

Influence of the shape of quantum dots on their far-infrared absorption

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We investigate the effects of the shape of quantum dots on their far-infrared absorption in an external magnetic field by a model calculation. We focus our attention on dots with a parabolic confinement potential deviating from the common circular symmetry, and dots having a circular doughnut shape. For a confinement where the generalized Kohn theorem does not hold, we are able to interpret the results in terms of a mixture of a center-of-mass mode and collective modes reflecting an excitation of relative motion of the electrons. The calculations are performed within the time-dependent Hartree approximation, and the results are compared to available experimental results. [S0163-1829(99)13947-X]

I. INTRODUCTION

Ever since the discovery that far-infrared radiation (FIR) can only be used to excite center-of-mass modes of electrons parabolically confined in circular quantum dots, the generalized Kohn theorem,¹⁻⁵ ingenious ways have been thought of to modify the confinement in order to excite internal modes. The modes caused by relative motion of the electrons would allow for an exploration of interaction and general many-body effects. It was demonstrated that in circular dots with a slight radial deviation from parabolic confinement, states with few electrons have a unique far-infrared spectrum.⁶ Dots with a higher number of electrons ($N > 10$) commonly only show a simpler structure of Bernstein modes^{6,7} and an energy shift.^{8,9} Early on, it became clear that a certain anti-crossing behavior discovered in the higher of the two Kohn's modes measured¹⁰ for quantum dots assumed to be circular is indeed a signature of a slight square deformation of the electron system.^{11,8} The far-infrared absorption of strictly square-shaped quantum dots with hard walls has been modeled by exact numerical diagonalization for two electrons,^{12,13} and by a real-time simulation of the density oscillations described by a local-spin-density approximation for many electrons.¹⁴ The generalized Kohn theorem has been further extended to describe elliptic¹⁵ quantum dots and related three-dimensional structures, ellipsoids.¹⁶ Magneto-plasmon excitations in an array of large quantum dots with square deviation have recently been studied within the Thomas-Fermi-Dirac-von Weizsäcker approximation,¹⁷ and the excitation spectrum of an electron gas on a disk with deviations from a circular shape have been calculated within time-dependent density-functional theory in the linear regime.¹⁸

The production of arrays of isolated angularly deformed quantum dots has been hampered by technical difficulties. The little there are of experimental results from FIR absorption measurements on quantum dots with angular deviation have not found their way into the general physics literature, but are to be found as thesis work.¹⁹⁻²¹

The FIR absorption of single quantum rings has been studied in relatively large systems with many electrons,^{22,23} but recently small quantum dots with few electrons and a

hole through their center have been produced and measured.^{24,25}

In this paper we model the FIR absorption of single quantum dots with a slight elliptic or square deviation, assuming the radial confinement to be parabolic. The model is general enough to be applicable for most angular deviations of the confinement. In a circular quantum dot the electron-electron interaction does not break the angular symmetry, but in our model it can strongly modify the shape of the dot as soon as the circular symmetry is broken by the initial confinement. We apply our model to FIR absorption of an elliptic dot in order to verify that it produces the results expected by Kohn's theorem.¹⁵ The calculated absorption spectra for a square-deviated quantum dot show both familiar results for a weak deviation,^{10,11} and effects reflecting internal electron motion that have not yet been observed in experiments. In addition, we calculate the FIR absorption for a small dot with few electrons with its center removed to compare with the results of Lorke and co-workers.^{24,25}

