COLLECTIVE SPIN STATES IN THE ELECTRON GAS IN DIFFERENT DIMENSIONS AND GEOMETRIES *)

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We study spin longitudinal and transverse linear response of the 3-dimensional electron gas, metal clusters and quantum dots. When the systems are spin unpolarized in the ground state, a low energy collective state emerges in finite size systems due to the discrete shell structure, whereas it is absent in the bulk due to the Landau damping. In the case of spin polarization of the ground state a collective state is present also in the bulk and a family of new collective states appears in finite size systems.

In this paper we study the excited states of electron systems which are induced by the external fields $F^{+,-,z} = \sum_{i=1}^{N} f(\boldsymbol{r}_i)\sigma_i^{+,-,z}$, where $\sigma_i^{+,-,z}$ are the spherical components of the vector of Pauli matrices which represents the spin operator. We use the following definitions: $\sigma^{\pm} = 1/2(\sigma_x \pm i\sigma_y)$, $\sigma^+|\uparrow\rangle = 0$, $\sigma^+|\downarrow\rangle = |\uparrow\rangle$, $\sigma^-|\uparrow\rangle = |\downarrow\rangle$, $\sigma^-|\downarrow\rangle = 0$, $\sigma^z|\uparrow\rangle = |\uparrow\rangle$, $\sigma^z|\downarrow\rangle = -|\downarrow\rangle$. These fields enter the interaction Hamiltonian of the electron systems with an oscillating magnetic field

$$H_{\text{int}} = \tfrac{1}{2}\mu_{\text{B}} g B \sum_{i=1}^{N} \vec{\sigma}_i \cdot (\hat{\boldsymbol{k}} \times \hat{e}) \mathrm{e}^{\mathrm{i}(\boldsymbol{k}\cdot\boldsymbol{r}_i - \omega t)}, \tag{1}$$

where $\mu_{\text{B}} = e\hbar/(2mc)$ and g is the gyromagnetic factor. The response induced by this interaction Hamiltonian has been recently observed in GaAs layers by means of scattering of polarized light in the backscattering geometry [1].

When the system is spin unpolarized in the ground state, it is the same to excite it with F^+, F^- or F^z since the three final states can be related by a rotation in spin space. However, when the system has a spin different from zero in the ground state, which can be reached by putting it into a static magnetic field along the z-direction, significant differences emerge among the final states and one observes the existence of splittings between the excited states with different ΔS_z. In the following we will study the linear response of the electron gas, metal clusters and quantum dots in the time dependent local spin density approximation (TDLSDA) in two cases: a) the longitudinal $\Delta S_z = 0$ channel in the case of zero magnetization $\langle\sum_i \sigma_i^z\rangle = 0$ in the ground state; b) the transverse $\Delta S_z = \pm 1$ channel in the case

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of a magnetization $\langle\sum_i \sigma_i^z\rangle = (N_\uparrow - N_\downarrow) \neq 0$ in the ground state, where $N_\uparrow$ ($N_\downarrow$) is the number of electrons with spin up (down).

In the $\Delta S_z = 0$ channel, for zero magnetization, the TDLSDA linear response to the external field $F^z = \sum_{i=1}^N f(r_i)\sigma_i^z$ is given [2] by $\alpha_0(f,\omega) = \int \mathrm{d}\boldsymbol{r} f^*(r)\delta m_0(r,\omega)$, where $\delta m_0(r,\omega)$ is the variation of the z-component of the magnetization density $\vec{m}(\boldsymbol{r}) = \langle\sum_i \vec{\sigma}_i\delta(\boldsymbol{r}-\boldsymbol{r}_i)\rangle$. It is induced by the external field on the system and is given by

$$\delta m_0(\boldsymbol{r},\omega) = \int \mathrm{d}\boldsymbol{r}'\,\chi_{00}(\boldsymbol{r},\boldsymbol{r}',\omega) f(\boldsymbol{r}')\,. \tag{2}$$

