-\zeta)/h}\left[\int v^2 e^{4\pi iv(k-\zeta)/h}dv\right]d\zeta=\frac{2}{h}\int \zeta^2 e^{-4\pi ix(k-\zeta)/h}\left(\frac{h}{4\pi i}\right)^2\delta''\left[\frac{2(k-\zeta)}{h}\right]d\zeta\\
&=\left(\frac{h}{4\pi i}\right)^2\int \zeta^2 e^{-4\pi ix(k-\zeta)/h}\delta''(k-\zeta)d\zeta=\left(\frac{h}{4\pi i}\right)^2\frac{d^2}{d\zeta^2}\left[\zeta^2 e^{-4\pi ix(k-\zeta)/h}\right]_{\zeta=k}\\
&=\left(\frac{h}{4\pi i}\right)^2\left[\left(\frac{4\pi i}{h}\right)^2 x^2k^2+\frac{4\pi i}{h}4xk+2\right].
\end{aligned}\tag{A1}$$

The composition with the opposite order of x^2 and k^2 is computed similarly:

$$x^2\sharp_h k^2=\left(\frac{h}{4\pi i}\right)^2\left[\left(\frac{4\pi i}{h}\right)^2 x^2k^2-\frac{4\pi i}{h}4xk+2\right].\tag{A2}$$

Appropriate combinations of the above results produce Eqs. (28) and (29) of Sec. III. For instance, the commutator in Eq. (30),

$$[x^2,k^2]_{\sharp_h}=x^2\sharp_h k^2-k^2\sharp_h x^2=-\frac{h}{2\pi i}4xk=4i\hbar xk\tag{A3}$$

contributes to the second term in Eq. (29). The Kohn-Nirenberg correspondence given by Eq. (33) requires even simpler calculations:

$$\begin{aligned}
k^2\sharp_h^{\mathrm{KN}}x^2&=\frac{2}{h}\int\int u^2v^2e^{4\pi i(x-u)(v-k)/h}du\ dv\\
&=\frac{2}{h}\int v^2\left[\int u^2e^{-4\pi iu(v-)/h}du\right]e^{4\pi ix(v-k)/h}dv\\
&=\frac{2}{h}\int v^2\left(\frac{h}{4\pi i}\right)^2\delta''\left[\frac{2(v-k)}{h}\right]e^{4\pi ix(v-k)/h}dv\\
&=\left(\frac{h}{4\pi i}\right)^2\frac{d^2}{dv^2}\left[v^2e^{4\pi ix(v-k)/h}\right]_{v=k}\\
&=\left(\frac{h}{4\pi i}\right)^2\left[\left(\frac{4\pi i}{h}\right)^2x^2k^2+\frac{4\pi i}{h}4xk+2\right].
\end{aligned}\tag{A4}$$

In the KN calculus the composition of $x^2\sharp k^2$ is just the product x^2k^2, and, for instance, the commutator appearing in right-hand side of Eq. (35) equals

$$\begin{aligned}
[x^2,k^2]_{\sharp_h}^{\mathrm{KN}}&=x^2k^2-\left(\frac{h}{4\pi i}\right)^2\left[\left(\frac{4\pi i}{h}\right)^2x^2k^2+\frac{4\pi i}{h}4xk+2\right]\\
&=-\frac{h}{4\pi i}4xk-2\left(\frac{h}{4\pi i}\right)^2=4i\hbar xk+\frac{\hbar^2}{2}.
\end{aligned}\tag{A5}$$

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