Collective excitation of spherical sodium cluster within a semiclassical discription*

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Abstract A microscopic semiclassical Vlasov equation approach is used to investigate the dipole giant resonances of spherical cluster Na_{14}. The main strength distributions of collective response function are located around 2 eV region, with a fair agreement with the experiment ones. The results are quite independent of the choice of parameters of the mean field.
Keywords Metal cluster, Vlasov equation, Dipole giant resonance

1 Introduction

Recently, cluster physics in general is a rapidly growing field which, touches a large number of physical phenomena bordering on atomic, molecular, surface, solid state and nuclear physics. Especially, because the cluster is a quantum many-body system, the properties of small metal clusters are dominated by the fermion nature of their constituents, with many parallels to what we have learnt from nuclear physics. Thus they are expected to exhibit the simultaneous existence of independent-particle and collective excitations and an interplay bewteen these two modes of motion. Although modern date acquisition techniques and computers are a considerable help, we can not produce nuclei heavier than 300 mass units. Between this upper limit and neutron star there is a vast gap in our knowledge. The cluster physics, however, bridges this gap.

By using the well-known Jellium model, both static and dynamic properties of small alkali-metal cluster spheres have been evaluated in a self-consistent way [1−7]. This model is essentially a mean field approximation in which the conduction electrons are assumed to move independently of each other. Such self-consistent microscopical calculations become quite time consuming when the size of the clusters increases, particularly to the collective excitations. Therefore, it is not easy to gain from these calculations an intuitive understanding of the most important physical effects which determine the final results. But, there is a very reliable framework, i.e. the microscopic semiclassical Vlasov equation approach for quantitative predictions. Such a novel approach has been recently developed for the study of collective motions in nuclei. The response functions obtained from such an approach show striking similarities with fully RPA (random-phase approximation) calculations, even in nuclei as light as ^{40}Ca and in the neutron halo nuclei as ^{11}Li[8~10]. The key element in the nuclear collective modes is the mean field, where nucleons move independently of each other in quantized orbits. This concept has proved to carry on well into the field of simple metal clusters and, more recently, also to provide the basis for a microscopic description of fullerence[11]. In the present paper, therefore, we will use the Vlasov equation, basing on the mean field theory, to study the collective excitation, i.e. the giant resonances of spherical metal clusters.

2 Outline of the theory and method

In Refs.[8~10], an almost analytic method is derived to solve the nuclear Vlasov equation for small amplitude collective oscillations, in the cases with and/or without particle-hole correlations. In particular, the test particle method used to directly solve the Vlasov equation is given in Ref.[10].

The Vlasov equation is an equation for

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single particle probability density function $f(\vec{r},\vec{p},t)$. It can be written in Poisson bracket form as

$$\frac{\partial f}{\partial t}+\{f,h+\beta Q\}=0 \qquad (1)$$

here $\beta(t)Q(\vec{r})$ is an external driving potential and h is the self-consistent single particle Hamiltonian,

$$h(\vec{r},\vec{p},t)=\frac{p^2}{2m}+U(\vec{r},t) \qquad (2)$$

with the self-consistent potential $U(\vec{r},t)$. It depends on the density f and introduces nonlinear terms into Eq.(1). For small amplitude variations about an equilibrium distribution function f_0,

$$f(\vec{r},\vec{p},t)=f_o(\vec{r},\vec{p})+g(\vec{r},\vec{p},t) \qquad (3)$$

we can expand Eq.(1) to the first order in g. We get

$$\frac{\partial g}{\partial t}=\{h_0,g\}+\{\delta W+\beta Q,f_0\} \qquad (4)$$

where $h_0=\frac{p^2}{2m}+U_0(\vec{r})$ is the equilibrium mean field, δW is the self-consistent variation of the mean field due to the residual interaction $u(\vec{r},\vec{r'})$ between particles:

$$\delta W(\vec{r},t)=\int \mathrm{d}\vec{r'}u(\vec{r},\vec{r'})\delta\rho(\vec{r'},t) \qquad (5)$$

where $\delta\rho$ is the transition density. Since

$$\delta\rho(\vec{r},t)=\int \mathrm{d}\vec{p}g(\vec{r},\vec{p},t) \qquad (6)$$

it is clear that Eq.(4) is actually a complicated integro-differential equation.

In a semiclassical picture, we can choose a Thomas-Fermi approximation for the equilibrium distribution f_0, that is

$$f_0(\vec{r},\vec{p})=F(h_0)=\frac{4}{(2\pi\hbar)^3}\Theta(E_\mathrm{f}-h_0) \qquad (7)$$

where E_f is the Fermi energy.