II. MODEL

As we allow for a very general angular shape of the quantum dot, neither the total angular momentum nor the angular momentum of the effective single-particle states in a mean field approach is conserved. The Coulomb interaction thus "mixes" up all elements of the functional basis used, and we limit ourselves to the Hartree approximation (HA) in order to be able to calculate the absorption. The quantum dot is modeled with the confinement potential

$$V_{\text{conf}}(\mathbf{r}) = \frac{1}{2} m^* \omega_0^2 r^2 \left[1 + \sum_{p=1}^{p_{\text{max}}} \alpha_p \cos(2p\phi) \right], \quad (1)$$

representing an elliptic confinement when $\alpha_1 \neq 0$ and $\alpha_p = 0$ for $p \neq 1$, and a square-symmetric confinement when $\alpha_2 \neq 0$ and $\alpha_p = 0$ for $p \neq 2$. We use the Darwin-Fock basis: the eigenfunctions of the circular parabolic confinement potential in which the natural length scale a is given by

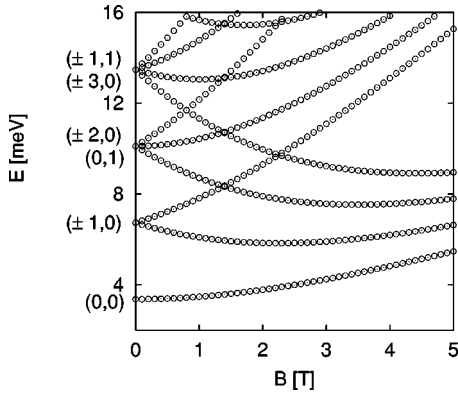


FIG. 1. The Darwin-Fock energy diagram of a single electron in a circular parabolic quantum dot. At $B=0$, we have the familiar two-dimensional harmonic oscillator. The states at $B=0$ are labeled with (M, n_r) .

$$a^2 = \frac{l^2}{\sqrt{1 + 4\left(\frac{\omega_0}{\omega_c}\right)^2}}, \quad l^2 = \frac{\hbar c}{eB}, \quad (2)$$

where $\omega_c = eB/m^*c$ is the cyclotron frequency of an electron with effective mass m^* in a perpendicular homogeneous magnetic field B . The states are labeled by the radial quantum number n_r and the angular quantum number M .⁸ The single-electron spectrum of these states is shown in Fig. 1.

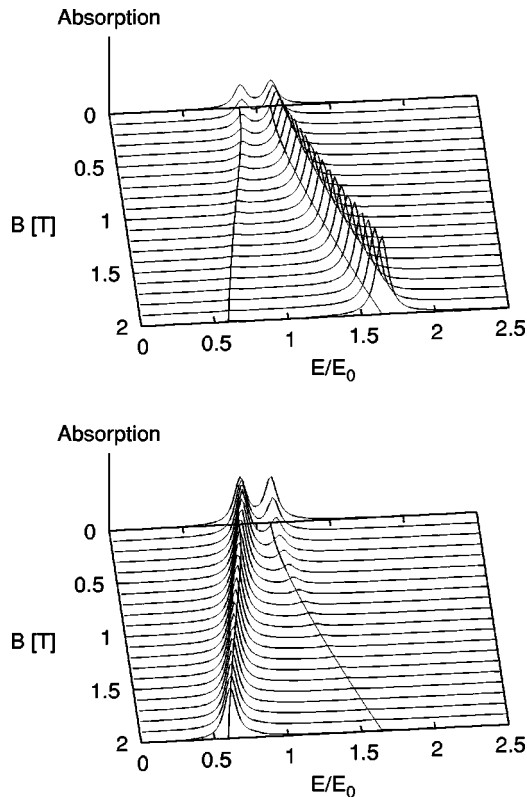


FIG. 2. The far-infrared absorption for $N_p = +1$ (above) and $N_p = -1$ (below) in the case of a two-electron elliptic quantum dot ($\alpha_1 = 0.20$). The solid lines indicate the dispersion relation for the absorption, with $\omega_x/\omega_0 = 1.10$ and $\omega_y/\omega_0 = 0.89$.

