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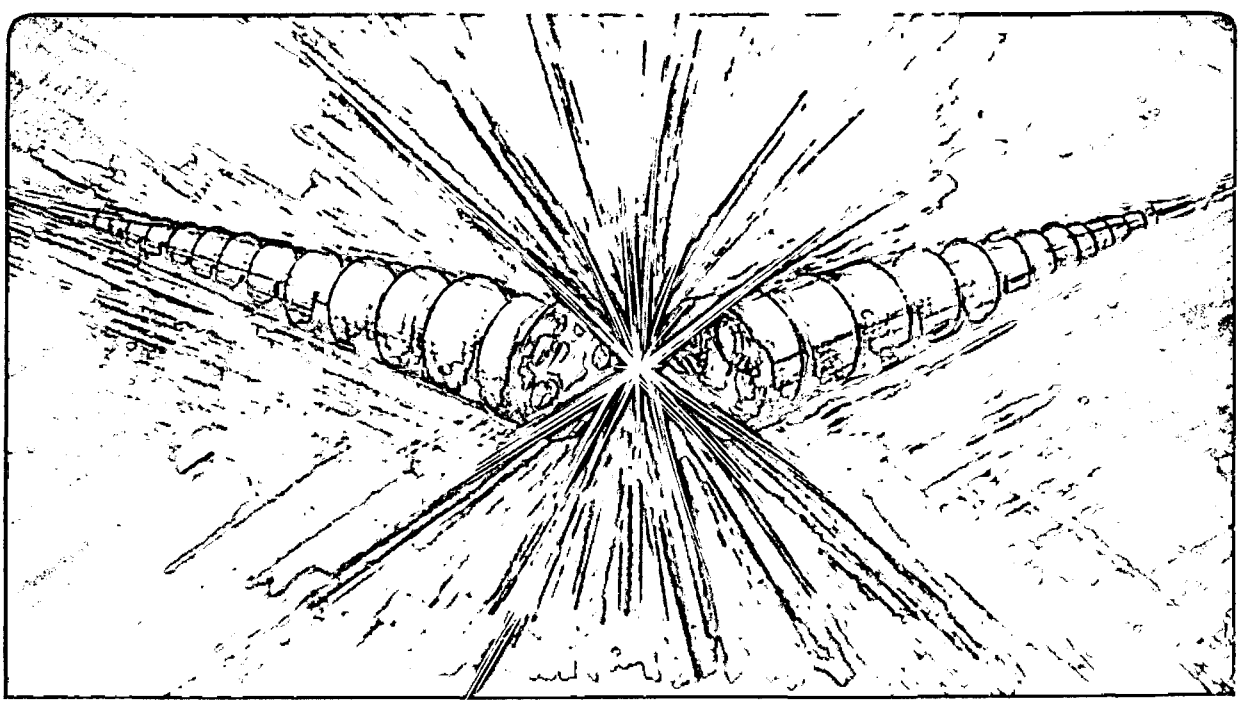

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Wave Packet Evolution and Quantization*

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Wave Packet Evolution and Quantization

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Abstract

A general relationship is established between wave packet evolution and quantization in classically integrable systems. Because of wave packet spreading, one cannot simply Fourier transform the time evolution of a wave packet. Instead, one must propagate the wave packet using the actions as Hamiltonians. The energy eigenvalues which result are the EBK values, and new forms for the eigenfunctions appear. These are free of caustic singularities, and represent averages of wave packets over the invariant torus.

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Wave packets provide methods for carrying out semiclassical calculations which have been attracting increasing attention in recent years. Especially notable are the theoretical and computational studies of Heller and coworkers,[1–5] who have used wave packets in a

number of chemical applications. Wave packet methods are appealing because they yield uniform results, they require no special attention at caustics, and they can be used in any number of dimensions. Nevertheless, they are somewhat less well developed theoretically than traditional approaches to WKB theory, which are usually based on an eikonal ansatz in configuration space. For example, the general relationship between the time-dependent problem of wave packet evolution and the time-independent problem of quantization has never been fully elucidated, even for classically integrable systems. In this Letter I shall address this question and remedy this shortcoming.

