Far-infrared spectroscopy of nanoscopic InAs rings

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Abstract

We have employed time-dependent local-spin density theory to analyze the far-infrared transmission spectrum of InAs self-assembled nano-rings recently reported [A. Lorke et al., cond-mat/9908263 (1999)]. The overall agreement between theory and experiment is good, which on the one hand confirms that the experimental peaks indeed reflect the ring-like structure of the sample, and on the other hand, assesses the suitability of the theoretical method to describe such small nanostructures. The addition energies of one- and two-electron rings are also reported and compared with the corresponding capacitance spectra.

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Recent progress in nanofabrication techniques has allowed to construct self-assembled nanoscopic InGaAs quantum rings occupied with one or two electrons each, and submitted to perpendicular magnetic fields \( B \) of up to 12 T. These are the first spectroscopic data available on rings in the scatter-free, few electrons limit in which quantum effects are best manifested. Previous spectroscopic studies dealt with microscopic rings in GaAs-Ga\(_{x}\)Al\(_{1-x}\)As heterostructures, fairly well reproduced by classical or hydrodynamical models.

In spite of the lacking of experimental information, the study of nanoring structures has already attracted a strong theoretical interest. We recall that due to the non-applicability of the generalized Kohn theorem, a very rich spectroscopic structure is expected to appear in few electrons nanorings, as anticipated by Halonen et al. and also found in recent works.

In this paper we attempt a quantitative description of some spectroscopic and ground state (gs) properties of the experimentally studied nanorings using current-density (CDFT) and time-dependent local-spin density (TDLSDFT) functional theories. The reason for such an attempt is twofold. On the one hand, to contribute to put on a firmer basis the interpretation of current experiments as manifestation of actual properties of few-electrons ring-shaped nanostructures. On the other hand, to disclose the capabilities and limitations of density functional methods to describe such small systems.

Following Ref. 6, we have modeled the ring confining potential by a parabola

\[
V^+(r) = \frac{1}{2} m \omega_0^2 (r - R_0)^2
\]

with \( R_0 = 14 \) nm and the frequency \( \omega_0 \) fixed to reproduce the high energy peak found in the far-infrared (FIR) transmission spectrum at \( B = 0 \). For \( N = 2 \) electrons this yields \( \omega_0 \approx 12.3 \) meV. The electron effective mass \( m^* = 0.063 \) (we write \( m = m^* m_e \) with \( m_e \) being the physical electron mass) and effective gyromagnetic factor \( g^* = -0.43 \) have been taken from the experiments, and the value of the dielectric constant has been taken to be \( \epsilon = 12.4 \).

To obtain the structure of the gs we have resorted to CDFT as described in Refs. 16, 17, and to obtain the charge density response we have used TDLSDFT as described in Ref. 18, which has been recently applied to the ring geometry. It is worthwhile to point out that we have not found any significant difference between using CDFT or LSDFT to describe the gs of the studied rings in the range of \( B \) values of the present work. The suitability of CDFT to describe such a small electronic system has been shown by Ferconi and Vignale comparing the results obtained for a dot with \( N = 2 \) electrons with exact and Hartree-Fock calculations. We refer the reader to the mentioned references for a detailed exposure of the methods.

The results obtained for the \( N = 2 \) ring are presented in Figs. 1-4. We have used a small temperature \( T = 0.1 \) K to work them out. Figure 1 shows that the ring becomes polarized near \( B = 3 \) T. Besides, two other \( B \)-induced changes arise in the gs at \( B \approx 8 \) T and, more weakly, at \( B \approx 14 \) T. These changes can be traced back to sp level crossings. As displayed in Fig. 2, the changes in the \( B \)-slope appear when an occupied sp level is substituted by an empty one. At \( B \approx 8 \) T, this involves the substitution of the \( l = 0 \) sp level by the \( l = 2 \) one, and at \( B \approx 14 \) T the \( l = 1 \) sp level is substituted by the \( l = 3 \) one. Other level crossings do not involve such substitutions, but a different ordering of the occupied levels and do not seem to produce a substantial effect (see for instance the crossings at \( B \approx 6 \) and \( \approx 11.5 \) T).
The experimentally observed change in the FIR spectrum around $B = 8$ T has been attributed to the crossing of $l = 0$ and 1 sp levels on the basis of a simple single-electron model (see also Fig. 2). A realistic description of the crossings requires to incorporate in the theoretical description the spin degree of freedom, of which single electron or Hartree models lack whereas CDFT or LSDFT do not. Yet, we confirm the finding that a magnetic induced transition takes place in the gs when approximately 1 flux quantum penetrates the effective interior area of the ring at $B \sim 8$ T, and predict another one at $B \sim 14$ T when this area is penetrated by $\sim 2$ flux quanta.