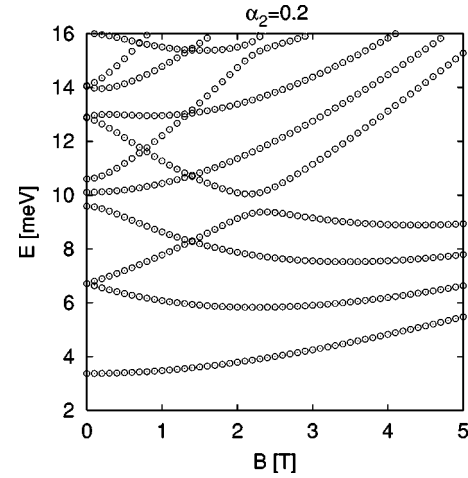


FIG. 3. The Darwin-Fock energy diagram of a single electron in a square-symmetric quantum dot with $\alpha_2 = 0.20$. The angular part of V_ϕ , $\cos(4\phi)$, couples states in the circular parabolic potential for which $\Delta M = \pm 4$. The degeneracy of states at $B=0$ fulfilling $\Delta M = \pm 4$ is lifted. An anticrossing occurs at $B \approx 2.3T$ due to coupling of the accidentally degenerate states $(+1,0)$ and $(-3,0)$ at this magnetic field.

To evaluate the FIR response of the quantum dot, the external potential is assumed to have the form

$$\phi^{ext}(\mathbf{r}, t) = \phi_0 r e^{i[N_p \phi - (\omega + i\eta)t]}, \quad (3)$$

where $N_p = \pm 1$ and $\eta \rightarrow 0^+$, representing a spatially constant external electric field. The FIR response is found by a self-consistent method in the linear-response regime: the time-dependent Hartree approximation. The power absorption is then given by

$$\mathcal{P}(\omega) \propto \omega \Im \left\{ \sum_{\alpha, \beta} f^{\beta\alpha}(\omega) |\langle \beta | \phi^{sc} | \alpha \rangle|^2 \right\}, \quad (4)$$

where

$$f^{\beta\alpha}(\omega) = \frac{1}{\hbar} \left\{ \frac{f_\alpha^0 - f_\beta^0}{\omega + (\omega_\alpha - \omega_\beta) + i\eta} \right\} \quad (5)$$

and f^0 is the equilibrium Fermi distribution. Self-consistency is obtained by calculating linear response not to the external perturbation, ϕ^{ext} , but rather to the total (self-consistent) potential $\phi^{sc} = \phi^{ext} + \phi_H^{ind}$, where ϕ_H^{ind} is the induced Hartree potential. For a circular quantum dot, the parabolic potential has proven to be a realistic approximation in many cases. For such a circular harmonic dot, the dipole selection rule for the center-of-mass angular momentum is $\Delta M = N_p$ and the resonance frequencies are given by

$$\omega_\pm = \frac{1}{2}(\Omega + N_p \omega_c), \quad \Omega = (\omega_c^2 + 4\omega_0^2)^{1/2}.$$

For an anisotropic harmonic confinement, the selection rule is still $\Delta M = \pm 1$, but there is absorption into both ω_+ and ω_- for each polarization. The resonance frequencies are then given by

$$\omega_\pm^2 = \frac{\omega_x^2 + \omega_y^2 + \omega_c^2 \pm [\omega_c^4 + 2\omega_c^2(\omega_x^2 + \omega_y^2) + (\omega_x^2 - \omega_y^2)^2]^{1/2}}{2}, \quad (6)$$

