# Wave-Packet Evolution and Quantization

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A general relationship is established between wave-packet evolution and quantization in classically integrable systems. Because of wave-packet spreading, one cannot simply take the Fourier transform of 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 Einstein-Brillouin-Keller 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 co-workers,[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 the Wentzel-Kramers-Brillouin 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 near the given trajectory $z(z_0;t)$.

In terms of these quantities, the wave-packet propagator is

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

The $T$ operators are Heisenberg-Weyl operators, defined by $T(z)=\exp[i(p\cdot\hat{p}-q\cdot\hat{p})]$. 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,

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

Finally, $M$ is the metaplectic operator[7] which is responsible for wave-packet spreading. [The metaplectic operators comprise a projective unitary representation of the classical symplectic group, $\mathrm{Sp}(2N)$. Their role in wave-packet evolution parallels that of the symplectic matrices in the evolution of localized distributions in phase space, i.e., in describing the linearized dynamics near some reference orbit.] 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 $M(S(t))$ is governed by two rules. First, at $t=0$ we have $S(0)=I$ and $M(S(0))=+1$. Next, $M(S(t))$ 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-wave 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] (basically, any conveniently chosen wave packet) satisfying $\langle 0|\hat{z}|0\rangle=0$. Then two Heisenberg

 

operators cancel, and one is led to the following kind of limit:

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

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, because of the wave-packet spreading governed by $M(S(t))$. 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 near 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 transformation of 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 $\lambda$, 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,

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

exactly as in Eq. (1), with $H,t$ replaced by $I_k,\lambda$. 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 $\lambda$, 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,

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

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 $\pm1$, so that $M(S(\lambda+2\pi))=e^{-i\mu\pi/2}M(S(\lambda))$, where the Maslov index[9] $\mu$ is an even integer. [Here the Maslov index is twice the homotopy class of a $2\pi$ period of $S(\lambda)$ in $\pi_1(\mathrm{Sp}(2N))$.] Finally, we have $\alpha(\lambda+2\pi)=\alpha(\lambda)+2\pi J$.

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

$$I_{k,n_k}=n_k+\mu_k/4, \quad (6)$$

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 Einstein-Brillouin-Keller 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 Einstein-Brillouin-Keller 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 $\hat{I}_k$, denoted by $|n_k\rangle$,

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

where I have subscripted $\lambda$ 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 take the Fourier transform in $\lambda_l$. Although the ring state $|n_k\rangle$ is not a wave packet, the integrand of Eq. (7) is, and 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

$$z(z_0;\lambda_k,\lambda_l) = z(z_1;\lambda_l) = z(z(z_0;\lambda_k);\lambda_l),$$

which is the position in phase space one obtains by starting at $z_0$, following an $I_k$ orbit for parameter value $\lambda_k$, then an $I_l$ orbit for parameter value $\lambda_l$. Since $\{I_k, I_l\} = 0$, these propagations can be applied in either order, with no effect on the final point. The two-dimensional surface swept out by varying $\lambda_k, \lambda_l$ is a two-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 end points, and can be written $\alpha(z_0;\lambda_k,\lambda_l)$. Finally, the two metaplectic operators can be combined, by use of 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 two-torus in phase space,

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

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

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

where now $\lambda$ represents $(\lambda_1, \ldots, \lambda_N)$. This state is free of caustic singularities. Its wave function is a uniform approximation to the exact energy eigenfunction, with the single formula (9) being valid at all points of configuration space, including caustic and evanescent regions. It is quite similar to some semiclassical eigenstates produced by Davis, DeLeon, and Heller,[2-5] and differs primarily in its treatment of wave-packet spreading and the Maslov phase shift. Those authors, recognizing that wave-packet spreading would make the formula (3) unworkable, artificially "froze" their wave packets, even though such a step is not directly justified on dynamical grounds. They also employed various reasonable but essentially *ad hoc* methods for incorporating the Maslov phase shifts. On the other hand, the wave packets in Eq. (9) follow the dynamics dictated by the action variables, with no further assumptions required. Thus, their spreading is quasiperiodic, and the Maslov phase shift is automatically incorporated.

The eigenstate (9) depends only weakly on the fiducial state $|0\rangle$, as long as the latter is reasonably chosen. For example, the approximation inherent in the propagator (1) will be invalidated if the Wigner function of $|0\rangle$ is very long and thin in phase space.

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 co-workers, especially those using "frozen Gaussians."[5] Third, the basis is laid for a semiclassical 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 Kolomogorov-Arnol'd-Moser 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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