Photoionization and Far Infrared Absorption of Single-Electron Transistors: Theoretical Results and Experiment Proposals

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We present theoretical results demonstrating that photoionization and far infrared absorption can be valuable tools to investigate single-electron transistors (SETs), and encourage experimentalists to use them to study nano-electronic devices. Photoionization permits to obtain information on how many electrons occupy a quantum dot and the charging energy in a direct manner. This is very important, because experiments carried out up to now, which measure the electric conductance, only allow to determine these quantities indirectly. It is worth emphasizing that in the photoionization processes considered by us, an electron absorbs a photon with energy of the order of the work functions (typically, \(\sim 1\) eV) and is ejected into the vacuum. This phenomenon is completely different from the widely studied photo-assisted tunneling considered by previous investigators, involving much lower photon energies (\(\sim 1\) meV). We give concrete suggestions on how to conduct experiments using photoionization alone or in combination with transport measurements. Monitoring zero kinetic energy (ZEKE) photoelectrons is especially advisable in view of the high-resolution of the ZEKE spectroscopy. We also suggest that far infrared absorption can be an even simpler method that photoionization to investigate SETs. [DOI: 10.1380/ejssnt.2010.1]

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I. INTRODUCTION

In the attempt to rationalize the impressive data available in many-electron systems, the single-particle picture turned out to be a very useful concept. It is based on the assumption that each electron moves independently in a average self-consistent field (SCF) created by the other electrons. The single-particle description is excellent in ordinary atoms, but it often fails in molecules, as revealed by well-documented studies on photoionization [1–3]. Isolated quantum dots (QDs) are often said to behave as “artificial” atoms [4]. Because the electron motion is confined within sizes \(R\) of the order of nanometers, electrons in QDs possess quantized levels \(\varepsilon_a = -A_a/R^2\), which are well separated energetically, similar to electrons in ordinary atoms. For sufficiently small sizes, the energy separation exceeds the thermal energy \(\Delta\varepsilon \gg k_B T\), and for many purposes one can restrict oneself to a single (nondegenerate) level, the uppermost (partially) occupied level \(\varepsilon_d\). This is the basic assumption of Anderson single-impurity model, to which we restrict ourselves throughout. Experimentally, the energy \(\varepsilon_d\) can be easily tuned by means of a “plunger” gate potential \(V_g\) [5, 6].

In assembled QDs, the important part played by electron correlations (i. e., departure from the SCF approximation) was demonstrated: experimentally, e. g., by the observation of a metal-insulator transition [7], and theoretically, e. g., by studies on photoionization [8–10] and photoabsorption [10–12].

The single-particle picture may break down even in a single QD because of the electron-electron interaction, which arises in the case where the level is occupied by two electrons of opposite spins. This can be quantitatively characterized by the the charging energy \(U\), which represents the energy required to bring an extra electron on a system where another electron is already present.

In extended systems, the charging energy is vanishingly small. Within classical electrostatics, \(U\) can be expressed in terms of the dot capacitance \(C\) as \(U = \varepsilon^2/(2C) \times 1/R\). For even one picofarad, of the order of the smallest capacitance values available for general use in electronic design of integrated circuits, \(U\) is of the order \(U \approx 0.1\mu eV\), which is three orders of magnitude smaller than the thermal energy for an electronic device operating at temperatures \(T \sim 1K\).

In a single-electron transistor (SET), a prototypic nanoelectronic device, a small quantum dot (QD) is weakly coupled via tunneling junctions to metallic electrodes. Because QDs are small (\(\sim nm\)), their charging energy is considerably larger. For the semiconducting QDs embedded into the SETs, \(U\) is of the order of a few meV [5, 6, 13]. Smaller metallic QDs are characterized by even higher values, e. g., \(U = 0.34\mu eV\), see Ref. 14. In the situations of interest, the dot charging energy \(U\) is larger than both the thermal energy \(k_B T\) and the finite level width \(\Gamma \sim t_d^2/D\), where \(t_d\) is the dot-electrode hopping integral and \(D\) the electrode bandwidth.