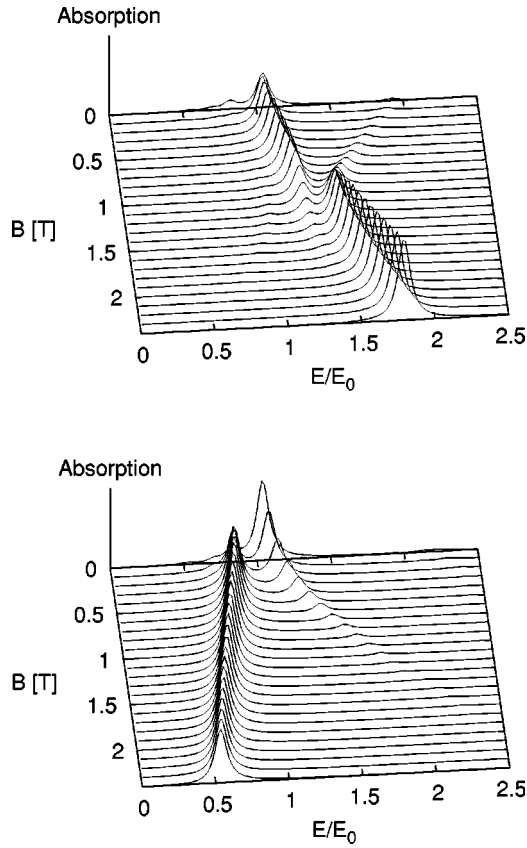


FIG. 4. The far-infrared absorption of a two-electron square-symmetric quantum dot ($\alpha_2=0.20$) for $N_p=+1$ (above) and $N_p=-1$ (below). $E_0=3.37$ meV, and $T=1.0$ K.

where ω_x and ω_y are the resonances at $B=0$ T,^{16,15} fulfilling $\omega_x=(1+\alpha_1)^{1/2}\omega_0$ and $\omega_y=(1-\alpha_1)^{1/2}\omega_0$ in our model.

III. RESULTS

In the calculations reported here we use GaAs parameters, the effective mass $m^*=0.067m_0$, and the dielectric constant $\kappa=12.4$. The FIR absorption of a quantum dot with an elliptic deviation is seen in Fig. 2. The induced density for the oscillation modes was used to confirm that only center-of-mass modes are excited, and the dispersion with respect to B can be reproduced as a simple difference of the dipole-active transfers in the Darwin-Fock energy spectrum for one electron in a parabolic elliptic confinement. The oscillation strengths do comply with known analytic expressions.¹⁵ Interestingly, the external circular polarized electric field leads to linear oscillations of the electron system parallel to the minor and major axes of the contours of the elliptic confinement potential.

Figure 3 shows the Darwin-Fock energy for a single electron in a confining potential with a square deviation. Clearly visible is the anticrossing of $(M,n_r)=(+1,0)$ and the state $(-3,0)$ that was found to be visible in the FIR absorption of quantum dots which were clearly of square shape in an electron micrograph.^{10,11,8} This anticrossing is present in the calculated absorption shown in Figs. 4 and 5 for two electrons in a dot. For this relatively large deviation ($\alpha_2=0.2$ or 0.4) from circular symmetry, the absorption at $B=0$ for both circular polarizations ($N_p=\pm 1$) is split into two or more

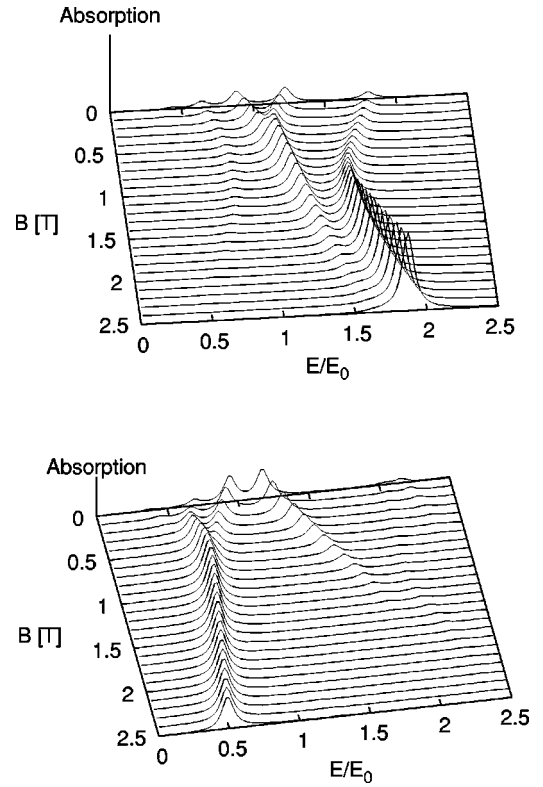


FIG. 5. The FIR absorption of a square-symmetric two electron quantum dot with $\alpha_2=0.40$, $N_p=+1$ (above) and $N_p=-1$ (below). $E_0=3.37$ meV and $T=1.0$ K.

