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Localization length enhancement in two-dimensional self-assembled systems

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Abstract

Disorder correlations lead to important effects on the localization of states in one-dimensional systems, as shown for a large number of correlation models in the context of polymers and macromolecules. The extension of the problem to two-dimensional systems has been less frequently discussed due to the lack, until recently, of realistic systems with properties that could be addressed to the effects of correlations in disorder. The advent of self-assembled quantum dots, which may show short-range ordering, has changed this scenario. In the present work we investigate the properties of a two-dimensional disordered lattice with short-range correlations in the disorder that reveals itself as a minimal model for self-assembled quantum dots. The short-range correlations in disorder may lead to significant enhancement of the localization length for wide energy windows.

1. Introduction

Low-dimensional systems of non-interacting electrons show only localized states in the presence of disorder, while in three dimensions a metal–insulator transition (MIT) would be expected. This landmark in condensed matter physics was established by scaling arguments in a seminal work [1] a few decades ago. In spite of numerical support for this statement [2], several one-and two-dimensional (2D) disordered systems have been investigated in which delocalization is achieved by adding extra ingredients to the problem. The first example is the integer quantum Hall effect, where extended states appear in 2D disorder systems in the presence of magnetic fields [3]. More recently it has been found that 2D disordered systems with sympletic symmetry also show mobility edges [4].

Several models also reveal delocalization of states in one-dimensional disordered systems with correlations in the disorder, for both short-range, like the random dimer model [5], or long-range correlations [6]. Such investigations have been triggered by the study of polymer conductivity [7]. On the other hand, the extension of these investigations to 2D systems has been much less intense. Indeed, the pioneering work on the subject, considering short-range correlation, is quite recent [8], although within a rather heuristic model framework. Another recent work shows that long-range correlated disorder may induce a MIT [9]. It should be noticed that the experimental evidences of MIT in 2D MOSFET-like systems [10] raised an avalanche of explanatory hypotheses, mostly related to the interplay of disorder and electron–electron interactions [11], as has been established. However, a MIT has also been observed in a different 2D disordered system; a self-assembled quantum dots array [12]. Here there is evidence of short-range order among the quantum dots and it has been suggested that such correlations in the disorder show signatures in the transport properties [13]. An even more promising system consists of adsorbed atoms on clean surfaces. Depending on the coverage, the adatoms show different ordering [14]. From the transport properties point of view, such systems may vary the conductivity from band-like to carrier localization regimes [15]. In this context, the investigation of correlations in the disorder in 2D systems of non-interacting electrons becomes relevant and necessary. The observation of such MIT in 2D systems not driven by interactions has been addressed theoretically by percolation-like descriptions [16] or power-law transfer terms within a tight-binding model [17].

In the present work we address the question of possible MIT from the point of view of short-range correlations in the disorder, motivated by experimental findings in self-assembled quantum dots [12]. The paper is organized as follows. In section 2, a minimal model for a quantum dot-like array, showing short-range order, is presented and a structural analysis of these systems is discussed. In section 3 the electronic structure of the system and the effect of the disorder correlations on the localization of the states is addressed. In the

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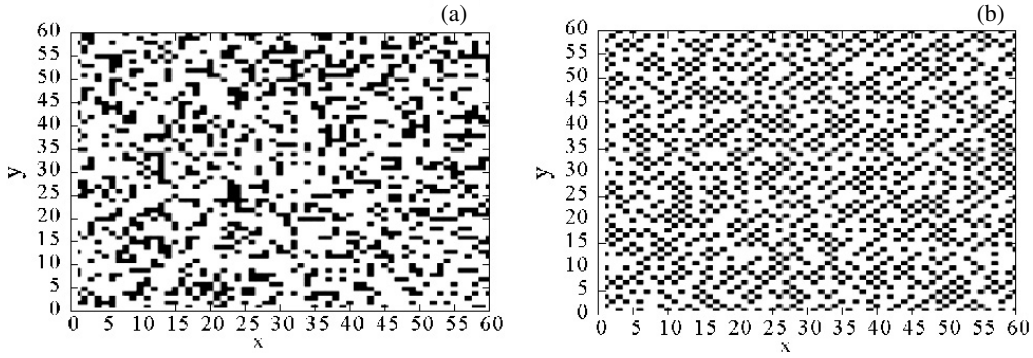

