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# Landau quantization of electrons on a sphere

H. Aoki and H. Suezawa
*Department of Physics, University of Tokyo, Hongo, Tokyo 113, Japan*
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The energy levels and wave functions of the electrons confined on the surface of a sphere in uniform magnetic fields $H$ are obtained from both the exact solution in terms of the spheroidal function and numerical study. The crossover points between the weak-$H$ (orbital Zeeman splitting) and strong-$H$ (Landau quantization) regimes are shown to delineate a series of envelopes, which, together with peculiar bunch of Landau levels, form a series of overlapping bands. The result is compared with the Landau quantization of tight-binding electrons on $C_{60}$ and $C_{240}$ fullerene structures. These spectra are shown to imply an interesting orbital magnetism.

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Landau quantization in two dimensions (2D) makes the energy levels coalesce into a discrete spectrum, which is the background to the quantum Hall effect [1,2]. An intriguing question would be whether yet another fascinating quantization takes place for the electrons on *curved surfaces* in magnetic fields. The simplest example would be an electron system on a sphere. Here we have solved the electronic structure for electrons on the surface of a sphere in uniform magnetic fields from both the exact solutions in terms of the spheroidal wave function and numerical results. Recent dramatic advances in fabricating microclusters such as fullerene families [3,4] are enabling us to realize spheres of microscopic dimensions in addition to mesoscopic fine particles. We have also obtained the electronic structure of tight-binding electrons on fullerenes to compare the results with those for the continuous surface of a sphere. These results are shown to have implications on the orbital magnetic moment and optical absorption of electrons on closed surfaces of mesoscopic dimension.

The Hamiltonian for a free electron on the surface of a sphere of radius $R$ in a uniform magnetic field $H$ is given by

$$\mathcal{H}=\frac{1}{2m^*}\left(\mathbf{p}+\frac{e}{c}\mathbf{A}\right)^2 =\frac{1}{2m^*}\left[\frac{\hat{\mathbf{L}}^2}{R^2}+\frac{eH}{c}\hat{\mathbf{L}}_z+\left(\frac{eH}{2c}\right)^2 R^2\sin^2\theta\right]. \quad (1)$$

Here $e$ ($m^*$) is the charge (effective mass) of an electron, $\hat{\mathbf{L}}$ is the angular momentum, $\mathbf{A}$ is the vector potential in the symmetric gauge, and $H$ is assumed to be in the $z$ direction. Since the system has a rotational symmetry, an eigenfunction can be written as $\psi(\theta,\phi)=\Theta(\theta)\exp(im\phi)$ in the spherical coordinates, where $m\hbar$ is the eigenvalue of $\hat{\mathbf{L}}_z$. The Schrödinger equation, $\mathcal{H}\psi=E\psi$, becomes

$$\frac{d}{dz}\left[(1-z^2)\frac{df}{dz}\right]+\left[\lambda+\Phi^2 z^2-\frac{m^2}{1-z^2}\right]f=0 \quad (2)$$

upon introducing $\Theta(\theta)\equiv f(z)$ with $z\equiv\cos\theta$, $\Phi\equiv\pi R^2 H/\phi_0$, $\lambda\equiv-\Phi^2-2m\Phi+E/\kappa$, $\kappa\equiv\hbar^2/2m^*R^2$. Here $\Phi\propto H$ denotes the magnetic flux encircled by the sphere in units of the flux quantum, $\phi_0=hc/e$, or, alternatively, $(2\Phi)^{1/2}$ corresponds to $R$ measured in units of the magnetic length, $L_H\equiv(c\hbar/eH)^{1/2}$, for a flat system. The equation is just the (oblate) spheroidal differential equation, so that the regular solution with $f(z)$ finite for $-1\le z\le 1$ is given by the (oblate) spheroidal wave function with discrete eigenvalue for $\lambda$ [5]. The spheroidal function, $f_{lm}$, of order $m$ and degree $l$ can be expressed as an expansion in terms of Legendre's bipolynomials, $P_l^{|m|}(z)$. In the Hamiltonian matrix, $\langle P_l^m|\mathcal{H}|P_{l'}^{m'}\rangle$, the elements are nonzero only for $m=m'$ and $l'=l,l\pm 2$.

