ARTICLES

Closed orbits and shell effects in semiclassical response functions

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The Vlasov kinetic equation can be viewed as the dynamical equivalent of the static Thomas-Fermi theory. By exploiting this analogy, the closed-orbit theory of the level density, which has been used by various authors for evaluating quantum corrections to the Thomas-Fermi level density, is generalized for calculating quantum corrections to the semiclassical propagator given by the Vlasov method. It is shown that the Vlasov strength function obeys a sum rule analogous to the well-known quantum energy-weighted sum rule. Explicit calculations of quantum corrections are performed for a simplified model of a large metal cluster (electrons in a spherical cavity).

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I. INTRODUCTION

The Vlasov equation has long been known to give a classical version of the random-phase approximation (see, for example, [1], p. 71). The linearized Vlasov equation gives an integral equation for the classical limit of the particle-hole propagator that is analogous to the RPA integral equation; symbolically

$$D=D^0+D^0uD \ . \tag{1.1}$$

Here D is the classical limit of the RPA propagator, D^0 the classical propagator in the static mean field, and u the two-body interaction between constituents of the many-body system being studied. Generally, Eq. (1.1) is an integral equation; it becomes an algebraic equation only for infinite homogeneous systems. The solution of (1.1) is a numerical problem that requires the zero-order propagator D^0 as an input. An important feature of the Vlasov equation is that it yields an explicit expression for the zero-order propagator D^0. Being entirely expressed in terms of classical quantities, this form of D^0 is more intuitive than the corresponding quantum propagator.

Bertsch [2] has pointed out that, since the Vlasov equation is just the Liouville equation of classical statistical mechanics with a self-consistent mean field, it can be applied also to quantum many-fermion systems because the Liouville theorem ensures that the Pauli principle can be respected within this classical theory. Thus the Vlasov theory is a reasonable starting point for the study of the dynamics of Fermi systems in the semiclassical regime.

Kirzhnitz, Lozovik, and Shpatakovskaya [3] have applied the Vlasov theory to nonhomogeneous systems [in this case Eq. (1.1) is an integral equation]. The expression that they derived for the classical zero-order propagator D^0 requires integrating the single-particle motion over an infinite time; hence it is not convenient for practical calculations.

In [4] it was pointed out that there is a whole class of systems for which the solution of the linearized Vlasov equation has a relatively simple form: all the systems for which the equilibrium mean-field Hamiltonian $h_0(\mathbf{r},\mathbf{p})$ is separable. In this case it is convenient to use angle-action variables for describing the single-particle motion and the zero-order propagator can be written explicitly as [4] (in momentum space)

$$D^0(\mathbf{q}',\mathbf{q},\omega)=(2\pi)^3\sum_{\mathbf{n}}\int d\mathbf{I}\,F'(h_0(\mathbf{I}))\frac{\mathbf{n}\cdot\boldsymbol{\omega}(\mathbf{I})}{\mathbf{n}\cdot\boldsymbol{\omega}(\mathbf{I})-(\omega+i\eta)} \times Q_{\mathbf{n}}^*(\mathbf{q}',\mathbf{I})Q_{\mathbf{n}}(\mathbf{q},\mathbf{I}) \ . \tag{1.2}$$

Here $\mathbf{I}$ denotes the action variables associated with the equilibrium Hamiltonian $h_0(\mathbf{I})$; they are related to the action variables $\mathbf{J}$ used in [4] by $\mathbf{I}=\mathbf{J}/(2\pi)$. The canonically conjugate angle variables Φ_α take values in the interval $[0,2\pi]$. The Fourier coefficients $Q_{\mathbf{n}}(\mathbf{q},\mathbf{I})$ are defined as

$$Q_{\mathbf{n}}(\mathbf{q},\mathbf{I})=\frac{1}{(2\pi)^3}\int d\boldsymbol{\Phi}e^{-i\mathbf{n}\cdot\boldsymbol{\Phi}}e^{i\mathbf{q}\cdot\mathbf{r}} \tag{1.3}$$

and they replace the quantum matrix elements of $e^{i\mathbf{q}\cdot\mathbf{r}}$ [5].

In Eq. (1.2) the vector $\mathbf{n}$ has integer components $n_\alpha=0,\pm1,\pm2,\ldots$, while the vector $\boldsymbol{\omega}(\mathbf{I})$, defined as

$$\boldsymbol{\omega}(\mathbf{I})=\nabla_I h_0(\mathbf{I}) \ , \tag{1.4}$$

gives the proper frequencies of the system. Generally, the value of these frequencies depends on the action vari-

ables so that the integration over $\mathbf{I}$ in Eq. (1.2) gives a spreading of the strength as ω changes with $\mathbf{I}$. Since the parameter η in (1.2) is infinitesimally small, the imaginary part of D^0 contains δ functions of the type $\delta(\omega-\mathbf{n}\cdot\omega(\mathbf{I}))$. If η is given a small but finite value, these δ functions are smeared out into Lorentzians of width η and the single-particle strength function becomes smoother.

The function $F'(h_0(\mathbf{I}))$ is related to the equilibrium distribution of particles in phase space. For zero-temperature electrons we take [4]

$$f_0(\mathbf{r},\mathbf{p})\equiv F(h_0)=\frac{2}{(2\pi\hbar)^3}\theta(E_F-h_0)\ , \tag{1.5a}$$

so that

$$F'(h_0)=\frac{-2}{(2\pi\hbar)^3}\delta(E_F-h_0)\ . \tag{1.5b}$$

E_F is the Fermi energy, which, in a first approximation, must be determined from the normalization condition

$$\int d\mathbf{r}\,d\mathbf{p}\frac{2}{(2\pi\hbar)^3}\theta(E_F-h_0(\mathbf{r},\mathbf{p}))=N\ , \tag{1.6}$$

and N is the total number of electrons.

Equation (1.2) for the zero-order propagator is reminiscent of the classical dispersion formula of the days before quantum mechanics (see [6], p. 31, and the references cited therein). The minor differences between the two formulas come from the different physical situation to which they are applied (here we deal with a distribution of particles in the ground state of a many-body system, while the dispersion formula usually refers to a single particle in a hydrogenic orbit).

