Rotational Magnetic State in Deformed Metal Clusters

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A magnetic state of orbital nature is predicted in deformed metal clusters. The restoring force of this state originates from a quantum effect associated with the kinetic Fermi motion. For Na clusters the frequency is predicted to be $\omega_{M1}=\delta 4.6N^{-1/3}$ eV, where δ is the deformation of the cluster.

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Collective electric dipole excitations have been recently observed in alkali-metal clusters.[1] These excitations correspond to the classical Mie[2] surface-plasmon oscillations and have revealed the importance of deformation effects.[3] In this Letter we suggest the existence of a new magnetic collective state in deformed clusters occurring at energies much smaller than the plasma frequency. Such an excitation has no classical counterpart and emerges from the quantum effect associated with the Fermi motion of valence electrons. A collective excitation of similar nature has been recently observed in deformed atomic nuclei via inelastic electron scattering as well as (γ,γ') reactions.[4]

A macroscopic illustration of the new magnetic state is suggested by the following form for the displacement field relative to the electronic motion:[5]

$$\mathbf{u}=\boldsymbol{\omega}\times\mathbf{r}+\frac{\delta}{1+\delta/3}\nabla(yz)\,. \tag{1}$$

In Eq. (1) $\boldsymbol{\omega}$ is the unit vector in the x direction (the cluster is assumed to be axially deformed along the z direction) and $\delta=\frac{3}{2}(R_z^2-R_y^2)/(R_z^2+2R_y^2)$ is the deformation of the electron density profile which is assumed to be of spheroidal shape: $\rho_e=\rho_e(x^2/R_x^2+y^2/R_y^2+z^2/R_z^2)$ (R_z and $R_x=R_y$ are the radii parallel and perpendicular to the symmetry axis, respectively).

The term $\boldsymbol{\omega}\times\mathbf{r}$ of Eq. (1) corresponds to a rigid rotation of the electrons with respect to the jellium background [scissor mode;[6] see Fig. 1(a)]. If only this term were included in the electronic motion, the electrons would experience a restoring force originating from the Coulomb interaction with the jellium, similarly to what happens in the dipole Mie oscillation. The cost in the Coulomb energy is minimized by including the quadrupole term in $\nabla(yz)$ in the displacement field. This is well understood by noting that the change in the electron density ρ_e,

$$\delta\rho_e=\nabla\cdot(\mathbf{u}\rho_e)\,, \tag{2}$$

is zero with choice (1) since $\mathbf{u}\cdot\nabla\rho_e=0$ in the spheroidal model. The resulting motion is illustrated in Fig. 1(b) where we have taken a sharp density profile. In this case one gets a rotation within a spheroid with a rigid surface. The relevant restoring force taking place during the motion goes not originate from the Coulomb interaction. Rather it is produced by the quadrupole component $\nabla(yz)$ of the velocity field which gives rise to a distortion of the Fermi sphere of the valence electrons. This effect of quantum nature has been extensively studied in the case of atomic nuclei and reveals an elastic behavior exhibited by Fermi systems.[7]

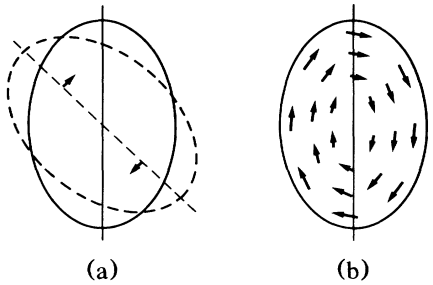
The frequency of the resulting mode can be estimated through the expression $\omega_{M1}=(K/\theta)^{1/2}$, where $K=2E(\mathbf{u})$ is fixed by the energy change $E(\mathbf{u})$ associated with the displacement field (1), and $\frac{1}{2}\theta=\frac{1}{2}m\rho_0\int dV\,\mathbf{u}^2$ is the collective mass parameter. In the limit of small deformations we find ($\hbar=c=1$)

$$\omega_{M1}=\delta\left(\frac{4\epsilon_F}{mr_s^2}\right)^{1/2}N^{-1/3}\,. \tag{3}$$

The Fermi energy ϵ_F entering Eq. (3) originates from the elastic behavior associated with the distortions of the Fermi surface discussed above. In fact, the shear contribution to the energy of an elastic medium is given by

$$E(\mathbf{u})=\tfrac{1}{4}\mu\int dV\sum_{k,l}(\nabla_k u_l+\nabla_l u_k)^2$$

and the Lame elastic constant μ is related to the Fermi energy[7] by $\mu=\frac{2}{5}\epsilon_F\rho_0$, where $\rho_0=3/4\pi r_s^3$ is the electron-



