LOW-ENERGY QUADRUPOLE MODES IN DEFORMED CLUSTERS

V.O. Nesterenko\textsuperscript{1,2}, W. Kleinig\textsuperscript{1,3}, P.-G. Reinhard\textsuperscript{4}, and D.S. Dolci\textsuperscript{1}

\textsuperscript{1} BLTP, Joint Institute for Nuclear Research, Dubna, Moscow region, 141980, Russia
E-mail: nester@thsun1.jinr.ru

\textsuperscript{2} Max Planck Institute for Physics of Complex Systems, 01187, Dresden, Germany

\textsuperscript{3} Technische Universität Dresden, Inst. für Analysis, D-01062, Dresden, Germany

\textsuperscript{4} Institut für Theoretische Physik, Universität Erlangen, D-91058, Erlangen, Germany

ABSTRACT

Properties of low-energy (infra-red) quadrupole modes (LEQM) of multipolarity $\lambda\mu = 20$, 21 and 22 in deformed sodium clusters are studied within the Kohn-Sham LDA RPA approach. Possible manifestations of LEQM in stimulated Raman adiabatic passage (STIRAP) reaction are discussed. It is shown that, in free light clusters, where the low-energy spectrum is delute, LEQM can be unambiguously identified as particular electron-hole pairs. This gives a chance to reconstruct the mean field level scheme near the Fermi surface. Moreover, due to the connection with electric $\lambda\mu = 21$ mode, the scissors mode can be detected. In heavy (supported) oblate clusters, LEQM are in general rather involved. Nevertheless, some interesting $\lambda\mu = 21$ and 22 structures determined by specific deformation effects can be resolved. The origin of the structures is discussed in detail.
I. INTRODUCTION

Collective oscillations of valence electrons in metal clusters manifest themselves in a variety of electric and magnetic (orbital) plasmons (see \[1\] for review). Till now only the electric dipole (E1) plasmon was thoroughly investigated \[2\]. Other plasmons were not still observed and our knowledge about them is scares. At the same time, they promise interesting physics and so devote careful study.

In the present paper, we consider low-energy electric quadrupole modes in axially deformed clusters. Quadrupole excitations of valence electrons in such clusters are collected into bunches $\Delta N = 0$ and 2, where $N$ is the principle shell quantum number \[3, 4\]. The low-energy bunch $\Delta N = 0$ corresponds to E2 transitions inside the valence shell. It is clear that this bunch exists only in clusters with partly occupied valence shell. Just these clusters exhibit a non-spherical shape. The high-energy bunch $\Delta N = 2$ corresponds to E2 transition through two shells. This bunch exists in clusters of any shape and exhausts most of the quadrupole strength. Though the bunch $\Delta N = 2$ shares the energy region with the dominant E1 plasmon, it can be possibly observed by means of angular-resolved electron energy-loss spectroscopy (AR-EELS) at electron scattering angles $\sim 6^\circ$ \[5\].

In this paper, we will consider the low-energy quadrupole modes (LEQM) with $\Delta N = 0$, whose properties, by our knowledge, were never earlier investigated. We will show that LEQM demonstrate spectacular structures connected with cluster deformation.

LEQM are probably too weak to be observed in AR-EELS but have a chance to be detected in two-photon processes like Raman scattering (RS) or resonance fluorescence (RF) where the competition with E1 modes is completely excluded. These processes start from excitation of an intermediate E1 electronic state (virtual or real). Then the coupling of the intermediate and low-energy states results in the population of the latter, again through E1 transitions (e.g., the electron-phonon coupling results in RS population of ionic phonon states).

As far as we know, there was only one attempt to observe LEQM by RS in nanoparticles \[6\]. In that experiment a broad bump was observed at $\sim 0.12$ eV in supported silver clusters. The electronic nature of the bump was justified by the inverse cluster size dependence of the bump energy. Recent data of this group hint at the deformation dependence of the bump energy \[7\]. Explanation of these data requests a strong theoretical support. Our study could
be a first guide in this new field.

Perhaps, a weak coupling of electron states does not favor their detection by RS. In this connection, the stimulated Raman adiabatic passage (STIRAP) seems to be more appropriate. This method uses partially overlapping pulses (from pump and Stokes lasers) to produce complete population transfer between two quantum states \( |1\rangle \) and \( |3\rangle \) through the intermediate state \( |2\rangle \). The combination of the pump and Stokes frequencies has to be resonant with the two-photon Raman transition. In fact, the Stokes pulse plays the same role as the coupling between the states \( |2\rangle \) and \( |3\rangle \) in RS.