Within a single-particle picture, the zero-bias conductance \(G\) of a SET would exhibit two (Coulomb blockade) peaks of width \(\sim \Gamma\) located at the \(\varepsilon_d\)-values tuned such that the energies of the lower and upper Hubbard bands become resonant with the Fermi energy \(\varepsilon_F\) of the electrodes (\(\varepsilon_d = \varepsilon_F\) and \(\varepsilon_d + U = \varepsilon_F\), respectively). This behavior is observed only at higher temperatures \((T > T_K)\). It does not hold anymore below the Kondo temperature \(T_K\). There, instead of the Coulomb blockade peaks, the zero-bias conductance exhibits a Kondo plateau, where it reaches the unitary limit (perfect transmission): \(G(\varepsilon_d) \approx G_0 \equiv 2e^2/h\) for \(\varepsilon_F - U + \Gamma < \varepsilon_d \leq \varepsilon_F - \Gamma\) [5, 6, 15]. The Kondo effect is a manifestation of strong electron corre-
lations. According to the widest accepted interpretation, it appears in the case where the dot contains an unpaired electron \((n_d = 1\) for the Anderson model), whose spin is screened out by other electrons belonging to an extended Kondo cloud formed around it.

Besides \(U\), the energy \(\varepsilon_d\) of the dot level plays a key role in a SET, because it controls the dot occupancy \(n_d\), which, according to the above interpretation, is related to the Kondo plateau. Unpleasantly, neither \(n_d\) nor \(U\) or \(\varepsilon_d\) can be directly extracted from the \(G\)-data. The determination of \(\varepsilon_d\) and \(U\) also requires the conversion factor \(\alpha\), which relates \(\varepsilon_d\) to the gate potential \(V_g\)

\[
\varepsilon_d = \alpha V_g + \text{const.} \quad (1)
\]

and which, as noted above, is affected by significant errors [16]. Even worse, \(n_d\) cannot at all be extracted from dc-transport data, and therefore inferring the formation of the Kondo cloud around an unpaired electron occupying the dot from the \(G(\varepsilon_d)\)-plateau only remains based on an interpretation without direct experimental support.

For the completeness of the physical description, in addition to dc-measurements, experimental tools able to provide direct information on the aforementioned quantities would be highly desirable. As we recently showed [17, 18], photoionization and photoabsorption can be employed to this aim, and this fact will be emphasized in the present work.

As mentioned above, photoionization and photoabsorption represent methods that turned out to be sensitive to electron correlations in various systems. In the present work, theoretical results are presented showing that they can also be valuable tools, to be used alone or in combination with dc-transport, to investigate single electron transistors (SETs), considered here as one of the simplest devices of interest for correlated molecular or nanoelectronics. The results we are going to present are similar to or based on those of Refs. 17 and 18.

The advantage of these methods is that they permit to determine the parameters relevant for SETs in a direct manner, and not indirectly, as done in dc-transport measurements.

II. THEORETICAL RESULTS FOR PHOTOIONIZATION

The theoretical results presented below have been obtained by exact numerical diagonalization of finite clusters described within the Anderson model Hamiltonian \(H\). Details can be found in Refs. 17–19 and will therefore be omitted here.

We shall first discuss the photoionization in more detail, and then briefly consider the photoabsorption. To avoid confusions, we note that the photoionization we consider amounts to eject an electron from the QD into vacuum, which is completely different from the photon-assisted tunneling through a QD discussed by previous investigators. In the latter, a photon of low energy \((\sim U)\) helps an electron to pass through the dot.