Before we turn to the exact solution, we first discuss the weak $H$ limit. When the $H^2$ term in the Hamiltonian is dropped to have $\mathcal{H}=(\kappa/\hbar^2)\hat{\mathbf{L}}^2+\mu_B H\hat{\mathbf{L}}_z/\hbar$ (with $\mu_B=e\hbar/2m^*c$ being the Bohr magneton), $P_l^{|m|}(z)$ becomes the solution, and we have a series of Zeeman-split $(2l+1)$ states with

$$\frac{E}{\kappa}=l(l+1)+2m\Phi+\frac{1}{2}\left[1+\frac{(2m-1)(2m+1)}{(2l-1)(2l+3)}\right]\Phi^2 \quad (3)$$

for $l=0,1,\ldots,$ where we have also indicated the perturbation term proportional to $\Phi^2$ which contains the term $H^2R^2\sin^2\theta$ in the Hamiltonian.

To look into the opposite limit of strong $H$, we can rewrite the spheroidal equation as

$$4\Phi\left\{x\frac{d^2}{dx^2}+(m+1-x)\frac{d}{dx}+\left[\frac{1}{4}\left(\frac{\lambda+\Phi^2}{\Phi}\right)-\frac{1}{2}(m+1)\right]\right\}u +\left[-x^2\frac{d^2}{dx^2}+[x^2-2(m+1)x]\frac{d}{dx}+(m+1)(x-m)\right]u=0 \quad (4)$$

 

by substituting $u_{lm}=(1-z^2)^{-m/2}e^{(1\pm z)\Phi}f_{lm}$ with $x\equiv 2(1\pm z)\Phi$. For large $\Phi$ we can only retain the first line proportional to $\Phi$. This just yields the Laguerre equation, and the solution is given by Laguerre's polynomial, $L_\nu^{(m)}$, with $\nu$ equated to the constant term in that line. We can follow the analysis of the spheroidal function by Flammer [5] to establish, by counting zeros, that $u_{lm}\rightarrow f_N\propto e^{-(1-z)\Phi}L_\nu^{(m)}(2(1-z)\Phi)$ for the northern hemisphere ($z>0$) or $f_S\propto e^{-(1+z)\Phi}L_\nu^{(m)}(2(1+z)\Phi)$ for the southern hemisphere ($z<0$), where $\nu=(l-m)/2$ [for $(l-m)$ even] or $\nu=(l-m-1)/2$ [for $(l-m)$ odd]. Thus the eigenenergy becomes

$$E\rightarrow[(\nu+m)+\tfrac{1}{2}]\hbar\omega_c - [2\nu(\nu+m+1)+m+1]\kappa+O(\Phi^{-1}). \quad (5)$$

Here $\hbar\omega_c\equiv\hbar eH/m^*c=4\kappa\Phi$ is the cyclotron frequency for the flat system, so that the levels are shuffled into families of peculiar "Landau levels," $(N+\frac{1}{2})\hbar\omega_c+\text{const}$ ($N=0, 1, \ldots$), each of which comprises all the combinations of $(l,m)$ satisfying $l+m=2N$ or $l+m=2N+1$. In Eq. (5) we have indicated the additional constant shift $\sim O((1/\Phi)^0)$, which comes from the "perturbation" energy, $\langle L_\nu^{(m)}|\mathcal{H}'|L_\nu^{(m)}\rangle$, with $\mathcal{H}'$ being the deviation [second line of Eq. (4)] from the Laguerre equation. Thus, even in the $H\rightarrow\infty$ limit (with $L_H\rightarrow 0$), there is no degeneracy, and we have instead a *parallel* bunch of lines for each value of $N$.

Physically, we have a unique realization of coupled oscillators here in the following sense. The effect of $H$ of general magnitude in Eq. (1) is to add a confining potential proportional to $H^2R^2\sin^2\theta$ parabolic in the distance from the polar axis. When $H$ is large, an electron is trapped to either of the north pole or south pole, and the state around each pole may be thought of as a two-dimensional harmonic oscillator (on the curved surface). The perturbation, $\mathcal{H}'$, just represents the effect that the oscillators are not isolated but connected along the equator.