LEQM seem to be most interesting in the cases of free light non-spherical clusters and supported heavy oblate clusters. In free light clusters, the low-energy spectrum is very dilute and thus LEQM can be safely resolved. We will show that LEQM can be unambiguously identified as particular electron-hole (1eh) pairs involving the single-particle electron states near the Fermi level. This allows to obtain, at least partly, the single-particle spectrum in the clusters. Since this spectrum is sensitive to many factors (deformation, temperature, influence of detailed ionic structure, ...), we thus handle the effective method to study diverse cluster’s properties. The spectrum can be also used as a robust test for theoretical models. Besides, due to the connection between LEQM and scissors mode \[8, 9, 10, 11\], the latter can be detected as well.

Heavy clusters in experiments are usually supported and range shapes from spherical to strongly oblate \[2\]. Size and shape of supported clusters can be monitored \[13\]. If heavy clusters are supported on dielectric (and porous) surfaces, then the interface effect is mainly reduced to shaping the cluster \[2\]. Such clusters can be simulated, in a first approximation, by free clusters at a certain oblate shape. In the present paper, we pretend only to a rough estimation of LEQM properties and so this simulation is reasonable for our aims.

As a simplest case, we will consider sodium clusters. However, LEQM properties described below are of a general character and so should also take place in other clusters exhibiting quantum shells, e.g. in clusters of noble metals.

II. CALCULATION SCHEME

LEQM are described in the linear regime within the random-phase-approximation (RPA) method \[3, 4\] based on the Kohn-Sham LDA functional \[14\]. The ions are treated in the soft
jellium approximation. High reliability of the method has been checked in diverse studies of E1 plasmon in spherical [15] and deformed [4, 16] clusters.

We consider axial prolate Na$_{15}^+$ as an example of a free light cluster. Equilibrium quadrupole and hexadecapole deformations of Na$_{15}^+$ ($\delta_2 = 0.57$ and $\delta_4 = -0.18$) are determined by minimization of its total energy [4, 16]. The neutral Na$_{118}$ at oblate deformations $\delta_2 = -0.3$ and -0.5 is used as a simulation of heavy supported clusters.

STIRAP is treated as a resonance fluorescence (RF) process running through $\lambda\mu = 10$ and 11 branches of the dipole plasmon, as intermediate states (see Fig. 1). In strictly deformed nuclei, where the branches are well separated by energy (see Figs. 2 and 3), independent measurements through every branch are preferable. The population picture is then simpler and allows easier interpretation. For example, if STIRAP runs only through $\lambda\mu = 10$ plasmon branch, then the population of the low-energy quadrupole 22 mode is forbidden and thus remaining 20 and 21 modes can be easier treated.

STIRAP population of LEQM is approximately calculated as a coherent sum of independent two-step processes, every one being a product of dipole photoabsorption and emission:

$$\sigma_{2\mu_2i_2} = \sum_{i_1} \sigma_{E1\uparrow}^{ab}(0 \rightarrow 1\mu_1i_1) \cdot \sigma_{E1\downarrow}^{em}(1\mu_1i_1 \rightarrow 2\mu_2i_2) \quad (1)$$

$$\Rightarrow \sum_{i_1} \omega_{1\mu_1i_1} |<Q_{1\mu_1i_1}|E1|0>|^2(\omega_{1\mu_1i_1} - \omega_{2\mu_2i_2})^3| <Q_{2\mu_2i_2}|E1|Q_{1\mu_1i_1}^\dagger > |^2.$$

Here $|1\mu_1i_1>$ and $|2\mu_2i_2>$ are RPA states of the dipole plasmon and LEQM, respectively. Index $i$ runs all the RPA states of the given multipolarity in the chosen energy interval. The operator of the dipole transition is electron-hole in the photoabsorption and electron-electron (hole-hole) in the photoemission.

Eq. (1) follows from the general expression for RS cross-section [17] if one neglects the interference between the neighbouring RPA states. This is the case for light clusters (see Fig. 2) but not for heavy ones whose spectrum is rather dense [4, 16]. However, the interference mainly leads to smoothing the response, which can be taken into account by the reasonable averaging the results. In the present paper, we weight the low-energy responses by the Lorentz function with the averaging parameter $\Delta = 0.1$ eV. This should simulate both the interference and temperature smoothing. We emphasize that the structures discussed below are strong enough to be completely smoothed out and so the main conclusions of the present study have to be valid in spite of the interference.
Spread of collective oscillations is known to increase with the excitation energy. So, for the dipole plasmon which lies much higher than LEQM, we use the larger averaging with $\Delta = 0.25$ eV. Such averaging was successfully used in our previous RPA calculations for the photoabsorption [4, 15, 16].

