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Study of the two-step tunnelling from a quantum object

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Recent studies of advanced materials have succeeded in obtaining a whole new class of composites such as nanometre sized particles [1] and structures [2] with significant potential for field emission (FE) applications. While bulk emissive materials, such as metals and in most cases semiconductors exhibit smooth upward current-voltage (I-V) characteristics, FE from nanostructured materials often exhibit a behaviour that can be traced back to the quantum confinement conditions of electrons residing in such cathodes. There are two basic types of FE peculiarities reported so far from these structures: (i) the resonant tunnelling, where electrons with some resonant values of the energy go ballistically through the nanostructures; and (ii) the sequential or two-step tunnelling, where electrons are first supplied in the lower potential energies (inside an energy well) of the nanostructures before their tunnelling towards the anode. Inelastic processes are usually involved in the two-step tunnelling that lead to quasi-equilibrium distributions of electrons over the metastable states in the potential well. Resonant tunnelling has been thoroughly investigated both from the theoretical [3, 4] and experimental [5] point of view. However, while the existing theoretical models account for pronounced peaks in the I-V characteristic followed by regions of negative differential resistance (NDR), the experimental evidence is scarce due to the special conditions required by the occurrence of resonant tunnelling during FE. A more common observation in FE experiments is the step-like I-V characteristic, which can be associated with a two-step tunnelling phenomenon. A multitude of examples can be found in literature where step-like I-Vs have been observed [6, 7]. The explanation behind this peculiar behaviour of the I-V characteristic lies in the existence of nanometre size particles (or layers) on the sample, that produce a corresponding non-monotonic spatial variation of the electronic potential energy. In order to gain a deeper understanding of the quantum confinement implications in FE from nanoparticles a two-step FE model is proposed in this study.

The structure considered is simplified to a metallic substrate, which acts as the cathode, on top of which a wide band gap (WBG) material is deposited. Quantum confining objects (QCO) (e.g. metallic nanoparticles, CNTs etc.) are further deposited on top of the WBG material. The potential energy within QCOs is assumed to be lower than its level in the cathode, so that the WBG layer plays the role of a barrier controlling the supply of charge from the bulk. FE takes place from the QCOs towards the vacuum through a second barrier whose transparency is controlled by the applied electric field.

Similar structures have been investigated both theoretically [8] and experimentally [1]; the obtained results being correlated mostly with the Coulomb blockade effect. Diamond-like carbon (DLC) superlattices have also been shown to exhibit step-like characteristics associated with quantum tunnelling when subjected to point contact electrical characterisation using an STM tip [9]. Similarly, discontinuities in the current-voltage (I-V) characteristics have been previously observed, however a different explanation has been provided for the mechanism of conduction [10]. Existing theoretical models explain the step-like shape of the I-V characteristics by the quantization of the number of elementary charges inside the potential well (no confinement-induced quantization required) [8]. In the present work the focus is on the effects of quantum confinement in the QCO on the FE characteristics. The energy diagram of the considered layout is presented in Fig.1. In order to further simplify the problem without lose of generality, we will assume the system as one-dimensional (1D). Given the above framework, the electron field emission from this heterostructure will be analyzed as a two-step tunnelling phenomenon: the tunnelling supply from the substrate produces a charge accumulation in the well, which determines a shift of its bottom energy (W_0).