$\chi_{00}(\boldsymbol{r},\boldsymbol{r}',\omega)$ is the correlation function, solution of the Dyson type integral equation

$$\chi_{00}(\boldsymbol{r},\boldsymbol{r}',\omega) = \chi_{00}^0(\boldsymbol{r},\boldsymbol{r}',\omega) + \int \mathrm{d}\boldsymbol{r}_1\mathrm{d}\boldsymbol{r}_2\,\chi_{00}^0(\boldsymbol{r},\boldsymbol{r}_1,\omega)\left.\frac{\delta W_{xc}}{\delta m}\right|_{m=0}\delta(\boldsymbol{r}_1-\boldsymbol{r}_2)\chi_{00}(\boldsymbol{r}_2,\boldsymbol{r}',\omega)\,, \tag{3}$$

and the independent-particle longitudinal response function χ_{00}^0 can be expressed in terms of the occupied single particle states $\varphi_{h\sigma}$ and energies $\varepsilon_{h\sigma}$ and the unoccupied ones $\varphi_{p\sigma}$ and $\varepsilon_{p\sigma}$ as

$$\chi_{00}^0(\boldsymbol{r},\boldsymbol{r}',\omega) = \sum_{\sigma\sigma'}\sum_{hp}\left[\frac{\varphi_{h\sigma'}^*(\boldsymbol{r})f(r)\sigma^z\varphi_{p\sigma}(\boldsymbol{r})\varphi_{p\sigma}^*(\boldsymbol{r}')f(r')\sigma^z\varphi_{h\sigma'}(\boldsymbol{r}')}{\omega+\mathrm{i}\eta-\varepsilon_p+\varepsilon_h} - \frac{\varphi_{p\sigma}^*(\boldsymbol{r})f(r)\sigma^z\varphi_{h\sigma'}(\boldsymbol{r})\varphi_{h\sigma'}^*(\boldsymbol{r}')f(r')\sigma^z\varphi_{p\sigma}(\boldsymbol{r}')}{\omega+\mathrm{i}\eta+\varepsilon_p-\varepsilon_h}\right]. \tag{4}$$

The single particle states are obtained as solutions of the LSDA equations

$$\left[-\frac{1}{2}\nabla^2 + \int \mathrm{d}\boldsymbol{r}'\frac{\rho(r')-\rho_j(r')}{|\boldsymbol{r}-\boldsymbol{r}'|} + V_{xc}(\rho,m) + W_{xc}(\rho,m)\sigma^z\right]\varphi_{i\sigma}(\boldsymbol{r}) = \epsilon_{i\sigma}\varphi_{i\sigma}(\boldsymbol{r})\,, \tag{5}$$

and in (3) $\delta W_{xc}/\delta m|_{m=0}\delta(\boldsymbol{r}_1-\boldsymbol{r}_2)$ is the residual interaction obtained by the spin dependent part of the exchange correlation potential of (5). The potentials in (5) are given by $V_{xc} = \delta E_{xc}(\rho,m)/\delta\rho$ and $W_{xc} = \delta E_{xc}(\rho,m)/\delta m$, where E_{xc} is the exchange correlation energy which has been taken from quantum Monte Carlo calculations, and satisfies the relations: $V_{xc}(\rho_0, m_0 = 0) \neq 0$, $W_{xc}(\rho_0, m_0 = 0) = 0$, $\delta W_{xc}/\delta m|_{m=0} \neq 0$, $\delta V_{xc}/\delta m|_{m=0} = 0$. The residual interaction in the spin channel is always negative (attractive).

In the bulk the integral equation (3) yields the following analytical expression for the polarizability

$$\alpha_0(q,\omega) = \frac{\alpha_0^{\text{free}}(q,\omega)}{1-\left.\frac{\delta W_{xc}}{\delta m}\right|_{m=0}\alpha_0^{\text{free}}(q,\omega)}\,, \tag{6}$$

where $\alpha_0^{\text{free}}(q,\omega)$ is the Lindhard free response function (notice that the free response is the same in the spin longitudinal and density channels) and $\delta W_{xc}/\delta m|_{m=0}$

is the residual interaction in momentum space. The eigenstates of the system are the poles of α_0, given by the solutions of the equation $\alpha_0^{\rm free}(q,\omega) = 1/(\delta W_{xc}/\delta m|_{m=0})$.