III. RESULTS AND DISCUSSION

Main results of the calculation are collected in Fig. 4. The first line of the figure contains photoabsorption for low-energy quadrupole states $\lambda \mu = 20, 21$ and 22. Photoabsorption is a useful step in any analysis of electron modes. The second line of Fig. 4 exhibits photoabsorption for the scissors magnetic dipole mode. The next two lines provide the STIRAP populations for the cases when the reaction runs separately through $\lambda \mu = 10$ and $\lambda \mu = 11$ branches of the dipole plasmon (these cases are marked in Fig. 4 as RF E10 and RF E11, respectively).

In the energy intervals given in Fig. 4 (0-1.5 eV for Na$_{15}^+$ and 0-1.0 eV for Na$_{118}$), we take into account all the quadrupole RPA states. As for the dipole intermediate RPA states used in the STIRAP calculations, we involved all the states in Na$_{15}^+$ and 40-50 of most collective states in Na$_{118}$, determined by the appropriate cut-off of the photoabsorption strength.

A. Light free clusters

The first plot in the left column of Fig. 3 shows that LEQM photoabsorption in Na$_{15}^+$ is dominated by two $\lambda \mu = 21$ peaks. Following the level scheme for Na$_{15}^+$ given in Fig. 5, these peaks can be associated with electron-hole pairs [211]-[202] and [211]-[200]. Our analysis shows that i) collective shifts for these peaks are very small and ii) just the above 1eh pairs provide dominant (up to 95%) contributions to the normalization conditions of the peaks. This justifies identification of the peaks as [211]-[202] and [211]-[200] 1eh pairs. The contributions into the optical response of other quadrupole modes, 20 and 21, given in Fig. 5 is negligible because of low values of their transition single-particle matrix elements.

Both [211]-[202] and [211]-[200] 1eh pairs include the Fermi level [211] whose energy can be obtained from the ionization potential data. Then one immediately gets the energies of electron levels [202] and [200]. So, STIRAP measurements together with other data
(ionization potential, photoemission data, etc) allow to obtain the single-particle spectrum in light deformed clusters. Since this spectrum is sensitive to many factors (deformation, temperature, influence of detailed ionic structure, ...), we can use it to study diverse cluster’s properties. Beside, the spectrum can serve as a robust test for theoretical models.

The next plot for Na$_{15}^+$ exhibits the photoabsorption cross section for the scissors mode. We see again two peaks corresponding to [211]-[202] and [211]-[200] states discussed above. While the lower state [211]-[202] favors the M1 response, the higher state [211]-[200] responds mainly to E21 field. Both photoabsorption plots display a close connection (coupling) between the scissors M1 and quadrupole E21 modes. The modes are characterized by one and the same set of 1ch pairs with quantum numbers $\Lambda^\pi = 1^+$ (where $\Lambda$ is the projection of the orbital moment to the symmetry axis $z$ and $\pi$ is the space parity of the state). The coupling of electric and magnetic modes with the same quantum numbers $\Lambda^\pi$ is a general feature of deformed quantum systems. For example, this feature is well known in atomic nuclei [19].

In fact, both scissors M1 and quadrupole E21 modes represent one and the same intrinsic electron motion in a deformed cluster. The scissors mode is more specific, while the E21 mode is more general. To illustrate this point, we expand the single-particle wave functions in a deformed mean field in terms of the spherical basis $(nL\Lambda)$

$$\Psi_{\nu=[N_n\Lambda]} = \sum_{nL} a_{nL}^\nu R_{nL}(r)Y_{LA}(\Omega).$$

This allows to evaluate the single-particle orbital M1 transition amplitude between hole ($\nu = h$) and particle ($\nu = p$) states

$$\langle \Psi_p | \hat{L}_x | \Psi_h \rangle \propto \delta_{\pi_p,\pi_h} \delta_{\Lambda_p,\Lambda_h+1} \sum_{nL} a_{nL}^p a_{nL}^h \sqrt{L(L+1)-\Lambda_h(\Lambda_h\pm 1)}.$$  