I base my analysis on a certain semiclassical wave packet propagator, which I have described in detail elsewhere.[6] When this propagator is applied to Gaussian initial states, it yields the same time evolution discovered originally by Heller.[1] I denote the $2N$ phase space coordinates collectively by $z = (q, p)$, and the corresponding quantum operators by $\hat{z} = (\hat{q}, \hat{p})$. I let $|\psi_0\rangle$ be some initial wave packet (not necessarily Gaussian), and I set $z_0 = \langle\psi_0|\hat{z}|\psi_0\rangle$ for the initial expectation values of $\hat{q}, \hat{p}$, which represent an initial point in the classical phase space. This serves as initial conditions for a classical trajectory $z(t)$ or $z(z_0; t)$, governed by the classical Hamiltonian $H(z)$, which is the Weyl symbol of the quantum Hamiltonian $\hat{H}(\hat{z})$. I shall also make use of the symplectic matrix $S_{ij}(t) = S_{ij}(z_0; t) = \partial z_i(z_0; t)/\partial z_{0j}$, which describes the behavior of orbits nearby the given trajectory $z(z_0; t)$.

In terms of these quantities, the wave packet propagator is

$$U(z_0; t) = e^{i\alpha(t) - iEt}\, T\big(z(t)\big) M\big(S(t)\big) T(z_0)^\dagger. \tag{1}$$

The T operators are Heisenberg-Weyl operators, defined by $T(z) = \exp[i(p \cdot \hat{q} - q \cdot \hat{p})]$. Throughout I set $\hbar = 1$. The energy E is that of the initial condition, $E = H(z_0)$, which is conserved along the classical orbit. The phase $\alpha(t)$ is a symmetrized Bohr-Sommerfeld phase,

$$\alpha(t) = \frac{1}{2} \int_{z_0}^{z(t)} p\,dq - q\,dp. \tag{2}$$

Finally, M is the metaplectic operator[7] which is responsible for wave packet spreading. The notation which suggests that M is a function of S is convenient but misleading, since there are actually two metaplectic operators for every symplectic matrix, differing by a sign. The choice of sign of $M\big(S(t)\big)$ is governed by two rules. First, at $t = 0$ we have $S(0) = I$ and $M\big(S(0)\big) = +1$. Next, $M\big(S(t)\big)$ is determined at all other times by continuity.

One can use Eq. (1) directly to advance wave packets in time, and thereby to solve initial value problems. For quantization problems, one might suppose that Eq. (1) could be Fourier transformed in time to obtain semiclassical energy eigenvalues and eigenstates. Although this approach does not work, I shall discuss it anyway for the useful features it reveals. For simplicity, I let the energy parameter of the Fourier transform be the same as the energy E of the classical orbit implied by Eq. (1), and I assume a discrete spectrum. I also let the propagator of Eq. (1) act on the state $|\psi_0\rangle = T(z_0)|0\rangle$, where $|0\rangle$ is some fiducial state[8] satisfying $\langle 0|\hat{z}|0\rangle = 0$. Then two Heisenberg operators cancel, and one is led to the following kind of limit:

$$\lim_{T \to \infty} \frac{1}{2T} \int_{-T}^{+T} dt\, e^{i\alpha(t)}\, T\big(z(t)\big) M\big(S(t)\big)\, |0\rangle. \tag{3}$$

This expression depends only on the initial condition z_0. On the basis of the properties of the exact propagator, one expects the limit to be nonzero only when $E = H(z_0)$ is an energy eigenvalue. In that case, the value of the limit should be an energy eigenstate.

Unfortunately, the limit (3) fails, in general, to yield sensible answers, due to the wave packet spreading governed by $M\big(S(t)\big)$. After a finite time, a wave packet will have spread so much that it can no longer be considered a wave packet, just as a localized bunch of particles will spread in classical mechanics. Furthermore, even if $z(t)$ is periodic, the integrand of (3) will not be, even to within a phase factor, again because of the spreading. The classical analog of this fact is that orbits which are nearby a given periodic orbit

do not usually have the same period as the given orbit, even when they themselves are periodic. Therefore, classical bunches of particles do not have periodic time behavior, even if individual particles do.

Nevertheless, one can extract semiclassical energy levels and eigenfunctions from the same kind of reasoning which went into the unworkable limit (3). The trick is to use the fact that if two operators commute, then they possess simultaneous eigenstates. Supposing that the classical motion is integrable, we can find action variables $I_1(z), \ldots, I_N(z)$, which have vanishing Poisson brackets with each other and with the Hamiltonian $H(z)$, i.e. $\{I_k, I_l\} = 0$ and $\{I_k, H\} = 0$. If these action variables are converted into quantum operators $\hat{I}_1, \ldots, \hat{I}_N$ by the Weyl correspondence, then, to within a relative error of $O(\hbar^2)$, we will have $[\hat{I}_k, \hat{I}_l] = 0$ and $[\hat{I}_k, \hat{H}] = 0$, in accordance with the Groenewold-Moyal[6] formula. Therefore, in a semiclassical sense, we expect the simultaneous eigenstates of the actions to exist, and to be simultaneously energy eigenstates. I shall denote these states by $|n_1, \ldots, n_N\rangle$, so that $\hat{I}_k|n_1, \ldots, n_N\rangle = I_{k,n_k}|n_1, \ldots, n_N\rangle$, where n_k is the quantum number of the operator $\hat{I}_k$.