peaks. This is in contrast to the Darwin-Fock energy diagram in Fig. 3, from which we have to conclude that the lowest energy center-of-mass mode should be unsplit at $B=0$. Certainly it has to be kept in mind that the states in Fig. 3 cannot

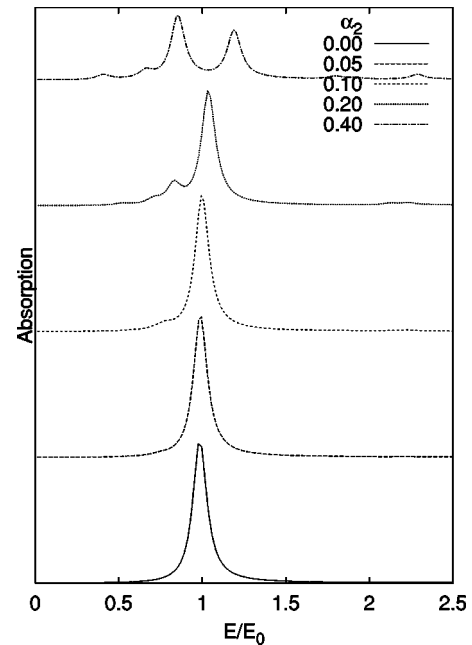


FIG. 6. The evolution of absorption peaks of a square-symmetric two-electron quantum dot at $B=0$ T as α_2 is increased. The influence of the square-symmetric deviation from the circular parabolic confinement starts to show up in the absorption spectrum for $\alpha_2 \geq 0.10$.

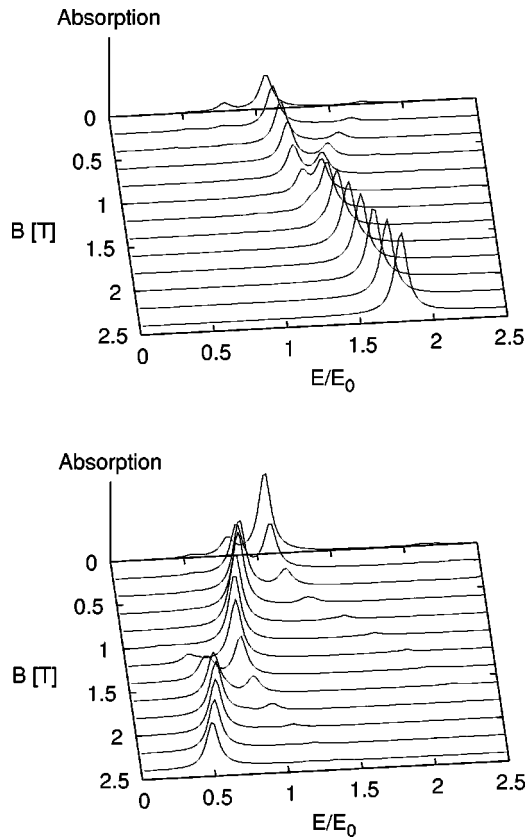


FIG. 7. The FIR absorption of a square-symmetric quantum dot. Same parameters as in Fig. 4, but $N=3$.

be assigned any unique quantum numbers n_r and M . The evolution from one absorption peak to many at higher deviation is seen in Fig. 6. Observation of the induced density shows that all the modes are a mixture of center-of-mass and relative modes to different extent here. Some peaks can be identified with depolarization-shifted transitions in the interacting Hartree energy spectrum, while others are very close to transitions in the noninteracting Darwin-Fock diagram reflecting center-of-mass modes.