Before concluding this section we write explicitly the RPA integral equation (1.1) in momentum space:

$$D(\mathbf{q}',\mathbf{q},\omega)=D^0(\mathbf{q}',\mathbf{q},\omega)+\frac{1}{(2\pi)^3}\int d\mathbf{k}\,D^0(\mathbf{q}',\mathbf{k},\omega)u(k)D(\mathbf{k},\mathbf{q},\omega)\ . \tag{1.7}$$

Here $u(k)$ is the Fourier transform of the two-body interaction $u(|\mathbf{x}_1-\mathbf{x}_2|)$.

The strength function describing the collective response of the system is given by

$$S(\mathbf{q},\omega)=-\frac{1}{\pi}\,\mathrm{Im}D(\mathbf{q},\mathbf{q},\omega)\ . \tag{1.8}$$

In situations where exact quantum calculations are not possible (for example, if the system is too large), Eq. (1.7) offers an interesting alternative to the fully quantum RPA calculations. Moreover, the physical insight allowed us by this classical approach would be difficult to gain in a quantum RPA calculation.

Equation (1.7) has been applied to the study of giant resonances in nuclei [7], surface plasmon in atomic clusters [8], and also to the search for possible collective excitations of the electrons in heavy atoms [9]. On the whole, the agreement with quantum RPA calculations is rather good, even when the quantum numbers involved are not particularly large.

The question of possible quantum corrections to the classical response has received some attention in the literature [3,4,10–14]. Here we study this problem from the point of view of the closed-orbit theory of the level density [15–17] (see also [18], p. 579, and [19], pp. 257 and 266). As we shall see, it is possible to establish a close analogy between the quantum corrections to Eq. (1.2) and the corrections to the Thomas-Fermi semiclassical level density.

II. SEMICLASSICAL PROPAGATOR

The *retarded* quantum propagator describing the single-particle response to an external field $Q(\mathbf{r})$ can be written as [20]

$$\Pi^{0R}(\omega)=\sum_{\mathbf{a},\mathbf{b}}\left\{\frac{[1-\theta(E_F-E_\mathbf{b})]\theta(E_F-E_\mathbf{a})}{\hbar\omega+E_\mathbf{a}-E_\mathbf{b}+i\eta}-\frac{\theta(E_F-E_\mathbf{b})[1-\theta(E_F-E_\mathbf{a})]}{\hbar\omega+E_\mathbf{a}-E_\mathbf{b}+i\eta}\right\}\times|\langle\mathbf{a}|Q|\mathbf{b}\rangle|^2\ , \tag{2.1}$$

where $\mathbf{a}$ and $\mathbf{b}$ are the sets of quantum numbers required in order to identify the single-particle states. By combining the two terms in (2.1), Π^{0R} can be written in a form that involves the difference between occupation probabilities of states $E_\mathbf{a}$ and $E_\mathbf{b}$:

$$\Pi^{0R}(\omega)=\sum_{\mathbf{a},\mathbf{b}}\frac{\theta(E_F-E_\mathbf{a})-\theta(E_F-E_\mathbf{b})}{\hbar\omega+E_\mathbf{a}-E_\mathbf{b}+i\eta}|\langle\mathbf{a}|Q|\mathbf{b}\rangle|^2\ . \tag{2.2}$$

In a semiclassical situation the matrix element $\langle\mathbf{a}|Q|\mathbf{b}\rangle$ depends mainly on the difference of quantum numbers $\mathbf{a}$ and $\mathbf{b}$ [21]. In the limit of large quantum numbers the WKB approximation to the quantum matrix element $\langle\mathbf{a}|Q|\mathbf{b}\rangle$ becomes the Fourier coefficient (1.3), with $Q(\mathbf{r})$ replacing $e^{i\mathbf{q}\cdot\mathbf{r}}$ (see [22], p. 167, and [10]).

Now we set

$$\mathbf{I}_b=\mathbf{I}_a+\mathbf{n}\hbar\ , \tag{2.3}$$

and write (2.2) as

$$\Pi^{0R}(\omega)\approx(2\pi)^3\int d\mathbf{I}_a\sum_\mathbf{n}\frac{F(h_0(\mathbf{I}_a+\mathbf{n}\hbar))-F(h_0(\mathbf{I}_a))}{h_0(\mathbf{I}_a+\mathbf{n}\hbar)-h_0(\mathbf{I}_a)-(\hbar\omega+i\eta)}\times|Q_\mathbf{n}(\mathbf{I}_a)|^2\ . \tag{2.4}$$

Moreover, using ([18], p. 579)

$$h_0(\mathbf{I}_a+\mathbf{n}\hbar)-h_0(\mathbf{I}_a)\approx\hbar\mathbf{n}\cdot(\nabla_I h_0)_{\mathbf{I}=\mathbf{I}_a} \tag{2.5}$$

and

$$F(h_0(\mathbf{I}_a+\mathbf{n}\hbar))-F(h_0(\mathbf{I}_a))\approx F'(h_0)\hbar\mathbf{n}\cdot\nabla_I h_0\ , \tag{2.6}$$

we have

$$\Pi^{0R}(\omega)\approx(2\pi)^3\int d\mathbf{I}_a\sum_\mathbf{n}\frac{F'(h_0(\mathbf{I}_a))\mathbf{n}\cdot\omega(\mathbf{I}_a)}{\mathbf{n}\cdot\omega(\mathbf{I}_a)-(\omega+i\eta)}|Q_\mathbf{n}(\mathbf{I}_a)|^2\ . \tag{2.7}$$

Thus we have proved that, in the limit of very large quan-

tum numbers, the retarded quantum single-particle propagator (2.1) equals the classical propagator (1.2) given by the Vlasov equation. In what follows we are concerned with the problem of evaluating quantum corrections to (2.7) when shell effects are not completely negligible.

