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COMPUTATIONAL METHODS FOR SUPERLATTICE STRUCTURES:

RENORMALIZATION AND RECURSION TECHNIQUES

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Lorenzo Resca(*)

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Department of Physics, The Catholic University of America

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1. INTRODUCTION

We summarize in this final report the main results and findings of the theoretical study of the electronic structure of superlattices which we have performed under Contract DAAB07-86-C-F114; research papers sponsored by this contract which detail all the technical results are quoted as Refs. 1-7.

In recent years there has been an explosion of interest in the experimental and theoretical study of the electronic properties of superlattices,⁸ related heterostructures and quantum wells, as well as two-dimensional structures and surfaces in general. The reason of such attention is certainly not a matter of fashion: there are deep motivations and ambitious perspectives.

The importance of the electronic and optical properties of superlattices is best appreciated within the general framework of the study of quantum electronic microstructures, i.e. systems where the electronic wavelength becomes comparable to the size of the alternating layers which form the superlattice structure. The ultimate goal of microstructure physics is to produce a new generation of electronic devices, whose operation is based on the very fundamental principles of quantum mechanics: these new quantum devices are already entering in competition with the more traditional electronic devices.⁹ Exploiting ingeniously new concepts in band engineering, quantum size effects, electron ballistic transport, tunneling and resonant tunneling, the new microstructures exhibit effects completely beyond those of more

traditional devices.

Furthermore, the importance of the study of electronic states in microstructures is not limited to applicative and technological aspects: in fact, microstructure physics has been stimulated by, and has itself spurred, very fundamental research. In this regard, we recall the unexpected and spectacular results of the quantum Hall effect,¹⁰ which has provided a new relationship among the fundamental constants, and, as a byproduct, the new standard of resistance.

The theoretical study of the electronic properties of superlattices is of major importance to interpret, understand, and possibly predict the wide range of structures and effects produced by the engineering of materials. After a preliminary study on the advantages and intrinsic limitations of the various methods of band structure calculations in superlattices, we decided to focus on the renormalization method; its formal procedure is basically related to the renormalization group introduced by Wilson to study second-order phase transitions, which produced a complete understanding of these phenomena for the first time.¹¹ At the beginning of this research the only existing calculation performed with the renormalization method in a superlattice referred to a one-dimensional highly simplified model.¹² Nonetheless, that pioneering work had given us encouragement to pursue the renormalization method to full-scale calculations in realistic superstructures. To this end, we have enriched the renormalization method with concepts and techniques peculiar to solid state physics, and we have now

developed it into a very powerful tool for the study of electronic properties of superlattices.

The main achievement of our work has been the development and firm establishment of the renormalization technique for the study of superlattice structures. A further step consists in including defects in this study. Defect levels are very important from both the fundamental and the applied point of view, as they influence very strongly the performance of electronic devices. We have already developed very efficient procedures to study microscopically impurity states in standard lattices. These are Green's function techniques based on the recursion method.^{5,6,13} A preliminary work with the recursion method in a superlattice structure has also been performed.¹⁴ The exact formal relation between the renormalization and the recursion method has also been established recently.¹⁵ A discussion of this and other perspectives is given in the third and final section of this report. But first we summarize in the following section the most relevant technical aspects of electronic-structure calculations in superlattices with the renormalization method.



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2. TECHNICAL DISCUSSION OF ELECTRONIC-STRUCTURE CALCULATIONS IN SUPERLATTICES WITH THE RENORMALIZATION METHOD

2.1 Motivation for implementing the renormalization method

In 1986, when this research program began, the state of the literature for calculating the electronic structure of superlattices was unsatisfactory in several aspects, despite the tremendous importance that superlattice studies were acquiring. In principle, all the standard methods of band-structure calculations¹⁶ can be extended to superlattices, since these are still periodic structures, and translational symmetry is included in Bloch form for the wavefunctions. However, in practice, only very thin superstructures can be satisfactorily treated this way.

Calculations for ultrathin superlattices (i.e. superlattices where the alternating slabs of the composing semiconductors consist of only a few atomic planes) have been performed both with the empirical tight-binding method and the empirical pseudopotential method. Sophisticated calculations of ultrathin microstructures have also been performed with linearized method, and the linearized muffin-tin-orbital method in particular. None of these methods, however, can be extended much beyond the case of ultrathin structures, except possibly the empirical pseudopotential method: in that method, the great simplicity of the matrix elements and the adoption of a folded-band scheme balances to some extent the complications due to a large number of atoms in

the supercell.

Before the introduction of the renormalization method, there were basically only two methods which dealt with superlattices of arbitrary length: namely, the envelope-function and the complex- k tight-binding method (the complex $k.p$ procedure is somewhat intermediate between the two, and borrows some features from both).

