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ORBITAL MAGNETISM AND DYNAMICS IN ALKALI METAL CLUSTERS

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1 Introduction

Orbital magnetism in atomic clusters is a subject of special interest. Clusters can contain many atoms and, therefore, single-particle orbital moments of valence electrons can reach very big values. This can result in huge orbital effects, for example, in strong orbital magnetic giant resonances (specific orbital oscillations of valence electrons, see review [1]).

Some of these resonances are of a general character and have to exist in different finite Fermi systems (atomic nuclei, atomic clusters, etc). M1 scissor mode (SM) in deformed systems [2]-[4] and M2 twist mode (TM) in systems of arbitrary shape [5]-[8] are most famous examples. They have been observed in atomic nuclei but not yet in clusters where they should manifest themselves as specific oscillations of valence electrons. We outline here both macroscopic and microscopic treatments of these two modes.

2 SCISSORS MODE

In the first approximation, SM in axially-deformed clusters can be treated as small-angle rotations of the spheroid of valence electrons against the spheroid of ions (see Fig.1, a) [3].

More precisely, the displacement field of this mode includes a rigid

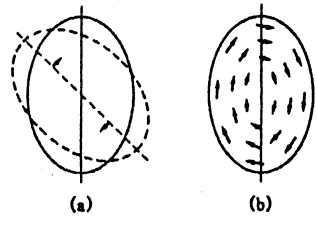

Figure 1: Macroscopic view of scissors mode: a) rigid rotation [2], b) rotation within a rigid surface [3]

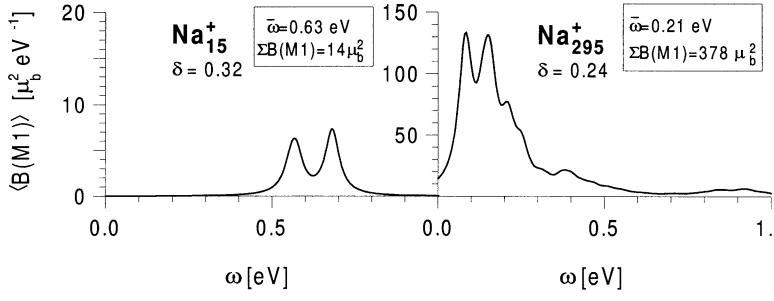

Figure 2: M1 strength calculated within the RPA in the interval 0-1 eV for axial deformed sodium clusters [4]. The deformation parameter δ, the energy centroid $\bar{\omega}$, and the summed M1 strength ($\sum B(M1)$) are given for each cluster. M1 strength is smoothed out with the Lorentz weight using the averaging parameter Δ =0.05 eV.

rotation plus a quadrupole term to vanish the velocity on the surface [3]:

$$\vec{u}(\vec{r}) = \vec{\Omega} \times \vec{r} + \delta(1 + \delta/3)^{-1} \nabla(yz). \tag{1}$$

SM is characterized by low-energy $K^\pi = 1^+$ states (K is magnetic quantum number) connected by strong magnetic dipole transitions with the ground state [3, 4]:

$$\omega_{M1} = \frac{20.7}{r_s^2} N_e^{-1/3} \delta \ eV, \tag{2}$$

$$B(M1) = 2 \mid \langle K^\pi = 1^+ \mid \sum_i \hat{l}_x(i) \mid 0 \rangle \mid^2 \mu_b^2 \simeq N_e^{4/3} \delta \ \mu_b^2, \tag{3}$$

where N_e is the number of valence electrons and δ is the parameter of the quadrupole deformation. Following the macroscopic estimations (2) and (3), both the exitation energy ω_{M1} and the reduced transition probability $B(M1)$ are proportional to δ. So, SM exists only in deformed systems. This mode can serve as a sensitive indicator of cluster's quadrupole deformation.

First RPA calculations for SM have been recently performed[4]. Some of the results for sodium clusters are presented in Fig. 2. Our analysis shows that in small clusters SM is promoted by one or two

particle-hole configurations. In heavy ones the resonance demonstrates a collective character. The reduced transition probabilities $B(M1)$ in these clusters can be huge. It can reach the impressive value of 350-400 μ_b^2 already at $N_e \sim 300$. The observation of such a strength is the challenge for future experiments.