The only degeneracy in the large-$H$ limit comes from the pair, $f_N$ with $l=-m+2N$ and $f_S$ with $l=-m+2N+1$, which have the same $\langle\mathcal{H}'\rangle$. From the degenerate perturbation an eigenfunction becomes either the "bonding" state, $f_B\propto f_N+f_S$, or "antibonding" state, $f_{AB}\propto f_N-f_S$, which conform to the constraint that each wave function has to be an eigenstate of the parity against the north-south exchange. Thus the levels corresponding to $f_B$ and $f_{AB}$ coalesce into a single line as $\Phi$ is increased, where the coalescence is shown to take place at the value of $\Phi$ that becomes smaller the smaller $(l,m)$ is.

The real problem is the crossover between these weak-$H$ and strong-$H$ limits. For the general value of $\Phi$ we have obtained the wave function by diagonalizing the Hamiltonian matrix, which is block diagonal in $m$. We have approximated each of the infinite-dimensional block matrices by a finite one, in which we have checked that the cutoff for $l$ ($\leq 540$) is large enough to give accurate eigenvectors.

The result for all the energy levels against $\Phi$ (Fig. 1) has a striking structure: The crossover points between the weak-$H$ regime (orbital Zeeman splitting) and the strong-$H$ regime (Landau levels) form a series of *envelopes*. For the characterization of the crossover point, we can look at the wave function in Fig. 2. The lowest envelope (locus of the ground-state energy) comes from the set of states with $m=-l(N=0)$. For these states the amplitude is peaked at the equator for small $H$. As $H$ is increased, the potential proportional to $H^2R^2\sin^2\theta$ pushes the amplitude towards the poles, and a competition between the kinetic and potential energies sets in. The wave function is seen to be significantly deformed from $P_l^{|m|}$ around the critical value, $\Phi_c$, at which $E(\Phi)$ curve for a given $l$ attains a minimum.

From Eqs. (3) and (5) we can roughly estimate $\Phi_c$ to be $\Phi_c\sim l$ (at which $E\sim\kappa l$) for large $l$, which may be thought of as a geometrical resonance at $\Phi\propto R^2/L_H^2\sim$ integer. Semiclassically, formation of the envelope is considered to originate from the following argument. For the electrons on the surface of a sphere, the component of $H$ perpendicular to the surface varies from place to place (depending on the latitude), so that there is no single

