Ground state of interacting quantum magnetic dipoles: Transition from a ferromagnetic Fermi liquid to an antiferromagnetic solid

Sudhanshu S. Jha
Department of Physics, Indian Institute of Technology Bombay, Mumbai 400 076, India

S. D. Mahanti*
Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824-2330, USA

(Received 30 August 2010; published 3 November 2010)

We show that the ground state of a system of magnetic dipoles, with no electric charge, is a ferromagnetic quantum Fermi liquid at high densities, driven by the dipolar exchange energy. As in the system of classical point dipoles, the direct dipole energy is zero in this case. With decreasing density, there is a transition to an antiferromagnetic lattice state. An addition of short range hard core repulsive potential will arrest the infinite density collapse of the ferromagnetic state, and possible melting of the low density antiferromagnetic lattice state.

DOI: 10.1103/PhysRevE.82.052101 PACS number(s): 05.30.Fk, 67.90.+z, 75.10.−b

The problem of the nature of the ground state of Fermi particles of mass m, magnetic moment μ_m and density $n=N/V_0=3/(4\pi r_0^3)$, with no electric charge, is of intrinsic interest in many body physics. Sauer [1] and Luttinger and Tisza [2] were among the first who studied the nature of lowest energy configurations of classical point magnetic dipoles localized on simple three dimensional lattices, at zero temperature. Although, their work was in the context of possible dipolar magnetism in solids, without any reference to forces responsible for the localization, their numerical results are of relevance to the present problem also. They found that the classical dipolar energy E_{cl} is given by $E_{cl}/V_0=n^2\mu_m^2 D_c$, i.e., $E_{cl}/N=(3/4\pi)(\mu_m^2/r_0^3)D_c$, where the energy constant D_c depends on the configurations (directions) of magnetic dipoles and the type of the lattice. The energy is lowest for the antiparallel antiferromagnetic alignment on a simple cubic(sc) lattice with dipoles directed along the cube axis (chains of dipoles oriented parallel and antiparallel to 001), having $D_c=-2.676$. No ferromagnetic state having energy lower than this is possible for point dipoles or spherical particles [2,3]. Parallel ferromagnetic alignments are possible for face centered cubic (fcc) and body centered cubic (bcc) lattices in thin elongated granules, with the maximum possible value of D_c being −2.094.

In our earlier work [4,5], we have investigated the nature of the ground state of N charge less spin-1/2 particles of mass m and magnetic moment $\vec{\mu}_m=\mu\vec{s}$, in volume V_0. For any comparison with the classical case discussed above, note that $\mu/2=\mu_m$ in our case. Within the framework of a single determinant variational Hartree-Fock (HF) calculation, we used plane waves for the spatial part of the single particle states, labeled by the wave vector $\vec{k}$ and spin $\sigma=\uparrow$ or $\downarrow$, to show that a fully polarized ferromagnetic state with prolate spheroidal occupation function $n_\uparrow(\vec{k})=\Theta(k_{F\uparrow}^2-k^2[1-\beta_2\{3\cos\theta_k^2-1\}/2])$, $0<\beta_2<1$, gives a good upper bound for the total energy E at high densities. Here, $\Theta(x)$ is the usual step function, β_2 is a variational deformation parameter, and $k_{F\uparrow}$ is determined from the relation $\Sigma_k n_\uparrow(\vec{k})=N$. For the trial occupation functions, $n_\sigma(\vec{k})=n_\sigma(|\vec{k}|)$ or $n_\uparrow(\vec{k})=n_\downarrow(\vec{k})$, the desired negative contribution from the dipolar exchange energy vanishes identically. As in the classical case, the direct contribution from the dipole interaction always vanishes. For the above spheroidal ferromagnetic state, called the JM ferromagnetic state for the purpose of identification, we obtained exact analytical results for the variational ground state energy for the system as a function of the deformation parameter β_2, and then minimized the energy to calculate the optimum value $\beta_2^*(r_0)$ of the variational parameter as a function of the density. We found β_2^* to be close to 1 at high densities, going over smoothly toward zero at low densities. At high densities, as $r_0\to 0$, we showed that in the JM ferromagnetic state the positive kinetic energy E_{kin}/N varies as $1/(r_0)^{2+4/7}$, and the negative contribution from the exchange term diverges as $E_{exch}/N\to\mu_m^2/r_0^3$, implying an infinite density collapse of the dipolar system. At high enough densities, the negative dipolar exchange energy is the dominant term in the total energy. We, however, showed that the addition of a suitable short finite-range repulsive hard core interaction between the particles, in addition to the magnetic dipole interaction, would always lead to a stable equilibrium density curve for the total energy E/N as a function of density.