The changes in $B$-slope of the total energy correlate well with these in the electronic chemical potential (the energy of the last occupied sp level in Fig. 2). The gross structure of the chemical potential and total energy displays the well known periodic, Aharonov-Bohm-type oscillation found in extreme sp models:

$$
\epsilon_l = \frac{\hbar^2}{2 m R_0^2} \left( l - \frac{e}{\hbar c} R_0 B \right)^2.
$$

The experimental FIR resonances have been grouped into different modes using a different symbol for each group. Here, we have used the same symbol to represent the experimental resonances in Figs. 3 and 4. Figure 3 shows the dipole charge density strength function in an arbitrary logarithmic scale as a function of the excitation energy. The curves have been offset for clarity. Charge density excitations (CDE) can be identified as ‘ridges’ in the plot, allowing to make a sensible comparison with experiment not only of the peak energies themselves, but also of the way the experimental modes have been grouped. A plot of the more intense CDE’s is presented in Fig. 4 as a function of $B$, which is qualitatively similar to that of Halonen et al for an $N = 2$ quantum dot with a repulsive gaussian impurity in its center. For completeness, we also show in Fig. 3 the longitudinal spin density strength function for the cases in which the ring is not fully polarized. As both strengths coincide in the non-interacting case, the observed shifts are a measure of the importance of the electron-electron interaction, which affects more the low energy peaks than the high energy ones.

These figures show that the FIR dipole response is splitted into two large groups of peaks. The low energy peaks correspond to transitions involving only $n = 0$ sp levels and are $\Delta n = 0$ transitions, whereas the high energy peaks involve $n = 0$ and 1 sp levels and are $\Delta n = 1$ transitions. One can easily distinguish two sets of resonances, a low-lying $\Delta n = 0$ one, and a high-lying $\Delta n = 1$ one exhibiting the usual Zeeman splitting when a magnetic field is applied. The intensity of the high energy resonance is more than one order of magnitude smaller than that of the low energy one. Experimentally, both sets have similar oscillatory strengths, whereas TDLSDFT yields a $\sim 90-10$ % share at most. The calculations in Ref. 9 also yield rather different absorption intensities to these resonances. We have checked that the computed spectrum fulfills the $f$-sum rule to within $\sim 98\%$, thus leaving no room for higher energy, $\Delta n > 1$ peaks to appear within TDLSDFT.

Besides these Zeeman-splitted resonances, several others show up in the spectrum. We have identified with a $+(-)$ sign these involving changes $\Delta |L| = 1(-1)$ in the total orbital angular momentum with respect to that of the gs.

At $B \sim 8$ T, the positive $B$-dispersion brach of the $\Delta n = 0$ resonance disappears, and a very low-lying, positive $B$-dispersion branch shows up. The origin of this transition is the
magnetic-induced change in the gs, as it can be easily inferred looking at the $n=0$ sp levels plotted in Fig. 2 and using the dipole selection rule to identify the ones involved in the non spin-flip excitation. A similar transition occurs at $B \sim 14$ T. They are the microscopic explanation of the appearance and disappearance of the 'ridges' shown in Fig. 3, also found for few electron nanorings. It is worthwhile to notice that the rich structure appearing in these nanorings (see below the $N=1$ case) is a peculiarity that has its origin in the smallness of $N$. When $N$ is just a few tens, many electron-hole pairs contribute to the building of the resonances and no drastic changes appear in the FIR spectrum.

We have also looked at the $N=1$ ring for which some experimental information is also available. As in the $N=2$ case, we have fixed $\omega_0$ so as to reproduce the high energy resonance at $B=0$. This yields $\omega_0 \sim 13.5$ meV.

Figure 5 shows several $n=0$ sp levels of the $N=1$ ring. For this system, the total energy $E(1)$ is simply the energy of the lowest sp level, $E(1) = \mu(1)$. This has been used to calculate the addition energy $\mu(2) = E(2) - E(1)$ shown in Fig. 8. The dipole charge density strength and the energy of the more intense $N=1$ CDE’s are plotted in Figs. 6 and 7 as a function of $B$.