Figure 1. Model quantum dot (black dots) in a wetting host layer (white region) system configuration. (a) Without short-range correlation showing QDs merging. (b) In the presence of the short-range correlation precluding QD coalescence. Both cases are for a 60×60 sites square lattice with $P(\mathrm{QD}) = 0.3$.

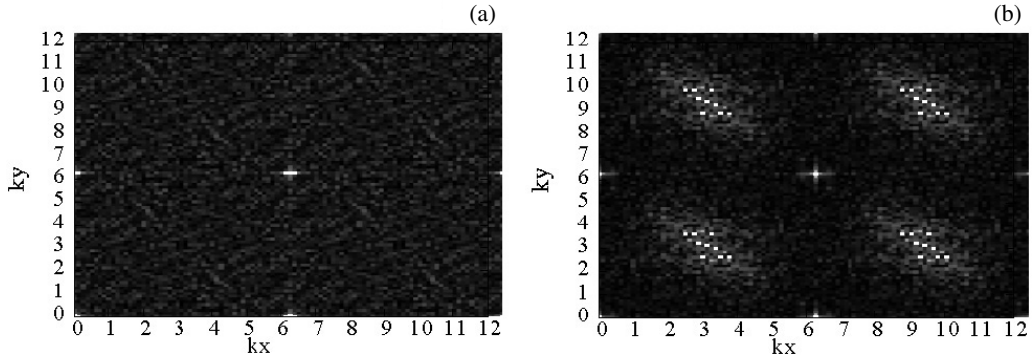

Figure 2. Fourier transforms of the systems depicted in figure 1. New structures in (b) are due to the short-range order.

following conclusions section, we summarize the results by observing that short-range order among quantum dots leads to an important enhancement of the localization length, but no true MIT can be seen in a non-interacting scenario.

2. Lattice model

The system studied in the present work is a square lattice of single s-like atomic orbitals, treated within a tight-binding approximation and considering nearest neighbor hopping only. We consider two kinds of orbitals, one of them emulating wetting layer (WL)-like sites, $\varepsilon_{\mathrm{WL}}$, and the second one emulating quantum dot (QD)-like sites, $\varepsilon_{\mathrm{QD}}$. In this approximation, the Hamiltonian assumes the following form:

$$H = \sum_{i,j}^{N} \epsilon_{i,j} |i, j\rangle\langle i, j| + V|i, j\rangle\langle i+1, j| + V|i, j\rangle \times \langle i-1, j| + V|i, j+1\rangle\langle i, j| + V|i, j\rangle\langle i, j-1|, \quad (1)$$

where the hopping parameter V can be either $V_{\mathrm{QD-WL}}$ or $V_{\mathrm{WL-WL}}$, for the two possible hoppings: between a QD site and a WL site or between two WL sites, respectively. The concentration of QD-like sites for a given disorder configuration, is given as a simulation input, $P(\mathrm{QD})$. The disorder correlation which mimics a self-assembled quantum dot layer is simply the restriction that sites assigned by $\varepsilon_{\mathrm{QD}}$ cannot be first neighbor of another QD-like site in the plane. Imposing this restriction leads to planes with isolated sites emulating self-assembled quantum dots, while removing this constraint will lead to clustering of QD-like sites. In figure 1 we show examples of disorder configurations with $P(\mathrm{QD}) = 0.3$: without correlation, figure 1(a); and with correlation, figure 1(b), both for a square lattice ($M = L \times L$) of 60×60 sites. We clearly see the effect of the correlation in the disorder: the formation of 'quantum dot polycrystals' (black squares) with short-range order, in particular, patterns of quantum dot wires can be identified. The uncorrelated configuration, figure 1(a), shows the expected site clustering of a binary alloy without any constraints. It should be noticed that since we are interested in the qualitative aspects of the effects of short-range correlation in 2D systems, we choose a heuristic model of a square lattice, although the experimental evidence points to a hexagonal short-range order in self-assembled semiconductor quantum dots [13]. On the other hand, the present approach could represent a more realistic modeling of adatoms systems, although here the experimental evidence is also local hexagonal ordering [14].