As already emphasized by us [4,5], the JM ferromagnetic state cannot be the ground state of the dipolar system at low densities, as r_0 becomes large. Even the unpolarized paramagnetic state with $n_\uparrow(\vec{k})=n_\downarrow(\vec{k})=\Theta(k_F-k)$, gives the HF energy $E_0/N=(\hbar^2/2m)(2.21/r_0^2)$ for the dipolar system, which is lower than the energy for the ferromagnetic state, at low densities. More recently, Fregoso and Fradkin [6] have used the same approach, of choosing the spheroidal form for the occupation function $n_\sigma(\vec{k})$ in the variational HF calculation, for the interesting case of the cold Fermi gas of magnetic dipolar atoms in the presence of an isotropic δ-function (contact) interaction. They allowed the possibility of occupation of both up and down spins. As expected, the result of finding the fully polarized ferromagnetic state in the high density limit, obtained earlier by us [5,4], is consistent with their

*mahanti@pa.msu.edu

phase diagram in the limit of vanishing isotropic contact interaction. In such a limit, they obtain the paramagnetic phase in the low density region. As shown by us explicitly [4,5], they would have also obtained a stable ferromagnetic phase in the high density limit, if they would have used a finite-range repulsive hard core interaction instead of the contact interaction. The question which we would like to ask here is whether the magnetic dipoles get localized at low densities to form a crystal lattice to lower the total energy because of vanishing positive kinetic energy contribution and the negative direct dipole interaction energy contribution arising, e.g., from the antiferromagnetic spin arrangement, and if so, at what density? In such a case, the total energy could be negative, much lower than the positive energy of the paramagnetic state. Fortunately, calculations of direct classical dipolar energy E_{cl}/N for various simple lattice configurations are already available [1,2]. We should, however, add the quantum correction to this classical energy, arising from spin fluctuations [7], and also possibly from lattice vibrations, before comparing the total energy E_L/N for the "localized" lattice case with the total energy E_{JM}/N in the itinerant JM ferromagnetic state, as a function of density. For this purpose, it is more convenient to define a dimensionless density parameter r_0/R_m in terms of a magnetic dipolar length $R_m \equiv 2m\mu_m^2/\hbar^2$. This parameter is independent of the spin S of quantum dipoles. It should be noted that R_m differs by a factor of 1/4 from r_m defined in our earlier work [4], with $r_0/R_m = 4r_0/r_m$. We find that at sufficiently high densities, the JM ferromagnetic state continues to have lower energy compared to the energy of the localized antiferromagnetic simple cubic lattice, even after the latter is corrected for the quantum spin fluctuations [7]. As r_0/R_m increases, there is a crossover at a certain value of r_0/R_m when the lattice state energy begins to be lower than E_{JM}. Any possible positive contribution from the zero-point lattice vibrations will only shift the transition point to higher values of the density parameter r_0/R_m.

The magnetic dipole Hamiltonian for the system of N spin $-1/2$ particles with magnetic moment $\vec{\mu}_m = 2\mu_m \vec{s}$ is given by [4,5]

$$H = \sum_i \frac{p_i^2}{2m} + \sum_{i<}^{N} \sum_{j}^{N} V(\vec{r}_i - \vec{r}_j; \vec{s}_i, \vec{s}_j), \tag{1}$$

$$V(\vec{r}_i - \vec{r}_j = \vec{R}_{ij}; \vec{s}_i, \vec{s}_j) = (4\mu_m^2/R_{ij}^3)[\vec{s}_i \cdot \vec{s}_j - 3\vec{s}_i \cdot \hat{R}_{ij} \vec{s}_j \cdot \hat{R}_{ij}]$$
$$= (1/V_0) \sum_{\vec{q}} \sum_{M=-2}^{+2} V^{(-M)}(\vec{q}) N_{i,j}^{(M)}(\vec{s}_i, \vec{s}_j) e^{i\vec{q}\cdot\vec{R}_{ij}} \{1 - \delta_{\vec{q},0}\}, \tag{2}$$

where

$$V^{(M)}(\vec{q}) = 4\mu_m^2 h_M Y_{2,M}(\hat{q}); \quad h_0 = (4\pi/3)(16\pi/5)^{1/2}, \tag{3}$$

$$N_{i,j}^{(0)} = s_i^{(0)} s_j^{(0)} - \tfrac{1}{4}(s_i^{(+1)} s_j^{(-1)} + s_i^{(-1)} s_j^{(+1)});$$

$$N_{i,j}^{(\pm 1)} = s_i^{(\pm 1)} s_j^{(0)} + s_i^{(0)} s_j^{(\pm 1)}, \tag{4}$$

$$N_{i,j}^{(\pm 2)} = s_i^{(\pm 1)} s_j^{(\pm 1)}; \quad s_i^{(0)} = s_{iz}, \; s_i^{(\pm 1)} = s_{ix} \pm i s_{iy}, \tag{5}$$

and where $R_{ij} = |\vec{r}_i - \vec{r}_j|$, $\hat{R}_{ij} = \vec{R}_{ij}/R_{ij}$.