Since the residual interaction is negative, one realizes immediately that collective solutions do not emerge from this equation, but only a continuum of single particle type solutions extending from zero to the energy $qv_{\rm F}$, where $v_{\rm F}$ is the Fermi velocity. These solutions correspond to the poles of the free response function. This should be contrasted with the case of the density channel where a collective state emerges as a solution of the TDLDA equation for the polarizability since in this channel the residual interaction is repulsive and equal to $4\pi/q^2$ in the small q limit.

The situation is, however, different in finite size systems like metal clusters and quantum dots due to the existence in these systems of a shell structure that gives rise to a gap in the single particle spectrum at low energy. An undamped low energy collective state can then survive in this case. It has been studied in metal clusters within the TDLSDA in Ref. 3, within the sum rule approach in Refs. 4, 5, and in quantum dots in Ref. 6. In Figs. 1 and 2 we compare the spin response of the metal cluster Na_{58} and the quantum dot of GaAs containing 12 electrons with the density and the free ones. From the figures one notices that in both channels the response is dominated by a single peak which exhausts a large fraction of the f-sum rule and that in the spin channel the energy of the peak is much lower than in the density channel. This is due to the effect of the residual interaction which is of opposite sign in the two channels and shifts the peak energy, with respect to the single particle response, in opposite directions. We also notice that this shift is much larger in the density channel than in the spin one.

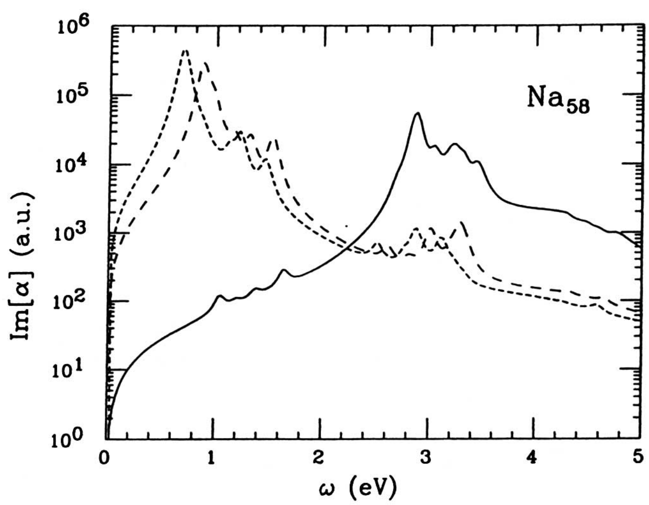

Fig. 1. Imaginary part of the polarizability in the spin (dashed) and density (solid) response of the Na_{58} cluster in atomic units. For comparison, the free response (long-dashed) is also shown.

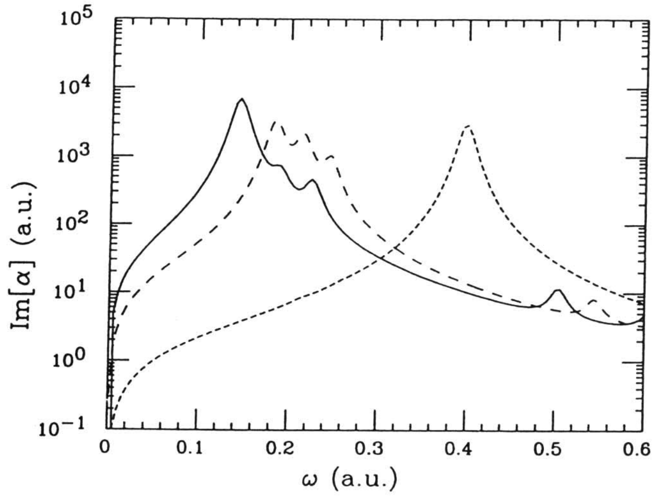

Fig. 2. Imaginary part of the polarizability in the spin (solid) and density (dashed) response of a spin unpolarized quantum dot of GaAs with 12 electrons in effective atomic units. For comparison, the free response (long-dashed) is also shown.