An order parameter can be given by the Fourier transform of these square lattice configurations. In figure 2 we show the Fourier transform of figure 1. For the uncorrelated case, figure 2(a), we identify only a sharp peak at the center of

the reciprocal lattice, related to the square host lattice of a completely random binary alloy. On the other hand, for the disorder correlated case in figure 2(b) broad structures of four satellite peaks are identified, which are related to the finite size clusters of binary alloys with short-range order ('quantum dot polycrystals'). The relative height of these satellite peaks gives an order parameter to the problem that permits us to follow the evolution of electronic structure and localization properties as a function of disorder. The distances between the satellite peaks are associated here to simply doubling the lattice parameter of the host lattice used in the simulations, but in real systems could be related to the reciprocal of the distance between a quantum dot and its nearest neighbors.

In what follows we describe the evolution of the electronic and localization properties for different systems, varying the quantum dot concentration and lattice sizes up to squares of 100×100 sites with Dirichlet boundary conditions. Since we are dealing with disordered finite systems, averages over different configurations are taken in all electronic quantity calculations shown below.

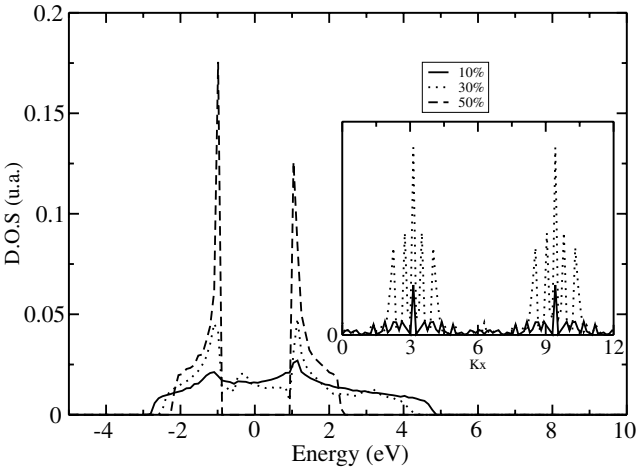

Figure 3. Densities of states for the QD lattice for increasing QD concentration. Inset: evolution of the extra peaks in the Fourier transform of correlated systems with increasing QD concentration.