The matrix element for the orbital M1 transition has a form

$$< \Psi^{\pi'}_{K'\nu'} | \hat{l}_{+1} | \Psi^{\pi}_{K\nu} >= \frac{1}{2} \delta_{\pi'\nu' K', \pi\nu K+1} \sum_{nl} a^{K+1\pi}_{nl} a^{K\pi}_{nL} \sqrt{l(l+1) - K(K+1)} \tag{4}$$

where the wave function of a deformed state $\Psi^{\pi}_{K\nu} = \Sigma_{nl}\, a^{K\pi}_{nL} R_{nl}(r) \cdot Y_{lK}(\Omega)\chi_{1/2\nu}$ is a superposition of spherical (nl)-configurations (n, l are the node number and orbital moment, respectively). Eq. (4) shows that SM is generated by the transitions between the components of the wave function, which belong to one and the same spherical nl-configuration but have the projections K differing by one. Certainly, such transitions cannot take place in spherical clusters where all the states belonging to one and the same (nl)-configuration are degenerated. The heavier the cluster, the larger orbital moments l of valence electrons and, so, the bigger the matrix element (4).

The realistic calculations [4] show that the SM energies and B(M1) values scale with δ and N_e basically according to the trends (2) and (3). However, strong fluctuations of B(M1) values around the trend (3) have been revealed for small clusters. These fluctuations reflect the single-particle nature of the transitions and can lead, as is shown below, to some new effects for the magnetic susceptibility.

The total orbital magnetic susceptibility is the sum of Langevin diamagnetic and van Vleck paramagnetic terms:

$$\chi_k = \chi_k^{dia} + \chi_k^{para} \tag{5}$$

where

$$\chi_k^{dia} = -\frac{e^2}{4m_e c^2} N_e < \rho_k^2 > \simeq -\mu_b^2 \theta_k^R, \tag{6}$$

$$\chi_k^{para} = 2\mu_b^2 \sum_n \frac{| < n|\hat{l}_k|0 > |^2}{\omega_n} \simeq \mu_b^2 \theta_k. \tag{7}$$

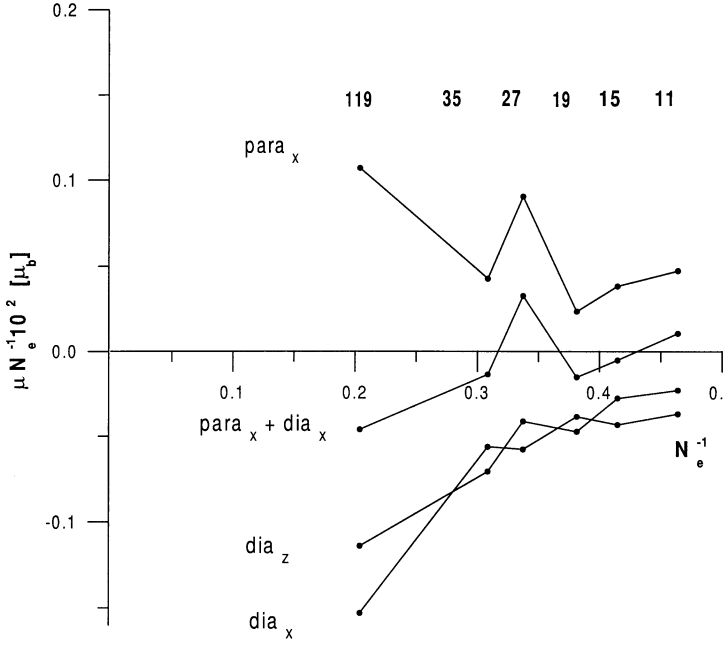

Figure 3: Normalized diamagnetic, paramagnetic and summed moments $\mu = \chi B$ ($B = 4T$) in axial deformed clusters $Na_9^+(\delta = 0.38)$, $Na_{15}^+(\delta = 0.32)$, $Na_{19}^+(\delta = -0.22)$, $Na_{27}^+(\delta = 0.21)$, $Na_{35}^+(\delta = -0.23)$, and $Na_{119}^+(\delta = 0.25)$. The moments are given for x- and z-directions.

Here $k = x, y, z$ are the coordinate axises (z is the symmetry axis), $\theta_{x,y} = 2\sum_n \frac{|<n|\hat{l}_{x,y}|0>|^2}{\omega_n}$ is the moment of inertia and $\theta_{x,y}^R = N_e < \rho_{x,y}^2 >$ is its rigid value.