The HF variational wave function Ψ_N for the JM ferromagnetic state is constructed by choosing N occupied single particle wave functions $\langle \vec{r}\vec{s} | \vec{k}\sigma \rangle = (1/V_0)^{1/2} \{\exp(i\vec{k}\cdot\vec{r})\} \chi_\sigma(\vec{s})$, with the occupation function

$$n_\sigma(\vec{k}) = \delta_{\sigma,\uparrow} n_\uparrow(\vec{k}); \quad n_\uparrow(\vec{k}) = \Theta(k_{F\uparrow}^2 - k^2[1 - \beta_2 P_2(\cos\, \theta_{\vec{k}})]) \tag{6}$$

and

$$k_{F\uparrow}^3 = 6\pi^2[(1 + \beta_2/2)(1 - \beta_2)^{1/2}]N/V_0; \quad 0 < \beta_2 < 1. \tag{7}$$

For any deformation parameter, $0 < \beta_2 < 1$, the total energy for the JM ferromagnetic state is

$$E_{JM} = E_{kin} + E_{exch}, \tag{8}$$

$$E_{kin} = E_0[2^{2/3} F_{kin}(\beta_2)],$$

$$F_{kin}(\beta_2) = (1 - \beta_2/2)(1 + \beta_2/2)^{-1/3}(1 - \beta_2)^{-2/3}, \tag{9}$$

$$E_{exch} = -E_0[(1/2.21)(R_m/r_0)] F_{exch}(\beta_2), \tag{10}$$

$$F_{exch}(\beta_2) = \frac{1}{\beta_2}[1 - (1 + \beta_2/2)(1 - \beta_2)^{1/2} \times [2/(3\beta_2)]^{1/2} (\sin^{-1}\{[3\beta_2/(2 + \beta_2)]^{1/2}\})], \tag{11}$$

where $E_0 = N(\hbar^2/2m)(2.21/r_0^2)$ is the HF energy for the paramagnetic state. The optimum value β_2^* of the deformation parameter at each density is obtained by minimizing the total energy E_{JM} with respect to β_2. The resulting plot for β_2^* as a function of $r_0/r_m = 4r_0/R_m$ is given in our earlier paper [4], which also gives the limiting analytical forms for β_2^* and the total energy for low as well as high densities. Essentially, at very low densities, as $r_0/R_m \to \infty$, $\beta_2^* \to 0$ and $E/E_0 \to 2^{2/3}$. At high densities, as $r_0/R_m \to 0$, $\beta_2^* \to 1 - 3.4552(r_0/4R_m)^{6/7}$, and

$$E_{kin}/E_0 \to (3)^{-1/3}(3.4552)^{-2/3}(R_m/4r_0)^{4/7},$$

$$E_{exch}/E_0 \to -(1/2.21)(R_m/r_0) \tag{12}$$

so that the high density limiting value for the total energy is

$$E_{JM}/E_0 \to -\left(\frac{1}{2.21}\right)\frac{R_m}{r_0} = -0.4525\frac{R_m}{r_0}. \tag{13}$$

A plot for the total energy in the JM ferromagnetic state as a function of r_0/R_m is shown in Fig. 1. If one rewrites the high density limiting expression for E_{JM}/V_0 in the form, $n^2\mu_m^2 D_{JM}$, i.e., $E_{JM}/N = (3/4\pi)(\mu_m^2/r_0^3) D_{JM}$, then the energy constant $D_{JM} = -(4\pi/3) = -4.1888$.

