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The effects of external magnetic field on the surface charge distribution of spherical nanoparticles

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Abstract

The finite element method has been implemented in order to investigate the electronic structure of spherical quantum dots (QDs) in an external magnetic field. The Schrödinger equation has been discretized by means of the Galerkin's weighted residue method with a nonuniform mesh of triangular elements. Unlike other approaches, the computational effort required to obtain converged results is independent of the strength of the magnetic field. The effects of the diamagnetic term on the energy levels and surface charge density distribution for semiconductor metal oxide QDs in alkaline aqueous colloids have been discussed.

Keywords: Magnetic field; Quantum dot; Nanocrystals

1. Introduction

Important theoretical and experimental efforts have lately been devoted to the study of the physical properties of the semiconductor quantum dots (QDs) [1]. In particular, the ability to manipulate and control spin states and modulate the Landé factor is at the present moment of fundamental importance in the study of electronic systems with potential applications in spintronics and quantum computation. Although the intense researches have been made in this area, the effects of magnetic fields on the optical properties of QDs still remain in discussion. The starting point for a serious study of any optical or spintronic property is the rigorous determination of the electronic structure of QDs. Several calculation procedures have been proposed in the literature to solve the problem of a single SQD in presence of an external magnetic field. Among them, the strong perturbation theory is widely used [2]. In this method, the accuracy of the solution depend on the number of basis functions and their functional forms. However, appropriate basis functions are difficult to obtain or are not available in closed form for most QDs with complex geometries. On the other hand, the $k{\cdot}p$ method is also used to determine the energy spectra in presence of magnetic fields. This method is of highly efficient if the solutions of the unperturbed problem are used as basis functions to diagonalize the $k{\cdot}p$ Hamiltonian. However, for the case of SQD under a magnetic field, the unperturbed problem has no analytic solutions. Therefore, some approximations should be used. For instance, in the regime of weak magnetic field, the magnetic length is larger than the spatial dimensions of SQD. As a result, the magnetic field can be considered as a perturbation that does not significantly deform the wave functions and the solutions of SQD in the absence of magnetic field can now be used as basis functions to expand the solution.

Recent applications of nanostructures in spintronics and quantum computation call for a detailed knowledge of the properties of the eigenvalues and eigenfunctions in any regime of applied magnetic field. Therefore, it is necessary to develop robust computation tools that allow the study of optical and transport properties of nanocrystals under external magnetic field. To our knowledge, an efficient method that allows the investigation of the effects of an external magnetic field on the electronic states in real 3D QDs has not yet been reported.

Finite elements method (FEM) is a convenient and efficient procedure for the calculation of electronic structures and other physical properties in semiconductor

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nanostructures. Its successful application on quantum mechanics problems was reviewed in Ref. [3] and references therein. In this paper, we extended FEM to the study of the electronic structure of nanocrystals in the presence of an external magnetic field.

2. Theory

In the presence of an external magnetic field parallel to the z-direction, the Hamiltonian of the system is described by

$$\hat{H} = [-\nabla^2/2m + U(r)] - \tfrac{1}{2}\omega_c \hat{L}_z + \tfrac{1}{8} m[\omega_c r \sin\theta]^2, \quad (1)$$

where $\omega_c = eB/m^*$ is the cyclotron frequency, $U(r)$ is potential energy of QD. L_z is the projection of the angular moment operator L onto the field axis-z. B is the magnetic field strength. From Eq. (1), we observe that the Hamiltonian commutes only with the operator L_z. In addition, the spherical symmetry of the energy potential is broken by the external magnetic field, while the cylindrical symmetry still holds. Thus, cylindrical coordinates (ρ, z) are more convenient to be used for the solution of Schrödinger equation. The wave function is required to vanish as $\rho \to \infty$ and $\pm z \to \infty$. Finally, the FEM is used to discretize the two-dimensional Schrödinger equation. Triangular element meshes are used because of their adaptability to various shapes of boundaries and interfaces. The Schrödinger equation in cylindrical coordinates is discretized using Galerkin's weighted residue method, and the wave function is approximated by an expansion over the FEM basis functions. For the kth element, a complex linear combination is given by