In Fig. 7 the FIR absorption is shown for three electrons

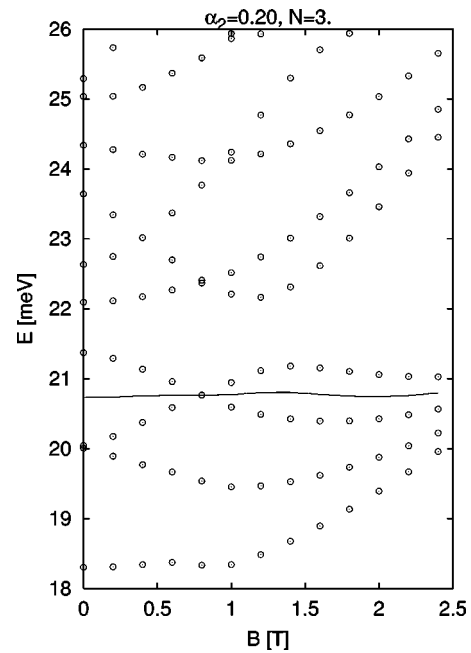


FIG. 8. The Darwin-Fock energy diagram for $N=3$ interacting electrons and $\alpha_2=0.20$. The chemical potential is denoted by the solid line.

($N=3$). A splitting or anticrossing occurs in the absorption for $N=3$ and $N_p=-1$, whereas it does not for $N=2$. This striking behavior can be understood from the Darwin-Fock energy diagram for three electrons (Fig. 8). The splitting can be identified as a transition from the third energy level to the fourth and fifth with a depolarization shift, showing its many-body character. This behavior cannot be seen for $N=2$, since the third level is then not occupied.

Generally the lower-energy branch (seen in $N_p=-1$ polarization in circular dots) is very stable against splitting, especially for dots with many electrons. One exception is a dot with some kind of a hole through its center. Such dots have been grown as self-organized InAs rings embedded into a GaAs/Al_xGa_{1-x}As heterostructure. Here we try to model simply the qualitative behavior of such dots by using GaAs

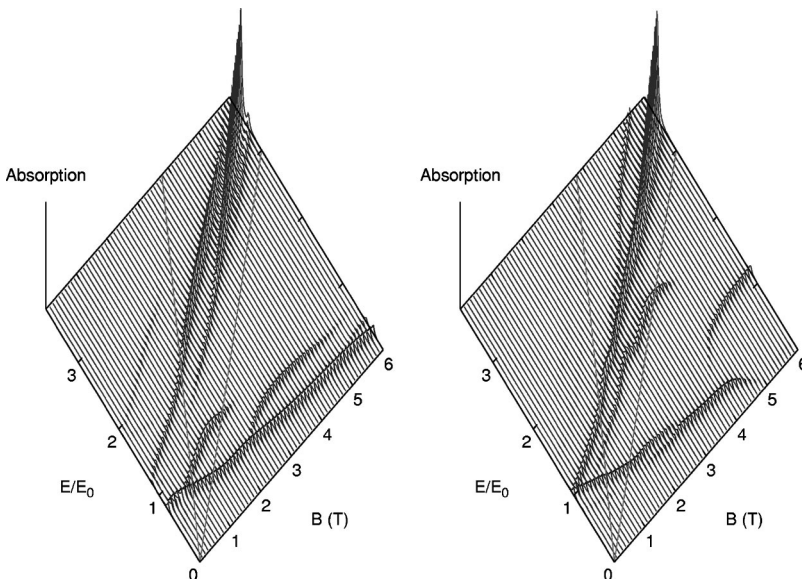


FIG. 9. The far-infrared absorption $P(E/E_0, B)$ in the case of $N=2$ (left), and $N=12$ (right) electrons in a circular symmetric quantum dot with the center removed. The straight lines indicate the cyclotron resonance and its first harmonic. Only the lowest branch in each case corresponds to $N_p=-1$ and all other branches belong to $N_p=+1$. $E_0=\hbar\omega_0=3.37$ meV and $T=1.0$ K.