To answer the question whether at low densities the magnetic dipoles will get localized to form a lattice, let us examine the value of the possible ground state energy E_L for such a system. From the work of Luttinger and Tisza [2], it is clear that the classical dipolar energy is lowest for a

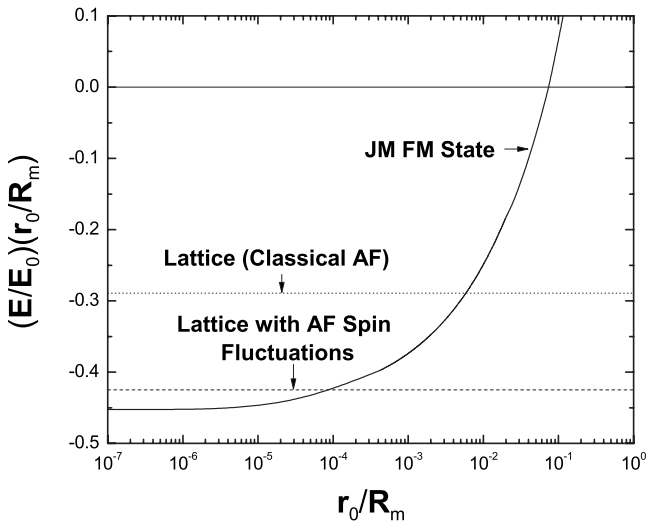

FIG. 1. Plots for the total energy E in the units of the paramagnetic free fermion energy $E_0=N(\hbar^2/2m)(2.21/r_0^2)$, as a function of the density parameter r_0/R_m, for the homogeneous JM ferromagnetic state (solid line), for the classical antiferromagnetic sc lattice (dotted line), and for the antiferromagnetic sc lattice with corrections for the zero-point energy of quantum spin fluctuations (dashed line). Note that the Y axis has been multiplied by r_0/R_m, for ease in plotting of values in the high density region, where $R_m \equiv (2m\mu_m^2/\hbar^2)$, $r_0=[3/(4\pi n)]^{1/3}$.

simple cubic lattice with antiferromagnetic configuration, with

$$E_{cl}/N = -(2.676)n\mu_m^2 = -(3/4\pi)(2.676)\mu_m^2/r_0^3. \quad (14)$$

The quantum correction arising from spin fluctuations has been calculated for this configuration by Corruccini and White [7] in the spin-wave approximation, for magnetic dipoles of spin S. They found a large correction to the classical energy, with $E_{LQS}=E_{cl}[1+(0.236/S)]$. This is almost 47% correction for $S=1/2$ system. Although, the spin-wave approximation is good only for large S, and it may overestimate the correction for a spin-1/2 system, we will use this result as an upper limit for the correction. In other words, for the lowest energy configuration, we take

$$E_{LSQ}/N = (E_{cl}/N)(1.47) = -(3/4\pi)(\mu_m^2/r_0^3)(3.9337), \quad (15)$$

which implies?

$$E_{LSQ}/E_0 = -(3/4\pi)(3.9337)\left(\frac{1}{2.21}\right)\frac{R_m}{r_0} = -0.4249\frac{R_m}{r_0}. \quad (16)$$

This is plotted in Fig. 1, where a plot of E_{cl}/E_0 is also given along with the plot of E_{JM}/E_0, for comparison. Note that at high densities, as $r_0/R_m \to 0$, the JM ferromagnetic state has lower energy [see Eq. (13)] than the case of the antiferromagnetic simple cubic lattice. However, as r_0/R_m increases, there is a transition at $r_0/R_m \sim 10^{-4}$, after which the lattice state has lower energy. Without the correction for the spin density fluctuations, the transition occurs at a higher value of $r_0/R_m \sim 10^{-2}$. For thin elongated ferromagnetic cubic lattices, the energy constant $D_{LSQ}=-3.9337$ in Eq. (16) (for the antiferromagnetic sc lattice) changes to, -3.2664 for the fcc case and to, -3.2960 for the bcc case [7], in the corresponding spin-wave calculations of Cohen and Keffer [8]. Note that these energies are higher than that of the antiferromagnetic sc case.

At this point, we may ask whether one should add a positive quantum zero-point energy arising from lattice vibrations of the lattice, as one does in the case of Wigner solid for the case of the electron gas problem? For the case of the Wigner lattice [9], the classical lattice energy varies as, $-1/r_0$, and the zero-point energy of lattice vibrations varies as, $+1/r_0^{3/2}$, which implies stability of the lattice for low densities. In the magnetic dipolar problem, the localization of the dipoles at the lattice points may arise only from additional forces, e.g., from a suitable repulsive hard core potential. Even if the lattice positions are equilibrium positions, it is not necessary that all vibrational modes arising purely from the dipolar interaction, which is not repulsive everywhere, are stable, i.e., some of the lattice modes may have imaginary frequency. In that case, one has to stabilize the lattice by using stronger repulsive forces such that all lattice vibration modes have real frequencies. To get some idea of the magnitude of zero-point energy for stable modes, one may consider the case of parallel spin-1/2 dipoles along the crystal axis (001) and long wavelength longitudinal vibrations along the (001) direction. Following the usual procedure [10], assuming that the dipoles are in equilibrium lattice positions, one can show that the long wavelength longitudinal phonon frequencies ω_{ql} are determined by