In the $\Delta S_z = \pm 1$ channel, for a magnetization different from zero in the ground state obtained by putting the system in a static magnetic field $\boldsymbol{B}$, the LSDA equations become:

$$\left[\frac{1}{2}\left(\boldsymbol{p}+\frac{e}{c}\boldsymbol{A}\right)^2 + \int \mathrm{d}\boldsymbol{r}'\frac{\rho(\boldsymbol{r}')-\rho_j(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|} + V_{xc}(\rho,m) + \left(f_{xc}\vec{m}+\frac{1}{2}g\mu_{\mathrm{B}}\boldsymbol{B}\right)\cdot\vec{\sigma}\right]\varphi_{i\sigma}(\boldsymbol{r}) = \epsilon_{i\sigma}\varphi_{i\sigma}(\boldsymbol{r}), \tag{7}$$

where $f_{xc} = W_{xc}/m$, $m = |\vec{m}\,|$ and in the ground state $m_x = 0, m_y = 0, m_z = m_0$ and the vector potential $\boldsymbol{A} = 1/2(-y, x, 0)B$ describing the magnetic field B in the z direction gives rise to diamagnetic effects, which in the following will be taken into account in the quantum dots and not in the electron gas. The spin dependent terms of (7) give rise to different and coupled equations for spin up and down single particle wave functions which are solved using iteration techniques. In the case of the bulk the magnetization induced in the electron system by the static magnetic field can be calculated analytically and is given by

$$\int \mathrm{d}\boldsymbol{r}(\rho_\uparrow - \rho_\downarrow) = N_\uparrow - N_\downarrow = \mu_{\mathrm{B}}B\frac{\dfrac{3\rho}{2\epsilon_{\mathrm{F}}}}{1+\dfrac{3\rho}{2\epsilon_{\mathrm{F}}}\dfrac{\delta W_{xc}}{\delta m}\Big|_{m=0}}, \tag{8}$$

where ϵ_{F} is the Fermi energy. When the system interacts with the external field $F^- = \sum_{i=1}^N f(r_i)\sigma_i^- \mathrm{e}^{-\mathrm{i}\omega t} \equiv \sigma_f^- \mathrm{e}^{-\mathrm{i}\omega t}$, the potential $2f_{xc}(m^+\sigma^- + m^-\sigma^+)$, entering the scalar product $f_{xc}\vec{m}\cdot\vec{\sigma}$ of (7) and statically equal to zero, changes due to the changes δm^{+} dynamically induced in the magnetization m^{+}. The transverse linear response function is defined by $\alpha_{\pm}(f,\omega)=\langle(\sigma_f^{-})^{\dagger}\rangle=\int \mathrm{d}\boldsymbol{r} f^{*}(\boldsymbol{r})\delta m^{+}(\boldsymbol{r},\omega)$, with

$$\delta m^{+}(\boldsymbol{r},\omega)=\int \mathrm{d}\boldsymbol{r}'\,\chi_{\pm}(\boldsymbol{r},\boldsymbol{r}',\omega)f(\boldsymbol{r}')\,, \tag{9}$$

and $\chi_{\pm}(\boldsymbol{r},\boldsymbol{r}',\omega)$ is the correlation function, solution of the Dyson type integral equation

$$\chi_{\pm}(\boldsymbol{r},\boldsymbol{r}',\omega)=\chi_{\pm}^{0}(\boldsymbol{r},\boldsymbol{r}',\omega)+\int \mathrm{d}\boldsymbol{r}_1\mathrm{d}\boldsymbol{r}_2\,\chi_{\pm}^{0}(\boldsymbol{r},\boldsymbol{r}_1,\omega)f_{xc}(m_0)\delta(\boldsymbol{r}_1-\boldsymbol{r}_2)\chi_{\pm}(\boldsymbol{r}_2,\boldsymbol{r}',\omega)\,, \tag{10}$$