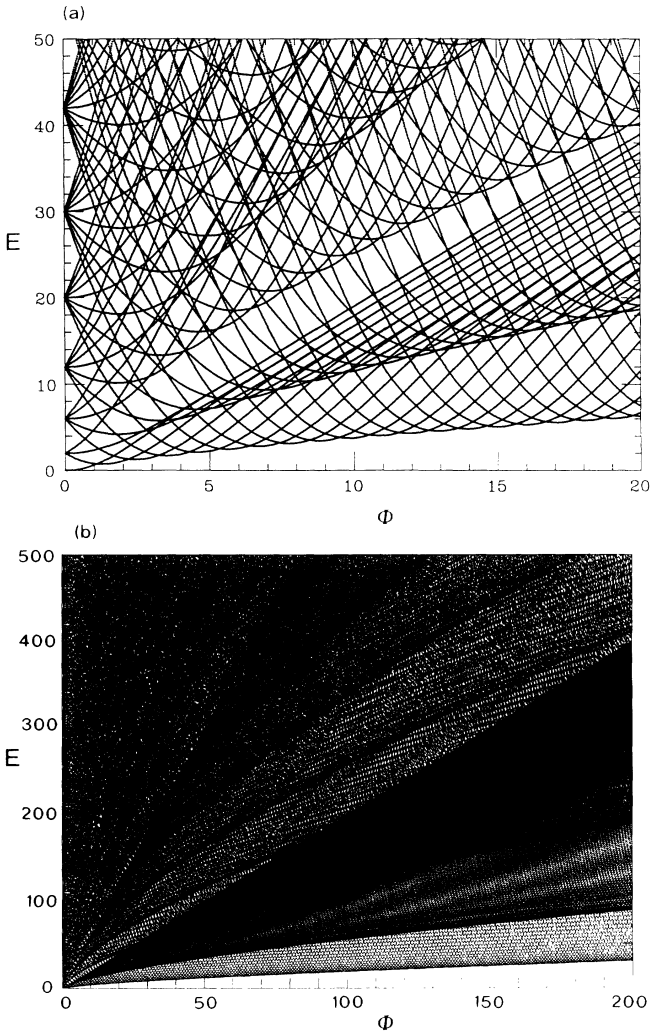


FIG. 1. Energy levels against the dimensionless magnetic field, $\Phi$, for an electron gas on a sphere in a uniform magnetic field. (a) is the (tenfold) enlargement of (b).

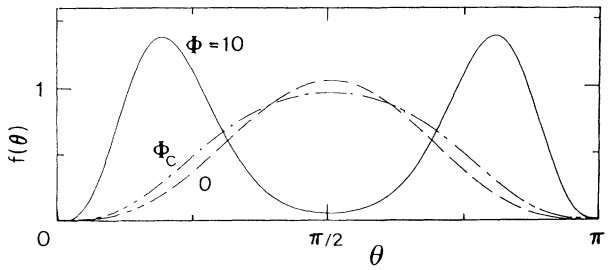


FIG. 2. Wave function, $f_{4,-4}(\theta)$, for various values of $\Phi$, where $\Phi_c$ [$\approx 3.5$ for $(l,m)=(4,-4)$] is the value of $\Phi$ that gives the lowest energy.

length scale characterizing the system unlike the flat system, which will give rise to a similarity among the states with progressively larger number, $l$, of cyclotron orbits accommodated on the sphere.

Similarly there are $m=-l+1$, $m=-l+2,\ldots$ families with $f(z)$ having increasing number of nodes, in which the above competition between kinetic and potential energies depends on both $l$ and $m$. In terms of the limiting behaviors above, the crossover position of the two behaviors is a definite function of $l$ that depends only on the deviation of $m$ from $-l$, so that the crossover points constitute distinct envelopes for various $(l,m)$ families. An indication that the crossover is right in the middle of the nonperturbative region is also seen from the fact that the crossover occurs well before the aforementioned coalescence of the pair takes place.

An effect of interlacing of the energy spectrum for various $(l,m)$ families appears in the total energy, $E_T$, of the system for a given number of electrons: while an individual level in the large-$H$ region, Eq. (5), has a diamagnetic contribution, $[2(\nu+m)+1]\mu_B$, the series of level crossings arising from the overlapping bands makes $E_T$ for a fixed $E_F$ (i.e., a fixed number of electrons) oscillate with $H$. This is the case even when we start from a closed shell (with $E_F$ in a gap between adjacent $l$ fans at low $H$) in the energy spectrum, and the oscillation exists both before and after $E_F$ plunges into the envelope region as $H$ is increased. This implies that the orbital magnetic moment (at $T=0$), $M=-\partial E_T/\partial H$, always oscillates strongly (Fig. 3). In some regions of $H$, $M$ becomes positive (ferromagnetic), which is consistent with the observation of Meier and Wyder [6] for rotationally symmetric systems.

If we look at the energy spectrum for wider ranges of $E$ and $\Phi$ [Fig. 1(b)], a striking full picture emerges: each family of given $N$ forms a *band,* of which the lower envelope comprises the crossover points for the $l=-m+2N$ bunch, the middle envelope is delineated by the crossover points of the partner states with $l=-m+2N+1$, and the upper boundary is the top of the bunch of Landau levels with $E=(N+1/2)\hbar\omega_c+\text{const}$, which corresponds to the pair containing $l=m=N$ state, and the whole spectrum is in fact a *superposition* of the bands for $N=0,1,\ldots$.