III. CLOSED-ORBIT THEORY

The closed-orbit theory of the level density [15–19] has recently scored a remarkable success when its prediction [23] of supershell effects in atomic clusters has been confirmed experimentally [24] by an analysis of the mass spectrum of sodium clusters. Here we follow closely Berry and Tabor [17] in order to establish a parallel between the calculation of quantum corrections to the semiclassical Thomas-Fermi level density and the problem of evaluating quantum corrections to the semiclassical Vlasov propagator (1.2). Thus we first briefly recall the approach of [17] to the quantum corrections of the level density. The quantum level density, defined as

$$n(E)=\sum_{\mathrm{m}}\delta(E-E_{\mathrm{m}})\ , \tag{3.1}$$

is first transformed into an infinite sum of integrals by means of the Poisson sum formula ([25], p. 52). The expression given by Berry and Tabor reads

$$n(E)=\frac{1}{\hbar^3}\sum_{\mathbf{M}}e^{-i(\pi/2)\alpha\cdot\mathbf{M}}\int_{+}d\mathbf{I}\,\delta(E-h_0(\mathbf{I}))e^{i(2\pi/\hbar)\mathbf{M}\cdot\mathbf{I}}\ , \tag{3.2}$$

with the integral extending over the positive quadrant in $\mathbf{I}$ space.

This is a general formula that requires some further specifications for a central potential. Because of rotation symmetry, the quantum levels of a spherical potential can be classified according to the two quantum numbers $n_r=0,1,2,\ldots$ and $l=0,1,2,\ldots$; thus

$$n(E)=\sum_{n_r,l,m}\delta(E-E_{n_r,l})=\sum_{n_r,l}(2l+1)\delta(E-E_{n_r,l})\ . \tag{3.3}$$

Approximate semiclassical values of the quantum energy levels are

$$E_{n_r,l}\approx h_0(\lambda=(l+\tfrac{1}{2})\hbar,I_r=(n_r+\alpha_r/4)\hbar)\ , \tag{3.4}$$

where λ is the classical angular momentum and I_r the radial action. For a cavity, the Maslov index $\alpha_r=3$, while for a smooth potential such as a harmonic oscillator or a Saxon-Woods, $\alpha_r=2$. Using the Poisson formula twice, the level density (3.3) can be written as

$$n(E)=\frac{1}{\hbar^3}\sum_{M_L=-\infty}^{+\infty}\sum_{M_S=-\infty}^{+\infty}\int_0^{\infty}d\lambda\int_0^{\infty}dI_r\int_{-\lambda}^{+\lambda}d\lambda_z e^{i(2\pi/\hbar)[M_L(\lambda-\hbar/2)+M_S(I_r-\alpha_r\hbar/4)]}\delta(E-h_0(\lambda,I_r))\ . \tag{3.5}$$

If we use the action variables (A1), this expression can be put in the form (3.2), provided that the first component of the vector $\mathbf{M}$ vanishes: $\mathbf{M}=(0,M_2,M_3)$. In other words we only need to quantize the "proper" actions I_2 and I_3 (or I_r) ([26], p. 383). The term with $\mathbf{M}=(0,0,0)$ gives the semiclassical Thomas-Fermi result

$$n_{\mathrm{TF}}(E)=\frac{1}{\hbar^3}\int d\mathbf{I}\,\delta(E-h_0(\mathbf{I}))\ , \tag{3.6}$$

while the remaining terms give quantum corrections to (3.6) [27]. Because of the δ function in the integrand, the oscillatory integrals

$$n_{\mathbf{M}}=\frac{1}{\hbar^3}\int d\mathbf{I}\,\delta(E-h_0(\mathbf{I}))e^{i(2\pi/\hbar)\mathbf{M}\cdot\mathbf{I}} \tag{3.7}$$

are of the kind $\int d\alpha\, g(\alpha)e^{i\psi(\alpha)}$ and can be evaluated by using the stationary phase approximation ([25], p. 192). The integration in (3.6) and (3.7) is an integration over all possible classical orbits with energy E. Within the stationary-phase approximation the main contribution to the integrals (3.7) comes from closed classical orbits, that is, orbits for which the components of the frequency vector (1.4) are in the ratio of two integers. These orbits correspond to particular values $\mathbf{I}_{\mathbf{M}}$ of the action vector. Then, within the stationary-phase approximation, Eq. (3.2) gives the following expansion for the level density of a separable Hamiltonian:

$$n(E)\approx n_{\mathrm{TF}}(E)+\sum_{\mathbf{M}}{}'\,A_{\mathbf{M}}e^{i[(2\pi/\hbar)\mathbf{M}\cdot\mathbf{I}_{\mathbf{M}}+\varphi_{\mathbf{M}}]} \tag{3.8}$$

(the prime on the summation means that it must exclude the term $\mathbf{M}=\mathbf{0}$; for spherically symmetric systems, the sum is only over M_2 and M_3). The Thomas-Fermi density $n_{\mathrm{TF}}(E)$ gives a smooth background that does not contribute to the shell structure, while the additional terms are oscillating functions of E that give rise to the shell and supershell structures in $n(E)$. The factors $A_{\mathbf{M}}$ give the amplitude of these contributions, while the phases $\varphi_{\mathbf{M}}$ contain both the Maslov indices and a contribution coming from the stationary-phase formula.

The closed-orbit theory of the level density can be extended to the response function of a system of zero-temperature electrons. In fact, using Eqs. (2.7) and (1.5b), we have, for the zero-order propagator,

$$\Pi^{0R}(\omega)=\frac{1}{\hbar^3}\sum_{\mathbf{M}}e^{-i(\pi/2)\alpha\cdot\mathbf{M}}\int d\mathbf{I}\,\delta(E_F-h_0(\mathbf{I}))\times f(\mathbf{I},\omega)e^{i(2\pi/\hbar)\mathbf{M}\cdot\mathbf{I}}\ , \tag{3.9}$$

with, from Eq. (1.2),

$$f(\mathbf{I},\omega)=-2\sum_{\mathbf{n}}\frac{\mathbf{n}\cdot\omega(\mathbf{I})}{\mathbf{n}\cdot\omega(\mathbf{I})-(\omega+i\eta)}|Q_{\mathbf{n}}(\mathbf{I})|^2\ . \tag{3.10}$$

Again, for spherically symmetric systems, we only need to quantize the proper actions I_2 and I_3. The term with $\mathbf{M}=\mathbf{0}$ in (3.9), which is analogous to the volume term of the Thomas-Fermi level density, is just the Vlasov propagator (1.2), while the remaining terms represent quantum corrections to the classical propagator given by the Vlasov equation.

