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Design of a Nanomagnet

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We design a two-dimensional ferromagnet of quantum spheres of radius $O(10\ \mathrm{nm}^3)$ made of *nonmagnetic* intrinsic semiconducting material, each connected to its neighbors by nano-cylinders made of identical material. Ferromagnetism ensues if there is precisely one electron trapped in each sphere; this is a direct consequence of a theorem by Elliott Lieb. We analyze and estimate the important materials parameters. At concentrations of less than 1 electron per sphere, ultimate collapse of the ferromagnetic state should yield a paramagnetic Fermi liquid. We point out the need for additional numerical and physical experimentation.

KEY WORDS: Nanotechnology; ferromagnetism; Hubbard model; Lieb theorem; nanocircuits; silicon arrays.

INTRODUCTION

One of the goals of "spintronics" has been the production and manipulation of spin-polarized electrons in microelectronic circuitry. Here I show how magnetically polarized arrays of arbitrary size can be constructed out of *non*magnetic semiconducting material such as *intrinsic silicon* into which electrons have been injected. This design implements in two dimensions a beautiful theorem by Lieb[2] relating the spontaneous spin-magnetism of a network to its connectivity and geometry. It also relies on a phenomenon of wave mechanics that has only been understood in recent years: the formation of localized states at singularities in *electron waveguides*.

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[2] The theorem proved by Elliott Lieb, *Phys. Rev. Lett.* **62**, 1201 (1989), on which our model of bubbles and channels is based, applies—strictly speaking—only to the Hubbard model. It generalizes earlier work by E. Lieb and D. Mattis, *J. Math. Phys.* **3**, 749 (1961), concerning the Heisenberg model (spins) in bipartite lattices with antiferromagnetic exchange interactions.

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In a separate publication I show how some of these results can be duplicated—or extended—even in *one*-dimensional arrays.

Lieb's theorem applies to interacting electrons on half-filled bipartite networks, regardless of their short- or long-range-order. For simplicity the network we shall analyze in this paper will be ideally ordered. "Bipartite" implies that an electron on an A site can only "hop" (i.e., be transferred,) to a nearby B site, and vice-versa. A to A or B to B hops are prohibited. Lieb proved that the ground-states of any such system include a state of total spin moment $S_{\text{tot}} = \frac{1}{2}|N_A - N_B|$ (in units of $\hbar$) N_A and N_B are respectively the number of A and B sites, not necessarily equal and not necessarily ordered.

I shall specify a nano-array that is conjectured, by virtue of this theorem, to exhibit spontanous ferromagnetism.[3] Physically it consist of identical, interconnected, *non*magnetic quantum dots organized into N cells in a two-dimensional network where $N_A = 2N_B = 2N$. For historical and other reasons I choose the architecture to be that of the "decorated" *sq* lattice of planar CuO_2. Indeed, this example of a bipartite lattice, specifically cited in footnote 2, is commonly associated with high-T_c superconductivity. In our variant, illustrated in Fig. 1, the Cu and O ions are replaced by a set of identical spherical objects that we name "bubbles" to distinguish them from the usual quantum dots. They are connected by solid cylinders or nanotubes *made of the same material as the bubbles*. We shall use the nomenclature "channels" for these solid cylinders, to distinguish them from the hollow *carbon nanotubes* which serve entirely different functions. Because spheres and cylinders are all constituted out of the same intrinsic, nonmagnetic, semiconductor material the work function is identical throughout. The radii, b, of the bubbles and the radii, a, and lengths l of the channels are picked to favor electron occupancy of bubbles over channels.

The entire array could be formed by *uv* nanolithography followed by selective etching. Instead of, or in addition to, using ion implantation or chemical doping to introduce electrons into the neutral semiconductor material, we propose their injection by tunneling, using a standard capacitative method such as that which informs the usual MOSFET devices.[4]

[3] Some may call it *ferri*magnetism because of the analogy to the Fe^{2+}/Fe^{3+} alternation in ferrites that results in a net magnetic moment.