We thus see that the experimental data on FIR transmission spectroscopy reflects that the surface ring morphology of the experimental samples has indeed being translated to a true underlying electronic ring structure, and that a fair quantitative agreement can be found between TDLSDFT calculations and experiment. Our calculations also give support to the way the experimental resonances have been grouped, with the only doubt of the 'dot' peak at $B=6$ T and $\omega \sim 16.1$ meV which could also be a 'triangle' peak of $(\pm)$ character because in this region both branches merge. To unambiguously arrange the peaks into branches and disentangle the $B$ dispersion of the modes, it would be essential to experimentally assign the polarization state to the main CDE’s, as is has been done for antidot array. This is crucial in the analysis of the theoretical FIR response, which otherwise does not allow us to distinguish between peak fragmentation and different plasmon branches in some cases.

From the theory viewpoint, the main shortcomings are the 'cross' peaks appearing at around 8 T for $N=2$, and 10 T for $N=1$, as well as clear overestimation of the peak energy of the $(\pm)$ high energy $\Delta n=1$ mode, which also lacks of some strength. These drawbacks are also qualitatively present in the calculations of Ref. 9. It is alike that using other possible confining potentials, like a jellium ring or that of Ref. 10 which yields analytical sp wave functions in the non-interacting case will improve the agreement in view of other possible sources of uncertainty, as for example the precise value of the ring radius $R_0$ (we have taken that of Ref. 2, but larger values would also be acceptable), and the values of $m^*$, $g^*$ and $\epsilon$ corresponding to InAs. In particular, the effective mass value seems to depend on whether it is extracted from capacitance of from FIR spectroscopy. We have checked that if we take $m^* = 0.08$ we achieve a better description of the 'dot' peaks in Figs. 3 and 4 at the price of spoiling the description of 'diamond' and 'triangle' peaks. Yet, the patterns look qualitatively similar.

Finally, we show in Fig. 8 the addition energies of both rings as compared with the gate voltage shift of the lowest capacitance maximum. It can be seen that the agreement between theory and experiment is rather poor. At $B \sim 12$ T the calculations underestimate the shift voltage around a factor of 3 for $N=2$, and of 2 for $N=1$. We recall that the agreement between capacitance spectroscopy experiments and exact diagonalization calcu-
lations of few electron quantum dots is also only qualitative. We cannot discard that using a different radius $R_0$ for each ring would not improve the agreement but have not tried this possibility to avoid too much parameter fitting in the calculation. The electron-electron interaction determines the energy difference between $\mu(1)$ and $\mu(2)$ at $B=0$. A small bump in $\mu(2)$ at $B \sim 2-3$ T is the signature of full polarization. A similar structure shows up in the experimental points but between 3-4 T. Interestingly however, the change in the electronic structure at $B \sim 8$ T is visible in the calculated addition energy $\mu(2)$.

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FIGURES

FIG. 1. Total energy of the \( N = 2 \) ring as a function of \( B \). The dashed line corresponds to an \( S_z = 0 \) gs, and the solid line to an \( S_z = 1 \) gs. The ring becomes fully polarized near \( B = 3 \) T.

FIG. 2. Several \( n = 0 \) sp energies for the \( N = 2 \) ring as a function of \( B \). The quantum labels \((n, l, \sigma)\) of the sp states are also indicated.

FIG. 3. Charge density strength function vs excitation energy (solid lines) for \( N = 2 \) and \( B = 0 \) to 15 T. The symbols represent the experimental peak energies\(^2\). The dashed line at \( B = 0, 1 \) and 2 T is the longitudinal spin density strength function.

FIG. 4. Energy of the more intense CDE’s as a function of \( B \) for \( N = 2 \). The dashed line represents the cyclotron frequency \( \omega_c \), and the solid lines are drawn to guide the eye. The thick symbols represent the experimental data\(^2\).

FIG. 5. Several \( n = 0 \) sp energies for the \( N = 1 \) ring as a function of \( B \). The lower energy state of each \((n, l)\) pair has spin up.

FIG. 6. Same as Fig. 3 for \( N = 1 \). The experimental data are from Ref. 20.

FIG. 7. Same as Fig. 4 for \( N = 1 \). The experimental data are from Ref. 20.

FIG. 8. Addition energies as a function of \( B \) (left vertical scale). The symbols are the experimental capacitance data\(^\\text{3, 4}\) (right vertical scale). Large dots correspond to \( N = 2 \), and small dots to \( N = 1 \).
$N = 2$

$S_z = 1$

$S_z = 0$
$\varepsilon_{n,l,\sigma}$ (meV)

$N = 2$

$B$ (T)
$N = 1$

- $\omega$ (meV)

- $B=0$

- $15 T$
- $14 T$
- $13 T$
- $12 T$
- $11 T$
- $10 T$
- $9 T$
- $8 T$
- $7 T$
- $6 T$
- $5 T$
- $4 T$
- $3 T$
- $2 T$
- $1 T$

- $\omega$ (meV)