Now, however, we are not allowed to use immediately the stationary-phase approximation in order to evaluate the oscillatory integrals in (3.9) because $f(\mathbf{I},\omega)$ is not a smooth function of $\mathbf{I}$. If we give a finite value to the parameter η in (3.10), then $f(\mathbf{I},\omega,\eta)$ is smooth. We can follow all the steps that lead to Eq. (3.8) and get, for the zero-order propagator,

$$\Pi^{0R}(\omega,\eta)\approx D^0(\omega,\eta)+\sum_{\mathbf{M}}{}' A_{\mathbf{M}} e^{i[(2\pi/\hbar)\mathbf{M}\cdot\mathbf{I}_{\mathbf{M}}+\varphi_{\mathbf{M}}]} \times f(\mathbf{I}_{\mathbf{M}},\omega,\eta) \ . \tag{3.11}$$

Here $D^0(\omega,\eta)$ is the Vlasov propagator (1.2), evaluated with a small but finite value of η. In practice $\hbar\eta$ should be of the order of the energy difference between two neighboring levels of the quantum system at the Fermi surface. The smoothing factor η, which is necessary in order to be allowed to apply the stationary phase approximation to the oscillatory integrals in (3.9), should not be confused with the smoothing factor γ introduced by Balian and Bloch [16] in order to damp the contribution of longer orbits.

Equation (3.11) is the main result of the present paper. It has several interesting features. First, the Vlasov propagator (1.2) gives a classical approximation to the (retarded) quantum single-particle propagator Π^{0R}, which is analogous to the Thomas-Fermi approximation for the quantum level density. Hence the Vlasov theory can be viewed as a dynamical generalization of the static Thomas-Fermi approach [29]. Second, in the case of zero-temperature fermions, quantum corrections to the Vlasov propagator can be evaluated by using the closed-orbit theory developed for the level density, but only if a sufficiently large smoothing factor η is introduced into (3.10). The last point worth noting is that the function $f(\mathbf{I}_{\mathbf{M}},\omega,\eta)$ in (3.11) depends only on the topology of a given closed orbit and not on repetitions of the same orbit.

IV. SUM RULE

The quantum strength function

$$S^0(\omega)=-\frac{1}{\pi}\,\mathrm{Im}\Pi^{0R}(\omega) \tag{4.1}$$

satisfies the well-known energy weighted sum rule

$$\int_0^\infty d\,\hbar\omega(\hbar\omega)S^0(\omega)=\sum_n (E_n-E_0)|\langle n|Q|0\rangle|^2 =\tfrac{1}{2}\langle 0|[Q^\dagger,[h_0,Q]]|0\rangle \ . \tag{4.2}$$

In order to determine the constraints on the quantum corrections due to this sum rule, we prove that the classical strength function given by the Vlasov propagator (1.2) satisfies a sum rule that is the classical version of (4.2):

$$\int_0^\infty d\,\hbar\omega(\hbar\omega)S^0_{\mathrm{Vlasov}}(\omega) =-\frac{\hbar^2}{2}\int d\mathbf{r}\,d\mathbf{p}\,f_0(\mathbf{r},\mathbf{p})\{Q^*,\{h_0(\mathbf{r},\mathbf{p}),Q\}\}_{r,p}, \tag{4.3}$$

with Poisson brackets replacing the commutators in (4.2) and the averaging over the equilibrium distribution (1.5a) replacing the ground-state expectation value.

Because we are dealing with separable systems, we use angle-action variables on the right-hand side of (4.3)

$$\int d\mathbf{r}\,d\mathbf{p}f_0(\mathbf{r},\mathbf{p})\{Q^*,\{h_0(\mathbf{r},\mathbf{p}),Q\}\}_{r,p} =\int d\mathbf{I}\,d\Phi\,F(h_0(\mathbf{I}))\{Q^*,\{h_0(\mathbf{I}),Q\}\}_{I,\Phi} \tag{4.4}$$

and the expansion

$$Q(\mathbf{r})=\sum_{\mathbf{n}} e^{i\mathbf{n}\cdot\Phi}Q_{\mathbf{n}}(\mathbf{I}) \ . \tag{4.5}$$

Since h_0 depends only on the action variables

$$\{h_0,Q\}_{I,\Phi}=-i\sum_{\mathbf{n}}(\mathbf{n}\cdot\boldsymbol{\omega})e^{i\mathbf{n}\cdot\Phi}Q_{\mathbf{n}}(\mathbf{I}) \ , \tag{4.6}$$

where $\boldsymbol{\omega}$ is the frequency vector defined in (1.4).

Working out the second Poisson bracket, with the help of Eq. (4.5), of its complex conjugate and of the orthogonality relation

$$\int d\Phi\, e^{i(\mathbf{n}-\mathbf{m})\cdot\Phi}=(2\pi)^3\delta_{\mathbf{n},\mathbf{m}} \ , \tag{4.7}$$

we get

$$\int d\mathbf{I}\,d\Phi\,F(h_0(\mathbf{I}))\{Q^*,\{h_0(\mathbf{I}),Q\}\} =\int d\mathbf{I}\,F(h_0(\mathbf{I}))\nabla_i\cdot\sum_{\mathbf{n}}\mathbf{n}[\mathbf{n}\cdot\boldsymbol{\omega}(\mathbf{I})]|Q_{\mathbf{n}}(\mathbf{I})|^2 \ . \tag{4.8}$$

The integration on the right-hand side can be performed by parts and, under the reasonable assumption that $F(h_0(\mathbf{I}))$ vanishes for very large values of I, we get

$$\int d\mathbf{I}\,d\Phi\,F'(h_0(\mathbf{I}))\{Q^*,(\mathbf{I}),Q\}\}_{I,\Phi} =-\sum_{\mathbf{n}}\int d\mathbf{I}\,F'(h_0)(\mathbf{n}\cdot\boldsymbol{\omega})^2|Q_{\mathbf{n}}(\mathbf{I})|^2 \ , \tag{4.9}$$

which is proportional to the Vlasov energy-weighted strength function since, from (1.2),

$$\int_{-\infty}^{+\infty} d\,\hbar\omega(\hbar\omega)\left[-\frac{1}{\pi}\,\mathrm{Im}D^0(\mathbf{q},\mathbf{q},\omega)\right] =\hbar^2\sum_{\mathbf{n}}\int d\mathbf{I}\,F'(h_0)(\mathbf{n}\cdot\boldsymbol{\omega})^2|Q_{\mathbf{n}}|^2 \ . \tag{4.10}$$

For the dipole operator, the right-hand side of Eqs. (4.2) and (4.3) are identical so that the Vlasov strength function satisfies the same sum rule as the quantum strength function. Then it is reasonable to expect that the quantum corrections to the Vlasov strength function should preserve the dipole sum rule.