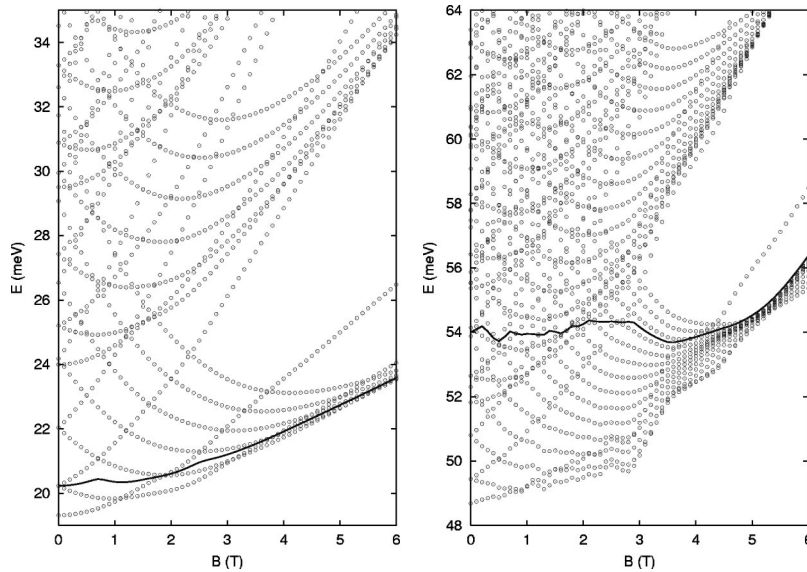


FIG. 10. The Darwin-Fock energy diagram in the case of $N=2$ (left) and $N=12$ (right) electrons in a circular-symmetric quantum dot with the center removed. The chemical potential μ is indicated by the solid curve.

parameters, and by replacing the hole with a repulsive Coulomb center in the middle of the dot. The confinement of the electrons is thus not of the same type as was used to study the far-infrared absorption of a finite two-dimensional electron gas with an embedded impurity.²⁶ The calculated absorption is seen in Fig. 9 for two and 12 electrons in the quantum ring. To understand the absorption, one has to consider the Darwin-Fock energy spectrum of the Hartree interacting electrons in the ring, shown in Fig. 10. In the case of two electrons in the ring the emptying of states with a low angular quantum number M close to the center with increasing magnetic field B results in low-energy “side branches” with polarization $N_p = +1$ to the $N_p = -1$ branch. The magnetic length a decreases with increasing B , causing the electrons to “see” in a sense more of the repulsive Coulomb center (and each other), resulting in the emptying of states of low M . It is our suggestion that Lorke and co-workers observed this same effect in their experiment.^{24,25} It should be mentioned here that the electron density in our calculation at the center of the ring is low for $B=0$, and vanishes as B increases. In the case of 12 electrons, screening effects and oscillating structure reminiscent of emptying of Landau bands is quite clear in the Darwin-Fock energy diagram, but again an emptying of the $M=0$ state at $B=4$ T is visible in the absorption together with a restructuring of the states with high M at the outer edge.

IV. SUMMARY

We have used a general two-dimensional multipolar expansion of the confinement potential of a single quantum dot

to study the effects of shape on its properties. We have shown that the far-infrared absorption spectrum of quantum dots with few electrons strongly reflects their shape and the exact number of electrons if the confinement potential does not allow the application of Kohn’s theorem. Elliptic dots have two low-energy peaks of equal strength at $B=0$, whereas dots with slight square symmetry reveal only one peak. This behavior is connected to the different effects of shape on the $(M, n_r) = (\pm 1, 1)$ degeneracy of circular dots at $B=0$. We find that this degeneracy can be lifted by the activation of relative modes in the far-infrared absorption spectrum of square-shaped dots, with increased deviation from the circular form.

