Interaction-driven distinctive electronic states of artificial atoms at the ZnO interface

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Abstract
We have investigated the electronic states of planar quantum dots at the ZnO interface containing a few interacting electrons in an externally applied magnetic field. The electron–electron interaction effects are expected to be much stronger in this case than in traditional semiconductor quantum systems, such as in GaAs or InAs quantum dots. In order to highlight that stronger Coulomb effects in the ZnO quantum dots, we have compared the energy spectra and the magnetization in this system to those of the InAs quantum dots. We have found that in the ZnO quantum dots the signatures of stronger Coulomb interaction manifests in a unique ground state that has very different properties than the corresponding ones in the InAs dot. Our results for the magnetization also exhibits behaviors never before observed in a quantum dot for a realistic set of parameters. We have found a stronger temperature dependence and other unexpected features, such as paramagnetic-like behavior at high temperatures for a quantum-dot helium.

Keywords: quantum dots, zinc oxide heterojunction, magnetization

For decades, creation of high-mobility two-dimensional electron gas (2DEG) in quantum confined semiconductor heterojunctions has paved the way for present-day electronics and quantum devices and was crucial for many seminal discoveries in correlated electron systems in a magnetic field. The most notable example was the fractional quantum Hall effect [1] and several other unique phenomena in various nanoscale systems, viz. the quantum dots (QDs) (or, the artificial atoms) [2–4] and quantum rings (QRs) [5, 6]. Similar phenomena have also been investigated in Dirac materials such as graphene [7, 8] and other graphene-like materials, such as silicene [9], germanene [10, 11] and black phosphorous [12, 13]. In recent years, very exciting developments have taken place with the creation of high-mobility 2DEG in heterostructures with insulating complex oxides. Unlike in traditional semiconductors, electrons in these systems are strongly correlated [14]. These should then exhibit effects ranging from strong electron correlations, magnetism, interface superconductivity, tunable metal-insulator transitions, among others, and of course, the exciting possibility of all-oxide electronic devices. Interestingly, odd-denominator fractional quantum Hall states were discovered in MgZnO/ZnO heterojunction [15], and surprising results were found for the even-denominator states [16, 17], and in a tilted magnetic field [18]. Preparation of various nanostructures in ZnO, such as nanorings, nanobelts, etc have been reported [19, 20]. It has been stressed there that electron correlation effects are strong in these systems due to the increased electron effective mass and reduced dielectric constant of ZnO. Given the enormous potential of this newly developed source of 2DEG, it is therefore imperative that the electronic properties of quantum confined systems at the oxide interfaces are thoroughly understood.

Here we report on our studies of the electronic states of artificial atoms in this new planar electron system with the hope that the strong correlation effects that are expected at the ZnO interface will manifestly alter the electronic states in...
second and three-electron QDs are present. It is clear from the figure that without the magnetic field for the ZnO QD, the energy values are lower and the levels are closer to each other due to the larger value of electron effective mass. For the ZnO QD the ground state is \( l = 0 \), \( s = -1/2 \) for all the values of magnetic field. In contrast to that, the ground state for the InAs QD has \( l = 0, s = 1/2 \). For the excited states many level crossings are visible. For the ZnO QD these crossings are shifted to lower values of the magnetic field.

At zero temperature the magnetization \( M \) of the QD is defined as \( M = -\frac{\partial E_0}{\partial B} \), where \( E_0 \) is the ground state energy of the many-electron system [24]. Here we report on our studies of the magnetic field dependence of \( M \) by evaluating the expectation values of the magnetization operator \( \mathbf{m} = -\frac{\partial \mathcal{H}}{\partial \mathbf{B}} \), where \( \mathcal{H} \) is the system Hamiltonian (1). We then need to evaluate the expectation values of magnetization operator \( \mathbf{m} \) using the wave functions of the interacting many-electron system. We have also studied the temperature effect on magnetization, following the thermodynamical model discussed in [30]. The
The temperature dependence of magnetization is evaluated from the thermodynamic expression \[ \mathcal{M} = -\sum_i \frac{\partial E_i}{\partial B} e^{-E_i/kT} \sum_i e^{-E_i/kT}, \] where the partial derivatives are evaluated as expectation values of operator \( \mathbf{\hat{r}_i} \) for the interacting state \( i \).

For a better understanding of the obtained results we have also calculated the electron densities for few electron states in the QD.

In figure 2, several low-lying energy levels for the ZnO and InAs QDs with two electrons are presented against the magnetic field for various values of total angular momentum \( L \). These figures clearly indicate that in the absence of the magnetic field for the ZnO QD, the energy values are lower and the levels are closer to each other due to the larger value of the electron effective mass in the former case. It is well known that for small values of the magnetic field the ground state of a two-electron QD is a singlet state with total angular momentum \( L = 0 \) and spin \( S = 0 \). With an increase of the magnetic field a singlet-triplet transition of the ground state is observed. For the InAs QD this transition occurs at \( B = 2.4 \) T and the ground state changes to triplet state with \( L = 1 \), \( S = 1 \). For the ZnO QD we can observe a similar transition but for a much smaller value of the magnetic field, \( B = 0.55 \) T which will change the ground state to the triplet state with \( L = 1 \), \( S = -1 \). The spin difference of the two triplet ground states can be explained by the sign of the g-factor in the two cases. Due to the strong Coulomb interaction, with further increase of the magnetic field, a second ground state transition is observed for the ZnO QD at \( B = 4.2 \) T which changes the ground state to \( L = 3 \), \( S = -3 \). This transition does not occur in the case of the InAs QD for experimentally observable ranges of the magnetic field.