3. Electronic structure and localization

The results shown below are qualitatively robust for a wide range of tight-binding parameters. The results shown here are for the following parameters: $\varepsilon_{\mathrm{WL}} = 1.0$ eV, $\varepsilon_{\mathrm{QD}} = -1.0$ eV, $V_{\mathrm{QD-WL}} = 0.5$ eV, and $V_{\mathrm{WL-WL}} = 1.0$ eV. The choice of these parameters is rather heuristic, since the experimental system which motivates the present work [12] has some characteristics worth mentioning. We are interested in the modification of electronic structure of a 2D system—the wetting layer—by the presence of a very high density of QDs (5×10^{10} dots cm^{-2}). With this remark in mind, the chosen parameters emulate a wide WL-like band with quantum dot levels in the lower energy range. Increasing the number of QDs leads to drastic changes in the electronic structure of the whole system, which are experimentally rather unknown. The densities of states for different concentrations of QD-like sites, $P(\mathrm{QD})$, are shown in figure 3 for correlated disorder cases of 60×60 site systems. As expected, for $P(\mathrm{QD}) = 0.0$ the densities of states show the signature of the 2D host square lattice, while $P(\mathrm{QD}) = 0.5$ represents an ordered binary alloy. Fingerprints of the van Hove singularities of the ordered binary alloy case are already present at low concentrations like $P(\mathrm{QD}) = 0.05$ (not shown here). In figure 3 we also show an intermediate concentration of $P(\mathrm{QD}) = 0.3$ where the van Hove singularities of ordered binary systems are already well developed, but the disorder is reflected in the finite density of states in the gap region. The density of states in the gap region is, nevertheless, not as clear a measure of disorder as the Fourier transform of the dot configuration, figure 2. In the inset of figure 3, the relative height of the satellite peaks in the Fourier transform is shown. One can see how this order parameter evolves, comparing two cases: $P(\mathrm{QD}) = 0.3$, with well developed satellite peaks (quite broad due to the finite size of the 'QD crystals'), and $P(\mathrm{QD}) = 0.1$, where there are only very incipient signatures of these structures. It should be mentioned that some of the important features that appear in our results are still not comparable to real self-assembled QD systems. Nevertheless, we consider of relevance the lower energy range, around the QD-like site energy, $\varepsilon_{\mathrm{QD}}$, since QD-like sites are those for which a local correlation is considered, while WL-like sites continue uncorrelated until concentrations approaching the ordered binary alloy limit, $P(\mathrm{QD}) = 0.5$.

There are no qualitative changes in the electronic structure, from the point of view of the calculated density of states, with the size of the system. If, on the other hand, one focuses on the degree of localization, such size effects should be evaluated. The degree of localization of a state is given here by the participation ratio (PR) [18], defined, in the tight-binding approximation, by:

$$\mathrm{PR} = \frac{1}{N^2 \sum_{i=1}^{N} \sum_{j=1}^{N} |a_{i,j}|^4} \tag{2}$$

where $a_{i,j}$ is the wavefunction amplitude in the (i, j) site.

The PR is close to zero for localized states when $N \to \infty$ (for maximum localization, i.e. the wavefunction localized on a single site, $|a_{i,j}| = \delta_{i,j}$, $\mathrm{PR} = 1/N^2$) and reaches the maximum value of $4/9$ for delocalized states in the case of a 2D ordered system. The expression above is explicit for a square 2D system N sites wide in each direction. Such measuring of the degree of localization is sometimes addressed also by an equivalent quantity, namely the inverse participation ratio, as discussed by Thouless [19].

In order to reveal the effect of our model correlation on the electronic localization, we compare in figure 4 the PR of the electronic states in a 2D system with $P(\mathrm{QD}) = 0.3$ with correlation (continuous line) and without correlation (large dots) for 60×60 site host lattices. Three different regions can be identified for the behavior of the PR. For lower energies, there is evidence of a noticeable enhancement (a factor of 3) of the localization length. The gap edges (at -1.0 eV and 1.0 eV) show clear fingerprints also in the averaged PR for both correlated and uncorrelated cases. It should be noticed

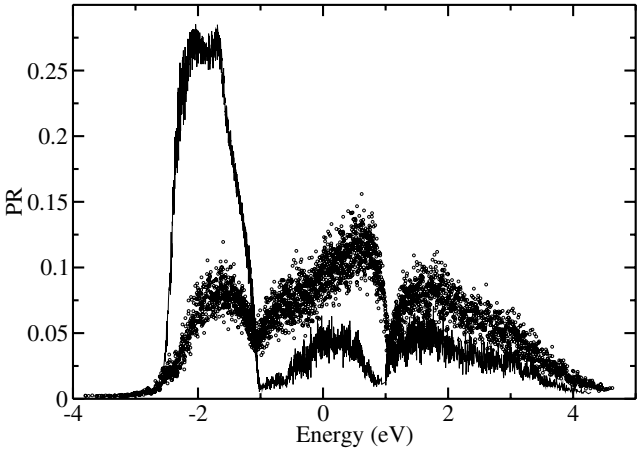

Figure 4. Participation ratio for a 60 × 60 lattice with a concentration $P(\mathrm{QD}) = 0.3$ with (continuous line) and without (large dots) correlation.