In quantum rings we observe a shift of occupation of the states labeled by the angular momentum quantum number M to higher values with increasing magnetic field. This is essentially caused by the changing ratio of the magnetic length a and the dots radii. We identify structures seen in experiments with this effect.²⁴

The Hartree approximation has been used here rather than the more desirable Hartree-Fock approximation in some unrestricted version, in order to manage the size of the absorption part of the calculation measured in GBytes and CPU time. On the other hand, the HA can be expected to give a valid description of a system whose modes of excitation are mainly of the center-of-mass type.

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¹P. A. Maksym and T. Chakraborty, Phys. Rev. Lett. **65**, 108 (1990).

²L. Brey, N. Johnson, and B. Halperin, Phys. Rev. B **40**, 10 647 (1989).

³P. Bakshi, D. Broido, and K. Kempa, Phys. Rev. B **42**, 7416 (1990).

⁴F. M. Peeters, Phys. Rev. B **42**, 1486 (1990).

⁵A. V. Chaplik and S. A. Gorov, Pis’ma Zh. Éksp. Teor. Fiz. **52**, 681 (1990) [JETP Lett. **52**, 31 (1990)].

⁶D. Pfannkuche, V. Gudmundsson, P. Hawrylak, and R. Gerhardt, Solid-State Commun. **37**, 1221 (1994).

⁷V. Gudmundsson, A. Brataas, P. Grambow, T. Kurth, and D.

- Heitmann, Phys. Rev. B **51**, 17 744 (1995).
- ⁸V. Gudmundsson and R. Gerhardts, Phys. Rev. B **43**, 12 098 (1991).
- ⁹Z. L. Ye and E. Zaremba, Phys. Rev. B **50**, 17 217 (1994).
- ¹⁰T. Demel, D. Heitman, P. Grambow, and K. Ploog, Phys. Rev. Lett. **64**, 788 (1990).
- ¹¹D. Pfannkuche and R. Gerhardts, Phys. Rev. B **44**, 13 132 (1991).
- ¹²R. Ugajin, Phys. Rev. B **51**, 10 714 (1995).
- ¹³R. Ugajin, Phys. Rev. B **53**, 6963 (1996).
- ¹⁴A. Puente and L. Serra, cond-mat/9905288 (unpublished).
- ¹⁵S. K. Yip, Phys. Rev. B **43**, 1707 (1991).
- ¹⁶Q. P. Li, K. Karrai, S. K. Yip, S. Das Sarma, and H. D. Drew, Phys. Rev. B **43**, 5151 (1991).
- ¹⁷B. P. van Zyl, E. Zaremba, and D. A. W. Hutchinson, cond-mat/9908235 (unpublished).
- ¹⁸C. A. Ullrich and G. Vignale, cond-mat/9907186 (unpublished).
- ¹⁹M. Hochgräfe, Master's thesis, University of Hamburg, Germany, 1998.
- ²⁰E. Vasiliadou, Ph.D. thesis, University of Hamburg, Germany, 1995.
- ²¹C. Dahl, Ph.D. thesis, Ludwig-Maximilians-Universität, München, Germany, 1992.
- ²²E. Zaremba, Phys. Rev. B **53**, R10 512 (1996).
- ²³A. Emperador, M. Barranco, E. Lipparini, M. Pi, and L. Serra, Phys. Rev. B **59**, 15 301 (1999).
- ²⁴A. Lorke and R. J. Luyken, Physica B **256-258**, 424 (1998).
- ²⁵A. Lorke, R. J. Luyken, A. O. Govorov, T. P. Kotthaus, J. M. Garcia, and P. M. Petroff, cond-mat/9908263 (unpublished).
- ²⁶V. Gudmundsson and Á. S. Loftsdóttir, Phys. Rev. B **50**, 17 433 (1994).