where

$$\chi_{\pm}^{0}(\boldsymbol{r},\boldsymbol{r}',\omega)=\sum_{\sigma\sigma'}\sum_{hp}\left[\frac{\varphi_{h\sigma'}^{*}(\boldsymbol{r})f^{*}(\boldsymbol{r})\sigma^{+}\varphi_{p\sigma}(\boldsymbol{r})\varphi_{p\sigma}^{*}(\boldsymbol{r}')f(\boldsymbol{r}')\sigma^{-}\varphi_{h\sigma'}(\boldsymbol{r}')}{\omega+\mathrm{i}\eta-\varepsilon_p+\varepsilon_h}\right.$$
$$\left.-\frac{\varphi_{p\sigma}^{*}(\boldsymbol{r})f^{*}(\boldsymbol{r})\sigma^{+}\varphi_{h\sigma'}(\boldsymbol{r})\varphi_{h\sigma'}^{*}(\boldsymbol{r}')f(\boldsymbol{r}')\sigma^{-}\varphi_{p\sigma}(\boldsymbol{r}')}{\omega+\mathrm{i}\eta+\varepsilon_p-\varepsilon_h}\right] \tag{11}$$

is the free transverse correlation function, built with the solutions of the LSDA equations (7). Notice that when the system is polarized $\chi_{\pm}^{0}\neq\chi_{00}^{0}$.

In the 3d electron gas, where $F^{\pm}\equiv\sigma_q^{\pm}=\sum_{i=1}^{N}\sigma_i^{\pm}\mathrm{e}^{\mp\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}$ one gets for the transverse response

$$\alpha_{\pm}(q,\omega)=\sum_n\frac{|\langle n|\sigma_q^{-}|0\rangle|^2}{\omega-\omega_{n0}+\mathrm{i}\eta}-\sum_n\frac{|\langle n|\sigma_q^{+}|0\rangle|^2}{\omega+\omega_{n0}-\mathrm{i}\eta}\,, \tag{12}$$

the TDLSDA expression

$$\alpha_{\pm}(q,\omega)=\frac{\alpha_{\pm}^{\mathrm{free}}(q,\omega)}{1-2f_{xc}(\rho_0,m_0)\alpha_{\pm}^{\mathrm{free}}(q,\omega)}\,, \tag{13}$$

where $\alpha_{\pm}^{\mathrm{free}}(q,\omega)$ is the free transverse response. In the $qv_{\mathrm{F}}\ll\epsilon_{\mathrm{F}}$ limit it is given by

$$\alpha_{\pm}^{\mathrm{free}}(q,\omega)=-\frac{3}{4}\frac{\rho_0}{\epsilon_{\mathrm{F}}}\left(1+\frac{\omega}{2qv_{\mathrm{F}}}\ln\frac{\omega-\omega_a-qv_{\mathrm{F}}}{\omega-\omega_a+qv_{\mathrm{F}}}\right)\,, \tag{14}$$

where $\omega_a=\omega_{\mathrm{L}}/(1+3\rho_0 f_{xc}(\rho_0,m_0)/(2\epsilon_{\mathrm{F}}))$ and $\omega_{\mathrm{L}}=eB/(mc)$ is the Larmour frequency, and is different from the Lindhard free response

$$\alpha_{00}^{\mathrm{free}}(q,\omega)=-\frac{3\rho_0}{2\epsilon_{\mathrm{F}}}\left(1+\frac{\omega}{2qv_{\mathrm{F}}}\ln\frac{\omega-qv_{\mathrm{F}}}{\omega+qv_{\mathrm{F}}}\right). \tag{15}$$

When the static spin polarizing field B is zero, then $\omega_{\mathrm{L}}=\omega_a=0$, $f_{xc}(\rho_0,m_0)=\delta W_{xc}/\delta m|_{m=0}$ and (13) coincides with (6) and the electron gas does not display