Spherical structures may be realized in covalent clusters with $\pi$ electrons as exemplified by the recently fabricated fullerene families. In finite clusters the tight-binding nature of electrons may influence the Landau quantization. To explore this point, we have numerically

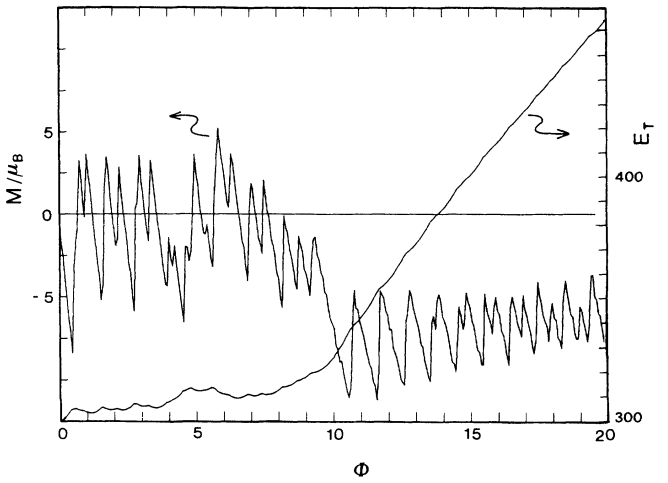


FIG. 3. Total energy, $E_T$, and the orbital magnetic moment, $M=-\partial E_T/\partial H$, against magnetic field for 25 electrons on a sphere. $E_F$ starts to run along an envelope around the middle of the figure.

diagonalized the tight-binding model on the idealized $C_{60}$ and $C_{240}$ fullerene structures, in which a constant transfer energy $t$ is assumed between nearest-neighbor atoms. The presence of uniform magnetic fields is incorporated as the Peierls phase in the transfer energy. The result for the energy spectrum (Fig. 4) shows that, for small clusters such as $C_{60}$, the zero-field spectrum [7,8] itself is dominated by the specific structure (icosahedral symmetry here), so that degeneracies and gaps in the levels due to the symmetry strongly interfere with the effect of uniform magnetic fields. There, the splitting of degenerate levels for finite $H$ should be observable by optical absorptions. We also notice a closure of the lowest unoccupied molecular orbital–highest occupied molecular orbital gap with increasing $H$.

The orbital magnetism is again of interest. The magnetic susceptibility of $C_{60}$ is intriguing as pointed out by Elser and Haddon [7], who have pointed out that the ring current effect only results in a weak diamagnetism for infinitesimal $H$. The total energy at general magnitude of the field, however, has also interesting structures. Although this aspect already appears for the sphere as mentioned above, an effect specific to a molecule is that the electron levels depend on the orientation of the system relative to $H$ as well, so that the molecular axis, when free to rotate, has preferred directions in $H$ with a stabilization energy (of typically $0.1t$ depending on $\Phi$).

As the size of the cluster is increased (to $C_{240}$ in Fig. 4), the region over which the result resembles the continuous-space result increases, and some envelopes become visible. For these molecules the magnitude of $H$ in the region of interest [$\Phi\sim 1$, i.e., the encircled flux $\sim O(\phi_0)$] is still orders of magnitude larger than those attainable experimentally at present. If we regard the problem more generally as a quantization in mesoscopic systems, we can alternatively consider clusters of larger sizes or fine particles with metallic coating. The effect of a finite thickness of the metallic surface is an interesting fu-

