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]

\[ \rho(E) = \rho_{TF}(E) + \sum_{M} A_M \cos \left[ 2\pi M \cdot \left( \frac{I_M}{\hbar} - \frac{\alpha_M}{4} \right) + \frac{\pi}{4} \right]. \]  

(1)

The first term is the Thomas-Fermi average level density, which does not contribute to the shell structure. \( M \) 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 \( I_M \) 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 \( \alpha_M \) denotes the Maslov index associated with the closed orbit \( M \). For the case of a smooth potential, this index is

\[ \alpha_M \cdot M = 2M_S + 2M_L. \]

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

\[ A_M = \frac{2}{h^{3/2}} \sqrt{\frac{\epsilon(M)\tau(L_M, E)}{\partial \Theta(L_M, E)/\partial L}}. \]

(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

\[ S = \int_{r_1}^{r_2} \sqrt{2m(E - V(r) - L^2/2m^2)} \, dr, \]

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 \( M = (M_L, M_S) \), 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 \( M \). 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 \( \Theta_{\text{min}} \), 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 \( M_L = (\Theta_{\text{min}}/\pi)M_S \). The quantity \( \Theta_{\text{min}} \) 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 \( \epsilon(M) \) is a degeneracy factor equal to

\[
\epsilon(M) = \frac{2L_M}{\hbar}.
\]

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

\[
2\pi M \cdot I_M = \oint p(r) \, dr.
\]

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

\[
| p(r) | = \left( 2m [E - V(r)] \right)^{1/2}.
\]

The integration is taken over one period of the closed orbit. Equivalent to the smearing done for the level energies, the quantity \( | k(r) | \) is replaced by \( | k(r) | + ik_i \), leading to a suppression factor \( e^{-k_i L_M} \), where \( L_M \) is the trajectory length of the corresponding closed orbit \[3]. The value of \( k_i \) 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).

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 assessment of the role played by higher polygons in $\rho(E)$ for large-size clusters.
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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References


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.