In the photoionization processes considered here, an electron of the dot absorbs a photon and is emitted into the vacuum. This brings the SET from its neutral ground state \(\Phi\) (case \(T = 0\)) into one of its excited ionized eigenstates \(\Psi_k\). Each ionization process is characterized by an ionization energy threshold \(\omega_k\) and a spectroscopic factor \(f_k\). The former represents the difference between the energies of the ionized and the neutral states, \(\omega_k = \langle \Psi_k | H | \Psi_k \rangle - \langle \Phi | H | \Phi \rangle\). This amounts to consider zero kinetic energy (ZEKE) photoelectrons, the case on which we focus here, because the resolution of the ZEKE-spectroscopy is superior to that of conventional photoemission, and a high resolution is important for the proposed experiments (see below). The spectroscopic factor is defined by

\[
f_k = | \langle \Psi_k | d_\sigma | \Phi \rangle |^2 \quad (2)
\]

and is important because it is related to the intensity of the ionization signal, more precisely, to the partial-channel ionization cross section. Above, \(d_\sigma\) is the annihilation operator of an electron of spin \(\sigma\) on the dot. From Eq. (2), the following exact sum rule can be deduced

\[
\sum_k f_k = \langle \Phi | d_\sigma^\dagger d_\sigma | \Phi \rangle \equiv n_{d,\sigma} = n_{d}/2. \quad (3)
\]

From the present point of view, Eq. (3) is very important: in the left hand side, the integrated ionization intensity (a quantity that is accessible experimentally) provides direct information on the dot occupancy \(n_d\).

Let us briefly consider the dot ionization in the limit of weak QD-electrode coupling \((t_d \to 0)\). The dot is doubly occupied \((n_d = 2)\) for \(\varepsilon_d - \varepsilon_F < -U\), singly occupied \((n_d = 1)\) for \(-U < \varepsilon_d - \varepsilon_F < 0\), and vacant \((n_d = 0)\) for \(\varepsilon_d - \varepsilon_F > 0\). In the first case, the photoionization occurs if the photon energy \(\omega\) is tuned to the upper Hubbard band, \(\omega = \omega^U_0 \equiv -\varepsilon_d - U\), while in the second case if it is tuned to the lower Hubbard band, \(\omega = \omega^L_0 \equiv -\varepsilon_d\).

The relevant results for photoionization obtained for nonvanishing \(t_d\) are collected in Figs. 1 and 2. The curve for the dot occupancy visible in Fig. 1, which according to Eq. (3) is nothing but the integrated ionization intensity, shows that \(n_d\) computed exactly for \(t_d \neq 0\) significantly differs from that in the limit \(t_d \to 0\) described above only in the narrow mixed valence regions of widths \(\sim \Gamma\) around \(\varepsilon_d \approx \varepsilon_F\) and \(\varepsilon_d \approx \varepsilon_F - U\).

But photoionization experiments can provide more information than the integrated intensity. Photons with tunable frequency \(\omega\) can be used to bring the nanosystem from its neutral ground state to various ionized eigenstates. Typical results of this kind, which can be seen in Fig. 2 of Ref. 19, look complicated, with numerous curves and avoided crossings. The latter appear because, instead of true intersections, the curves for exact quantum eigenstates exhibit avoided crossings. An example of avoided crossing is visible on the curves of Figs. 1 and 2 around \(\varepsilon_d \approx \varepsilon_F\).

Fortunately, out of the numerous ionized states with nonvanishing spectral weights, only two diabatic ionized states are of experimental relevance (see Ref. 19 for details). Roughly, they correspond to the separate ionization from the lower \((\omega = \omega_k \approx \omega^L_0 \approx -\varepsilon_d)\) and the upper \((\omega = \omega_k \approx \omega^U_0 = -\varepsilon_d - U)\) Hubbard bands. The curves for the corresponding spectroscopic factors \(f_{\omega_k}\) are shown in Fig. 1 as a function of the dot energy \(\varepsilon_d\), while the ionization energies \(\omega^\pm_1\) are presented in Fig. 2. Similar to the
In Ref. 17, we proposed several ways to conduct experiments for investigating SETs by photoionization. The analysis presented below is complementary to that of Ref. 17 and mainly emphasizes aspects relevant for the proposed experiments. As a rule, the experiments should be performed in the following way. For a given value of the gate potential $V_g$, the dot should be shone by a flux of monochromatic photons of tunable frequency $\omega$. By progressively increasing $\omega$ starting from a sufficiently low value, one should monitor the point $\omega = \omega(V_g)$ where photoelectrons of zero kinetic energy will be emitted. In view of the analysis of Sec. II, this value $\omega(V_g)$ would be either the ionization energy $\omega_d$ or $\omega_l$. To discriminate between the ZEKE-photoelectrons emitted from the dot and those ejected from electrodes is important but fortunately easy [17]: the signals related to the latter do not depend on the gate potential and should be disregarded, while those related to the former do depend and should be analyzed.

