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LETTER TO THE EDITOR

SUPERSHELLS IN LARGE Na-CLUSTERS FROM A SEMICLASSICAL APPROACH

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Semiclassical level densities are calculated for metal clusters up to 10^4 valence electrons moving in a mean field jellium potential. Supershell structures are predicted by the calculation. We investigate in detail the role played by triangular, square and higher polygon orbits in the interference pattern.

Semiclassically, the shells observed in finite Fermi systems, that is the periodic enhancements of level density from its mean value, as a function of energy or wave number, are associated with the occurence of periodic orbits in the mean field.

The foundation of this theory is found in the work of Gutzwiller [1] and of Balian and Bloch [2]. They have shown for a spherical cavity that the single particle level density, as a function of energy (or momentum), shows a long wavelength oscillating pattern (supershells) that envelopes short period oscillations (corresponding to ordinary shells). Balian and Bloch explained the origin of this phenomenon through the semiclassical treatment of the single-particle level density. Supershells are the beating pattern of two interfering waves associated semiclassically with triangular and square closed orbits inside the cavity. In Ref. 3 Nishioka et al. pointed out that metal cluster is the first real system for the application of the semiclassical theory of level density. Recently, the corresponding predictions for Na-clusters have received experimental confirmation [4,5]. In this paper we present results for semiclassical level densities, for electrons moving in a Woods-Saxon potential which simulates the self-consistent mean-field potentials of clusters [6]. The structure of the supershells, as well as the effects of permitted classical orbits, are studied in systems with up to 8000 atoms.

An accurate expression of the level density for smooth spherical potentials is the one derived by Berry and Tabor [7]

$$\rho(E) = \rho_{TF}(E) + \sum_M A_M \cos\left[2\pi\mathbf{M}\cdot\left(\frac{\mathbf{I}_M}{\hbar} - \frac{\boldsymbol{\alpha}_M}{4}\right) + \frac{\pi}{4}\right] . \tag{1}$$

The first term is the Thomas-Fermi average level density, which does not contribute to the shell structure. $\mathbf{M}$ is a vector of two positive (non zero) integer components M_L, M_S. Each set (M_L,M_S) represents a classical closed orbit where M_L rounds in the angular degree of freedom and M_S oscillations in the radial direction. The quantity $\mathbf{I}_M$ is a set of two actions of the corresponding invariant torus with the frequency ratio $\omega_l/\omega_s = M_L/M_S$. The quantity α_M denotes the Maslov index associated with the closed orbit $\mathbf{M}$. For the case of a smooth potential, this index is

$$\boldsymbol{\alpha}_M \cdot \mathbf{M} = 2M_S + 2M_L .$$

The amplitude A_M is positive for all values of M_L and M_S and is given by

$$A_M = \frac{2}{\hbar^{3/2}} \frac{\epsilon(M)\tau(L_M, E)}{\sqrt{M_S \mid \partial\Theta(L_M)/\partial L \mid}} . \tag{2}$$

The quantity

$$\tau = \frac{\partial S(L,E)}{\partial E}$$

is the arc time between apogee and perigee. The radial action in the expression above is defined as

$$S = \int_{r_1}^{r_2} \sqrt{2m(E - V(r) - L^2/2mr^2)}\mathrm{d}r,$$

where r_1 and r_2 are the radial limits of the motion for given E and L.

The quantity

$$\Theta(L,E) = -\frac{\partial S(L,E)}{\partial L},$$

is the arc angle between apogee and perigee and the curvature is expressed in term of $\partial\Theta/\partial L$. For a given orbit of topology $\mathbf{M} = (M_L, M_S)$, the closure occurs after $2M_S$ arcs and a total rotation angle of $2\pi M_L$. Therefore the arc angle must be

$$\Theta(L_M) = \pi \frac{M_L}{M_S},$$

that defines the angular momentum L_M of the contributing torus of orbits with topology $\mathbf{M}$. As L increases along the energy contour from 0 to its maximal classically allowed value, $\Theta(L)$ decreases from $\pi/2$ to a minimum value Θ_{min}, so that in the topological lattice only a particular sector of points contributes. This sector is closed between the line $M_L = (1/2)M_S$ and the line $M_L = (\Theta_{min}/\pi)M_S$. The quantity Θ_{min} decreases with E and with the cluster size(N_e) so that sector opens up and new topological structures contribute to $\rho(E)$. The quantity $\epsilon(\mathbf{M})$ is a degeneracy factor equal to

$$\epsilon(\mathbf{M}) = \frac{2L_M}{\hbar}.$$

The degeneration arises from the different values of the z-components L_z of the angular momentum.

