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Optical spectroscopies of electronic excitations in quantum dots

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Abstract

Calculations of the electronic and optical properties of quantum dots (QD) reveal that optical absorption and emission and inelastic light scattering may probe a rich spectrum of ground and excited states of QD in a magnetic field. Comparisons are made with Raman measurements for deep-etched GaAs/AlGaAs modulation doped QD in a magnetic field.

Keywords: Electron density, excitation spectra calculations; Gallium arsenide; Light scattering; Many body and quasi-particle theories; Molecular beam epitaxy; Photoluminescence; Photon emission; Quantum effects; Quantum wells; Raman scattering spectroscopy

1. Introduction

We discuss here electronic and optical properties of dots with soft (parabolic [1–4]) and hard-wall (disks [5]) confinement and present preliminary results of inelastic light scattering from deep-etched GaAs/AlGaAs modulation doped disks in a magnetic field. The disks with radii R, ($50\ \text{nm} < R < 100\ \text{nm}$), were etched from a modulation doped multi-quantum well structure [5] with carrier density, $n_s = 8.5 \times 10^{11}\ \text{cm}^{-2}$.

2. Theory

The electronic states of a quantum dot in a magnetic field are determined by the competition between kinetic, Zeeman, and potential energy due to confinement V_c and positive charges V_+, and by electron–electron interactions and correlations.

For quantum dots with soft (parabolic) confinement we can replace the net confining potential $V_c(r_i) + V_+(r_i)$ by an effective parabolic potential [4]. For deep-etched dots the confining potential is modelled by a disk of radius R with infinite walls and the positive potential is a potential of a disk of positive charge with density $N/\pi R^2$ at a distance D from the plane of the dot.

Electronic states of QD's in a magnetic field with N electrons ($N = 1,..,130$) were calculated using exact diagonalization techniques for $N < 15$, and Hartree and Hartree–Fock methods for large number of electrons. All states are labelled by a total angular momentum J and excitations are labelled by an excess angular momentum M.

Results of calculations are illustrated in Fig.1.

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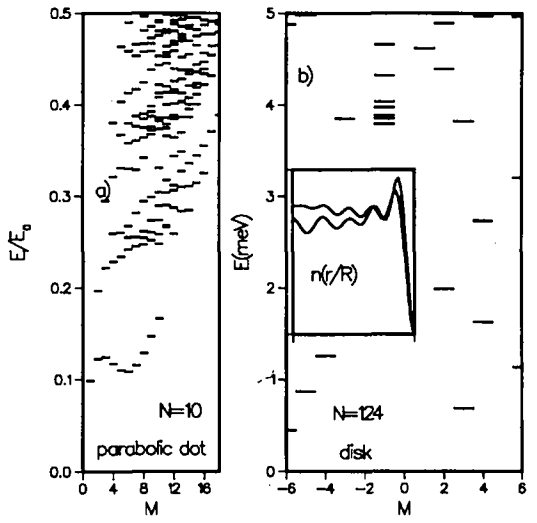

Fig. 1. Electronic excitation energies as a function of excess angular momentum M at magnetic field $B=2$ tesla: (a) $N=10$ compact droplet in a parabolic dot (E_c is the exchange energy of a lowest Landau level), (b) Hartree energies of a disk with $N=124$ electrons.

Fig. 1a shows excited state energies $E(M)$ as a function of the excess angular momentum M of a $N=10$ spin polarized compact parabolic dot with $\omega_N=2.1$ meV in the magnetic field $B=2$ tesla. The well separated low lying excitations of the droplet, the edge magneto-rotons, are clearly visible, while the higher excited states form pairs of edge magneto-rotons. Increasing the magnetic field and the strength of e–e interactions leads to a softening of edge magneto-roton dispersion and to a reconstruction of the dot's charge density. The reconstruction of the dot in terms of freezing of edge magneto-rotons is equivalent to "magic" angular momentum transitions in a few electron dots [4].

Fig. 1b shows the low angular momentum Hartree excitation spectrum of a disk with $R=$ 70 nm and $N=124$ electrons ($n_s\approx 8\times 10^{11}$ cm^{-2}) in a magnetic field $B=2$ T. There is a low lying branch of excitations followed by a band of excitations at $\approx$ 4 meV. The inset shows charge density profiles $n(r/R)$ of a non-interacting disk (almost uniform charge density) and an interacting disk. Because of the spatial separation $D\approx 300$ Å of the positive background from the disk, electrons repel each other very effectively and the electron charge density is depressed inside the disk and enhanced at the edges of the disk.

3. Absorption/emission

In absorption/emission, an exciton probes the magnetic field induced reconstruction of charge density of the parabolic dot and repopulation of quantum levels in a disk. The detailed calculations of a magneto-exciton in an N electron parabolic droplet were carried out using exact diagonalization techniques. In Fig. 2 we illustrate our results by showing the absorption peaks for the $N=3$ electron dot as a function of the magnetic field. Insets show the charge density of the ground state of the dot. For low values of the magnetic field the dot is compact and excitons can be added outside the dot. When the dot undergoes reconstruction, vacancies (holes) appear in the dot, and a photo-excited electron can annihilate these holes. A new, low energy, absorption line (b) appears then as a direct signature of a phase transition in the dot. The phase transition is also visible in the main absorption line (a) which undergoes discontinuous jumps whenever the ground state changes its angular momentum.