One should start with a sufficiently positive gate (sufficiently low $\varepsilon_d$), such that the dot is doubly occupied and sufficiently away from the mixed-valence situation. By making the gate potential more and more negative ($\varepsilon_d$ increases), the ionization intensity (the number of ZEKE-photoelectrons) $I_u \propto f_u$ remains nearly constant, and reaches a plateau value $I_u \simeq \bar{T}_u$ ($f_u \simeq \bar{T}_u \simeq 1$, see Fig. 1) up to the point where the mixed valence point $V_g,2$ that separates the double occupied and single occupied dot is approached. In this process, the recorded ionization energy $\omega(V_g) \simeq -\varepsilon_d - U$ decreases linearly with decreasing $V_g$. By choosing two points I and J in this range (see Fig. 2), one can use Eq. (1) to determine the conversion factor as $\alpha = \frac{\omega(V_g^I) - \omega(V_g^J)}{(V_g^I - V_g^J)}$, because all the quantities entering the expression of the r.h.s. can be experimentally measured.

By subsequent decrease of $V_g$, the mixed-valence regime will be reached ($V_g = V_{g,2}$), where two ionization signals should be detectable, one that rapidly falls off $I_u \propto f_u$, another that rapidly increases $I_l \propto f_l$. From the width $\delta V_{g,2}$ of this mixed-valence region one can extract information on the hybridization parameter $\Gamma \sim |\alpha|\delta V_{g,2}$.

Further on ($V_g < V_{g,2}$), the intensity of the second ionization signal increases and reaches a constant value $I_l \simeq \bar{T}_l$ ($f_l \simeq \bar{T}_l \simeq 1/2$), which represents half the value $\bar{T}_u$. This $f_l$-plateau, which overlaps with the Kondo plateau of the zero-bias conductance $G$, will last until one draws near the other mixed-valence regime $V_g \approx V_{g,1}$, which separates the single occupied and the vacant dot. Away from the mixed-valence situations, the recorded ionization energy should be now $\omega(V_g) \simeq -\varepsilon_d$. Again, one can choose two points L and M in this range (see Fig. 2), and use Eq. (1) to determine the conversion factor as

$$\alpha = \frac{\omega^M - \omega^L}{V_{g,1}^M - V_{g,1}^L}.$$ (4)

From the width $\delta V_{g,1}$ of this mixed-valence regime one can similarly estimate the hybridization parameter $\Gamma \sim |\alpha|\delta V_{g,2}$. Beyond $V_{g,1}$ the ionization intensity becomes vanishingly small, which indicates a negligible dot occupancy.

Once $\alpha$ is known, the charging energy can be calculated as

$$U = \alpha (V_{g,1} - V_{g,2}).$$ (5)

Alternatively, one can deduce $U$ directly from the linear extrapolations of the two ionization energies, as indicated at the end of Sec. II. So, one can even make a self-consistency test.

Most importantly, the observation of a ratio between the two plateau intensities of $\bar{T}_u/\bar{T}_l \simeq 2$ would represent the clearest evidence that the dot occupancy in the first plateau is twice the occupancy of the second plateau.
Such a behavior would give direct support of the current physical interpretation of the Kondo effect in SETs, which assigns the Kondo plateau in the zero-bias conductance to an unpaired electron on the dot, which polarizes the surrounding electrons of a Kondo cloud acting to screen out the spin of the lone electron.