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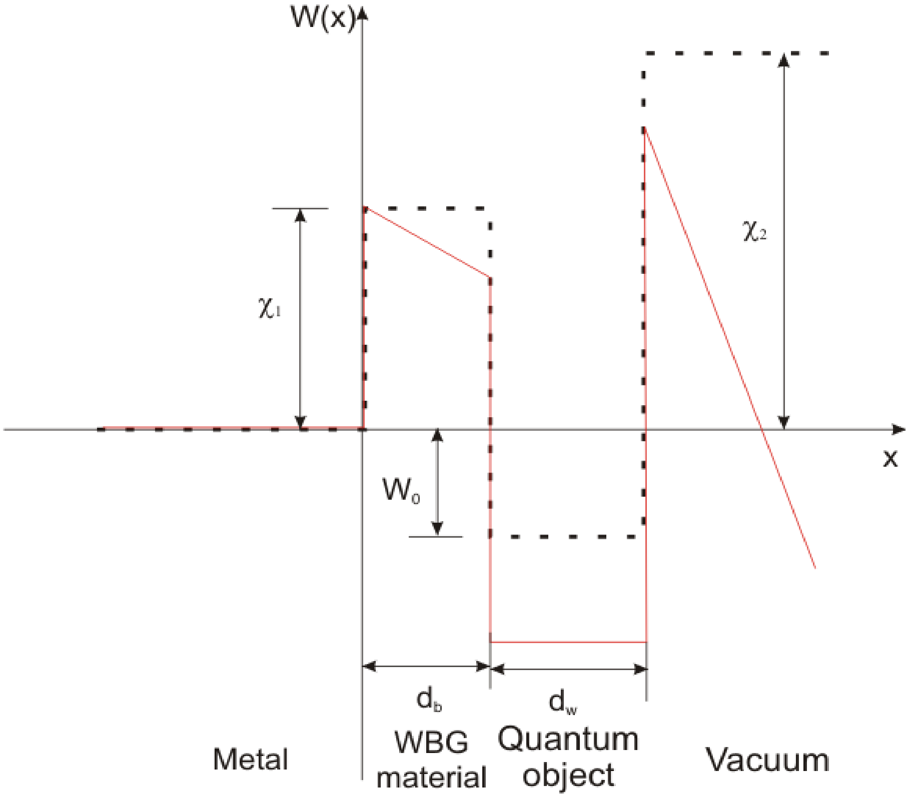

Figure 1: The potential energy diagram for the studied system (the figure is not drawn to scale). The spatial ranges are of the order of few nanometers and the energies extend to few electronvolts. The origin for the energy scale is set at the Fermi level of the metallic substrate.

The electrons can further tunnel out of the well, both into the vacuum (producing the FE current) and back into the substrate. The essential assumption of the present model is that a balance occurs between the incoming and outgoing electron fluxes from the QCO. No electron current from the vacuum into the well is assumed. The balance equation can be written as a function of the average number of electrons inside the potential well n and the applied external field F as:

$$I_{BW}\left(n,F\right)=I_{WV}\left(n,F\right)+I_{WB}\left(n,F\right) \tag{1}$$

where, I_{BW}, I_{WV} and I_{WB} are the electron current from the bulk metal towards the QCO, the electron current from the QCO towards the vacuum and the electron current from the QCO back inside the bulk, respectively. Equation (1) can, in principle, provide a stationary value of the average number of electrons in the QCO for each value of the applied field. Such a balance condition will normally alter the electrical neutrality of the QCO, so that a charging energy must be considered therein. This will indirectly affect the energy of the well's bottom and will react to the configuration of the corresponding energy levels. As a consequence, for each value of the applied field, the well's energy configuration will be altered. The balance between the incoming and outgoing currents will finally decide the value of some internal parameter of the electronic system, which, in this case, has been chosen to be the number of electrons populating the well. Once the electron population is derived for a given value of the applied field, the FE current is obtained through the semi classical concept of attempt-to-escape frequency and using the WKB approximation for the vacuum barrier [10]. As a consequence of the electron tight confinement in the QCO, the allowed energy levels are well separated. Thus, as their electron population is described by a Fermi-Dirac statistics, the average number of electrons on the levels most favoured for tunnelling into the vacuum will have a step like variation with the total number of electrons.

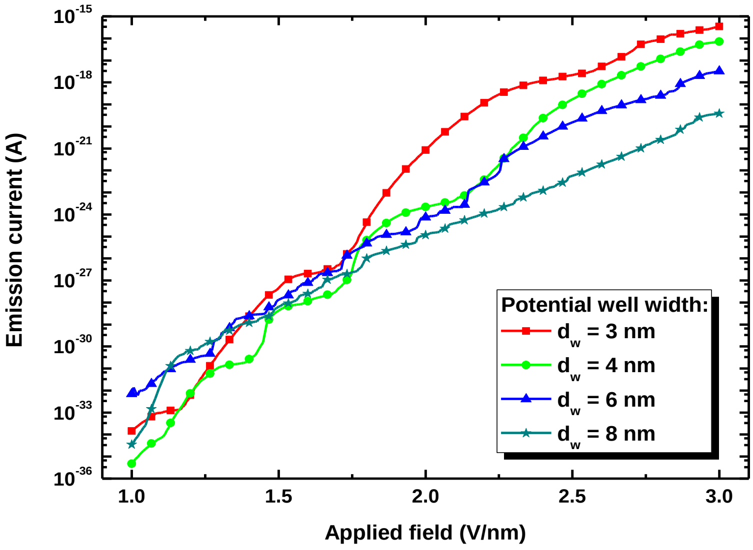

Figure 2: I-V characteristics plotted for different values of the potential well's width.