$$\psi_{n,m}^{(k)}(z,\rho) = \sum_{i=1,N'} c_i^{(k)} \phi_i(z,\rho), \quad (2)$$

where $N' = 3$. We obtain a generalized eigenproblem for the coefficients c_i and eigenvalues $\lambda_{n,m}$ which determine the approximate eigenfunctions and eigenvalues of the variational formulation, and thus the original problem is written as $Hc = \lambda_{n,m} Dc$, where

$$H_{ij}^{(k)} = \frac{1}{2m(r)} \int\int_{(k)} [\nabla\phi_i \nabla\phi_j + 2m(r) V_{\text{eff}}(z,\rho)\phi_i\phi_j]\rho \, \mathrm{d}\rho \mathrm{d}z \quad (3)$$

is a structured sparse symmetrical matrix of Hamiltonian (1) with

$$V_{\text{eff}} = V_c(\rho, z) + \frac{1}{2m(r)}\left[\left(\frac{\rho}{2l_B^2}\right)^2 + \frac{m^2}{\rho^2}\right],$$

and

$$D_{ij}^{(k)} = \int\int_{(k)} \phi_i\phi_j\rho \, \mathrm{d}\rho \mathrm{d}z.$$

Here, $V_c(\rho, z)$ is hard wall energy potential and l_B is the magnetic length. It is important to note that the elements of the Hamiltonian matrix (3) include (i) the potential energy $V_c(\rho, z)$ that depends strongly on the actual geometry and structure of the studied system, and (ii) the interaction with an external uniform magnetic field B (linear Zeeman and quadratic diamagnetic terms). It is also worthwhile to note that FEM with the cylindrical representation is also useful in the study of nonspherical nanocrystals (hemispherical, lens-shape, quantum rings, two or more aligned and interacting nanocrystals).

3. Results

Fig. 1 shows the influence of magnetic field on the energy spectrum of ZnO/H_2O SQDs for d =200 Å. We found that the effects of magnetic field on electron energy levels depend strongly upon the size of SQD. For the smaller SQDs the field dependence of the energy levels is, like in atoms, almost exclusively linear even for fields as strong as B =50 T (linear Zeeman regime). Actually, the effects of the diamagnetic term, which is proportional to B^2, start to be seen only at fields $B > 70$ T in a single SQD with a diameter d =100 Å. In a larger SQD, however, the diamagnetic effect is seen already for laboratory fields ($B \sim 20$ T). Moreover, the signatures of Landau levels can also be observed between the states 1P ($m = 1$) and 1D ($m = -1$), as shown in Fig. 1. Other important effect is related to the dependence of the $m = 0$ levels with the magnetic field. A decrease of the energy is observed for increasing magnetic fields. This behavior is due to the diamagnetic contributions and the peculiar symmetry

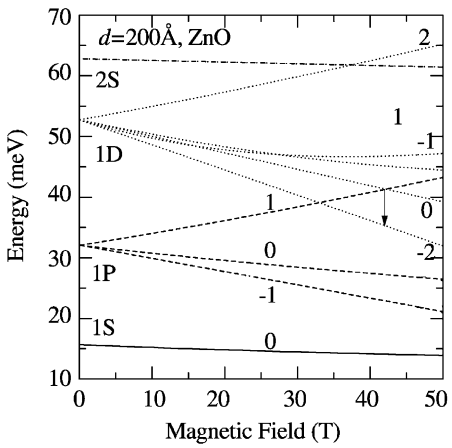

Fig. 1. Electron energy spectra as a function of a magnetic field d = 200 Å ZnO/H_2O SQD. A band offset V_0 = 2.0 eV was used. The numbers near the energy levels represent the corresponding quantum number m.