If we introduce some change of variables to a new set which includes constants of motion of h_0, we can reduce the number of independent variables. Because there is a spherical symmetry for the equilibrium function f_0, in Refs.[8~9] the transformation $(\vec{r},\vec{p})\rightarrow(E,\lambda,r,\alpha,\beta,\gamma)$ has been suggested. Here E is energy, λ the angular momentum,r the radius and (α,β,γ) the Euler angles. Apart from E and λ, also α and β are constants of motion since the plane of orbit does not change and $\cos\beta=(\lambda_z/\lambda)$. γ represents the angle variable on the orbit plane, with $\dot{\gamma}=\lambda/mr^2$. The γ-dependence can be eliminated by using a multipole expansion for both the external field and the self-consistent term, i.e. $Q(\vec{r})=\sum_{LM}Q_{LM}(r)Y_{LM}(\theta,\phi)$ (and a similar expansion for δW), with (θ,ϕ) the polar coordinates of $\vec{r}$ in the laboratory frame. The solution $g(\vec{r},\vec{p},t)$ of the Eq.(4) can then be obtained exactly following the procedure discussed in Refs.[8~10]. Some important points should be stressed:

(1) All parameters, which will be given later, must be fixed in order to have the correct number of conduction electron in metal cluster. We remind that in the semiclassical approach, up to the Fermi surface the number of particle, i.e. the conduction electron number inside a cluster, is given by

$$A=\frac{4}{\pi}\int_0^{\lambda_{max}}\lambda\mathrm{d}\lambda[2m_e\int_{r_1(\lambda)}^{r_2(\lambda)}v(r)\mathrm{d}r], \qquad (8)$$

with the velocity field

$$v(r)=[\frac{2}{m_e}(E_\mathrm{f}-V_\mathrm{eff}(r,\lambda))]^{1/2}, \qquad (9)$$

where m_e is the mass of electron, $r_1(\lambda)$ and $r_2(\lambda)$ are the classical turning points for (λ,E) orbit, V_eff is the effective radial potential including Coulomb and centrifugal terms, i.e.

$$V_\mathrm{eff}(r,\lambda)=U_0(r)+U_c+\frac{\lambda}{2m_er^2} \qquad (10)$$

Here the angular momentum λ is a continuous variable in Vlasov approach, but we can quantize it by means of $\lambda\rightarrow\hbar\sqrt{l(l+1)}$ according to the quantum theory.

(2) The Fermi energy is obtained by

$$E_\mathrm{f}=V_\mathrm{eff}(r,\lambda_\mathrm{max})|_{r=r_\mathrm{min}} \qquad (11)$$

where $r=r_\mathrm{min}$ means the above expression has minimum value at r_min.

(3) From the knowledge of solution g, we can construct the strength function of collective excitation associated with any operator of multipolarity L and radial dependence of external driving field $Q_L(\vec{r})$. Let's first ignore the residual interaction between electrons. After solving the linearized Vlasov equation, Eq.(4), we

obtain the uncorrelated (without $u(\vec{r},\vec{r'})$) giant resonance poles(energies) as

$$\omega_n(N) = n\frac{2\pi}{T} + N\frac{\Gamma}{T}, \tag{12}$$

with

$$T = 2\int_{r_1}^{r_2}\frac{dr}{v(r)}, \tag{13}$$

radial period and

$$\Gamma = 2\int_{r_1}^{r_2}\frac{dr}{v(r)}\frac{\lambda}{m_e r^2}, \tag{14}$$

angular "period" for each orbit (λ, E) where we can find electrons for a given mass, Fermi energy and unperturbed mean field. Here n, N are integer number, $-\infty < n < \infty$ and $-L \leq N \leq L$ with $(-1)^N = (-1)^L$. Correspondingly,the uncorrelated strength function for any external field with multipolarity L and radial dependence $Q_{LM}(r)$ has the structure:

$$S_L^0(\omega) = -\frac{8\pi^2}{2L+1}\sum_{nN}\int \mathrm{d}EF'(E)\int \lambda \mathrm{d}\lambda\omega_n(N)T|Y_{LN}(\frac{1}{2}\pi,\frac{1}{2}\pi)|^2|Q(n,N)|^2\delta(\omega-\omega_n(N)) \tag{15}$$

Here Y_{LN} is the spherical harmonic function and $Q(n,N)$, the residues at poles,are given by

$$Q(n,N) = \frac{2}{T}\int_{r_1}^{r_2} dr'\frac{Q_{LM}(r')}{v(r')}\cos[S_n(N,r')] \tag{16}$$

with the phase

$$S_n(N,r) = \omega_n(N)\tau(r) - N\gamma(r) \tag{17}$$

where

$$\tau(r) = \int_{r_1}^{r}\frac{dr'}{v(r')} \tag{18}$$

$$\gamma(r) = \int_{r_1}^{r}\frac{\mathrm{d}r'}{v(r')}\frac{\lambda}{mr'^2} \tag{19}$$

are, respectively,the time elapsed and angle spanned to reach the position r on orbit (λ, E).

