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Quantum box energies as a route to the ground state levels of self-assembled InAs pyramidal dots

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A theoretical investigation of the ground state electronic structure of InAs/GaAs quantum confined structures is presented. Energy levels of cuboids and pyramidal shaped dots are calculated using a single-band, constant-confining-potential model that in former applications has proved to reproduce well both the predictions of very sophisticated treatments and several features of many experimental photoluminescence spectra. A connection rule between their ground state energies is found which allows the calculation of the energy levels of pyramidal dots using those of cuboids of suitably chosen dimensions, whose solution requires considerably less computational effort. The purpose of this work is to provide experimentalists with a versatile and simple method to analyze their spectra. As an example, this rule is then applied to successfully reproduce the position of the ground state transition peaks of some experimental photoluminescence spectra of self-assembled pyramidal dots. Furthermore the rule is used to predict the dimensions of a pyramidal dot, starting from the knowledge of the ground state transition energy and an estimate for the aspect ratio Q . © 2000 American Institute of Physics. [S0021-8979(00)03921-9]

I. INTRODUCTION

A huge quantity of experimental data is available on InAs/GaAs self-assembled quantum dots (QDs) since the fabrication of samples with increasingly narrow size and uniform density distribution has been made easy to achieve by the Stranski–Krastanov growth method.¹ Islands of various sizes and shapes have been reported, depending on the growth conditions, such as temperature, dot material coverage, growth rate, time delay before cap regrowth, etc.

The energy levels of such structures cannot be easily calculated, both because of the finite potential confining barrier (often of the order of 200–500 meV) and the nontrivial geometry of the dot. The Schrödinger equation must thus be solved by means of a numerical method.

All these methods, although based on different theoretical approaches, have a common feature: they are usually very complex and their computational demands are often very high. Therefore, most of the time they are inaccessible to the experimentalists, who have to resort to a sort of *interpolation* of the available (i.e., published) theoretical data in order to interpret and analyze their spectra. The dimensions of the experimental dots are, however, almost never the ones which the theoretical calculations are made for. An example is the case of the samples grown by Sauvage *et al.*² which are lens shaped islands with typical height of 3 nm.

The estimate for their mean lateral size has been obtained by a comparison of the photoluminescence (PL) spectra with the theoretical calculations reported by Grundmann *et al.*³ relative to square based pyramids with aspect ratio $Q=1$. According to this estimate, however, the experimental structures all have different Q s (i.e. $Q=1.6, 1.38, 0.916$, and 1.53 , respectively), and, more importantly, all $Q \neq 1$. The problem, here, does not lay therefore in the pyramidal ap-

proximation (in a previous article⁴ we have shown that the transition energies of lens shaped structures can be successfully reproduced by using pyramids with the same dimensions. In fact, as illustrated by Andreev *et al.*,⁵ the strain distribution is similar for both shapes throughout most of the dot, differing only in the upper part, due to the sharp edges of the pyramid and smooth boundary of the hemisphere), the problem lies in the use of theoretical data relative to structures with a given $Q=Q_{th}$ to fit the spectra of structures with different aspect ratios ($Q_{exp} \neq Q_{th}$). A small difference in only one of the dimensions results in dramatic differences in the electronic energy levels, (as it is shown in Table I for square-based pyramids with the same base length $b=200 \text{ \AA}$ but different heights $h=100 \text{ \AA}$ and $h=70 \text{ \AA}$. $Q=1$ and 1.428 , respectively), so that the conclusions drawn from the comparison are at least inaccurate, if not misleading.

The aim of this article is to apply an extension of the method developed by Gershoni *et al.*⁶ originally for a rectangular quantum wire (and only very recently successfully applied by the authors⁴ to the study of pyramidal shaped dots), to determine a relationship between the ground state energy levels of self-assembled InAs pyramidal quantum dots and those of cuboids with the same characteristics, i.e., effective masses and confining potentials.

The objective being to obtain the energies of the former using those of the latter, (whose solution requires a much reduced code complexity resulting in a considerably less computational effort), and therefore provide experimentalists with a useful and simple (but not necessarily less reliable) tool to analyze their spectra.

Section II gives an overview of the method and the theory involved. In Sec. III results are presented and discussed. A comparison of the prediction (using both the cuboidal approximation and the full pyramidal calculation),

TABLE I. Comparison between electronic energy levels of square-based pyramidal dots with the same base length $b=200 \text{ \AA}$ and different heights $h=70$ and $h=100 \text{ \AA}$. The difference is not only in the energy eigenvalues (about 50 meV for each level), but in the number of bound states as well: 6 for the 70 \AA height pyramid (where n.b. stands for not bound), and 9 for the 100 \AA height one.

Level n	$E_n^{\text{el}}(h=70 \text{ \AA})$	$E_n^{\text{el}}(h=100 \text{ \AA})$
0	-259	-305
1	-155	-204
2	-155	-204
3	-77	-125
4	-60	-117
5	-37	-81
6	n.b.	-56
7	n.b.	-31
8	n.b.	-27
9	n.b.	0

with experimental spectra of self-assembled pyramidal dots is given. The summary and conclusions are given in Sec. IV.