(a) (b)

FIG. 1. Displacement field for the $M1$ low-lying rotational state; (a) corresponds to a rigid rotation (scissor mode) of the electrons with respect to the jellium background and (b) corresponds to a rotation within a rigid surface.

ic density. To derive Eq. (3) we have used the approximation $\langle r^2\rangle = \frac{3}{5} r_s^2 N^{2/3}$. Differently from the plasma frequency, the energy (3) exhibits a $N^{-1/3}$ dependence. It is worth noting also that for light clusters this frequency lies much below the Mie dipole mode $\omega_{\rm Mie} = \omega_p/\sqrt{3}$, where $\omega_p = (3e^2/mr_s^3)^{1/2}$ is the plasma frequency. For example, for Na clusters ($r_s = 4$ a.u., $\epsilon_F = 3.1$ eV) we find

$$\omega_{M1}({\rm Na}) = \delta 4.6 N^{-1/3}\ {\rm eV}\,, \tag{4}$$

whereas the Mie frequency occurs at $\omega_{\rm Mie} = 3.4$ eV.

The collective state (3) carries a considerable amount of orbital magnetic strength which can be evaluated in terms of the frequency (3) and of the mass parameter θ through

$$B(M1) \simeq \mu_B^2 \omega_{M1}\theta = \tfrac{4}{5}\mu_B^2 \delta(\epsilon_F m r_s^2)^{1/2} N^{4/3}\,, \tag{5}$$

where $\mu_B = e/2m$ is the Bohr magneton. For Na clusters Eq. (5) yields

$$B(M1) \simeq \mu_B^2 \delta N^{4/3}\,.$$

It can be useful to evaluate the ratio between the magnetic dipole strength (5) and the electric strength associated with the Mie dipole frequency. We find

$$\frac{B(M1)}{B(E1)} = \frac{\sum_k |\langle 0|\sum_{i=1}^N \mu_B l_i^k |M1\rangle|^2}{\sum_k |\langle 0|\sum_{i=1}^N e r_i^k |E1\rangle|^2} \simeq \tfrac{1}{15}\omega_{M1}\omega_{\rm Mie} R^2\,. \tag{6}$$

In Eq. (6) only the terms $k = x,y$ contribute to the magnetic strength. The value of this ratio turns out to be of the order of 10^{-6}.

The above results can be also derived using a microscopic calculation based on the random-phase approximation (RPA) with a separable quadrupole-quadrupole force. The equation to solve is

$$2\sum_i \frac{\epsilon_i |\langle 0|F|\epsilon_i\rangle|^2}{\epsilon_i^2 - \omega^2} = \frac{1}{\chi}\,, \tag{7}$$

where ϵ_i and $\langle 0|F|\epsilon_i\rangle$ are the single-particle excitation energies and matrix elements that we calculate with the deformed harmonic-oscillator Hamiltonian

$$H_0 = \sum_i \left[\frac{p_i^2}{2m} + \tfrac{1}{2} m[\omega_y^2(x_i^2+y_i^2) + \omega_z^2 z_i^2]\right], \tag{8}$$

where $\omega_y = \omega_0(1+\frac{1}{3}\delta)$ and $\omega_z = \omega_0(1-\frac{2}{3}\delta)$. Here the value of δ is fixed by the usual minimization procedure.[8] The RPA coupling constant χ associated with the excitation operator $F = \sum_{i=1}^N y_i z_i$ is given by (in the following, higher-order effects in δ will be neglected)

$$\chi = -\frac{3}{5}\frac{m\omega_p^2}{N\langle r^2\rangle} + 3\frac{m\omega_0^2}{N\langle r^2\rangle}\,. \tag{9}$$

The first term (in ω_p^2) originates from the direct (long range) part of the Coulomb electron-electron interaction, while the second one comes from the zero-range local-density approximation interaction accounting for exchange and correlation effects (an explicit derivation of the separable quadrupole-quadrupole force will be presented in a future work[9]). Note that the direct electron-electron Coulomb interaction does not affect the static Hamiltonian because the resulting static potential is screened by the jellium background.