In figure 3, the dependence of the low-lying energy levels of the ZnO and InAs QDs with three interacting electrons are presented against the magnetic field for various values of the total angular momentum \( L \). Usually in QDs for small values of the magnetic field the three electron ground state has total angular momentum \( L = 1 \) and total spin \( S = 1/2 \) which was observed by many authors in the case of InAs, GaAs and other QDs. But our present study indicates that due to the strong Coulomb correlation effects in ZnO QD, for small values of the magnetic field the three-electron ground state has the total angular momentum \( L = 0 \) and total spin \( S = -3/2 \). With the increase of the magnetic field, at \( B = 1.3 \) T again a ground state transition can be observed to the state with \( L = 3 \) and \( S = -3/2 \). Similar ground state transition is also observed for InAs QD with three electrons, but for a larger value of the magnetic field, viz., at \( B = 3.4 \) T. The reason of these interesting results is the interplay of three most important parameters of ZnO: effective mass, Lande factor and the dielectric constant. In order to be sure that the observation of the new ground state in ZnO QD is not accidental and does not depend on the size of the QD we have recalculated these results also for larger QDs (figures 3(c) and (d) with \( h_0 = 1 \) meV and \( h_0 = 7 \) meV) and for smaller QDs (figures 3(e) and (f) with \( h_0 = 2 \) meV and \( h_0 = 10 \) meV).
with $\hbar\omega_{\text{ZnO}} = 2\,\text{meV}$ and $\hbar\omega_{\text{InAs}} = 10\,\text{meV})$. These results indicate that the new three-electron ground state with angular momentum $L = 0$, observed for ZnO QD, always occurs also for other dot sizes, but never occur for the InAs QD. This suggests that the main reason of this new ground state is the strong coulomb interaction in ZnO. These interesting results will manifest themselves in optical and magnetic characteristics of the ZnO QDs. In particular, we have considered here the magnetization of the ZnO QD with few electrons and compared our results with similar ones for the InAs QD.

In figure 4 the magnetization of the ZnO and InAs QDs with two electrons are presented against the applied magnetic field for various values of the temperature from 0.1 to 4 K. Similar results but for three-electron ZnO and InAs QDs are presented in figure 5. In all cases, the magnetic field dependencies of the magnetization at $T = 0.1\,\text{K}$ have step like behaviors. These jumps in magnetization can be explained by the ground state oscillations of few electron QDs [23]. In case of the two-electron ZnO QD (figure 4(a)) there are two jumps in the magnetization for magnetic fields $B = 0.55\,\text{T}$ and $4.2\,\text{T}$. This result is to be contrasted to the case of the two-electron InAs QD, where only one jump is observed at $B = 2.4\,\text{T}$. Also it should be noted that the magnetization of ZnO QD has a very strong temperature dependence as compared to that of the InAs quantum dots. Due to the large value of the electron effective mass in ZnO the excited energy levels of the few electron QD are very close to ground state and the increase of temperature populates these states that explains the smoothening and averaging of the magnetization curves. Furthermore, for $T = 4\,\text{K}$ a surprising paramagnetic-like behavior of the magnetization is observed for the two-electron ZnO QD. In contrast to the ZnO QD, the magnetization curves of the InAs QDs have a weak temperature dependence (figure 4(b)). Similar behaviors are also observed for three-electron ZnO and InAs QDs (figures 5(a) and (b)). Here again a step like behavior of the magnetization is observed for both QDs. At zero temperature the first jump is caused by the lifting of fourfold degeneracy of the ground state due to the magnetic field. The second jump for the ZnO QD at $B = 1.4\,\text{T}$ is caused by the change of the ground state.

In figure 6 the ground state density is presented for two- and three-electron ZnO QDs and InAs QDs for various values of the magnetic field. In a InAs QD, the electrons are mostly located in the central part of the dot, while for the ZnO QD, due to weaker confinement and stronger Coulomb interaction, electrons are repelled from the central part of the dot. With an increase of the magnetic field the ground state electron densities for the ZnO QD and the InAs QD exhibit completely different behaviors which can be explained by the different ground state changes discussed above. For example, at $B = 0$ the two-electron ground state for both QDs is with $L = 0$, but at $B = 5\,\text{T}$ the ground state for the ZnO QD has $|L| = 3$ and for the InAs QD $|L| = 1$. For three electron QDs the ground states are different even at zero magnetic field.

To summarize, we have presented here our studies of the electronic states and magnetization of the ZnO quantum dot with few interacting electrons and compared our results with similar ones for the InAs QDs. Our results have been compared with the results for the InAs QDs. We have shown that electron–electron interaction exerts a very strong influence on the electronic states and on magnetization of the ZnO QD. In particular, the energy levels of a two-electron ZnO QD display more level
crossing for finite values of the magnetic field. Additionally, 
in the case of the three-electron QD, even in the absence 
of the magnetic field the ground state is changed to \( L = 0, \) 
\( S = -3/2, \) as compared to the usual state at \( |L| = 1, \) and 
\( S = \frac{1}{2}, \) that is found, for example, for the InAs QD for a 
realistic set of parameters (size, confinement potential, effective 
mass, etc) which is in good agreement with the experimental 
results. These interesting and unexpected results will manifest 
itself in optical and magnetic characteristics of the ZnO QDs. 
Further, we have shown that the magnetization curves of the 
ZnO QDs have the expected step-like behavior, but in contrast 
to the InAs QDs, the corresponding jumps in magnetization 
are observed for much smaller values of the magnetic field. 
Therefore the ZnO QDs are suitable for low-field magnetiza-
tion measurements. The magnetization of the ZnO QDs has a 
very strong temperature dependence, and surprisingly, at high 
temperatures, the two-electron ZnO QD shows a paramagn-
etic-like behavior.

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