SEMICLASSICAL APPROACH TO SURFACE PLASMONS IN SPHEROIDAL CLUSTERS *)

A. Dellafiore

Istituto Nazionale di Fisica Nucleare, Sezione di Firenze, L.go E. Fermi 2, I-50125, Firenze, Italy

F. Matera

Dipartimento di Fisica, Università degli Studi di Firenze, L.go E. Fermi 2, I-50125, Firenze, Italy

F.A. Brieva

Departamento de Fisica, Facultad de Ciencias Fisicas y Matematicas, Universidad de Chile, Casilla 487-3, Santiago, Chile

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The linearized Vlasov equation gives a semiclassical version of the random phase approximation. The solution of this equation is studied for electrons moving in a deformed equilibrium mean field which is approximated by a cavity of a spheroidal shape (both prolate and oblate). Contrary to spherical systems, there is a coupling between excitations of different multipolarity induced by the interaction between constituents. The dipole response presents a typical double-peaked profile with a strong dependence on the deformation.

The linearized Vlasov equation offers an interesting semiclassical alternative to the fully quantum RPA calculations in the study of collective oscillations in many-body systems. If the equilibrium mean-field Hamiltonian is integrable, the zero-order propagator given by the Vlasov equation is relatively simple [1, 2]. An integral equation analogous to the RPA integral equation for the ph propagator [3] can be obtained from the linearized Vlasov equation. In momentum space this equation reads

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

Here D is the semiclassical limit of the RPA propagator, and D^0 corresponds to the single-particle propagator. Following [1, 2],

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

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with the Fourier coefficients

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

taking the place of the quantum matrix elements. The angle variables $\boldsymbol{\Phi}$ are canonically conjugate to the action variables $\boldsymbol{I}$. The components of the vector $\boldsymbol{\omega}$ are the fundamental frequencies of the multiply-periodic particle motion in the equilibrium mean field.

For spheroidal systems a partial-wave expansion of the propagators in (1) leads to the following set of coupled integral equations for the dynamic polarizability tensors

$$\begin{aligned} D_{L'LM}(q',q,\omega) = {} & D^0_{L'LM}(q',q,\omega) \\ & + \frac{1}{(2\pi)^3}\sum_{\ell=|M|}^{\infty}\int_0^{\infty}\mathrm{d}k\,k^2 D^0_{L'\ell M}(q',k,\omega)u(k)D_{\ell LM}(k,q,\omega) \end{aligned} \tag{4}$$

with

$$\begin{aligned} D^0_{L'LM}(q',q,\omega) = & -\frac{2}{(2\pi\hbar)^3}\sum_{n_u,n_v}(2\pi)^3\int\mathrm{d}\lambda_z\int\mathrm{d}\epsilon\left|\frac{\partial(I_v,I_u)}{\partial(E,\epsilon)}\right| \\ \times & \frac{n_u\omega_u+n_v\omega_v+M\omega_\varphi}{n_u\omega_u+n_v\omega_v+M\omega_\varphi-(\omega+\mathrm{i}\eta)}Q^{(L'M)*}_{n_u,n_v,M}(E,\epsilon,\lambda_z;q')Q^{(LM)}_{n_u,n_v,M}(E,\epsilon,\lambda_z;q)\,. \end{aligned} \tag{5}$$

The frequencies and the coefficients in the last equation can be expressed in terms of elliptic integrals if the equilibrium mean field is approximated by a spheroidal cavity (details will be published elsewhere). Note that, since we do not consider "pear-shaped" clusters, $D^0_{L'LM} = 0$ unless $(-)^{L'} = (-)^L$, only multipoles with the same parity are mixed in (4). Clearly Eq. (4) is useful only if the sum over l can be truncated at some relatively small $l_{\max}$. Fortunately this turns out to be the case, even at rather large deformations. In Figure 1 we show the photoabsorption cross section in arbitrary units (proportional to $\omega R(q,\omega)$, with $R(q,\omega) = -\mathrm{Im}\left[\sum_M D_{11M}(q,q,\omega)\right]/\pi$) for various values of the deformation parameter $\eta = R_>/R_<$, corresponding to a prolate sodium cluster containing 254 atoms ($\eta = 1$ corresponds to spherical shape). The peaks in the region of the Mie resonance correspond to the two M components $M = 0$ and $M = \pm 1$, while the peaks around $\omega \approx 0.2\omega_{\mathrm{M}}$ are the analogous peaks in the zero-order propagator D^0.

Since we have not taken into account the electron "spill-out", the plasmon peaks shown in Figure 1 are slightly blue shifted compared to the corresponding Mie frequencies. From the classical Mie theory one has the following expression for the frequency of the surface plasmon corresponding to oscillations along the i-axis:

$$\omega_i = \sqrt{n_i}\,\omega_p\,, \tag{6}$$

where ω_p is the bulk plasmon frequency and n_i the appropriate depolarizing factor [4]. For spherical symmetry $n_x = n_y = n_z = 1/3$, while for a prolate spheroid [4]

$$n_z = \frac{1-e^2}{2e^3}\left(\log\frac{1+e}{1-e}-2e\right), \qquad n_x = n_y = \tfrac{1}{2}\left(1-n_z\right), \tag{7}$$

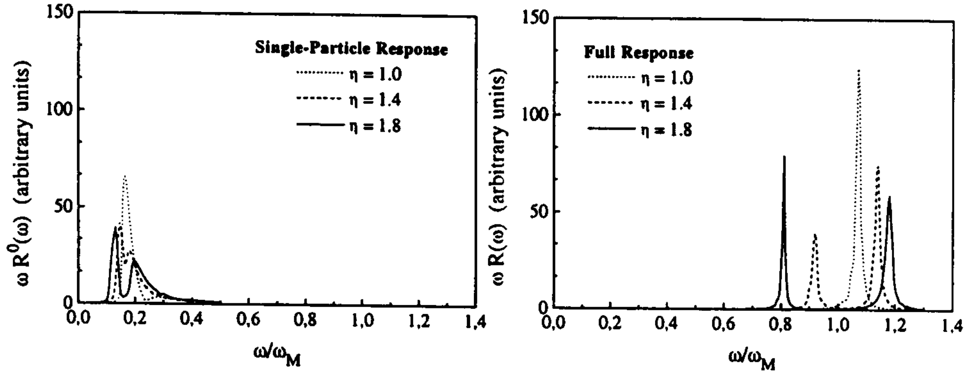

Fig. 1. Dipole response for prolate spheroidal sodium clusters and different deformations, as a function of frequency, expressed in units of the Mie frequency for spherical geometry ω_M. The lefthand side figure displays the single-particle response. The righthand side figure displays the full response.

where $e = \sqrt{1 - 1/\eta^2}$ is the eccentricity. For $\eta = 1.8$, Eq. (6) gives $\omega_0 \approx 0.76\omega_M$, and $\omega_1 \approx 1.10\omega_M$. Our plasmons in Figure 1 are slightly blue shifted.

The analysis for the oblate geometry is slightly complicated by the fact that in an oblate cavity there are two types of three-dimensional orbits: the orbits that never cross the focal circle (W orbits), and the orbits that always cross the focal circle (B orbits). The two kinds of orbits are characterized by the value of λ_z, the component of the particle angular momentum along the symmetry axis. Since the two kinds of orbits never mix, for each kind we can define the zero-order propagators W or B similar to (5). Thus, for the oblate equilibrium geometry, we have

$$D^0_{L'LM} = B_{L'LM} + W_{L'LM}, \tag{8}$$

and the collective response is still described by the solution of Eq. (4).

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