Two single-particle levels contribute to the sum of Eq. (7). They occur at the energies $\epsilon_0 = \omega_y - \omega_z = \delta\omega_0$ and $\epsilon_2 = \omega_y + \omega_z = 2\omega_0$ and have matrix elements given by

$$|\langle 0|F|\epsilon_0\rangle|^2 = N\langle r^2\rangle\delta/12m\omega_0$$

and

$$|\langle 0|F|\epsilon_2\rangle|^2 = N\langle r^2\rangle/6m\omega_0\,,$$

respectively.[10] Two solutions then emerge from the RPA equation (7). The highest in energy is the quadrupole plasmon excitation occurring at

$$\omega_{2+} = (2\omega_0^2 + \tfrac{2}{5}\omega_p^2)^{1/2}\,. \tag{10}$$

This frequency coincides with the classical quadrupole plasma value apart from the term in $2\omega_0^2$ which originates from the quantum Fermi motion.[11] The lowest one occurs at the energy

$$\omega_{M1} = \sqrt{2}\omega_0\delta(1 + 5\omega_0^2/\omega_p^2)^{-1/2}\,. \tag{11}$$

The magnetic strength carried by this state can be easily evaluated in the RPA model. We find

$$B(M1) = 2\left|\langle 0|\mu_B \sum_{i=1}^N l_i^x |\omega_{M1}\rangle\right|^2 = \mu_B^2 2\frac{\sqrt{2}}{3} m\omega_0 N\langle r^2\rangle\delta\,. \tag{12}$$

It is easy to show that the state (11), differently from the quadrupole one [Eq. (10)], has in practice no quadrupole strength. This follows from the coupling between the rotational and quadrupole motions already discussed in the macroscopic model [see Eq. (1)]. It is worth noting that the effect of the RPA correlations is to renormalize not only the frequency of the high-lying solution (10), but also the one of the low-lying mode (11), with respect to the predictions ($2\omega_0$ and $\delta\omega_0$, respectively) of the independent-particle picture.

The equivalence between the results of the RPA calculation and of the macroscopic model discussed in the first part of the work is straightforwardly obtained using the relation

$$\omega_0 = (2\epsilon_F/mr_s^2)^{1/2} N^{-1/3} \tag{13}$$

obtained by identifying the kinetic energy per particle given by the harmonic-oscillator model with the one of the Fermi gas and by approximating $\langle r^2\rangle$ with $\frac{3}{5} r_s^2 N^{2/3}$ [the correction $5\omega_0^2/\omega_p^2$ entering Eq. (11) can be safely

ignored].

Equations (11) and (12) are easily generalized to the case of triaxially deformed clusters. In this case three distinct excitations are predicted corresponding to the three different excitation operators

$$F^1=\sum_{i=1}^{N} y_i z_i\,,\quad F^2=\sum_{i=1}^{N} x_i z_i\,,\qquad F^3=\sum_{i=1}^{N} x_i y_i\,. \tag{14}$$

The resulting frequencies are

$$\omega^1_{M1}=[\cos\gamma+(1/\sqrt{3})\sin\gamma]\omega_{M1}\,,$$
$$\omega^2_{M1}=[\cos\gamma-(1/\sqrt{3})\sin\gamma]\omega_{M1}\,, \tag{15}$$
$$\omega^3_{M1}=(2/\sqrt{3})\sin\gamma\,\omega_{M1}\,,$$

where γ characterizes the triaxial nature of the deformation according to the usual expression for the deformed radius:[8]

$$R(\vartheta,\varphi)=R_0[1+\tfrac{1}{3}\delta\cos\gamma(3\cos^2\vartheta-1)+(1/\sqrt{3})\delta\sin\gamma\sin^2\vartheta\cos^2\varphi]$$

and ω_{M1} is given by Eq. (11). The magnetic strength relative to the above excitations is given by $B^i(M1)=\frac{1}{2}\mu_B^2\omega^i_{M1}\theta$.

In conclusion, we have predicted a magnetic state of orbital nature in deformed clusters. The frequency of this mode exhibits a bulk $N^{-1/3}$ dependence which distinguishes it from the surface-plasmon mode. For clusters with $N=10$–100 and typical deformations $\delta=0.2$–0.4 the frequency of this state is predicted to occur at the energy $\omega=0.2$–0.6 eV. This state lies below the threshold for particle emission and carries a considerable amount of magnetic strength. Clearly a more microscopic investigation of this state, based, for example, on a quasiparticle RPA calculation[12] with realistic single-particle wave functions, would be very important for a better understanding of its collective nature and of the possible mechanisms of fragmentation of the magnetic strength.

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