It is easy to see that just the low-lying SM mainly contributes to $\chi_{x,y}^{para}$. The schematic model [3] gives $\theta_{x,y} = \theta_{x,y}^R$, which results in the complete compensation of dia- and para-terms in $\chi_{x,y}$. Due to the symmetry of the problem, one also has $\chi_z^{para} = 0$. Finally, the total susceptibility has to be strictly anysotropic [3]:

$$\chi_x = \chi_y = 0, \qquad \chi_z = \chi_z^{dia}, \tag{8}$$

i.e. runs from zero to diamagnetic values. As it is seen from Fig. 3,

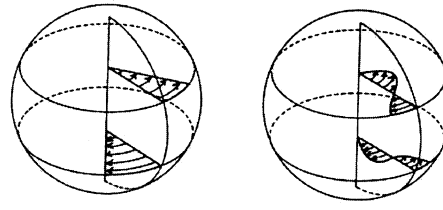

Figure 4: Nodeless (left) and one-node (right) branches of twist mode [5].

the fluctuations in $B(M1)$ values modify this result. Namely, some particular small clusters (Na_{11}^{+} and Na_{27}^{+}) may demonstrate the dia-para anysotropy. This result is rather sensitive to the calculations scheme and has to be check in more involved studies. However, it is very desirable to check this exciting prediction in experiment.

3 TWIST MODE

TM is generated [5] by the operator $\hat{T} = e^{-i\alpha z l_z} = e^{\alpha \vec{u} \cdot \vec{\nabla}}$ with the velocity field $\vec{u} = (yz, -xz, 0)$ [5]. This mode can be viewed as a rotation of different layers of the system against each other with a rotational angle proportional to z (projection to the axis of rotation). Fig. 4 demonstrates nodeless and one-node branches of TM.

The twist can exist in both spherical and deformed systems. It represents one of the most bright examples of elastic-like excitations in finite Fermi systems. TM has been predicted [5] and then observed experimentally [8] in atomic nuclei. Without any doubts, TM must exist in atomic clusters as well [9].

The external field $zl_z \propto r(Y_{10}l_z)$ is a part of the orbital term in the operator of $M2(\mu = 0)$ transition:

$$\hat{F}(M2, \mu = 0) = \mu_b \sqrt{10} r [g_s \{Y_1 \hat{s}\}_{20} + \frac{2}{3} g_l \{Y_1 \hat{l}\}_{20}]. \qquad (9)$$

So, it is natural to consider TM as a part of the orbital M2 resonance.

Investigations of orbital ML resonances in atomic nuclei show that they very slightly depend on spin-multipole residual interaction and

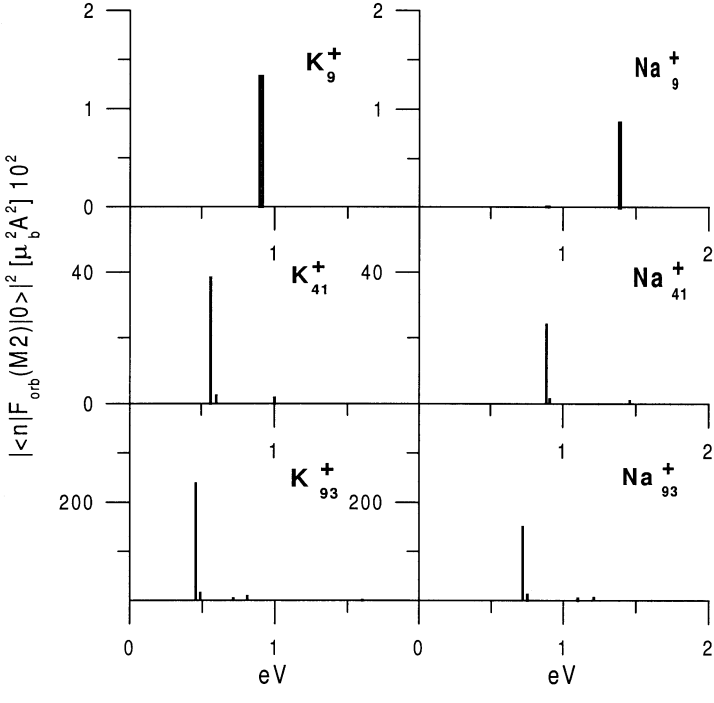

Figure 5: M2 orbital resonance in K and Na spherical clusters.[9].

this dependence is mainly caused by spin-orbital coupling. This is even more the case for small atomic clusters where the spin-orbital coupling can be safely neglected. Our study for SM shows that the influence of multipole residual forces also should not be strong. For these reasons, the simple particle-hole consideration of TM seems to be quite realistic. The corresponding results are presented in Fig. 5. It is seen that the heavier the cluster, the lower the energy of the resonance. It lies safely below the overwhelming dipole plasmon, which favours its experimental observation. It is remarkable that the resonance is dominated by one kind of a particle-hole peak. The analysis shows that in all the cases this peak corresponds to the transition between nodeless levels which are placed in the vicinity of Fermi level and have

maximal orbital momenta. These transitions are $1p \to 1d, 1f \to 1g$ and $1h \to 1i$ in Na_9^+, Na_{41}^+ and Na_{93}^+, respectively. They exhaust from 100% (Na_9^+) to 80% (Na_{93}^+) of the total M2 strength and correspond to the nodeless branch of TM (see Fig. 4, left side). A few weak peaks in Fig. 5 represent one-node, two-node, and etc. TM branches. The heavier the cluster, the larger the contribution of these branches. Due to the strict domination of the nodeless branch, TM can serve as a valuable source of information about single-particle levels with maximal orbital momenta near the Fermi surface.