V. SPHERICAL CAVITY

As an illustration of the theory discussed in the previous sections, we study here a simplified model for a sodi-

um cluster. We consider a gas of $N=254$ electrons occupying the lowest quantum levels of a spherical cavity of radius

$$R=r_S N^{1/3} , \tag{5.1}$$

with $r_S=4a_0$ for sodium (a_0 is the Bohr radius). The quantum energy levels are

$$E_{n_r l}=\frac{\hbar^2}{2m}k_{n_r l}^2 \approx X_{n_r l}\times 0.0212 \text{ eV} , \tag{5.2}$$

where $X_{n_r l}$ is the n_rth zero of the spherical Bessel function $j_l(x)$, excluding the origin. The highest occupied state is a $2h$ level ($n_r=2, l=5$), at an energy $E_{25}\approx 3.564$ eV. The sequence of levels near the Fermi surface is shown in Fig. 1.

The quantum single-particle strength function is a sum of δ functions corresponding to transitions between the various quantum levels. The main contributions to the dipole single-particle strength function come from transitions $\Delta n_r=0$, $\Delta l=1$ between the states shown in Fig. 1. These transitions exhaust about 76% of the energy-weighted sum rule. In Fig. 2 we show the quantum dipole single-particle strength function, smoothed according to the replacement

$$\delta(E-E_n)\rightarrow -\frac{1}{\pi}\,\mathrm{Im}\frac{1}{E-E_n+i\eta} . \tag{5.3}$$

Also shown in Fig. 2 is the corresponding Vlasov strength function, smoothed in the same way.

The calculation of the Vlasov zero-order propagator requires the determination of the Fermi energy as a preliminary step. A first estimate of the Fermi energy is obtained using Eq. (1.6), which is equivalent to taking the approximate level density

$$n(E)\approx n_{\mathrm{TF}}(E)=\frac{1}{3\pi E}x^3 , \tag{5.4}$$

with $x\equiv(1/\hbar)\sqrt{2mE}\,R$. The Vlasov strength function shown in Fig. 2 (dotted line) has been obtained with the Fermi energy calculated in this way. However, since we want to include quantum corrections into the Vlasov propagator, in view of the parallel established in Sec. III, it seems desirable to keep the same degree of approximation in both the level density and the semiclassical propagator. This is because quantum corrections affect the Vlasov propagator in at least two ways: through the additional terms in (3.8), which modify the Fermi energy evaluated using (5.4), as well as more directly through the additional terms in (3.11). Thus we need to define more explicitly the degree of approximation that we intend to keep in both (3.8) and (3.11).

In a cavity the term $(2\pi/\hbar)\mathbf{M}\cdot\mathbf{I}_{\mathbf{M}}$ in the exponent of

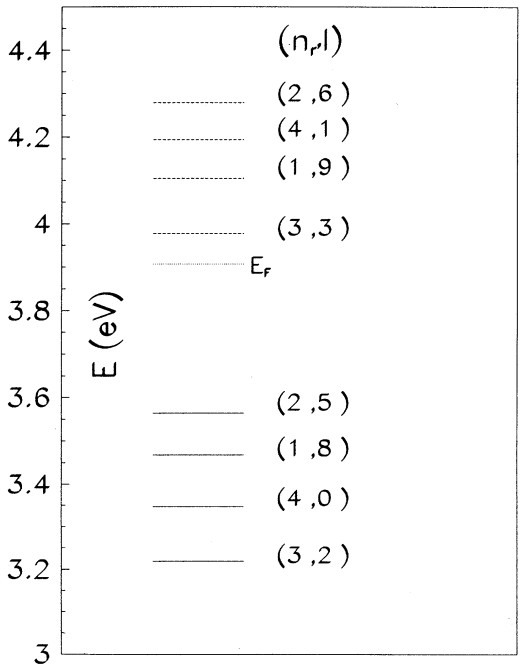

FIG. 1. Highest occupied (solid lines) and lowest unoccupied (dashed lines) levels in a spherical cavity containing 254 zero-temperature electrons. The dotted line shows the value of the Fermi energy given by Eq. (5.7).

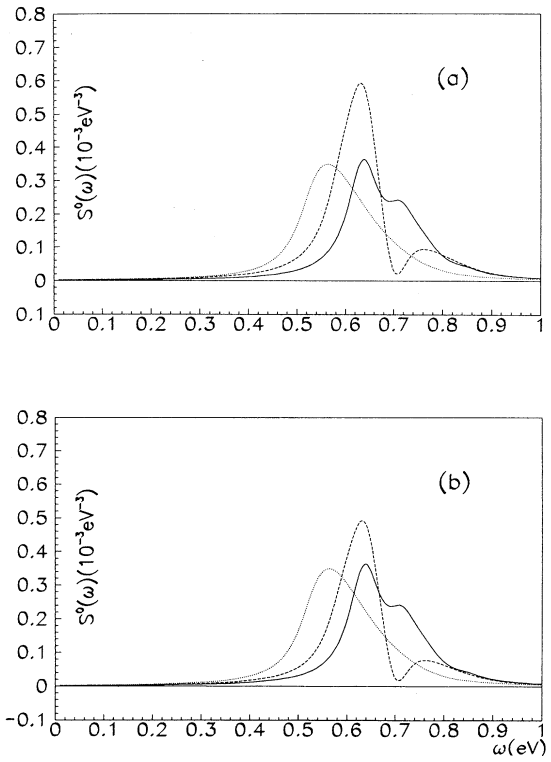

FIG. 2. Dipole component of a single-particle response function of electrons in a spherical cavity in various approximations: smoothed quantum response (solid line), pure Vlasov response (dotted line), and Vlasov plus closed-orbit corrections (dashed line). The dotted curve has been obtained with the Fermi energy $E_F=3.318$ eV, given by Eq. (1.6) or (5.4). In (a) the dashed curve does include the oscillatory closed-orbit corrections, but not the smooth surface and curvature corrections. In (b) the dashed strength function is renormalized by the smooth corrections according to Eq. (5.17).