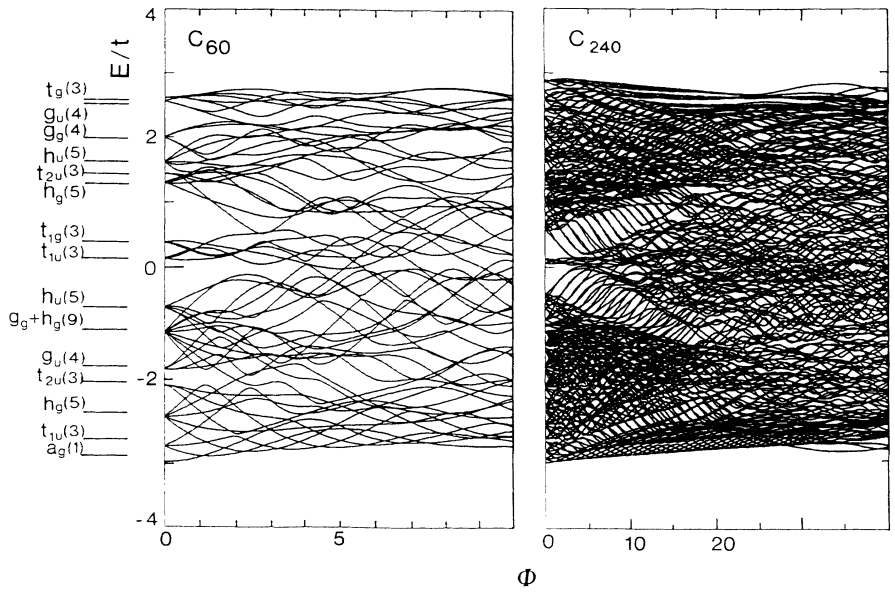


FIG. 4. Energy levels of a tight-binding system on $C_{60}$ (left panel) and $C_{240}$ (right panel) fullerene structures against the strength of the uniform magnetic field. Here $\Phi$ is the total magnetic flux encircled by the sphere circumscribing each fullerene in units of $\phi_0$ and $E$ is measured in units of the transfer energy ($t$) in the tight-binding model. Here $H$ is applied parallel to the sixfold axis of the fullerene structures. For $C_{60}$ the symmetry and degeneracy of each level for $H=0$ is indicated after Ref. [7].

ture problem. Another extension would be to consider free electrons on solid $C_{60}$ crystals [4] in a uniform magnetic field. Also, the effect of curvature may be probed in a structure such as a negative-curvature fullerene [9].

The message obtained in this work is that, although such phenomena as the quantum chaos in the hydrogen atom in a uniform magnetic field [10] are absent for electron systems confined on surfaces, the application of magnetic fields does give rise to interlacing "shell" structures in the energy spectrum with an interesting orbital magnetism.

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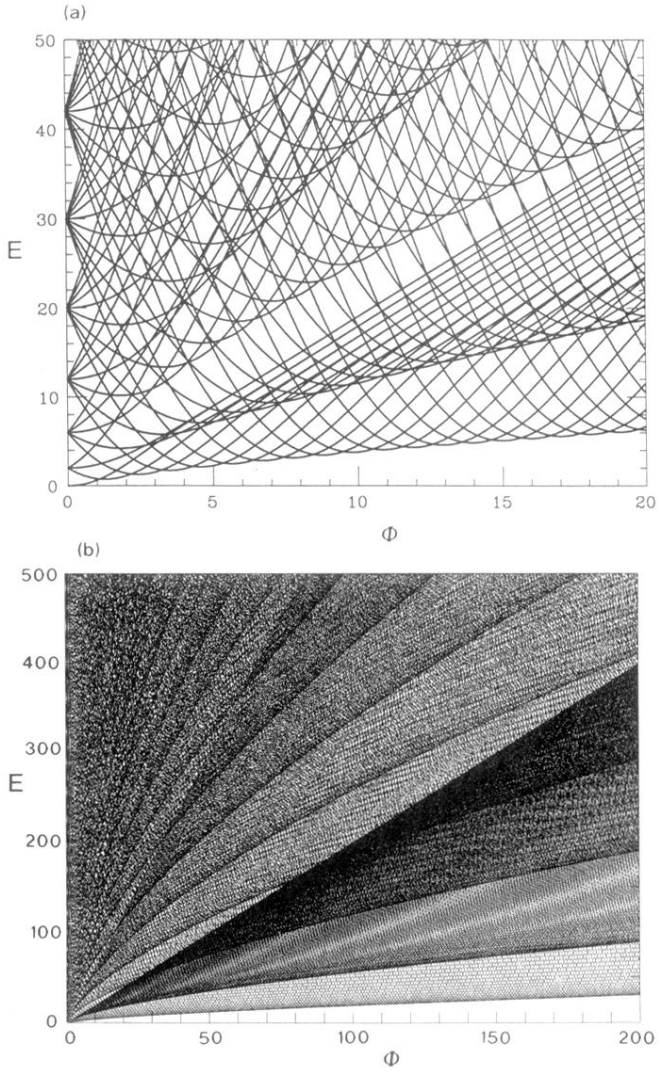


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