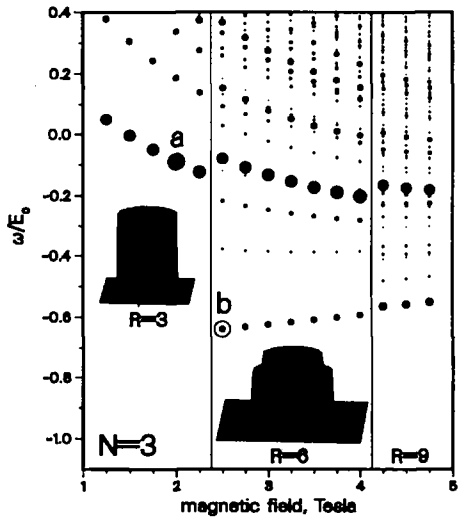

Fig. 2. Absorption spectrum $A(\omega)$ of a $N=3$ electron dot as a function of the magnetic field. Insets show approximate charge density of the dot.

4. Raman scattering

Resonant inelastic light scattering is a useful probe of the spectrum of electronic excitations [5,6] since it does not involve adding or subtracting particles [7]. Slightly off-resonance, the Raman cross section $I(q,\omega)$ corresponding to the transfer of energy ω and momentum q is proportional to the imaginary part of the dynamical structure factor $S(q,\omega)$ of a quantum dot [7].

The Raman spectrum and the dynamical structure factor for few-electron quantum dots has been calculated in Ref. [7]. In a strong magnetic field magnetic field induced transitions correspond to the closing and opening of gaps in the excitation spectrum. It was shown that the Raman spectrum should exhibit the appearance of soft modes corresponding to the collapse of gaps and freezing of edge-magnetorotons as a signature of magnetic field induced phase transitions.

We now turn to inelastic light scattering from deep-etched quantum dots. Photoluminescence and resonant electronic Raman spectra of deep-etched GaAs/GaAlAs quantum dots were measured in magnetic fields up to 17 tesla. The rich spectrum of excitations with Raman shifts in the range 1–35 meV was observed. The spectra show level-splitting, level-crossing, and mode-softening with increasing magnetic field. The spectra for $B=$ 0,1 and 2 T are shown in Fig. 3 for a dot with radius $R=75$ nm.

Since the spectra do not show a clear polarization dependence, they are a mixture of spin density (SDE), charge density (CDE), and single particle (SPE) excitations. Broad features visible in the spectra are on the scale of several meV, yet single particle 0D excitations in Fig. 1 show quantization on the scale of a fraction of a meV. Thus interpretation of the experimental spectra is difficult, and we initially restrict ourselves to SPE based on Hartree calculations.

The effect of the magnetic field is to remove the degeneracy of states with angular momenta $\pm M$ and to change the Hartree fields and the equilibrium charge distribution. This results in a splitting and rearrangement of transition energies and hence in changes of the Hartree SPE spectra.

We show in Fig. 3 the evolution of the calculated Hartree SPE spectra $E(\omega)=$

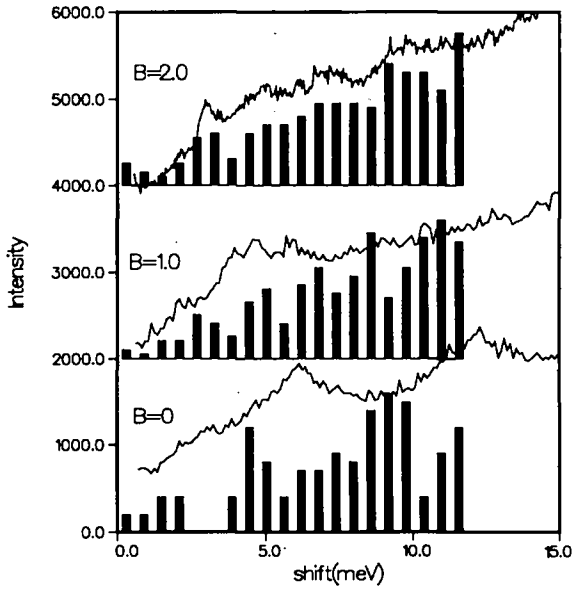

Fig. 3. (a) Raman spectrum $I(q\approx 0,\omega)$ of an array of deep-etched disks with nominal carrier density $n_s=8.5\times10^{11}$ cm^{-2} and radius $r=75$ nm for magnetic fields $B=0$–2 tesla. (b) Hartree Single Particle excitations of a disk with carrier density $n_s=8.0\times10^{11}$ cm^{-2} and radius $R=70$ nm for magnetic fields $B=0$–2 tesla.

$\Sigma_{i,j}(1-f_i)f_j\delta(E_i-E_j-\omega)$ with the magnetic field B (f_i is the Fermi occupation function for a Hartree state E_i) superimposed with the measured spectra. We assumed in our model a slight loss of the carrier density (from 8.5×10^{11} cm^{-2} to 8.0×10^{11} cm^{-2}) and a reduction in the dot size from $R=75$ nm to $R=70$ nm due to the etching. We do not resolve transitions that are less than 0.5 *meV* apart, and do not include matrix elements nor finite state interactions. We see that the spectrum consists of many transitions with a typical spacing of $\approx$ 0.5 meV. The magnetic field induces the splitting of transition energies and some transitions move to lower energies with increasing magnetic field. The envelope of the spectrum shows magnetic field tunable modulations, with a number of bands spaced by several meV. This situation is qualitatively consistent with experiment but more work, and in particular a realistic calculation of edge magno-plasmons and spin density excitations, is needed for a quantitative interpretation of experiment.

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