Eq. (3) shows that the scissors mode is generated by $\Lambda_p = \Lambda_h \pm 1$ transitions between the components of one and the same spherical $(nL)$-level. Such selectivity is explained by the fact that the scissors operators, $\hat{L}_x$ and $\hat{L}_y$, do not depend on the space coordinate $r$ and so, due to orthogonality of the radial wave functions $R_{nL}(r)$, cannot connect the components originating from different $(nL)$-subshells. Instead, the quadrupole operator $r^2Y_{21}$ does connect the components with different $(nL)$. So, the scissors operators are more selective than the E21 operator. Every $\Lambda^\pi = 1^+$ state responds to both M1 and E21 external fields. Magnitudes of the responses depend on the wave function of the state.
The third and forth plots show STIRAP populations when the reaction runs through \( \lambda \mu = 10 \) and \( \lambda \mu = 11 \) branches of the dipole plasmon (RF E10 and E11 cases, respectively). Like in the photoabsorptions, \( \lambda \mu = 21 \) mode dominates over 20 and 22. In RF E10 case, the populations of both [211]-[202] and [211]-[200] 1eh states are equally strong. Detection of these states provides energies of electron levels [202] and [200]. Besides, the scissors mode associated with the state [211]-202 can be observed. In RF E11 case, only [211]-[200] 1eh state is well populated. The differences between RF E10 and E11 cases can be of use (for example, to distinguish [211]-[202] and [211]-[200] 1eh states).

B. Heavy oblate supported clusters

The low-energy spectrum in heavy clusters is expected to be involved. In general, this is indeed the case. However, as is seen from Fig. 4 (second and third columns), the picture essentially depends on the reaction. Some responses reveal pronounced structures. We will analyse these structures by using Fig. 5 as a guide. Though Fig. 5 shows the level scheme for the small cluster, it hints the principle trends.

The first line of Fig. 4 displays the quadrupole photoabsorption. We see bunching of the strength at 0.5-1.0 eV. The bunching involves two kinds of quadrupole transitions: i) between neighbouring subshells (like E20([220]-[200]) in Fig. 5) and ii) inside the valence subshell (like E22([220]-[202]) in Fig. 5). The second line of Fig. 4 exposes photoabsorption for the scissors mode. The scissors peak rises and blue-shifts with the deformation. Unfortunately, both LEQM and scissors modes cannot be detected in photoabsorption and their plots are given here mainly for better understanding of the subsequent STIRAP results.

The RF E10 plots demonstrate the overwhelming dominance of \( \lambda \mu = 21 \) mode over 20. This can be explained by statistical arguments. First, the mode \( \lambda \mu = 21 \) covers two projections \( \mu = \pm 1 \) instead of one in \( \lambda \mu = 20 \). Second, the mode \( \lambda \mu = 21 \) usually has much more transition single-particle matrix elements than 20.

The RF E10 response is presented by two distinct peaks. The first one originates from the deformation splitting (like E21([211]-[202]) in Fig. 5) and exhibits a blue-shift with increasing \( |\delta_2| \). This peak is not seen in E2 photoabsorption. At the same time, it displays the correlation with the scissors mode. The second peak at \( \sim 0.6 - 0.7 \) eV is mainly determined by E21 transitions between neighbouring subshells (like E21([211]-[200]) in Fig.
5) and its energy does not depend on the deformation.

In RF E11 case, the mode 22 comes to play. The response is entangled at $\delta_2 = -0.3$ but exhibits a pronounced low-energy ($\sim 0.1$ eV) $\lambda\mu = 22$ structure at $\delta_2 = -0.5$. For the first glance, this structure looks puzzling since it cannot be explained by any option discussed above. Indeed, its energy does not visibly depend on the deformation splitting (like E22([220]-[202] in Fig. 5 or 22 bump in Fig. 4, first line). It also has too low energy to be explained by transitions between the neighbouring subshells. Our analysis has revealed very specific origin of this structure. It is explained in Fig. 6 as a result of bunching $[Nn_z\Lambda]$ levels with the same $n_z$ (see E22 transitions $[523] \rightarrow [521]$ and $[532] \rightarrow [530]$). The bunching signifies that in large systems with strong deformation the asymptotic Nilsson-Clemenger quantum number $n_z$ (number of quants along the symmetry axis $z$) becomes exact.

C. STIRAP discussion

Some essential points concerning detection of LEQM in STIRAP should be commented. The lifetime of the dipole plasmon is mainly determined by the Landau fragmentation while the radiative decay of the plasmon usually plays a minor role. The advantage of the STIRAP is that it strongly enhances the population of the desirable low-energy states in spite of a weak natural radiative decay to them from intermediate states.

To distinguish in STIRAP the electron LEQM from possible excitations of other nature (contributions of impurities, ionic modes, etc), one can use the feature of electronic excitations to exhibit the inverse dependence of their energy on cluster size.

It worth noting that STIRAP deals with both free and supported systems.