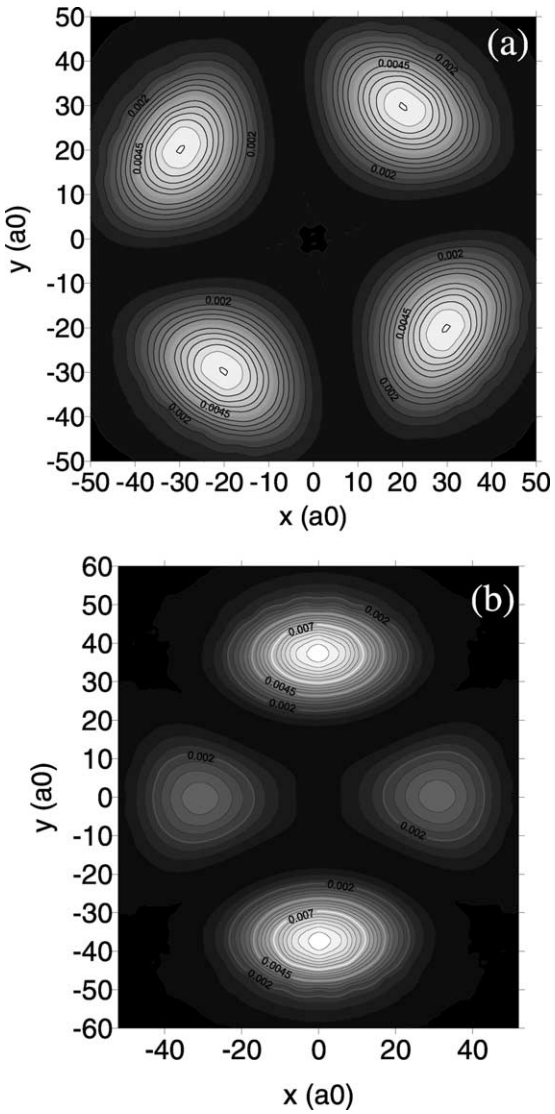

Fig. 2. Contourn of electron probability of D-orbit for a single ZnO/H_2O SQD with $d = 100$ Å for (a) $B = 0.05$ and (b) 150 T.

of the states with $m = 0$. When the magnetic field increases, the confinement introduced by magnetic field plays a more and more important role in determining energy spectrum and the term proportional to B^2 should produce a general increase in the energy levels. This effect, only visible for the level 1D ($m = 1$), requires intense magnetic fields. It is also important to put attention to the crossover produced between the levels 1D ($m = 0$) and 1D ($m = -1$) for $B \approx 14$ T and between the levels 1D ($m = -1$) and 1D ($m = 1$) near $B = 25$ T. This crossing structure of the levels produces a reordering of the energy levels for intense magnetic fields. Experiments in magneto-absorption could verify these theoretical results since the interband selection rules $\Delta m = 0$ allow us to study the evolution of a state with well-defined quantum number m.

Fig. 2 shows the magnetic field dependence of the electron probability in D-orbit for a single ZnO/H_2O SQD with $d = 100$ Å for (a) $B = 0.05$ and (b) 150 T, respectively. As known, surface charge density of electron in SQD is directly related to electron probability. Thus, Fig. 2 indicates that the surface charge distribution can be tuned by external magnetic field.

4. Conclusion

We present an approach for nanoparticle electronic structure calculations based on FEM. In this method, the basis functions are strictly local, piecewise polynomials. Therefore, the method is completely general and its convergence can be controlled systematically. Because the basis functions are local in real space, the method allows for variable resolution in real space; produces sparse, structured matrices, enabling the effective use of iterative solution methods. Within the framework of FEM, the effects of spherical nanocrystal size and external magnetic field on the energy spectra of electron in SQDs have been numerically investigated. The diamagnetic contribution to Zeeman effect has been observed in large dots for relatively low magnetic fields.

Acknowledgements

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