By contrast, the incoming electron flux from the substrate will be almost unaffected by the well's quantization. Therefore by varying the applied field, the balance equation given by (1) of the electron fluxes will give a step-like shape (or at least some discontinuities) to the device's current-voltage characteristics. It must be stressed that although this behaviour seems similar to the Coulomb blockade, the two phenomena differ essentially through the main cause of producing the ladder-shape of the characteristics. In the case of the Coulomb blockade, this is mainly an effect of the electric charge quantization. In the present model, the quantization of the electron energy in the QCO plays an essential role along with the local electrical charging effects.

In Fig. 2 the I-V characteristics have been plotted for four values of the potential well width. As expected, the step-like features gradually disappear as the width is increased. The mechanism of step formation in the I-V is a direct consequence of Eq. (1) and it can be better explained with the help of Fig. 3. We have plotted the electron currents for three values of the external field in two circumstances. As it can be seen the electron current incoming from the substrate has little dependence on the applied external field and is not influenced by the quantisation of the energy states in the potential well (hence the lack of steps). On the other hand, the electron current from the potential well in vacuum has a high dependence on the external field, an upward shift being noticed as the field is increased. A solution for Eq. (1) can be found at an intersection point, hence for each value of the applied field one should find in principle an average number of electrons in the potential well (the back-tunnelling current from the QCO into the substrate is neglected for this illustration). In the first instance (Fig. 3a) all the intersections are placed on one of the stairs that is shifted vertically by the increase in the external field and moves the intersection point upwards on the I_{BW} curves. This would determine a leap in the net current flowing through the structure. On the contrary, in Fig. 3b, there is a situation where the intersection points occur on a vertical jump between two stairs. Therefore, the solutions of Eq. (1) in all three instances will be the same or will differ slightly. Translating this case in the I-V diagram of Fig. 2, for those values of the field where the intersection in Fig. 3b) takes place the current will be almost constant, thus creating a lower slope or even horizontal I-V dependence.

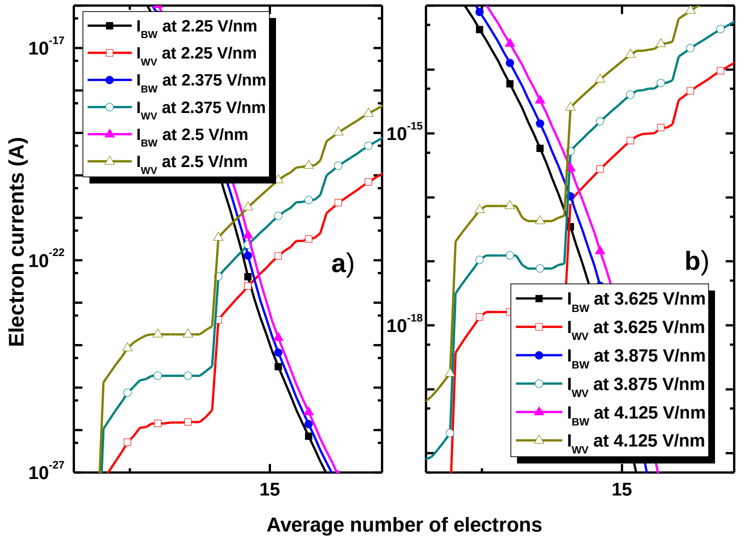

Figure 3: Electron currents plotted against the average number of electrons in the potential well: a) the intersection takes place on a vertical jump of one of the steps, b) the intersection takes place on the horizontal region of the step.

In conclusion, a simple and powerful method has been constructed to explain step-like characteristics arising from nanometre-size quantum objects through a two-step tunnelling mechanism. The method accounts for the influence of the energy quantization and geometrical dimensions of the QCO. Various types of QCO can be used in the model, by simply modifying the energy spectrum and the barriers.

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