Impurity effects in few-electron quantum dots: 
Incipient Wigner molecule regime

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Abstract. – Numerically exact path-integral Monte Carlo data are presented for \( N \leq 10 \)
strongly interacting electrons confined in a 2D parabolic quantum dot, including a defect to break rotational symmetry. Low densities are studied, where an incipient Wigner molecule forms. A single impurity is found to cause drastic effects: (1) The standard shell-filling sequence
with magic numbers \( N = 4, 6, 9 \), corresponding to peaks in the addition energy \( \Delta(N) \), is
destroyed, with a new peak at \( N = 8 \), (2) spin gaps decrease, (3) for \( N = 8 \), sub-Hund’s rule
spin \( S = 0 \) is induced, and (4) spatial ordering of the electrons becomes rather sensitive to
spin. We also comment on the recently observed bunching phenomenon.

During the past few years, the electronic properties of 2D quantum dots have been the
subject of intense theoretical studies \cite{1,2}. A particularly interesting aspect of such artificial
atoms is the wide experimental tunability of the electron number \( N \) and the Brueckner
interaction strength parameter \( r_s \) in high-quality semiconductor dots \cite{3,4,5}. In particular, it has
been established both theoretically \cite{17} and experimentally \cite{8,9} that a “Wigner molecule” of
crystallized electrons forms already at surprisingly high densities corresponding to \( r_s \gtrsim 2 \) \cite{10}.
In this paper, we present numerically exact path-integral Monte Carlo (PIMC) \cite{11} simulation
results for \( N \leq 10 \) electrons in the most challenging incipient Wigner molecule regime, choosing
\( r_s \approx 4 \). Due to the breakdown of effective single-particle descriptions, this parameter
regime is notoriously difficult to treat within approximation schemes such as Hartree-Fock
theory \cite{1}, density functional theory \cite{10,12,13}, semiclassical \cite{14} or classical \cite{15}
approaches. Especially concerning disordered dots, in the absence of benchmark calculations, their
accuracy has so far been largely unclear.

The incipient Wigner molecule regime also exhibits unexpected and interesting physics, in
particular rather dramatic effects when including just one impurity in order to break rotational
symmetry. Below we shall demonstrate this in three different ways. First, despite the presence
of strong interactions, the clean system still exhibits shell structure, with exceptional stability of the
dot for “magic numbers” \( N = 4, 6, \) and \( 9 \). The stability is reflected in peaks in the
corresponding addition energy,

\[
\Delta(N) = E(N+1) - 2E(N) + E(N-1),
\]

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Table I – Quantum dot energies for fixed $N$ and $S$ at $k_B T = 0.125$ from PIMC simulations with and without impurity.

| $N$ | $S$ | $E_{imp}/\hbar\omega_0$ | $E_{clean}/\hbar\omega_0$ | $N$ | $S$ | $E_{imp}/\hbar\omega_0$ | $E_{clean}/\hbar\omega_0$ |
|-----|-----|------------------|-----------------|-----|-----|-----------------|-----------------|
| 1   | 1/2 | 0.51(1)          | 1.00            | 7   | 1/2 | 52.49(2)        | 53.71(2)        |
| 2   | 0   | 3.911(7)         | 4.893(7)        | 7   | 3/2 | 52.555(23)      | 53.80(2)        |
| 2   | 1   | 3.960(6)         | 5.118(8)        | 7   | 5/2 | 52.72(4)        | 53.93(5)        |
| 3   | 1/2 | 9.857(8)         | 11.055(8)       | 8   | 0   | 67.12(2)        | 68.52(2)        |
| 3   | 3/2 | 9.880(8)         | 11.050(10)      | 8   | 1   | 67.18(2)        | 68.44(1)        |
| 4   | 1   | 17.89(2)         | 19.104(6)       | 8   | 2   | 67.30(5)        | 68.51(5)        |
| 4   | 2   | 18.05(1)         | 19.34(1)        | 9   | 3/2 | 83.22(4)        | 84.45(6)        |
| 5   | 1/2 | 27.75(1)         | 29.01(2)        | 9   | 5/2 | 83.37(17)       | 84.61(17)       |
| 5   | 3/2 | 27.84(2)         | 29.12(2)        | 10  | 1   | 100.59(11)      | 101.96(14)      |
| 5   | 5/2 | 28.00(3)         | 29.33(2)        |       |       |                 |                 |
| 6   | 0   | 39.30(2)         | 40.53(1)        |       |       |                 |                 |
| 6   | 1   | 39.37(2)         | 40.62(2)        |       |       |                 |                 |
| 6   | 2   | 39.48(2)         | 40.69(2)        |       |       |                 |                 |
| 6   | 3   | 39.84(7)         | 40.83(4)        |       |       |                 |                 |