[4] Without discussing this technology any further than to point out that the reqired density of electrons in our model is quite low and therefore achievable, of the order of or less than $10^{12}/cm^2$, we point out that an alternative way and perhaps simpler of reaching the same goal uses the techniques of Pepper and collaborators, as we shall elaborate elsewhere.

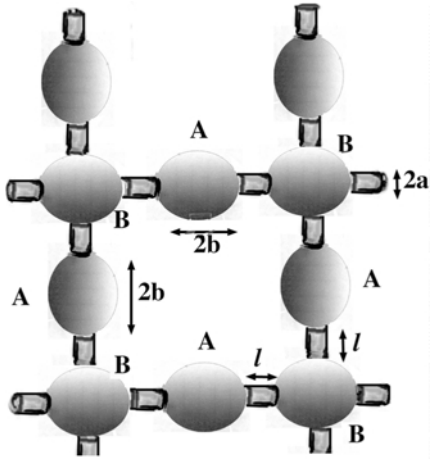

Fig. 1. Indicates the layout of spheres and the cyclinders that connect them ("bubbles" and "channels") in the model array discussed in the text. For our example we have chosen spherical radius $b = 1.5a$, (a = cylinder radius) and an optimal cylinder length $l = 1.683a$. The magnitude of a is arbitrary—until Coulomb forces and effective exchange interactions are taken into account and optimized. This reveals that a must be in the 10 nm range.

As shown in Fig. 1, each bubble is connected by channels to either 2 or 4 nearest-neighbor spheres. Spheres that "decorate" the square lattice are assigned to the A sublattice while the corner spheres constitute the B sublattice, an ordinary *sq* lattice. In an infinite array the unit cell can be chosen to include one B and its two A nearest-neighbors (one immediately above it and the other immediately to its right.) Lieb's theorem tells us that for $3N$ electrons (one per bubble and none in the channels,) the ground state spin *per* unit cell can be as large as $\sigma = S_{\text{tot}}/N = \frac{1}{2}$ for N in the thermodynamic limit $N \to \infty$.[5]

In what follows we optimize values for the parameters a, l, and b so that: (1) electrons reside principally in the bubbles and *not* in the channels that connect them and such that, (2) on average only a single electron occupies each bubble. For the chosen geometry, this amounts to 3 electrons per cell. We have calculated the various dimensions to yield the maximum possible correlations among electrons. The idealized ferromagnet that results in this design is an electrical insulator, because the on-site Coulomb

[5] For a sample with finite N and a rectangular perimeter the average magnetization *per* cell σ is somewhat less (because, on the perimeter, the number of A's *equals* the number of B's hence overall $N_A < 2N_B$). For a single decorated square $\sigma = 0$, for two contiguous squares it is $\sigma = 1/4$ in each, etc., rapidly approaching the saturation value $\sigma = 1/2$.

interaction is sufficiently strong to discourage charge fluctuations. In other words, we rely on a mechanism called the "Coulomb blockade" in the example of a single microdot and "Mott insulator" for an array.

Small changes in electron density or Fermi level can transform our array into a nonmagnetic conductor, while changes in the architectural design can produce an ordered *anti*ferromagnet instead. The latter, again depending on position of the Fermi level, could also be made into a metal and possibly into a superconductor.[6]

Over-all contractions or expansions of the length scales affect various important parameters differently, therefore there is an optimum length scale to be calculated for each desirable physical property, given a geometry and a particular many-body system. Here we concentrate on the Hubbard–Lieb ferromagnet.

SPHERES AND CYLINDERS

Let all the channels have the same radius a (cross-sectional area πa^2,) length $l > a$ and all spheres radius b, with $b > a$. A variational upper bound on the internal one-electron energy levels E and a first approximation to the corresponding stationary states Ψ of each *physical* (i.e., connected) bubble may be obtained by the following procedure: insert 1 electron into an idealized sphere of radius b and set its wavefunction $\Psi = 0$ on the surface, ignoring the embedded channels. We shall assume that the material properties (mobility, dielectric constant κ, carrier mass m^*) are those of the bulk semiconductor.