(3.8) equals $k(E)L_{\mathbf{M}}$, where $k(E)=(1/\hbar)\sqrt{2mE}$ and $L_{\mathbf{M}}$ is the length of the closed orbit specified by the vector $\mathbf{M}$. By considering a smoothed level density, obtained by replacing the δ functions in (3.1) with Lorentzians of width γ (this is a way of accounting for finite-temperature effects), Balian and Bloch [16] give arguments in favor of keeping only the shortest orbits contributing to (3.8); they also argue that the contribution from orbits along the diameter are less important and can be neglected in a first approximation. Thus the most important contributions to the sum in (3.8) come from orbits of triangular and square shapes. Since these orbits have similar lengths, the interference between these two terms is sufficient to reproduce the supershell structure experimentally observed in sodium clusters containing a few thousand atoms [23,24].

Then, for a spherical cavity (infinite square-well potential), Eq. (3.8) gives

$$n(E)\approx n_{\mathrm{TF}}(E)+A_3(E)\cos(3\sqrt{3}x+\tfrac{3}{4}\pi)+A_4(E)\cos(4\sqrt{2}x+\tfrac{5}{4}\pi)\ , \tag{5.5a}$$

with

$$A_3(E)=\frac{3}{4\pi E}x^3\left[\frac{2\pi}{3\sqrt{3}x}\right]^{1/2}\ , \tag{5.5b}$$

$$A_4(E)=\frac{1}{\sqrt{2}\pi E}x^3\left[\frac{2\pi}{4\sqrt{2}x}\right]^{1/2}\ , \tag{5.5c}$$

and $n_{\mathrm{TF}}(E)$ is given in (5.4). However, for a spherical cavity, this is a poor approximation [28]. The smooth surface and curvature corrections [16] are essential in order to get a reasonable value of the Fermi energy (these smooth corrections might be less important for other kinds of potentials [28]), so we use instead

$$\tilde{n}(E)=\tilde{n}_{\mathrm{TF}}(E)+A_3(E)\cos(3\sqrt{3}x+\tfrac{3}{4}\pi)+A_4(E)\cos(4\sqrt{2}x+\tfrac{5}{4}\pi)\ , \tag{5.6a}$$

with

$$\tilde{n}_{\mathrm{TF}}(E)=n_{\mathrm{TF}}(E)-\frac{1}{4E}x^2+\frac{1}{3\pi E}x\ , \tag{5.6b}$$

including the smooth surface and curvature corrections to the volume Thomas-Fermi term n_{TF} [16].

To summarize, we determine the Fermi energy through the requirement

$$2\int_0^{E_F}dE\,\tilde{n}(E)=254\ , \tag{5.7}$$

(the factor 2 in front of the integral accounts for the spin degeneracy). Then we obtain $E_F\approx 3.907$ eV ($x=13.575$). This value of the Fermi energy is shown in Fig. 1 by a dotted line and is seen to lie in the gap between the last occupied and first unoccupied level. We turn now to the single-particle propagator and evaluate it in the same approximation as (5.5a). From Eq. (3.9), making the replacement $I_2=\hbar x\cos\alpha$, we have

$$\Pi^{0R}(\omega,\eta)=\sum_{\mathbf{M}}\int_0^{\pi/2}d\alpha\,G(E_F,\alpha,\omega,\eta)e^{i\psi_{\mathbf{M}}(\alpha)}\ , \tag{5.8}$$

with $\mathbf{M}=(0,M_L,M_S)$,

$$\psi_{\mathbf{M}}(\alpha)=2\pi x\left[M_L\cos\alpha+\frac{M_S}{\pi}(\sin\alpha-\alpha\cos\alpha)\right]-\pi M_L-\tfrac{3}{2}\pi M_S\ , \tag{5.9}$$

and $G(E_F,\alpha,\omega,\eta)$ is determined from the condition that the term $\mathbf{M}=\mathbf{0}$ must give the Vlasov propagator

$$D^0(E_F,\omega,\eta)=\int_0^{\pi/2}d\alpha\,G(E_F,\alpha,\omega,\eta)\ . \tag{5.10}$$

For spherical systems it is convenient to expand in multipoles, as in Eq. (A16). For the dipole component ($L=1$, $Q_L=r$), Eq. (A17) gives

$$G_{L=1}(E_F,\alpha,\omega,\eta)\approx\frac{3}{4\pi}n_{\mathrm{TF}}(E_F)\alpha^2\cos\alpha\left[\frac{\sin\alpha}{\alpha}\right]^5\times\frac{\omega_F}{(\omega+i\eta)-\dfrac{\alpha}{\sin\alpha}\omega_F}R^2\ , \tag{5.11}$$

where $\omega_F\equiv v_F/R$ and v_F is the Fermi velocity. We have used the following explicit expressions of the various quantities appearing in (A17):

$$\frac{1}{T}\oint\frac{dr}{v_r(r)}e^{-is_{nN}(r)}r=(-1)^nR\left[\frac{\sin\alpha}{n\pi+N\alpha}\right]^2\ , \tag{5.12}$$

$$T(\alpha)=\frac{2R\sin\alpha}{v_F}\ , \tag{5.13}$$

$$\omega_3(\alpha)=\frac{2\pi}{T(\alpha)}\ , \tag{5.14}$$

$$\omega_2(\alpha)=\frac{\alpha}{\sin\alpha}\omega_F\ . \tag{5.15}$$

Moreover, since the transitions shown in Fig. 2 are of the kind $\Delta n_r=0$ and $\Delta l=1$, we have taken only the terms $n=0$ and $N=1$ in (A17).

Then, the approximation analogous to Eq. (5.5a) for the dipole propagator is

$$\Pi^{0R}_{L=1}(\omega,\eta)\approx D^0_{L=1}(E_F,\omega,\eta)+G_{L=1}(E_F,\pi/3,\omega,\eta)\left[\frac{2\pi}{3\sqrt{3}x}\right]^{1/2}2\cos(3\sqrt{3}x+\tfrac{3}{4}\pi)+G_{L=1}(E_F,\pi/4,\omega,\eta)\left[\frac{2\pi}{4\sqrt{2}x}\right]^{1/2}2\cos(4\sqrt{2}x+\tfrac{5}{4}\pi)\ . \tag{5.16}$$

All the quantities on the right-hand side of (5.16) are evaluated at the Fermi energy, which is determined from the normalization condition (5.7).