The other peculiarity of the orbital M2 resonance is that its total energy-weighted strength can be estimated by an accurate and simple way. Just (and only) in the case of M2, the corresponding sum rule does not include any radial integrals and, therefore, can be derived by a *model-independent* way. As a result, the following novel energy-weighted sum rule can be proposed [9]:

$$\begin{aligned} EWSR &= \sum_n \omega_n | < n|\hat{F}_{orb}(M2)|0 > |^2 \\ &= \frac{1}{2} < 0|[\hat{F}_{orb}(M2), [H, \hat{F}_{orb}(M2)]]|0 > \\ &\simeq \frac{1}{2} < 0|[\hat{F}_{orb}(M2), [T, \hat{F}_{orb}(M2)]]|0 > \\ &= \frac{25}{3}\frac{\hbar^2}{\pi m_e} \sum_{nl}^{N_{Fermi}} (2l^3 + 3l^2 + l). \end{aligned} \tag{10}$$

Here, $\hat{F}_{orb}$ is the orbital part of the M2 operator (9) and n, l are node number and orbital moment, respectively. The calculation of the EWSR is extremely simple: it is enough to know the orbital moments l of occupied single-particle levels. Since different models predict the same set of occupied levels, the estimation (10) indeed is model-independent. It gives the values 121, 728, and 2546 $\mu_b^2 \mathring{A}^2 eV$ for Na_9^+, Na_{21}^+ and Na_{41}^+, respectively. The same values take place for corresponding potassium clusters.

4 CONCLUSIONS

Orbital M1 (scissors) and M2 (twist) modes are predicted in alkali metal clusters [4, 9].

The scissors mode (SM) exists only in deformed clusters and can serve as a reliable fingerprint and measure of cluster quadrupole deformation. The larger the cluster, the stronger the M1 strength. Already in clusters with the number of valence electrons $N_e \simeq 300$ the summed low-energy M1 strength can reach $350 - 400\mu_b^2$. SM in small clusters provides a valuable information on the single-particle spectrum. SM gives a dominant contribution to the van Vleck paramagnetism and leads to a strong anysotropy of the magnetic susceptibility. Strong fluctuations of M1 strength predicted in small clusters can, in principle, lead to dia-para anysotropy in some particular clusters.

The twist mode (TM) constitutes a part of the M2 orbital resonance and can exist in clusters of any shape. In spherical clusters of small and moderate size up to 80-100% of the total M2 strength is provided by one transition between the nodeless (n=1) single-particle levels. This transition has lowest energy and involves levels with maximal orbital momenta in the Fermi region. As a result, TM can provide a valuable information about these particular levels. Since the twist mode involves orbitals with highest orbital moments, it represents essentially a surface mode.

The novel model-independent energy-weighted sum rule (EWSR) has been derived for M2 orbital excitations in spherical atomic clusters [9]. It provides very simple and, at the same time, explicit estimations. Both low-energy SM and TM lie safely below the overwhelming dipole plasmon and so have a good chance to be observed experimentally.

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Нестеренко В.О. и др. E17-2000-14
Динамические эффекты орбитального магнетизма
в кластерах щелочных металлов

Предсказано существование в кластерах двух орбитальных магнитных резонансов — M1 «ножничного» (в деформированных кластерах) и M2 твистового. Показано, что эти резонансы позволяют получить ценную информацию о многих свойствах кластеров (квадрупольная деформация, магнитная восприимчивость, одночастичный спектр и т.д.).

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Orbital Magnetism and Dynamics in Alkali Metal Clusters

Two remarkable orbital magnetic resonances, M1 scissor mode and M2 twist mode, are predicted in deformed and spherical metal clusters, respectively. We show that these resonances provide a valuable information about many cluster properties (quadrupole deformation, magnetic susceptibility, single-particle spectrum, etc.)

The investigation has been performed at the Bogoliubov Laboratory of Theoretical Physics, JINR.

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