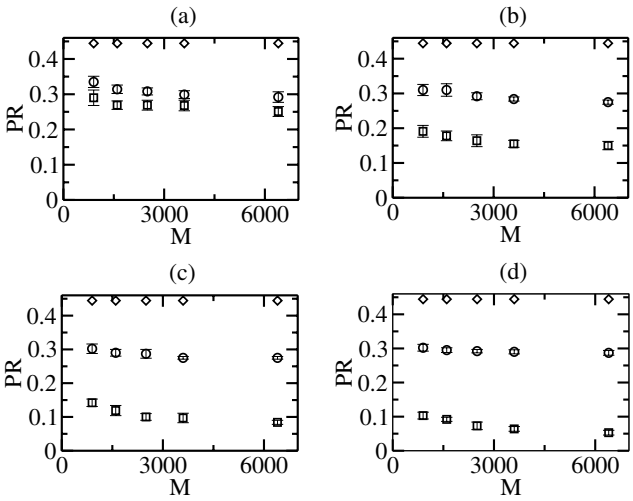

Figure 5. Participation ratios at $E \approx -1.5$ eV for correlated (open circles) and uncorrelated (open squared) systems as a function of lattice size and for different QD-like site concentrations: (a) $P(\mathrm{QD}) = 0.1$, (b) $P(\mathrm{QD}) = 0.2$, (c) $P(\mathrm{QD}) = 0.3$ and (d) $P(\mathrm{QD}) = 0.4$. The PR for a state completely delocalized is also shown (diamond).

that in the gap region, the PR for the uncorrelated case is now higher than for the correlated system. Nevertheless, this difference is not so pronounced as the inverse one for lower energies. For the higher energy band region no significant differences are introduced by the presence of short-range order. All these characteristics are robust as a function of system size and become more pronounced with increasing QD-like sites up to $P(\mathrm{QD}) = 0.45$: (i) formation of an effectively delocalized band at a lower energy band -2.5 eV $< E < -1.0$ eV, (ii) suppression of the density of states in the gap region for the correlated cases with the concomitant diminution of the PR, compared to the uncorrelated systems and, (iii) no qualitative differences at higher energies.

The main result found here is the appearance of a band of effectively delocalized states (seen here as a state with a localization length greater than the system size [20]), although we find no evidence of a true MIT [8] for the present model at the explored parameter ranges. We call attention to the fact that a band of effectively delocalized states occurs at lower energies, associated to the QD-like sites that are correlated, while the higher energy range is associated to the WL-like sites, which are not correlated. However, the effective delocalization could be of relevance in actual self-assembled QD systems [13], as can be seen from PR calculations as a function of QD concentration and system size ($M = L^2$), figure 5. When a low QD concentration is considered, $P(\mathrm{QD}) = 0.1$ and $P(\mathrm{QD}) = 0.2$, figures 5(a) and (b), the dependence on the system size for the maximum PR ($E \approx -1.5$ eV) is similar for both cases, correlated and uncorrelated, and the difference between their absolute values is rather small, up to two times higher for the correlated system. There should be few 'QD crystals' regions, even when the correlation is imposed, due to the low concentration. The size of such regions has also to be rather small. When higher concentrations are considered, $P(\mathrm{QD}) = 0.3$ and $P(\mathrm{QD}) = 0.4$, as shown in figures 5(c) and (d), new important qualitative and quantitative differences between the correlated and uncorrelated cases appear at the system sizes investigated. While the PR for uncorrelated systems continuously decrease with the increase of the system size, it tends to saturate when correlation is considered. This saturation seems to occur even for small system lengths. Indeed, in figure 5(c) one can see that the correlation enhances the PR by a factor of two even for a system of 30 × 30 sites. Moreover, for large systems, the PR values of correlated systems are now six times higher than for the uncorrelated cases, a significant difference. These results suggest that the enhancement of the localization length, one possible mechanism for a metal–insulator crossover—due to a local spatial correlation—is present already at low quantum dot concentration, becoming more pronounced with increasing dot density [12].