IV. CONCLUSIONS

The properties of the low-energy quadrupole modes (LEQM) in deformed clusters were analysed within the Kohn-Sham LDA RPA approach. The stimulated Raman adiabatic passage (STIRAP) was considered as the most suitable experimental method to detect LEQM. The population of the LEQM in STIRAP was calculated for two special cases: free light deformed clusters and supported heavy strongly-oblate clusters. In the first case, LEQM spectrum is dilute and can be resolved in STIRAP. The spectrum is easily associated with
particular electron-hole pairs. This finally allows to determine the single-particle energies near the Fermi level. Besides, close connection between the $\lambda\mu=21$ and scissors modes allows to observe the latter.

In heavy clusters, STIRAP population through the $\lambda\mu=10$ branch of the dipole plasmon is strictly dominated by two $\lambda\mu=21$ peaks. The nature of the peaks is explained. In the population through the $\lambda\mu=11$ branch, the soft $\lambda\mu=22$ mode devotes a special attention. It originates from the fact that in large systems with strong deformation the asymptotic Nilsson-Clemenger quantum number $n_z$ becomes exact.

It worth noting, that the LEQM properties described above are of a general character and so should take place not only in sodium clusters considered here but also in other clusters with quantum shells, for example in clusters of noble metals. Since any other information on electron LEQM in deformed clusters is absent, our study can be used as a first guide in the field.

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**Figure captions:**

**Figure 1:** Scheme of population of the low-energy quadrupole states $\lambda\mu = 20, 21$ and 22 in RS, RF and STIRAP reactions through $\lambda\mu = 10$ (left) and 11 (right) branches of the dipole plasmon.

**Figure 2:** Photoabsorption cross section for the dipole plasmon in Na$_{15}^+$, Na$_{19}^+$ and Na$_{27}^+$. Parameters of quadrupole and hexadecapole deformations are given in boxes. The experimental data [18] (triangles) are compared with our results given as bars (for every RPA state) and the strength function smoothed by the Lorentz weight (with averaging parameter 0.25 eV). Contributions of $\lambda\mu = 10$ and 11 dipole branches (the latter is twice stronger) are given by dashed curves. The bars are given in eV\(\AA^2\).

**Figure 3:** The dipole plasmon in Na$_{118}$ at oblate deformations $\delta_2 = -0.1$ (first line), $\delta_2 = -0.3$ (second line), and $\delta_2 = -0.5$ (third line). Contributions of $\lambda\mu = 10$ and 11 dipole branches (the latter is twice stronger) are given by dashed curves. The photoabsorption is smoothed by the Lorentz weight with $\Delta = 0.25$ eV.

**Figure 4:** Low-energy electron modes in Na$_{15}^+$ (left column) and Na$_{118}$ at $\delta_2 = -0.3$ (middle column) and $\delta_2 = -0.5$ (right column). The plots exhibit LEQM photoabsorption (first line), scissors M1 photoabsorption (second line), STIRAP population of LEQM through $\lambda\mu = 10$ (third line) and 11 (forth line) dipole branches. LEQM are depicted by solid ($\lambda\mu = 20$), dashed ($\lambda\mu = 21$), and dotted ($\lambda\mu = 22$) curves. All the responses are smoothed by the Lorentz weight with $\Delta = 0.1$ eV.

**Figure 5:** The electron level scheme for Na$_{15}^+$ in the spherical limit (left) and at the equilibrium deformation (right). The Fermi level is [211]. Arrows depict the possible low-energy quadrupole hole-electron $E2\mu$ transitions.

**Figure 6:** Deformation splitting of the electron subshell $2f$ in Na$_{118}$. The oblate deformations are listed below the plot. The levels are marked by Nilsson-Clemenger quantum numbers [$Nn\Lambda$]. The plot demonstrates the origin of the soft E22 transitions.
E10  E11

E10

E10

E11

E11

E11  E10

20  22  21

20  22  21

g. s.
$\sigma(E_1)/N_e \text{ [Å}^2\text{]}$

| Na$^+$ |
|-------|
| $\omega$ [eV] |
| 1.5 | 2.5 | 3.5 |
| Na$_{15}^+$ | δ$_2$ = 0.57 | δ$_4$ = -0.18 |
| Na$_{19}^+$ | δ$_2$ = -0.285 | δ$_4$ = -0.09 |
| Na$_{27}^+$ | δ$_2$ = 0.33 | δ$_4$ = 0.08 |
\[
\sigma(E1)/N_e [\text{Å}^2] \\
\delta_2 = -0.1 \\
\delta_2 = -0.3 \\
\delta_2 = -0.5 \\
\omega [\text{eV}]
\]
\[ \text{Na}_{118} \]

\[ E_2 = 0 \quad E_2 = -0.1 \quad E_2 = -0.3 \quad E_2 = -0.5 \]