These states are to be found by Fourier transforming the propagators corresponding to the N operators $\hat{I}_k$. Let us first pick one of these operators $\hat{I}_k$, for fixed k, and denote the corresponding parameter of evolution by λ, so that the exact propagator is $V_k(\lambda) = \exp(-i\lambda\hat{I}_k)$. This propagator is converted into a semiclassical, wave packet version,

$$V_k(z_0; \lambda) = e^{i\alpha(\lambda) - i\lambda J}\, T\big(z(\lambda)\big) M\big(S(\lambda)\big) T(z_0)^\dagger, \tag{4}$$

exactly as in Eq. (1), with H, t replaced by I_k, λ. The quantity J is $I_k(z_0)$, and $z(\lambda)$ is the orbit obtained by treating $I_k(z)$ as a Hamiltonian.

As before, Eq. (4) is applied to a state $|\psi_0\rangle = T(z_0)|0\rangle$, and Fourier transformed in λ, with the action parameter of the Fourier transform being taken, for simplicity, to be $J = I_k(z_0)$. Again, one is led to a limit,

$$\lim_{\Lambda \to \infty} \frac{1}{2\Lambda} \int_{-\Lambda}^{+\Lambda} d\lambda\, e^{i\alpha(\lambda)}\, T\big(z(\lambda)\big) M\big(S(\lambda)\big)\, |0\rangle. \tag{5}$$

Now, however, the limit does make sense. Action variables have the property that all orbits generated by them are periodic, with period $\lambda = 2\pi$. Furthermore, since the period is independent of initial conditions, nearby orbits have the same period as any given reference orbit, and therefore the symplectic matrix $S(\lambda) = \partial z(z_0;\lambda)/\partial z_0$ is itself periodic. Thus, we have $z(\lambda + 2\pi) = z(\lambda)$ and $S(\lambda + 2\pi) = S(\lambda)$. The metaplectic operator is periodic to within a phase of ± 1, so that $M\big(S(\lambda+2\pi)\big) = e^{-i\mu\pi/2}\, M\big(S(\lambda)\big)$, where the Maslov index[9] μ is an even integer. Finally, we have $\alpha(\lambda + 2\pi) = \alpha(\lambda) + 2\pi J$.

Therefore the integral of (5) over a single 2π period of the parameter λ is a simple phase factor times the integral over the previous period. This factor is $\exp(2\pi i J - \mu\pi i/2)$, and the limit (5) can be nonzero only if this factor is unity. Therefore we obtain a quantization condition,

$$I_{k,n_k} = n_k + \frac{\mu_k}{4}, \tag{6}$$

where I have identified J with the eigenvalue of $\hat{I}_k$, and subscripted the Maslov index, since it is a characteristic of the orbits generated by $I_k(z)$.

Equation (6) represents the EBK quantization conditions. Once the actions are quantized, the energy eigenvalues are determined by expressing the Hamiltonian as a function of the actions, $H = H(I_1, \ldots, I_N)$, and writing $E_{n_1,\ldots,n_N} = H(I_{1,n_1}, \ldots, I_{N,n_N})$. Thus, the wave packet evolution developed by Heller implicitly contains the EBK quantization conditions,[10] if only one propagates in the actions instead of the Hamiltonian. Although this is the same result obtained by eikonal approaches, one should note that no considerations of caustics have entered into this analysis. Furthermore, it is simpler and more stable numerically to determine the Maslov index directly from $S(\lambda)$ than it is to count caustics.

The energy eigenvalues are the same by this method as with the Keller-Maslov[11–12] method, but the eigenstates are different. Equation (5) taken over a single period gives an

eigenstate of $\hat{I}_k$, which I denote by $|n_k\rangle$,

$$|n_k\rangle = \int_0^{2\pi} d\lambda_k \, e^{i\alpha(z_0;\lambda_k)} \, T\big(z(z_0;\lambda_k)\big) M\big(S(z_0;\lambda_k)\big) \, |0\rangle, \tag{7}$$

where I have subscripted λ and carefully indicated dependencies. The initial conditions z_0 are chosen so that $I_k(z_0) = I_{k,n_k}$. The state $|n_k\rangle$ is an average of the wave packet $|0\rangle$ over the closed orbit of I_k passing through z_0, and its Wigner function is concentrated on this orbit in phase space, in the manner of a ring.