Making use of the uncorrelated poles and strength function,and taking the residual interaction between electrons into account, the correlated results i.e. the correlated "new" poles and correlated strength functions then can be obtained through the well known dispersion relation as discussed in Ref.[12]. For a general separable residual interaction,we can define the multipole-multipole residual interaction as:

$$u_L(r,r') = k_L r^L r'^L \tag{20}$$

where k_L is the strength of residual interaction and L is the multipolarity. For the more details about the giant poles and strength functions for both the uncorrelated and correlated cases,one can find in Refs.[8~10].

3 Results and discussion

We have studied the dipole giant resonance in spherical cluster Na_{14} within the framework mentioned in Sec.2. We remark that the dipole giant resonance excitation of small metallic cluster is the electrons oscillating collectively relative to the heavy positive charges in cluster. We are, therefore, exclusively concerned with the dipole properties, i.e. we need to specialize the response equations to the case $L = 1$. The potentials that simulate the self-consistent mean field potentials for electrons in Na cluster had been derived by Ekardt[2]. Such potentials can here be approximated by a simple analytic expression in terms of a spherical Woods-Saxon potential:

$$U_0(r) = \frac{U_0}{\exp[(r-r_0)/a_0]+1} \tag{21}$$

where U_0 is the depth of potential, r_0 is the effective radius of the cluster sphere and assumed to be $r_s N_c^{1/3}$, where r_s is the radius of a sphere containing one electron in the bulk, and N_c the number of atoms per cluster. a_0 is the surface thickness of potential. Table 1 presents parameter sets in our calculations which lead to Fermi energies $\sim$ 3 eV and the number of atoms per cluster $N_c \sim 14$. There are three choices for parameter r_s, where r_s = 3.93 u and r_s = 2.25 u had been used to reproduce well the electronic shell and supershell structures of sodium clusters[13,14]. r_s=3.93 and 2.25 are slightly larger than the Wigner-Seitz radius for bulk sodium (2.08). This is due to the so-called "spill out" resulting from the extension of electrons into the region outside the positive

charge. A typical correlated result of the collective response strength distributions for dipole giant resonance mode is shown in Fig.1. One can see from Fig.1 that the strength distributions (in percent of energy weight sum rule, EWSR) of the dipole giant resonance in cluster Na_{14} have no substantial changes for using different parameters sets. The main distributions locate around 2 eV region. A fair agreement with the experimental observations is obtained [15,16]. Since there is very weak residual interaction between electrons, in Fig.1 we take the strength parameter of k_1 (where $L = 1$) as 0.015. We have tested different k_1 of 0.01~0.02 and found no significant changes in the final results.

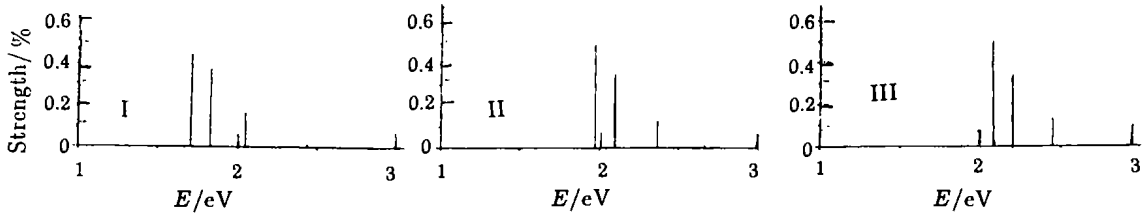

Fig.1 Strength distributions of dipole giant resonances in Na_{14}
Based on parameter sets I, II, III in Table 1

Table 1 Parameter sets used in calculation and calculated values of electron Fermi energy E_f and of atom number per cluster N_c

set	λ_{max} /$\hbar$	U_0 /eV	r_s /u	a_0 /u	k_1 /eV·u^{-1}	E_f /eV	N_c
I	3.110	-5.93	3.93	1.50	-0.015	-4.06	14.00
II	3.125	-6.00	2.25	0.74	-0.015	-2.26	13.97
III	3.105	-7.50	2.08	0.74	-0.015	-3.02	14.16

In conclusions, we have studied the collective excitations of dipole mode in sodium cluster within a microscopic semiclassical Vlasov approach, getting response functions in close agreement with experiments. The strength distributions display that the collective response of dipole mode is laid in region from 2 eV to 2.5 eV. Both resonance energies and response strengths are not very sensitive to the choice of the strength of residual interaction. Finally, we would like to stress that although the Vlasov approach is based on mean field theory, one can easily go beyond mean field, by extending Vlasov equation to include the particle-particle collisional terms. It will be quite interesting to study the interplay between Laudau damping (one body damping) and collision damping (two body damping), since such an interplay between two damping mechanisms is very important to account for the observed widths of giant resonances. Further researches along this line are proceeding, and results will be presented in near future.

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