The zeros of the relevant Bessel functions then yield the energies for the spheres and also for the transverse energy in the channels (the bottom of a continuum once the longitudinal energies are added in). It is convenient to express all energies in terms of the *threshold channel energy* $E_a(0, 0)$.[7] In these units, states of energy $\varepsilon < 1$ live in the spheres while states of energy $\varepsilon > 1$ (*scattering states* of the spheres) are localized mainly in the channels.

One extracts the ideal range for the ratio $(b/a)^2$ using the following criteria. Requiring $(b/a)^2 > 1.7021$ ensures there is at least 1 state of the sphere with $\varepsilon < 1$. If the ratio also satisfies $(b/a)^2 < 3.4923$ then there is *at most* 1 such state.[8] The ground state wave function is of course nodeless,

[6] Details will be given in separate publications: D. Mattis, to be submitted.

[7] This unit compares to the Rydberg (binding energy of an Hydrogen atom,) as follows: $E_a(0, 0) = 2.9(m/m^*)(a_o/a)^2 \times 13.6$ eV, where $a_o =$ Bohr radius $\approx 1/20$ nm. After determining the optimal cylinder radius a (in nm) we obtain $E_a(0, 0)$ in eV from this formula.

[8] This last is not as important a requirement as the first.

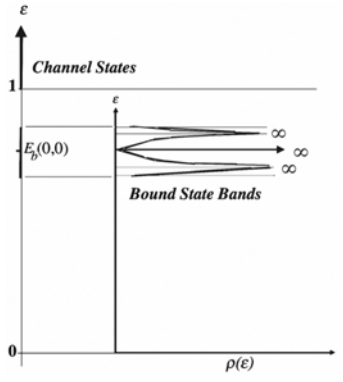

Fig. 2. Sketch of the energies and energy bands in the free-electron approximation for the given layout and parameters. In plotting the densities of state $\rho(\varepsilon)$ we show 3 non-overlapping bands. The central one is sharp (all its states are localized) and the two that straddle it exhibit van Hove singularities, as indicated. But note that this band structure does not survive in strong coupling.

with angular and radial quantum numbers $l = 0$, $n = 0$. A mid-range compromise adopted here and in the rest of this paper has $a \approx 2b/3$; it corresponds to $\varepsilon_b(0, 0) = 0.7564$ ($\approx 3/4$ to graphical accuracy.) We shall examine other possibilities in future studies.

SPIN PARAMAGNETISM OF (HYPOTHETICALLY) NONINTERACTING ELECTRONS

Next we examine the connections that allow an electron to tunnel from one sphere to the next. In any extensive periodic array ($N \gg 1$,) Bloch's theorem applies and the individual levels broaden into the energy bands sketched in Fig. 2 and detailed below. The *bandwidths* are multiples of an "effective" hopping parameter t, itself a function of a, b, and l and closely related to the decay length λ (of the bound state into a channel.) Let us pursue this in the 1-electron picture, temporarily ignoring the Coulomb interactions of 2 or more electrons. We examine the 2-body Coulomb forces in the next section.

The exponential decay of the sphere's lowest bound state into a connecting channel is characterized by a length λ,

$$\lambda \approx a \sqrt{\frac{1/5.7985}{1 - 1.7021(a/b)^2}} = \frac{0.4153a}{\sqrt{1 - 1.7021(a/b)^2}} \tag{1}$$

from which we deduce[9] the value of t for two bubbles separated by a channel of length l:

$$t = \frac{\hbar^2}{4m^*\lambda^2} \exp -(l/\lambda). \tag{2}$$

At fixed a we alter λ in Eq. (1) by varying the ratio a/b. For fixed l the hopping parameter t in Eq. (2) vanishes at both small and large values of λ and is maximal at $\lambda = l/2$. Evaluating Eq. (1) using $a/b \approx 2/3$ and replacing λ by $l/2$ in that formula, we find the optimal cylinder length to be $l = 1.683a$. Insertion into Eq. (2) show that the latter scales with $E_a(0, 0)$, thus yielding an optimal dimensionless hopping parameter $\tau \equiv t/E_a(0, 0) = 0.0165$, independent of a.