To find a simultaneous eigenstate of $\hat{I}_k$ and another action, say $\hat{I}_l$, denoted by $|n_k n_l\rangle$, we can apply the propagator $V_l(\lambda_l) = \exp(-i\lambda_l \hat{I}_l)$ to $|n_k\rangle$, and Fourier transform in λ_l. Although the ring state $|n_k\rangle$ is not a wave packet, the integrand of Eq. (7) is, so this exact propagator can be brought inside the integral and replaced by its semiclassical, wave packet equivalent. This is just as in Eq. (4), with k replaced by l and z_0 replaced by $z_1 = z(z_0;\lambda_k)$. The Heisenberg operator $T(z_1)^\dagger$ cancels with the Heisenberg operator in Eq. (7), bringing two metaplectic operators adjacent to each other. In the remaining Heisenberg operator, there appears the function $z(z_0;\lambda_k,\lambda_l) = z(z_1;\lambda_l) = z\big(z(z_0;\lambda_k);\lambda_l\big)$, which is the position in phase space one obtains by starting at z_0, following an I_k orbit for parameter value λ_k, then an I_l orbit for parameter value λ_l. Since $\{I_k, I_l\} = 0$, these propagations can be applied in either order, with no effect on the final point. The 2-dimensional surface swept out by varying λ_k, λ_l is a 2-torus. It is also an "isotropic" submanifold,[13] which means that the line integral for $\alpha(\lambda)$, with the same integrand as in Eq. (2), is invariant with respect to continuous deformations of the path on this surface. Therefore $\alpha(z_1;\lambda_l) + \alpha(z_0;\lambda_k)$ depends only on the endpoints, and can be written $\alpha(z_0;\lambda_k,\lambda_l)$. Finally, the two metaplectic operators can be combined, by using the rule $M(S_1)M(S_2) = M(S_1 S_2)$. The product of the two symplectic matrices which results is just the derivative $\partial z(z_0;\lambda_k,\lambda_l)/\partial z_0$, which I denote by $S(z_0;\lambda_k,\lambda_l)$. Altogether, one obtains a state which is an average of the wave packet $|0\rangle$ over the 2-torus in phase space,

$$|n_k n_l\rangle = \int_0^{2\pi} \int_0^{2\pi} d\lambda_k \, d\lambda_l \, e^{i\alpha(z_0;\lambda_k,\lambda_l)} \, T\big(z(z_0;\lambda_k,\lambda_l)\big) M\big(S(z_0;\lambda_k,\lambda_l)\big) \, |0\rangle, \tag{8}$$

where now z_0 is assumed to lie on quantizing contours of both I_k and I_l.

Proceeding in this manner, we can finally construct the simultaneous eigenstates of all the actions, as an average of a wave packet over the invariant N-torus. It is

$$|n_1, \ldots, n_N\rangle = \int d^N\lambda \, e^{i\alpha(z_0;\lambda)} \, T\big(z(z_0;\lambda)\big) M\big(S(z_0;\lambda)\big) \, |0\rangle, \tag{9}$$

where now λ represents $(\lambda_1, \ldots, \lambda_N)$. This state is quite similar to some semiclassical eigenstates produced by Davis, DeLeon and Heller,[2–5] although it is free of certain *ad hoc* elements present in those studies. If analytic expressions are known for the action-angle variables, such as can sometimes be obtained by Lie transforms, then it would be straightforward to evaluate (9) directly. On the other hand, if analytic expressions for the actions are not known, it still appears to be practical to evaluate the symplectic matrix $S(z_0;\lambda)$ numerically and to replace the λ integral by a time integral over an ergodic orbit.

There are several significant aspects to these results. First, the theoretical foundation of wave packet techniques is extended. Second, a deeper understanding is provided for some of the computational successes of Heller and coworkers, especially those using "frozen Gaussians".[5] Third, the basis is laid for a semiclasical theory of symmetries and invariants, since little of the analysis above has depended on there being a complete set of actions in involution. Less than complete sets often occur in applications, as in systems with rotational symmetry, or with some degrees of freedom which possess good KAM tori, and others which do not. I will elaborate on the analysis above and further explore these issues in subsequent publications.

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