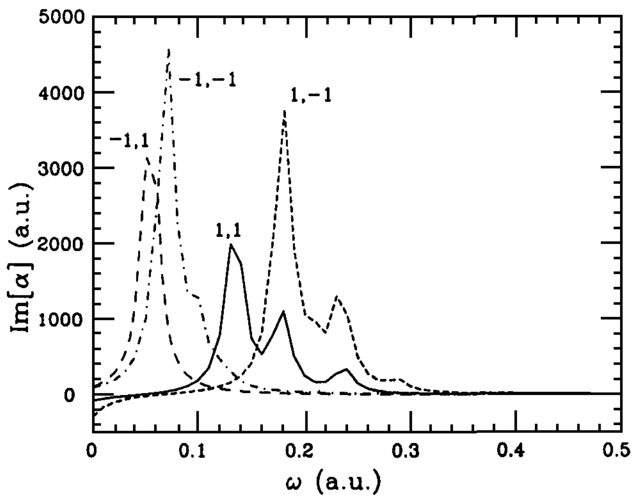

Fig. 3. Imaginary part of the transverse polarizability of a spin polarized quantum dot of GaAs with 6 spin up and 5 spin down electrons in effective atomic units. The full line is the $\Delta L_z = 1, \Delta S_z = 1$ response, the dotted line is the $\Delta L_z = 1, \Delta S_z = -1$ response, the dashed line is the $\Delta L_z = -1, \Delta S_z = 1$ response, the dotted-dashed line is the $\Delta L_z = -1, \Delta S_z = -1$ response.

a collective state in the spin channel. However, when B is different from zero and sufficiently high, new solutions emerge.

From (12) one sees that the poles of $\alpha_\pm(q,\omega)$ with positive frequency are excitations induced by σ_q^- and the poles of $\alpha_\pm(q,\omega)$ with negative frequency are excitations induced by σ_q^+. Furthermore, taking the $\omega \to \infty$ limit of (12), one gets $\alpha_\pm(q,\omega)|_{\omega\to\infty} \simeq s_0/\omega$ with $s_0 = \sum_n |\langle n|\sigma_q^-|0\rangle|^2 - \sum_n |\langle n|\sigma_q^+|0\rangle|^2 = N_\uparrow - N_\downarrow$, a model independent sum rule. The poles of (12) are solutions of the equation

$$1 - 2f_{xc}(\rho_0, m_0)\alpha_\pm^{\text{free}}(q,\omega) = 0\,, \tag{16}$$

that in the $q \to 0$ limit and for $qv_{\text{F}}/\omega_a < 1$ (high magnetic field) has a collective undamped solution at the energy

$$\omega(q) = \omega_{\text{L}} + \frac{1}{3}\frac{q^2 v_{\text{F}}^2}{\omega_a}\frac{F}{1+F}\,, \tag{17}$$

with $F = 3\rho_0 f_{xc}(\rho_0, m_0)/(2\epsilon_{\text{F}})$.

This solution is an elementary excitation induced by the operator σ_q^- since it exhausts completely the sum rule s_0 giving $|\langle\text{coll.}\,|\sigma_q^-|0\rangle|^2 = N_\uparrow - N_\downarrow$, the strength $\sum_n |\langle n|\sigma_q^+|0\rangle|^2$ being zero as a consequence of the Pauli blocking in the $q \to 0$ limit. This collective solution is the transverse spin wave of the electron gas predicted by the Landau theory [7] and experimentally observed in alkali metals [8].

In the quantum dots the electrons are confined in the (x,y) plane and we have solved the Kohn-Sham equations (7) including the terms arising from the vector potential in the so called current density functional theory (CDFT) [9, 10]. The CDFT ground state is an eigenstate of S_z and L_z, whose eigenvalues are predicted by the theory, and the external operators $F^{\pm} = \sum_i x_i \sigma_i^{\pm}$ excite states with $\Delta S_z = \pm 1$ and $\Delta L_z = \pm 1$. The result of our preliminary calculation [11] for the transverse polarizability, obtained from the solution of Eq. (10), in the case of a dot with 11 electrons in a static field of 0.7 T, is given in Fig. 3. From the figure, one sees that in dots a family of collective states emerge in the transverse response of the system. These states can be studied experimentally with inelastic scattering of polarized light in a backscattering geometry.

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