The action term in Eq. (1) is given as

$$2\pi \mathbf{M} \cdot \mathbf{I}_M = \oint \mathbf{p}(\mathbf{r}) d\mathbf{r}.$$

where $\mathbf{p}(\mathbf{r})$ is the classical momentum of the particle. The magnitude of the momentum is given by

$$\mid \mathbf{p}(\mathbf{r}) \mid = \{2m\,[E - V(r)]\}^{1/2}.$$

The integration is taken over one period of the closed orbit. Equivalent to the smearing done for the level energies, the quantity $\mid \mathbf{k}(\mathbf{r}) \mid$ is replaced by $\mid \mathbf{k}(\mathbf{r}) \mid + ik_i$, leading to a suppression factor $\mathrm{e}^{-k_i L_M}$, where L_M is the trajectory length of the corresponding closed orbit [3]. The value of k, was chosen to be $0.13\, N_e^{-1/3} \times 10^8$ cm^{-1} which represents about a quarter of spacing between successive shells. Among the closed orbits, pendulating and circular orbits ($M_S = 2, \infty$) contribute to the level density only to higher order in $\hbar^{1/2}$, and consequently their contribution should be negligible in the limit $\hbar \to 0$.

Selfconsistent potentials were calculated [5] for sodium clusters using the density functional method with a jellium ionic background. These potentials have an approximately flat bottom with a depth almost independent of the number of electrons N_e.

A good parametrization of these potentials is given by a Fermi function

$$V(r) = \frac{V_0}{1 + \exp[(r - R_0)/a_0]}$$

where $V_0 = -6.0$ eV, $R_0 = r_s N^{1/3} \cdot 10^{-8}$ cm, $r_s = 2.25 \cdot 10^{-8}$ cm, $a_0 = 0.74 \cdot 10^{-8}$ cm. In Fig. 1 we show the level density as a function of the wave number with $N_e = 2000$, calculated using the semiclassical formula for level density $\rho(E)$ given in Eq. (1).

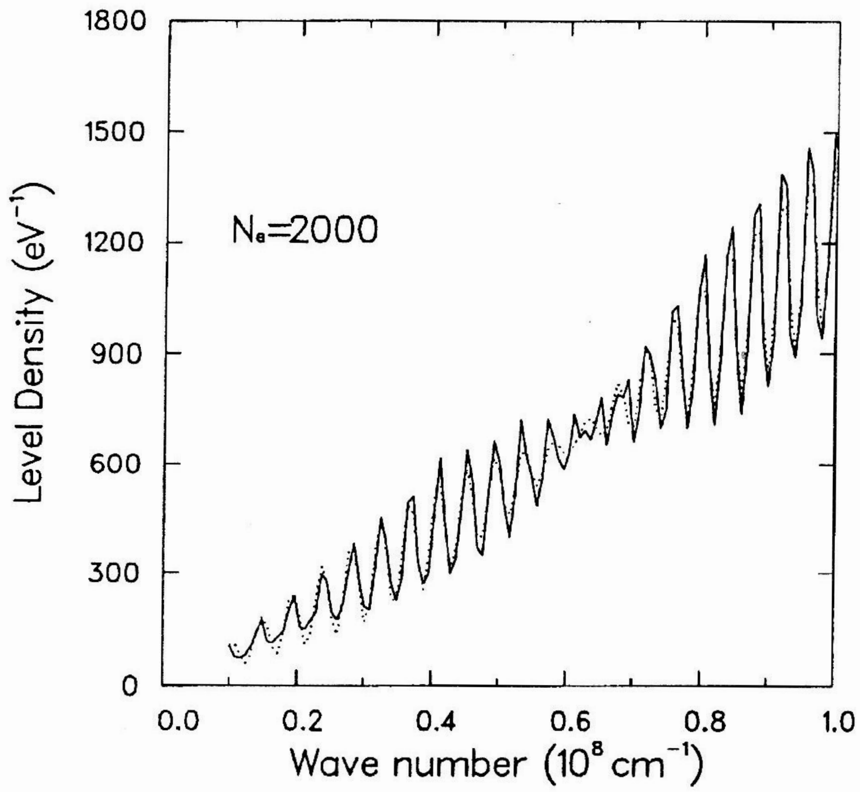

Fig. 1. Semiclassical level for a sodium cluster of $N_e = 2000$*. Dotted line represents the semiclassical level density including only the contributions of triangular and square orbits. The full line shows the results when all permitted polygonals contributions were included.*

Supershells are clearly observed. The dotted line shows the level density calculated considering only the contributions coming from triangular and square orbits, the full line gives $\rho(E)$ including contributions from all closed orbits, in the topologically permitted region. As can be seen, the main supershell structures are accurately accounted by the interference between triangular and square orbits [3].