The present results are also interesting in the scenario of conductivity dependence on adatom coverage of a given surface [15]. In this context, the increase of QD concentration may be seen as an increase of the surface coverage. In our model, the highest coverage $P(\mathrm{QD}) = 0.5$ corresponds to an ordered 2D system and all states become delocalized (not shown here). A slightly lower coverage ratio, $P(\mathrm{QD}) = 0.4$, figure 5(d), reveals an important difference between the presence or absence of correlation in the disorder.

4. Conclusions

Here, we have presented a minimal model for correlation in 2D self-assembled QDs in which there is no QD–QD coalescence. Due to that correlation, there is a formation of finite 'QD crystals' that leads to the appearance of four additional broad peaks in the Fourier transform of the lattice. Moreover, the main effect of such correlation is the raising of a band of effectively delocalized states. This effect is more pronounced for higher concentrations of QDs as reflected by the developing of the van Hove peaks in the density of states. In this work, however, there is no definitive evidence of a metal–insulator

transition as predicted theoretically for other models [8], or seen experimentally in related systems [12]. Nevertheless, that effective delocalization due to correlations could be an important mechanism in determining the transport properties of self-assembled quantum dots or adatom systems.

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References

[1] Abrahams E, Anderson P W, Licciardello D C and Ramarkrisshnan T V 1979 *Phys. Rev. Lett.* **42** 673
[2] Kramer B and Mackinnon A 1993 *Rep. Prog. Phys.* **56** 1469
[3] Klitzing K v 1986 *Rev. Mod. Phys.* **58** 519
[4] Assada Y, Sleivin K and Ohtsuky T 2002 *Phys. Rev. Lett.* **89** 256601
[5] Dunlap D H, Wu H-L and Phillips P W 1990 *Phys. Rev. Lett.* **65** 88
[6] de Moura F A B F and Lyra M L 1998 *Phys. Rev. Lett.* **81** 3735
[7] Huang W S, Humphrey B D and MacDiarmid A G 1986 *J. Chem. Soc. Faraday Trans.* **82** 2385
Zuo F, Angelopuolos M, MacDiarmid A G and Epstein A J 1987 *Phys. Rev.* B **36** 3475
[8] Hilke M 2003 *Phys. Rev. Lett.* **91** 226403
[9] Liu W-S, Liu S Y and Lei X L 2003 *Eur. Phys. J.* B **33** 293
[10] Kravchenko S V, Kravchenko G V, Furneaux J E, Pudalov V M and D'lorio M 1994 *Phys. Rev.* B **50** 8039
[11] Abrahams E, Kravchenko S V and Sarachik M P 2001 *Rev. Mod. Phys.* **73** 251
[12] Ribeiro E, Jäggi R D, Heinzel T, Ensslin K, Medeiros-Ribeiro G and Petroff P M 1999 *Phys. Rev. Lett.* **82** 996
[13] Heinzel T, Jäggi R, Ribeiro E, Waldkirch M v, Ensslin K, Ulloa S E, Medeiros-Ribeiro G and Petroff P M 2003 *Europhys. Lett.* **61** 674
[14] Liu C, Yamazaki S, Hobara R, Matsuda I and Hasegawa S 2005 *Phys. Rev.* B **71** 041310(R)
[15] Matsuda I and Hasegawa S 2007 *J. Phys.: Condens. Matter* **19** 355007
[16] Meir Y 1999 *Phys. Rev. Lett.* **83** 3506
[17] Potempa H and Schweitzer L 2002 *Phys. Rev.* B **65** 201105(R)
[18] Bell R J and Dean P 1970 *Discuss. Faraday Soc.* **50** 55
[19] Thouless D J 1974 *Phys. Rep.* **13** 93
[20] Parshin D A and Schober H R 1999 *Phys. Rev. Lett.* **83** 4590
Evers F and Mirlin A D 2000 *Phys. Rev. Lett.* **84** 3690