The envelope-function method is based on an overall description of the bulk properties of the two crystals composing the superlattice, rather than their detailed microscopic nature. More or less appropriate boundary conditions^{17,18} are then established at the interfaces, and determinantal-compatibility equations yield superlattice eigenvalues and eigenfunctions. The applicability of the envelope-function method is quite restricted. The two composing semiconductors must be chemically alike and have similar band structures: extensions to multivalley band structures are not quite justified. Beyond such a restricted range of applicability the envelope function can at best be used for qualitative considerations: this is generally inadequate to interpret quite accurate experimental results, especially optical data and transport properties.

The other previous method capable of treating superlattices of possibly large periodicity is tight-binding with a complex- k procedure.¹⁹ From a computational point of view this method is very complicated, since it involves a cumbersome energy-dependent procedure. One starts with a given energy E and

determines the complex- \underline{k} vectors compatible with it. Then, all the evanescent and stationary generalized plane-waves of each component crystal are used as energy-dependent basis-functions. Matrix elements of the one-electron crystal Hamiltonian are determined, and the matrix (which is energy dependent) is then diagonalized. The procedure is repeated until the input and output energies coincide. This procedure, which is already very cumbersome for idealized superlattices, becomes prohibitive for treating disrupted interfaces. Those may be treated in principle with an appropriate use of the transfer-matrix method,^{20,21} but that is not a routine procedure at all.

In summary, the envelope-function method is unsatisfactory because it is not based on a genuine microscopic description, while the complex- \underline{k} tight-binding method is unnecessarily cluttered with computational difficulties. These were the only previous methods which can in principle treat superlattices of arbitrary periodicity.

2.2 Basic features of the renormalization method and relevant aspects of the implemented computer programs

The renormalization method consists basically in a systematic reduction of the degrees of freedom of a given system. The reduction may be such that the Hamiltonian at each decimation step maintains the same form. If that is the case, it is possible to repeat the reduction process iteratively until a large number of

degrees of freedom of the system is eliminated.

We have shown that it is possible in the case of a superlattice to maintain the Hamiltonian invariance through decimation by properly exploiting the intralayer symmetry. Technically, one adopts a \mathbf{k} -space representation for layered Bloch sums and a real-space representation for interactions among different layers, i.e. families of planes perpendicular to the growth direction. The microscopic description of the crystal is then done in terms of two-dimensional Bloch sums, called layer orbitals, and atomic planes are grouped into layers such that inter-layer interactions are limited to the nearest neighbors. These "sites" are then decimated in the renormalization procedure.

There are several distinctive features of the renormalization procedure that we have developed for superlattices, which make it very appealing from both the physical and the computational point of view. They are:

(a) The decimation process only requires at each step inversion of matrices of small order, namely the order of the degrees of freedom at a given site.

(b) It is possible to take advantage of the local symmetry to drastically reduce the number of decimations performed in bulk-like regions. In those regions, whenever layers are equivalent, one can eliminate all alternate layers in a single stroke.

(c) For superlattices with N sites (in a slab of either component crystal) it is possible to show that the first N moments do not depend on the boundary conditions. Hence, the renormalization

process can be organized in such a way that only the very last renormalization depends on the boundary conditions. Then, the whole superlattice energy-band dispersion in the growth direction can be obtained with a minimal additional effort, once a given q -vector of the parallel two-dimensional Brillouin-zone has been fully investigated.

(d) The knowledge of the energy bands can be supplemented, when this is required, by calculation of the superlattice wavefunctions. This can be done by simply repeating the renormalization procedure, in correspondence to any given eigenvalue, preserving different layers. Alternatively, one can propagate the Green's function from any pair of layers to the adjacent ones and so on: this procedure requires, however, a careful check of the numerical stability when layers sufficiently faraway are reached.

We have developed various efficient computer codes which perform all the operations mentioned above. For superlattices with composing crystals with diamond or zinc-blend structures we used a tight-binding parametrization for the starting Hamiltonian. That has typically ten orbitals per atom, i.e. the product of the s , p_x , p_y , p_z , s^* atomic orbitals²² and spin-up and spin-down states, since spin-orbit interaction is typically included. Our current codes assume only nearest-neighbor interactions and stacking of layers along a crystallographic axis, but these restrictions could easily be removed. Exact lattice-constant matching between the two types of composing semiconductors has been assumed, but we have also initiated the study of strained superlattices. The computer code

automatically recognizes and eliminates equivalent sites. Subroutines for calculation of wavefunction-amplitude at all anion and cation sites are included in the program.