Equation (5.16) includes the corrections due to triangular and square orbits; however, there remains the problem of the smooth, surface and curvature, corrections. They can be included in a somewhat empirical fashion. It can be noticed from Eq. (5.11) that the Vlasov propagator is proportional to the Thomas-Fermi level density. It is reasonable to replace $n_{\rm TF}$ in (5.11) with $\tilde{n}_{\rm TF}$, which includes the surface and curvature corrections. Then the propagator (5.16) is renormalized by the smooth corrections

$$\tilde{\Pi}^{0R}_{L=1}(\omega,\eta)=\frac{\tilde{n}_{\rm TF}(E_F)}{n_{\rm TF}(E_F)}\Pi^{0R}_{L=1}(\omega,\eta)\ . \tag{5.17}$$

Equations (5.5a) and (5.16) include only contributions due to triangular and square orbits. However, Nishioka, Hansen, and Mottelson [23] find that these are sufficient to generate the most important shell effects in a wide range of electron number.

The results of including the periodic orbit contributions are shown in Fig. 2 and are compared with the simple Vlasov response function and the exact (smoothed) quantum result. The dashed curve in Fig. 2(a) is obtained from the imaginary part of the propagator (5.16), while the dashed curve in Fig. 2(b) is obtained from Eq. (5.17). Thus Fig. 2(b) includes renormalization due to surface and curvature effects. The smoothing parameter η used in Fig. 2 is $\hbar\eta=0.04$ eV, which is about one-third of the distance between two successive peaks in the quantum response function.

The quantum corrections modify the Vlasov results shown by the dotted curve in Fig. 2 in the following ways.

(i) The Fermi energy calculated from Eq. (5.7) is affected by the surface and curvature corrections in (5.6b) and by the contribution of periodic orbits in (5.6a). The calculations presented here include both these effects and give an upward shift of the Fermi energy by about 0.6 eV compared with the pure volume Thomas-Fermi value. This shift of the Fermi energy also changes the Fermi velocity v_F and the frequency ω_F.

(ii) The quantum corrections introduce additional terms in the propagator (5.16), coming from the periodic orbits.

(iii) There is a question as to which expression should be used for the density of states $n_{\rm TF}(E_F)$ in Eq. (5.11). One possibility is to use the volume Thomas-Fermi value (5.4). The dashed curve in Fig. 2(a) is calculated in this way. Another possibility is to use $\tilde{n}_{\rm TF}$ given by Eq. (5.6b), which includes the smooth surface and curvature corrections. This does not change the position of the peak, but reduces the overall strength and is in better agreement with the exact quantal result [see Fig. 2(b)]. A further possibility would be to replace $\tilde{n}_{\rm TF}$ in Eq. (5.11) with $\tilde{n}$ given by Eq. (5.6a), but in this case there is a possibility of double counting and it would also lead to a very critical dependence on the position of the Fermi energy.

Even if we do not make this last replacement, because of the relatively large value of the parameter x, the shape of the semiclassical response function depends rather critically on the value of the Fermi energy. In the present case ($x=13.575$) the contribution of triangular orbits is negative and is responsible for the dip around 0.7 eV, while square orbits give a positive contribution that adds to the large peak around 0.6 eV.

VI. CONCLUSION

We have established a parallel between the problem of evaluating shell effects in the level density and in the single-particle response function of large quantum systems. The level density is not the only quantity where an interesting connection can be made between quantum shell effects and a property of classical orbits. Closed classical orbits play exactly the same role in determining shell effects in the linear response function. Quantum corrections affect the Vlasov propagator in different ways: through the value of the Fermi energy parameter, through the additional terms that can be related to closed classical orbits, and also through the renormalization due to the surface and curvature corrections, which have been included somewhat empirically.

The results obtained here extend the possibility of applying the semiclassical theory of linear response based on the Vlasov equation to systems in which shell corrections are not completely negligible. A most interesting result of the closed-orbit theory is the possibility of relating features of the excitation spectrum of quantum systems to a simple property of classical orbits.

APPENDIX

In this appendix we show that, for spherical systems, Eqs. (1.2) and (1.3) give the same propagator obtained in [4]. A spherically symmetric Hamiltonian $h_0(\mathbf{r},\mathbf{p})=p^2/2m+U_0(r)$ is always separable in polar coordinates; however, the choice of action-angle variables is not unique. For our purpose it is convenient to choose the components of the vector $\mathbf{I}$ to be ([26], p. 476)

$$I_1=\lambda_z\ , \tag{A1a}$$

$$I_2=\lambda\ , \tag{A1b}$$

$$I_3=\lambda+I_r=\lambda+\frac{1}{2\pi}\oint dr\,p_r\ , \tag{A1c}$$

where λ_z is the z component of the particle angular momentum, λ the magnitude of the angular momentum vector, and p_r the radial component of the particle momentum. In a central potential the Hamiltonian does not depend on I_1, so that $h_0=h_0(I_2,I_3)$; consequently,

$$\dot{\phi}_1=\frac{\partial h_0}{\partial I_1}=0\ , \tag{A2a}$$

$$\dot{\phi}_2=\frac{\partial h_0}{\partial I_2}=\omega_2, \tag{A2b}$$

$$\dot{\phi}_3=\frac{\partial h_0}{\partial I_3}=\omega_3\ . \tag{A2c}$$

The frequency ω_3 (denoted by ω_0 in [4]) is just the frequency of radial motion in the effective potential $U_{\text{eff}}(r,\lambda)=U_0(r)+\lambda^2/2mr^2$. The frequency ω_2 instead is the precession frequency of the periapsis in the plane of the orbit ([26], p. 509) and it was denoted by ω_0/ν_γ in [4]. The angle variable ϕ_1 is a constant of the motion, it is the longitude of the ascending node ([26], p. 479). The variables ϕ_2 and ϕ_3 are linear functions of time

$$\phi_2(t)=\phi_2^0+\omega_2 t \ , \tag{A3a}$$

$$\phi_3(t)=\phi_3^0+\omega_3 t \tag{A3b}$$

and can be expressed in terms of r as (taking $\phi_3^0=0$)

$$\phi_3(r)=\omega_3\tau(r) \ , \tag{A4a}$$

$$\phi_2(r)=\phi_2^0+\omega_2\tau(r) \ , \tag{A4b}$$

with

$$\tau(r)\equiv\int_{r_1}^{r} dr' \frac{1}{v_r(r')} \ , \tag{A5}$$

the time required for the particle to move from the inner turning point r_1 to the generic point r (v_r is the radial velocity). Then, since $\omega_1=0$, Eq. (1.2) gives

$$D^0(\mathbf{q}',\mathbf{q},\omega)=(2\pi)^3\sum_{n_2,n_3}\int dI_3\int dI_2 F'\left[h_0(I_2,I_3)\right]\frac{n_3\omega_3+n_2\omega_2}{n_3\omega_3+n_2\omega_2-(\omega+i\eta)}\sum_{n_1}\int d\,I_1 Q_{\mathbf{n}}^*(\mathbf{q}',\mathbf{I})Q_{\mathbf{n}}(\mathbf{q},\mathbf{I}) \ . \tag{A6}$$