In tight-binding three energy bands emerge for this particular geometry: one of zero width at precisely $E_b(0, 0)$ and two that straddle it, $E_{(\pm)}(q_x, q_y) = E_b(0, 0) \pm 2t\gamma(q_x, q_y)$, where $\gamma = (\cos^2 q_x d/2 + \cos^2 q_y d/2)^{1/2}$. (Here $d/2 = l + 2b$ is the distance separating the centers of two neighboring spheres.) As shown in Fig. 2, for our choice of $b/a = 1.5$ the dimensionless energies of *all* the Bloch states in the three bands shown in Fig. 2 lie below the threshhold, $\varepsilon(q_x, q_y) < 1$, and therefore they all live on the bubbles. The density of states ρ in the central band of zero width is a Dirac delta function. The finite-width energy bands have bandwidths $\Delta\varepsilon = 2\sqrt{2}\,\tau = 0.0466 \approx 0.05$ each. Their densities of state $\rho(\varepsilon)$ exhibit logarithmic van Hove singularities, as sketched.

A word on the unit of energy[7] is in order at this point. Express $E_a(0, 0)$ in eV and assume a fixed $m^*/m = 0.01$ for the sake of definiteness. Then for $E_a(0, 0)$ to exceed a given value in eV—say x eV—necessitates $a < \frac{10}{\sqrt{x}}$ nm.

In the (purely hypothetical!) case that there are no Coulomb repulsions, the multiple many-body ground states of $3N$ electrons are constructed as follows: $2N$ occupy the lowest band and disappear from consideration. A number $N_\downarrow$ ranging from $N/2$ to N, have spin "down" in the degenerate band, with the remaining $N_\uparrow = N - N_\downarrow$ having spin "up." This yields a ground state degeneracy $W_o = 2^N$. Total spin S_{total} ranges from 0 to a maximum value $S_{\max} = \frac{N}{2}$ while, for each S_{total}, S_z ranges from $-S_{\text{total}}$ to $+S_{\text{total}}$.

Thus, even though the total number of electrons is fixed, σ, the magnetization *per* cell $= S_{\text{total}}$, can vary from 0 to a maximum value $1/2$ in the

[9] This formula is obtained by calculating the splitting between bonding and antibonding "molecular" orbitals and setting it equal to $2t$ in an equivalent tight-binding model.

ground state(s). The N fermions in the dispersionless band are in effect *paramagnetic*, just as a collection of N noninteracting spins $1/2$ would be.[10]

FERROMAGNETISM AS A RESULT OF COULOMB REPULSIONS

However, once 2-body repulsive forces are introduced, Lieb's theorem ensures that only the *most* magnetic configurations survive in the ground state. The system acquires a sturdy, macroscopic, moment. The most interesting questions concern repositioning of the many-body states in the presence of Coulomb interactions. Physically, the physics of the bubbles and channels maps onto a sort of tight-binding model,

$$H = -t \sum_{\vec{j},\sigma} (b^{+}_{\vec{j},\sigma}(a_{\vec{j}+(0,\frac{1}{2}),\sigma} + a_{\vec{j}+(\frac{1}{2},0),\sigma}) + H.C.)$$

$$+U \sum_{\vec{j}} (n_{\vec{j},\uparrow} n_{\vec{j},\downarrow} + n_{\vec{j}+(0,\frac{1}{2}),\uparrow} n_{\vec{j}+(0,\frac{1}{2}),\downarrow} + n_{\vec{j}+(\frac{1}{2},0),\uparrow} n_{\vec{j}+(\frac{1}{2},0),\downarrow}), \qquad (3)$$

a species of Hubbard model on a decorated lattice in which only the *intra*-bubble electrostatic interaction terms $\propto U$ are retained. The truncation of the Coulomb interactions presupposes that the most important correlations are local, i.e., that an electron repels a second electron strongly if within the same sphere but in a negligible way when it is on neighboring spheres. The formulation in Eq. (3) takes the intra-atomic exchange interaction among bound states in bubbles into account *exactly*, provided there is only 1 bound state per bubble (just as in the familiar Hubbard model.) Neglect of the Coulomb force between 2 electrons on neighboring bubbles requires us to neglect nearest-neighbor direct exchange correction, which is ferromagnetic but too weak to interefere with antiferromagnetic correlations. Just as in the Hydrogen molecule, nearest-neighbor electrons in this construction are always correlated *anti*ferromagnetically, never ferromagnetically.