where $E(N)$ is the energy of the $N$-electron dot [5]. Strikingly, a single impurity is able to completely alter this shell structure. Only the filled-shell peak at $N = 6$ survives, and a new peak at $N = 8$ emerges. A similar peak has also been observed in recent experiments [6, 4]. A second example concerns the ground-state spin $S$, where a high value of $S$ for certain $N$ can lead to a “spin blockade” for transport through the dot [16]. The role of impurities on the spin polarization of interacting quantum dots has recently been studied by various groups, suggesting either a tendency towards spin polarization [17], depolarization [18], or strongly $N$-dependent behaviors without general trend [13]. As energy differences between different spin states are typically very tiny, approximations are particularly prone to predict false spin. We find that “spin gaps” between the ground state and the next-higher spin state are typically decreased by the impurity, indicating a trend towards polarization, but without change in the ground-state spin for the $N$ under study. However, there is a notable exception to this rule: For $N = 8$, the rare case of sub-Hund’s rule spin $S = 0$ is found in the presence of the impurity, while the clean dot has $S = 1$. Finally, as third example, we show that the electron crystallization pattern shows a strong dependence on total spin $S$, especially if rotational symmetry is broken. To give a typical example, for $N = 6$ electrons with $S = 0$, electrons basically arrange on a ring, but for the fully polarized $S = 3$ state, the amount of charge in the central region increases by about 40% compared to $S = 0$. Therefore, there is no clear separation of energy scales for spin and charge fluctuations. Such behavior is only found in the incipient Wigner molecule regime, and invalidates the commonly used lattice-spin models appropriate in the deep Wigner-crystallized limit [14].

We start by discussing the model underlying our work. A closed 2D quantum dot with parabolic confinement of frequency $\omega_0$ and an additional impurity potential has been studied. Measuring energy in units of $\hbar\omega_0$ and distance in oscillator lengths $l_0 = \sqrt{\hbar/m^*\omega_0}$, the Hamiltonian for the $N$-electron system at zero magnetic field is

$$H = \sum_{j=1}^{N} \left(\frac{1}{2} \mathbf{p}_j^2 + \frac{1}{2} \mathbf{r}_j^2 + V_{imp}(\mathbf{r}_j)\right) + \sum_{i<j}^{N} \frac{\lambda}{|\mathbf{r}_i - \mathbf{r}_j|},$$

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where $\lambda = l_0/a$ for effective Bohr radius $a$. Throughout this study, we set $\lambda = 4$, which puts the quantum dot into the incipient Wigner molecule regime with $r_s \approx 4$. Here $r_s$ is half the nearest-neighbor distance of the electrons measured in units of $a$. For GaAs-based dots, this corresponds to $\bar{\hbar}\omega_0 \approx 0.7$ meV \cite{8, 9}. We allow for an attractive impurity of strength $w$ centered at $x$. The impurity potential is taken in the form \cite{13}

$$V_{\text{imp}}(r) = -w \exp[-2(r-x)^2/\sigma^2], \quad (3)$$

with parameters chosen as $w = 4$, $x = (0, 1.5)$, and impurity range $\sigma = 0.75$. Qualitatively similar results were also observed for other parameter choices. For the above parameters, the impurity is close enough to the center to influence the electrons. Typically, when integrated over a circle of radius $\sigma$ around $x$, about $3/4$ of an electron charge is trapped by this impurity. However, the impurity location is also sufficiently far away from the center to break rotational symmetry.