It is convenient to expand D^0 in partial waves. This is achieved by means of the expansion

$$e^{i\mathbf{q}\cdot\mathbf{r}}=\sum_{L,M} Q_L(qr)Y_{LM}(\theta,\varphi)Y_{LM}^*(\hat{\mathbf{q}}) \ , \tag{A7a}$$

with

$$Q_L(qr)=4\pi i^L j_L(qr) \ . \tag{A7b}$$

Then the Fourier coefficients (1.3) become

$$Q_{\mathbf{n}}=\sum_{L,M} Y_{LM}^*(\hat{\mathbf{q}})Q_{\mathbf{n}}^{(LM)}(q) \ , \tag{A8}$$

with

$$Q_{\mathbf{n}}^{(LM)}\equiv\frac{1}{(2\pi)^3}\int_0^{2\pi}d\phi_1\int_0^{2\pi}d\phi_2\int_0^{2\pi}d\phi_3 e^{-i(n_1\phi_1+n_2\phi_2+n_3\phi_3)}Y_{LM}(\theta,\varphi)Q_L(qr) \ . \tag{A9}$$

In order to evaluate these integrals, it is useful to perform the same rotation of the reference frame considered in [4]. The frame is rotated so that the z axis is aligned with the vector $\boldsymbol{\lambda}$ and the y axis with the radius vector $\mathbf{r}$. Then the spherical harmonics become

$$Y_{LM}(\theta,\varphi)=\sum_N D_{MN}^L(\alpha,\beta,\gamma)^* Y_{LN}\left[\frac{\pi}{2},\frac{\pi}{2}\right] \ , \tag{A10}$$

where D_{MN}^L are the rotation matrices and (α,β,γ) are the Euler angles that specify the new position of the reference frame. It is easy to realize that

$$\alpha=\phi_1 \ , \tag{A11a}$$

$$\cos\beta=\lambda_z/\lambda \ , \tag{A11b}$$

$$\gamma=\phi_2^0+\gamma(r)-\pi/2=\phi_2-\omega_2\tau(r)+\gamma(r)-\pi/2 \ , \tag{A11c}$$

with

$$\gamma(r)\equiv\int_{r_1}^{r} dr' \frac{1}{v_r(r')}\frac{\lambda}{mr'^2} \ . \tag{A11d}$$

Thus, by writing the rotation matrices as

$$D_{MN}^L(\alpha,\beta,\gamma)=e^{-iM\alpha}d_{MN}^L(\beta)e^{-iN\gamma} \ , \tag{A12}$$

the integration over ϕ_1 and ϕ_2 can be performed and the coefficients (A9) become

$$Q_{\mathbf{n}}^{(LM)}(q)=\sum_N Y_{LN}\left[\frac{\pi}{2},0\right]d_{MN}^L(\beta)\delta_{M,n_1}\delta_{N,n_2}\times\frac{1}{T}\oint\frac{dr}{v_r(r)}e^{-is_{n_3N}(r)}Q_L(qr) \ ; \tag{A13}$$

T is the period of radial motion and

$$s_{n_3N}(r)\equiv n_3\omega_e\tau(r)+N[\omega_2\tau(r)-\gamma(r)] \tag{A14}$$

coincides with the definition (5.18) of [4]. The integral in (A13) (divided by T) is the classical limit of the radial matrix element for the operator $Q_L(qr)$ [10], while the factors in front of it are classical limits of Clebsh-Gordan coefficients. The whole Eq. (A13) is the classical version of the following quantum relation between the full and radial matrix elements of a multipole operator in a central potential:

$$\langle n_b,l_b,m_b|Q_L(qr)Y_{LM}(\hat{\mathbf{r}})|n_a,l_a,m_a\rangle \propto\langle l_aLm_aM|l_bm_b\rangle\langle l_aL00|l_b0\rangle\times\int dr\,u_b(r)Q_L(qr)u_a(r) \ . \tag{A15}$$

The rotation matrix d_{MN}^L is the classical limit of the Clebsh-Gordan coefficient $\langle l_aLm_aM|l_bm_b\rangle$, while

$Y_{LN}(\pi/2,0)$ is the classical limit of $\langle l_a L 00|l_b\rangle$.

Finally, by using that $\int dI_1=\lambda\int d\cos\beta$ and the orthogonality of the rotation matrices, the propagator (A6) becomes

$$D^0(\mathbf{q}',\mathbf{q},\omega)=\sum_{L,M} D_L^0(q',q,\omega)Y_{LM}(\hat{\mathbf{q}}')Y_{LM}^*(\hat{\mathbf{q}}) , \quad \text{(A16)}$$

with

$$D_L^0(q',q,\omega)=\frac{8\pi^2}{2L+1}\sum_{n=-\infty}^{+\infty}\sum_{N=-L}^{L}\left|Y_{LN}\left[\frac{\pi}{2},0\right]\right|^2 \times\int dE\,F'(E)\int d\lambda\,\lambda T\frac{n\omega_3+N\omega_2}{n\omega_3+N\omega_2-(\omega+i\eta)}\left[\frac{1}{T}\oint\frac{dr}{v_r(r)}e^{-is_{nN}(r)}Q_L(q'r)\right]^* \times\left[\frac{1}{T}\oint\frac{dr}{v_r(r)}e^{-is_{nN}(r)}Q_L(qr)\right] , \quad \text{(A17)}$$

in agreement with Eq. (5.22) of [4].

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