In fact, the main effect of the Coulomb interaction $U = (e^2/\kappa b)\, f(b)$ is to inhibit double occupancy of a given sphere. (Here e is the charge on an electron, κ is the dielectric constant of the semiconductor and f is a slowly

[10] A similar conclusion applies to varying numbers $\mathbb{N}$ of electrons in the range $2N \leqslant \mathbb{N} \leqslant 4N$. The Fermi level remains pinned to the level $E_b(0, 0)$ while σ ranges between zero and a maximum $\sigma_{\max}(\mathbb{N}) = 1/2(1 - |\mathbb{N} - 3N|/N)$. Because the Bloch energies exhibit no dispersion, there is neither electrical conductivity nor orbital diamagnetism in this range of occupation numbers within the independent electron model.

varying function $O(1)$ related to the solution of the 2-body problem within a sphere.) This same inhibition causes the Coulomb blockade in ordinary microdots.

In units of $E_a(0, 0)$ the dimensionless Coulomb repulsion is,

$$u \equiv \frac{e^2 f/\kappa b}{\hbar^2 5.7985/2m^* a^2} = \left(\frac{a}{a_o}\right)\left(\frac{a}{b}\right)\left(\frac{m^*}{m}\right)\left(\frac{f}{2.9\kappa}\right), \tag{4}$$

where a_o is the ordinary Bohr radius ($\approx 1/20$ nm). The second of 4 factors can be replaced by the chosen value $2/3$.

If for the factors m^*/m and $f/2.9\kappa$ we arbitrarily assume 0.01 and 0.1 respectively, we obtain $u \approx (4/300) \times a$ where a is expressed in nm.[11] With a channel radius as small as $a = 7.5$ nm this expression yields $u = 0.1$, an amount that is quite significant by comparison with the free-electron bandwidths $\Delta\varepsilon = 0.05$. While larger a leads to an *even greater* value of u, both τ and $\Delta\varepsilon$ are rigorously constant w.r. to a. Thus the *relative* importance of the Coulomb force relative to the hopping increases with increasing a up to some maximum value.[11] (Note that the physical t decreases with increasing a, proportionally to a^{-2}, while the physical U also decreases, but only proportionally to a^{-1} so that *both* tend to zero as the length scale increases beyond the desirable 10 nm range.)

In analyzing $3N$ electrons with $u \gg \tau$ it might be tempting to choose a product state of *exactly* 1 electron *per* sphere, avoiding double-occupancy. But the energy of this state is 0, which is not optimal. Moreover it leaves the total spin indeterminate, even more so than in the previously examined case of noninteracting fermions. Perturbation theory suggests that one should allow the electrons on the A and B sites to carry *opposite* spins with probability amplitudes $O(\tau/u)$ of hopping onto an already occupied nearby site. Then perturbation theory allows us to estimate the ground state energy as $-4Nt^2/U = -4NE_a(0, 0)\,\tau^2/u$, a substantial improvement.[12] As shown in footnote 2, this system of fermions maps onto a *ferrimagnet* of spins $1/2$ on the very same bipartite lattice, having a ground state belonging to total spin $N/2$. This is apparent from Fig. 3 indicating $S_z = N/2$.

[11] Strictly speaking, once U given by this formula exceeds the ionization potential of the sphere $\Delta W \equiv E_a(0, 0) - E_b(0, 0)$ it must be replaced by a fixed ΔW and u ceases to increase with a. In our example this occurs once a has reached or exceeded 19 nm, so we shall restrict the analysis in this paper to $a < 19$ nm.

[12] Actually, the ground state energy *per* unit cell is found to be approximately $E_0/N \approx -2J$, where $J = 2t^2/U$.