$$m\omega_{ql}^2 = (n/4)[V^{(0)}(q\hat{z})]q^2, \quad (17)$$

where the required Fourier component of the dipolar interaction can be obtained from Eq. (3). This leads to

$$\tfrac{1}{2}\hbar\omega_{ql} = \tfrac{1}{2}\hbar sq, \quad \text{with}$$

$$s = (8\pi n\mu_m^2/3m)^{1/2} = (\hbar/m)(R_m/r_0)^{1/2}(1/r_0). \quad (18)$$

A Debye-like model then gives the corresponding zero-point energy of the form,

$$E_{LV}/N \sim (R_m/r_0)^{1/2}(\hbar^2/2m)(1/r_0^2); \quad E_{LV}/E_0 \sim (R_m/r_0)^{1/2}. \quad (19)$$

This crude estimate has been made here only to show that the lattice zero-point energy will vary as, $+1/r_0^{5/2}$, whereas the static part of the lattice energy E_{LSQ} varies as, $-(1/r_0^3)$. Thus, in this case there is no binding of the system at low densities as $r_0 \to \infty$, in the absence of any other interaction. Even for the case of the homogeneous dipolar quantum Fermi liquid, there is no binding at low densities, and the ground state will then be the paramagnetic state with energy E_0.

In conclusion, we find that the ground state of the magnetic dipolar system is the fully polarized homogeneous JM ferromagnetic state at high enough densities. At low densities, the dipoles will get localized into an antiferromagnetic lattice state, with the transition at $(r_0)_{tr}$ somewhere between $10^{-4}R_m$ and $10^{-2}R_m$, where $R_m \equiv (2m/\hbar^2)\mu_m^2$, depending on

the actual quantum correction due to quantum spin fluctuations to the classical dipolar energy constant D_c (see Fig. 1). To get an idea about the magnitude of R_m, it is to be noted that for heavy magnetic atoms, it is about 10^{-8} cm. In the presence of a suitable short finite-range repulsive hard core potential [4], the high density JM ferromagnetic phase is stable against any possible high density collapse, and the low density magnetic solid phase will be stable against vibrational melting of the lattice. In the absence of such a hard core interaction, which leads to stable vibrational modes, the system will, of course, go over to the paramagnetic gas phase at low densities. If the range of the suitable hard core repulsive potential is less than $(r_0)_{tr}$, the ground state of the system is the JM ferromagnetic state, whereas if the range of the repulsive hard core potential is greater than $(r_0)_{tr}$, the ground state will be the low density antiferromagnetic sc lattice state. To obtain a more accurate value of the crossover point, we will need more precise calculations of the spin density fluctuation contribution and the vibrational zero-point energy. We hope to address this problem in the future, in which we have to add a suitable repulsive hard core potential from the very beginning. At very high densities the system will always be in the homogeneous fully polarized ferromagnetic JM state with elongated needle like Fermi occupation function in the $\vec{k}$-space.

One of us (S.S.J.) would like to acknowledge support from the condensed matter theory group of the Department of Physics and Astronomy, Michigan State University. We thank Khang Hoang and River Huang for assistance.

[1] J. A. Sauer, Phys. Rev. **57**, 142 (1940).
[2] J. M. Luttinger and T. Tisza, Phys. Rev. **70**, 954 (1946).
[3] E. Z. Meilikhov and R. M. Farzetdinova, J. Exp. Theor. Phys. **94**, 751 (2002).
[4] S. D. Mahanti and S. S. Jha, Phys. Rev. E **76**, 062101 (2007). Note that the values of C_0 and h_0 given after Eq. (3) in that paper are misprinted. Correct values [5] are: $C_0=-(16\pi/5)^{1/2}$, $h_0=(4\pi/3)(16\pi/5)^{1/2}$.
[5] S. D. Mahanti and S. S. Jha, J. Phys. A **39**, 1239 (2006); S. S. Jha and S. D. Mahanti, Pramana, J. Phys. **68**, 819 (2007).
[6] B. M. Fregoso and E. Fradkin, Phys. Rev. Lett. **103**, 205301 (2009).
[7] L. R. Corruccini and S. J. White, Phys. Rev. B **47**, 773 (1993).
[8] M. H. Cohen and F. Keffer, Phys. Rev. **99**, 1135 (1955).
[9] E. P. Wigner, Trans. Faraday Soc. **34**, 678 (1938); W. J. Carr, Jr., Phys. Rev. **122**, 1437 (1961).
[10] D. Pines, *Elementary Excitations in Solids* (W. A. Benjamin, New York, 1964), Chap. 2.