Up to statistical error bars, PIMC data presented below are numerically exact. For $\lambda = 4$ and $k_B T = 0.125$, the fermion sign problem \cite{11} permits us to extract reliable results only for $N \leq 10$. The simulations yield accurate estimates, at given $N$ and total spin $S = (N_\uparrow - N_\downarrow)/2$, for the energy, the charge or spin density, and other correlation functions of interest. Typically, $5 \times 10^7$ MC samples were accumulated for each parameter set. Errors due to finite Trotter time discretization $\Delta \tau$ were carefully eliminated by extrapolating results for different discretization down to $\Delta \tau \to 0$ with a linear regression fit, using the fact that the expectation value of any Hermitian observable has finite-$\Delta \tau$ corrections vanishing $\propto \Delta \tau^2$ \cite{11}. This extrapolation procedure is essential for achieving high accuracy, as Trotter convergence is very slow except for fully spin-polarized clean dots \cite{20}. PIMC results for the spin-dependent energy are listed in table \cite{11}, where bracketed numbers denote one standard deviation error bar for the last digit, which includes stochastic MC errors and extrapolation uncertainties. When comparing our
data in table I to $T = 0$ results obtained from other methods such as exact diagonalization [21,22,23]. One has to take into account finite temperature, which can cause sizeable differences for not fully spin-polarized states. For instance, for two electrons and $S = 0$, PIMC predicts $E_{\text{clean}} = 4.893(7)$, while the $T = 0$ result is $E = 4.848$. Such differences arise because PIMC proceeds at fixed total $z$-component of the spin and therefore receives contributions from higher spin states [7]. Note that this effect is rather tiny and does neither affect the (finite-temperature) addition energies (1) nor the prediction of ground-state spins.

Let us first discuss the addition energies displayed in fig. 1a). Remarkably, in a clean dot, one can still observe the standard shell-filling sequence also found experimentally in small vertical dots [4,24]. The filled-shell configuration $N = 6$ and the half-filled cases $N = 4,9$ are exceptionally stable. The peaks in the addition energy at these magic numbers signal that the Wigner molecule is not yet fully established, since the classical Wigner limit is characterized by different and much less pronounced magic numbers [15]. By simultaneously monitoring the charge density (see below), however, clear traces of spatial ordering can be observed, reflecting the onset of Wigner crystallization. The energetic shell structure is then destroyed once the impurity is present, see fig. 1a). Apart from a new peak at $N = 8$, the addition energies $\Delta(N)$ now show a monotonic decrease with $N$. Only the filled-shell peak at $N = 6$ is still visible, yet considerably diminished. We note that the peak at $N = 2$ is always absent for this interaction strength [1,6].

Next we address spin properties in this regime, see table I and fig. 1b). Except for $N = 8$, the spin $S$ was not altered by the impurity [25]. While $S$ mostly takes the minimal value ($S = 0$ or $S = 1/2$), for $N = 4$ ($N = 9$), we have Hund’s rule spin $S = 1$ ($S = 3/2$). Furthermore, for $N = 10$, also a partially spin-polarized state $S = 1$ is found. For the clean $N = 3$ dot, at $\lambda_c \approx 4.34$ a spin transition $S = 1/2 \rightarrow 3/2$ occurs at $T = 0$ [7,13,22]. We
are already very close to this transition point, and within error bars, we cannot decide the ground state spin. The case \( N = 8 \) is particularly noteworthy. For the clean dot, the spin is \( S = 1 \) according to Hund’s rule, but the \( S = 2 \) state is actually very close, see table I. Strikingly, with impurity, we then find sub-Hund’s rule spin \( S = 0 \). Since this effect requires sufficiently strong interactions (low density), for a weakly disordered dot with \( N = 8 \), one can expect a transition from the usual Hund’s rule \( S = 1 \) ground state at high density (small \( r_s \)) to the unpolarized \( S = 0 \) state at low density. Since for \( N = 9 \), we have spin \( S = 3/2 \), this transition could be observed experimentally as spin blockade for the 9th electron to enter the dot. By monitoring spin-spin correlations between the electrons at the impurity location and the central region (data not shown here), strong antiferromagnetic couplings are visible. Hence it is likely that local moment formation at the impurity location and in the central region is important for this sub-Hund’s rule behavior. This is also confirmed by noting that charge and spin densities do not follow each other, as they do in the deep Wigner-molecule regime \[7, 23\]. Since except for \( N = 8 \) the spin \( S \) is not changed, the impurity’s overall effect on the spin polarization seems small. Nevertheless, our results show a decreasing “spin gap” \( \delta E(N) = E(S+1) - E(S) \), see fig. I(b), indicating a weak tendency towards polarization. Note that the spin excitation energies are very small, again stressing the need for highly accurate calculations.