2.3 Impurity effects in lattices and superlattices

The role of defects in superlattices is a major one, just as it is in ordinary lattices. At this point, our renormalization procedure cannot be directly applied when a defect breaks the periodic symmetry of the superlattice. There is, however, the possibility of applying the recursion method. Formally, that is equivalent to the renormalization method, and both can be derived within the same framework by partitioning differently the Hilbert space of the total Hamiltonian.¹⁵ Both methods represent iterative procedures which yield immediately the Green's function of the Hamiltonian operator, without requiring its direct diagonalization. Each method has nonetheless different advantages in practical situations. With the recursion method we have been able to give for the first time a unified description of shallow and deep, bound and resonant, defect states in both a cubic model and in silicon, including short- and long-range contributions to the defect potential consistently and non-perturbatively.^{5,6} A preliminary application of the recursion method to a model superlattice has also been attempted.¹⁴ A theoretical model of the DX center and its experimental investigation in $\text{Ga}_{1-x}\text{Al}_x\text{As}$ alloys and superlattices has been proposed.⁷

3. SUMMARY OF RESULTS, CONCLUSIONS AND RECOMMENDATIONS

The results mentioned in this report are described in detail in Refs.1-7: we summarize here the main findings.

We begin with the study of silicon superlattices, with a superimposed external potential in the [100] direction.¹ This can be considered as a prototype of group-IV superlattices. It also represents a simplified model of the very interesting Si-Si_{1-x}Ge_x system, and other composition or doping-graded elemental group-IV superlattices. The study of this superlattice was indeed challenging, due to the complicated multivalley structure of silicon, and represented a severe test of our renormalization-group approach. With that formalism we have been able to calculate the dependence of the energy gap on the applied external potential for various superlattice widths. We have evidenced level-quantization effects, which are large for small superlattice widths, and we have examined the possibility of tuning the energy gap down to a semiconductor-semimetal transition. A very interesting feature is the transition from an indirect-gap to a direct-gap material, as the width and the value of the potential are changed. That may have far-reaching consequences for the optical properties of this material, which await an experimental confirmation. Another interesting and peculiar effect of the superlattice band structure that we have evidenced is the doublet structure of the lowest states of the conduction band; this is due to the folding of the valleys in the [0,0,1] and [0,0,-1] directions. Such effect has

been compared with the intervalley effects which arise similarly in impurity problems.^{4-6,23} These calculations, beside the direct interest in this particular system, have definitely shown the capability of the renormalization method to treat multivalley components, thus opening new perspectives for the vast class of superlattices composed by materials with complicated energy-band structures.

Another system that we have studied with the renormalization method is the InAs-GaSb superlattice.^{2,24} This is a typical type-II superlattice, where the bottom of the InAs conduction band is lower in energy than the top of the GaSb conduction band. Because of quantum confinement, for small periods the superlattice is a semiconductor: but for large enough unit supercells (InAs thickness around 100 Å) the system becomes semimetallic. The superlattice band structure exhibits high non-parabolicity, mixing and anticrossing, and significant spin-orbit effects: yet, all that could be accurately accounted for by the renormalization procedure. The interpretation of the semimetallic regime is a delicate problem. The renormalization method has allowed a new explanation in terms of an intrinsic semimetallic behavior, an effect that could not be obtained with the envelope function method.¹⁸ Another subtle and quite interesting effect is the remarkable asymmetry of the wavefunction amplitude within the GaSb layers: that is due to the difference between the two interfaces, which are In-Sb and Ga-As, respectively. This somewhat surprising effect persists even at considerably large superlattices

widths.

In the study of impurity problems,^{5,6} we have used the recursion method, which reduces the initial Hamiltonian to a chain-like form. That can be directly diagonalized, or used to get immediately a continuous fraction expressing the Green's function matrix elements. We have approximated an infinite crystal of Si with a finite Si cluster, supplemented by periodic boundary conditions. We have considered clusters containing 32768 atoms (or more in some instances), corresponding to 163840 orbitals when the s , p_x , p_y , p_z , s^* microscopic description is adopted. Our results have clearly shown, for the first time, that the recursion method can provide an accurate description of both shallow and deep, bound and resonant impurity states, including self-consistently both short- and long-range defect-potential contributions, as well as the complete multivalley band structure. An original model of the DX center, which explains consistently both electronic and relaxation effects, has been proposed for $Ga_{1-x}Al_xAs$ alloys and superlattices.⁷

In conclusion, we have proved the effectiveness that Green's function techniques based on the renormalization and recursion methods can provide in the study of superlattices and defect levels. We expect that these methods of calculation will play a major role in interpreting the wealth of experimental data which are increasingly being collected on these new materials. In particular, we have made considerable progress in a thorough study of HgTe-CdTe superlattices, which are of great interest from both

points of view of fundamental research and infrared-detector technology. We recommend a completion of this study. We also recommend completion of the proposed study of the DX centers with the recursion method,⁷ since these defects affect crucially the performance of modulation-doped field-effect transistors (MODFET's) made of $\text{Ga}_{1-x}\text{Al}_x\text{As}/\text{GaAs}$, the structure of devices used in high-speed digital and analog circuits (the DX is believed to be responsible for slow transients in the switching characteristics of these MODFET'S).

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