An open question that can be answered with the present semiclassical calculations is the assesment of the role played by higher polygons in $\rho(E)$ for large-size clusters.

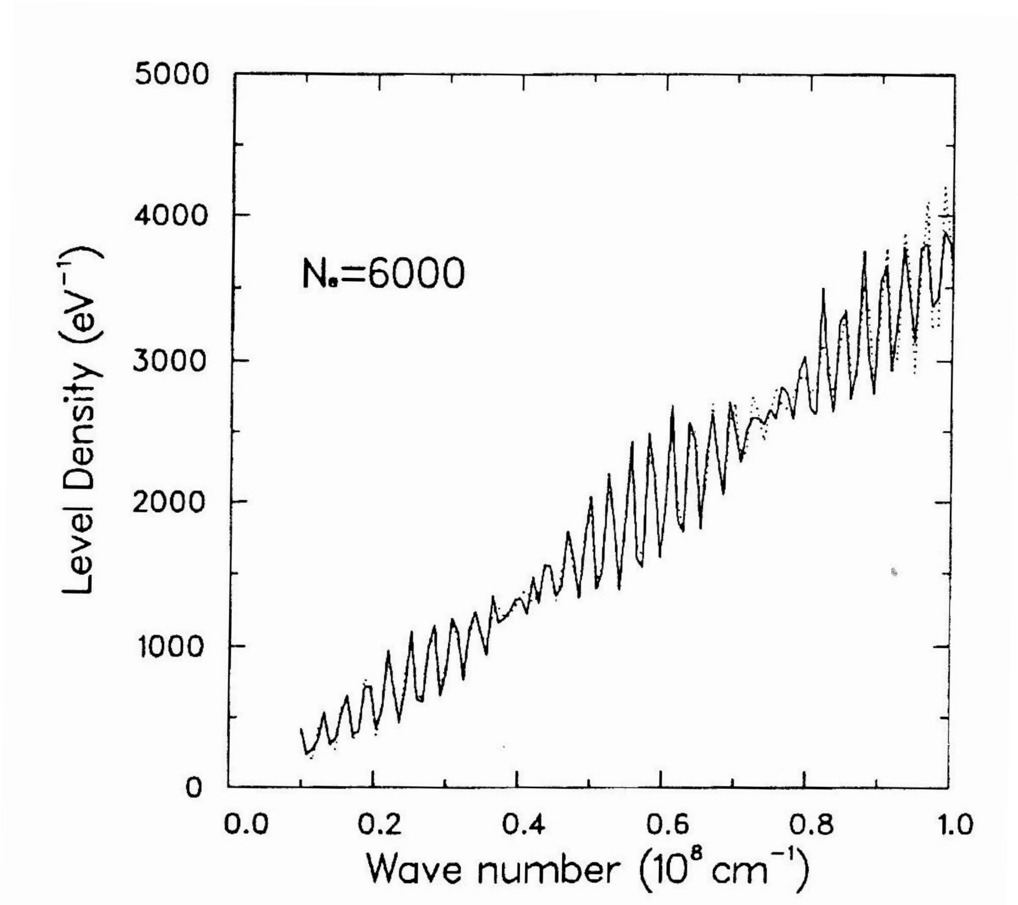

Fig. 2. Semiclassical level for a sodium cluster of N_e = 6000. Dotted line represents the semiclassical level density including only the contributions of triangular and square orbits. The full line shows the results when all permitted polygonals contributions were included.

In Fig. 2, we give the level density calculated with Eq. (1) for a $N_e = 6000$ sodium cluster including triangular and square orbits (dotted line), and all the permitted topological contributions (full line). The results obtained show that, at least for Na-clusters, the level density is affected weakly by the inclusion of the higher polygonal contributions. We can conclude that the origin of the supershells and of the beating pattern is essentially due to the interference between two orbits alone.

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SUPRALJUSKE U VELIKIM GROZDOVIMA Na U POLUKLASIČNOM PRISTUPU

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Poluklasične gustoće nivoa izračunate su za metalne grozdove do 10^4 valentnih elektrona koji se gibaju u srednjem polju žele potencijala. Na temelju računa predviđa se supraljuskasta struktura. Posebno je ispitan doprinos trokutnih, kvadratnih i poligonskih staza.