The most challenging task in the experiment proposed above is probably to detect the ZEKE-photoelectrons. Therefore, for the determination of the SET parameters we have also put forward an alternative method [17], which consists of simultaneous measurements of the dc-transport and photoionization. The experimental procedure is analogous to that described above, but, in addition, a constant low source-drain voltage should be always kept applied on the SET, and the gate voltage should be chosen in the range $V_{g,1} < V_g < V_{g,2}$. The values $V_g = V_{g,1}$ and $V_g = V_{g,2}$ can be extracted from a standard dc-transport experiment, by monitoring the location of the ends of the Kondo plateau (for for temperatures $T < T_K$). (Alternatively, one can examine the location of the Coulomb blockade peaks for $T \gtrsim T_K$).

Again, for a given value of the gate potential $V_g$ the dot should be irradiated with a well focused monochromatic flux of photons, whose frequency should be gradually increased up to the frequency value $\omega = \omega(V_g')$ where ionization occurs. What makes the difference from the experiment discussed above is that, instead of monitoring the emitted ZEKE-photoelectrons, one should monitor the source-drain current $J$. The reason is that, when the (single) electron occupying the dot in the Kondo regime is removed, the prerequisite for the occurrence of the Kondo effect disappears, and, consequently, $J$ should exhibit a drop, which can be evidenced by performing time-resolved dc-measurements. In this way, one would be able to get the curve $\omega_l(V_g)$ of Fig. 2 corresponding to the ionization from the lower Hubbard band. Form this curve, one can deduce the conversion factor and the charging energy from Eqs. (4) and (5), respectively.

IV. FAR INFRARED ABSORPTION

One important issue in the context of the proposed experiments of SET-photoionization is that the ionization energies, which are of the order of work functions ($\sim 1$ eV), vary within ranges of the order $U \sim 1$ meV [5, 6, 13]. Achieving such a high experimental resolution may be challenging.

Therefore, as an alternative method to photoionization, and less demanding than it, we have recently suggested to experimentally investigate SETs and other nano electronic devices by far infrared (FIR) absorption [18]. Calculations show that in the Kondo regime $\varepsilon_F - U \lesssim \varepsilon_d \lesssim \varepsilon_F$ ($V_{g,1} \lesssim V_g < V_{g,2}$) two photoabsorption signals are important [18]. Away from the mixed-valence regimes, these absorption frequencies are well approximated by $\omega_u \approx \varepsilon_F - \varepsilon_d$ and $\omega_u \approx \varepsilon_d + U - \varepsilon_F$. From the measurements of these absorption frequencies one can determine the SET-parameters $U$, $\alpha$, and $\Gamma$, analogous to the manner of using the frequencies $\omega_l$ and $\omega_u$ in the case of photoionization.

In particular, we note that the particle-hole symmetric point ($\varepsilon_d = \varepsilon_F - U/2$), which corresponds to the middle of the Kondo plateau $[V_g = V_g^* \equiv (V_{g,1} + V_{g,2})/2]$, the two absorption frequencies are degenerate, and their common value is $\omega^* = U/2$. Therefore, the easiest way to deduce the charging energy is just from this value ($U = 2\omega^*$).

As can be seen from the above expressions, the photoabsorption frequencies are of the order of the charging energy. In view of the typical values $U \sim 1$ meV [5, 6, 13], one should utilize FIR radiation. Importantly, carrying out FIR experiments on SETs should be less demanding than photoionization. What one has to measure here are photon frequencies in the FIR-range (and this can be done with reasonable accuracy), a much easier task than to measure frequencies of the order $\sim 1$ eV with an accuracy $\sim 1$ meV.

In the end of this section we mention that, similar to the case of photoionization, instead of measuring FIR-absorption intensities, one can monitor the dc-current in a combined FIR-absorption–dc-transport experiment.

V. CONCLUSION

In conclusion, in this work we have presented results demonstrating that the photoionization and the far infrared absorption represent valuable tools, which can be used alone or in combination with dc-transport measurements, to investigate SETs and other correlated nanoelectronic devices.

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