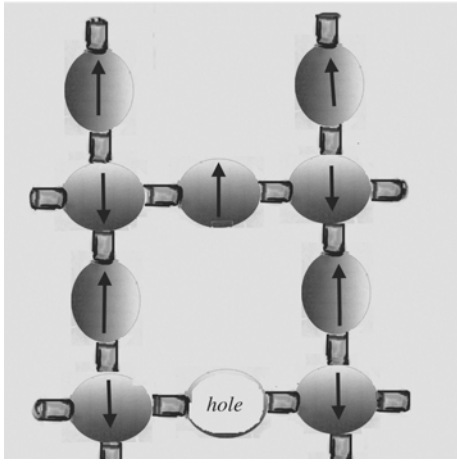

Fig. 3. Spin correlations on the model Hubbard–Lieb ferrimagnet, when the occupation density v is slightly less than 3. A "hole" (missing electron of spin $\uparrow$), shown on an A site, cannot survive on a B site but tunnels to a neighboring A site with a greatly *reduced* matrix element $t_{\text{eff}} \approx \mathrm{O}(t^2/U)$ (see text). The exchange parameter J that stabilizes the antiparallelism becomes negligible once the length scales become too great, e.g., for $a \gg 10$ nm, therefore some sort of electronic conductivity or diffusivity should occur at finite temperature for any occupancy $v < 3$.

EFFECTIVE EXCHANGE PARAMETER

An effective "exchange parameter" $J \approx 2t^2/U = (2\tau^2/u)\, E_a(0, 0)$ governs the effective spin-spin correlations in the ferrimagnet, important if we wish to map (3) onto some sort of $t-J$ model. It measures the maximum thermal fluctuation kT that the magnetic order can withstand. In our units, the dimensionless exchange parameter is $j = J/E_a(0, 0)$,

$$j = \frac{0.041}{a} \qquad (\text{with } a \text{ in nm}). \tag{5}$$

To stabilize the ferromagnetism it is desirable for j to be as large as possible. Assuming $E_a(0, 0) = 1$ eV and $a = 10$ nm, we have: $j \approx \tau/4$, $J/k_B = 44\ K$ and $\Delta\varepsilon = j > 540$ K. Although this suffices for a reasonable prototype, the design parameters will have to be chosen more precisely if one is to stabilize a maximum magnetic moment at room temperature.

DISCUSSION AND SUMMARY

Several features distinguish physical, interacting electrons from their simpler (but unrealistic) noninteracting counterparts:

(a) For $3N$ *noninteracting* electrons the ground state is exponentially degenerate whereas the ground state of $3N$ physical electrons belongs only to maximum spin, i.e., exhibits only the trivial inherent degeneracy $2S_{max}+1$. *Both* interacting *and* noninteracting systems attain their maximum spin density at $3N$ electrons and are *non*magnetic (i.e., have a ground state with $S=0$) at $2N$ or fewer electrons, or at $4N$ or greater number of electrons.

(b) The ground state energy of $3N$ noninteracting electrons is relatively large, $-O(N|\tau|)$, (in units of $E_a(0,0)$,) while that of the strongly interacting electrons is *very much reduced* by an extra factor τ/u, to $-O(N\tau^2/u)$.

(c) The occupancy factor v (number of electrons *per* cell) of *noninteracting electrons* can range from 2 to 4 all the while the Fermi level remains pinned to $E_b(0,0)$. Hence there is no transport in this range of occupancy. In contrast, the more physical system of *interacting* electrons *can* exhibit some "hole" conductivity or diffusion over a portion of the range[13] $v \leqslant 3$. The carriers have positive charge relative to the overall original negative charge $-3Ne$. But because the motion of 1 hole (*vide* Fig. 3) leaves a wake of re-arranged spins, or must tunnel through a barrier of height u such hopping is inhibited by a factor of $O(\tau/u)$.

In forthcoming works, we shall discuss various related topics such as (1) the photoconductivity in this model, (2) the construction of a formal bubble-and-channel equivalent of CuO_2 (it requires the A bubbles to be substantially larger than the B's, or to be replaced by dimers), (3) and various small-cluster calculations on which to base the dimensional estimates on a firmer footing. In this paper we have established that a purely theoretical construct by Elliott H. Lieb might some day change our notions of physical reality.

[13] For strongly interacting electrons only the range $v \leqslant 3$ is easily accessible, as *each* extra particle over $3N$ "costs" an extra energy u. See also footnote 11.