We now turn to the onset of electron crystallization in real space. Figure 2 shows density plots for \( N = 7 \) to 10 electrons in their respective ground-state spin configuration. At the defect, the density is about three times larger than elsewhere in the dot. Although there are large quantum fluctuations, a spatial shell structure is already discernible. The classical prediction for clean dots at these electron numbers is as follows \[14\]. For \( N = 7 \) and \( N = 8 \), just one electron is in the center, while for \( N = 9 \) and \( N = 10 \), the spatial filling is \((2-7)\) and \((2-8)\), respectively, with the configurations \((1-8)\) and \((3-7)\) very close in energy.
Quantum-mechanically, for $\lambda = 4$, the radial ordering is not yet very pronounced in the clean dot $[7, 26]$. With impurity, however, our data in fig. 2 displays spatial shells, although with a rather different filling sequence. Detailed examination of the central region shows that only one to two electrons are accommodated in this part. Therefore incoming electrons enter the outer ring for $N = 7, 8, 9$, and 10, with the ring expanding and, for $N = 10$, nearly embracing the impurity. While for very large $r_s$, the crystal structure is $S$-independent and effective Heisenberg-type lattice models for the spin physics can be constructed, for $r_s \approx 4$, the spatial charge ordering is strongly dependent on total spin $S$; see also $[23, 26]$. This is seen in fig. 3 for $N = 6$, where the densities for $S = 0$ and $S = 3$ are compared, both with and without the defect. Interestingly, for $S = 0$, electrons tend to basically arrange on a ring, while for the spin-polarized case, the classical configuration with one electron in the center $[15, 26]$ is approached. This effect, in particular concerning the influence of the impurity, can be quantified by computing the fraction $n_c$ of electron charge localized within a radius $\sigma = 0.75$ around the center of the dot. The result is shown in fig. 4 and illustrates that more and more charge goes into the central region when $S$ is increased. For the clean dot, comparing $S = 0$ and $S = 3$, this amounts to a $\approx 25\%$ increase in $n_c$, but with the defect, this increase is $\approx 40\%$. That implies that the spin sensitivity of the incipient Wigner crystallization process is enhanced by the impurity, probably due to a local spin at the impurity site.

Finally, let us briefly discuss the relation of our results to the experimentally observed “bunching” phenomenon found in the Wigner molecule regime $[9, 27]$, where the addition energy was found to vanish at certain values of $N$. Bunching was also reported in a recent numerical diagonalization study for $N = 7$ and $N = 8$ $[28]$, where the dot was modeled by a $3 \times 4$ Hubbard-type model containing only short-range interactions. In contrast to these results, for the more realistic continuum Hamiltonian $[2]$ with long-range interactions, bunching is not observed for $N < 10$, see fig. 1b). Since bunching is believed to be a generic effect $[27]$, a consistent explanation probably has to assume strong disorder or other mechanisms not contained in our model.

To conclude, we have presented a PIMC study of impurity effects in 2D quantum dots. We have focused on the most difficult yet most interesting regime of an incipient Wigner molecule. Large effects are caused already by a single impurity. When the rotational symmetry of the clean dot is broken, magic numbers for the energetic shell-filling sequence, observed as peaks in the addition energy, are changed. Instead of the usual peaks at $N = 4, 6, 9$, with impurity only a (weak) filled-shell peak at $N = 6$ and a new peak at $N = 8$ are observed. In addition,
for \( N = 8 \), we find a sub-Hund’s rule spin \( S = 0 \). Finally, the incipient Wigner crystallization process was found to be strongly spin-dependent, in particular when the impurity is present. We hope that these results will also be valuable as reference data to assess the validity of computationally less demanding yet approximate calculations.

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