II. THEORY

In the effective mass approximation the Schrödinger equation for the envelope function can be written as

$$-\frac{\hbar^2}{2} \left(\nabla \frac{1}{m^*(x,y,z)} \nabla \right) \Psi(x,y,z) + V(x,y,z) \Psi(x,y,z) = E \Psi(x,y,z). \quad (1)$$

The envelope function of the quantum confined system of interest (cuboid or pyramid), $\Psi(x,y,z)$, can be expanded in terms of a complete orthonormal set of solutions of the cuboidal problem with infinite barrier height, i.e.,

$$\Psi(x,y,z) = \sum_{lmn} a_{lmn} \psi_{lmn}, \quad (2)$$

where

$$\psi_{lmn} = \sqrt{\frac{2}{L_x}} \sin \left[l \pi \left(\frac{1}{2} - \frac{x}{L_x} \right) \right] \times \sqrt{\frac{2}{L_y}} \sin \left(m \pi \frac{y}{L_y} \right) \sqrt{\frac{2}{L_z}} \sin \left[n \pi \left(\frac{1}{2} - \frac{z}{L_z} \right) \right]. \quad (3)$$

The domains $[-L_x/2, L_x/2]$, $[-L_z/2, L_z/2]$ have been chosen for the variation of x and z , and $[0, L_y]$ for that of y .

Care has been taken to move the boundaries L_x, L_y, L_z away from the dot system, so that the energy eigenvalues are essentially independent of their choice. Since there is no need to explicitly match wave functions across the boundary between the barrier and dot materials, this method is easily applicable to an arbitrary confining potential. After substituting expression (2) into Eq. (1), multiplying on the left by $\psi_{l'm'n'}^*$, and finally integrating over the cuboid $L_x L_y L_z$ (where use is made of the orthonormality of the wave functions), obtain the matrix equation

$$(M_{lmnl'm'n'} - E \delta_{mm'} \delta_{ll'} \delta_{nn'}) a_{lmn} = 0. \quad (4)$$

The matrix elements $M_{lmnl'm'n'}$ are given by (more details can be found in Ref. 4)

$$M_{lmnl'm'n'} = \frac{\hbar^2}{2} \int \frac{1}{m^*} \nabla \psi_{l'm'n'}^* \nabla \psi_{lmn} dx dy dz + \int \psi_{l'm'n'}^* V \psi_{lmn} dx dy dz = \left[\frac{\hbar^2 \pi^2}{2} \frac{1}{m_B} \left(\frac{ll'}{L_x^2} + \frac{mm'}{L_y^2} + \frac{nn'}{L_z^2} \right) + V \right] \times \delta_{ll'} \delta_{mm'} \delta_{nn'} + \frac{\hbar^2}{2} \left(\frac{1}{m_W} - \frac{1}{m_B} \right) \times \int_W \nabla \psi_{l'm'n'}^* \nabla \psi_{lmn} dx dy dz - V \int_W \psi_{l'm'n'}^* \psi_{lmn} dx dy dz, \quad (5)$$

where the subscript W in the integrals means that the integration is over the dot (well) region. A very relevant feature of this method is that all the integrals in Eq. (5) can be performed analytically.

For expanding the envelope function, we have used a base of 19 wave functions in each direction for the pyramids and 14 eigenfunctions for the cuboids which is the minimum number of functions required to achieve convergence for the electronic energy eigenvalues to within less than 1 meV. Standard mathematical software such as LAPACK⁷ is then used to solve Eq. (4), where $M_{lmnl'm'n'}$ is a 6859×6859 (respectively, 2744×2744) matrix.

III. RESULTS AND DISCUSSION

In the self-assembled pyramidal dot material the effective masses differ from the unstrained ones due to the compressive stress which alters the curvature of the bulk bands. We have used the value of $0.04m_0$ ⁸ for the effective mass of InAs in the conduction band (the unstrained value is $0.023m_0$), to account for the strain as suggested by Cusack *et al.*⁹

Most authors use two different values for the hole effective masses, one along the symmetry axis z and the other along the plane xy normal to that axis, to account for the mass anisotropy. This choice nevertheless, increases the (computational) complexity of the treatment, without improving the approximation, since the mass for the motion along transverse directions (where the holes spend the most of their time) remains undefined. Furthermore, the in-plane masses m_{xy} , of electrons, light and heavy holes have been revealed by accurate calculations in quantum wells (using the pseudopotential method),¹⁰ and in quantum dots,³ to be similar to those commonly accepted for the motion along the z axis. Therefore, in our calculations the hole effective mass has been restricted to one value $m_{hh} = m_{hh,z} = 0.590m_e$. This value was estimated by Cusack *et al.*¹¹ through empirical pseudopotential and *ab initio* local density calculations,

TABLE II. Calculation parameters: m_W well region effective mass; m_B barrier region effective mass (both in units of m_0); V_0 carrier confining potential (in meV).

Electron			Heavy hole		
m_W	m_B	V_0	m_W	m_B	V_0
0.040	0.0665	450	0.59	0.3774	266

along the [001] (z) direction near the center of strained InAs pyramidal structures with an aspect ratio of half base to height, $Q_p = b/2h = 1$.

Also the confining potential of the carriers is affected by the strain and differs from the square well formed by the difference in the absolute energy of the conduction or valence band edges in the bulk barrier and dot material.⁹ The square well (constant potential) approximation however still gives good results for the conduction band,³ thus the strained value of the electron confining potential has been taken as the average over the QD, i.e., $V_0 = 450$ meV.³ The same treatment has been adopted for the heavy-hole confining potential, whose average value has been deduced (see Ref. 4 for further details) by fitting the theoretical data for strained pyramids of Cusack *et al.*⁹

The same values for effective masses and potentials (see Table II) have been used throughout the calculations for both cuboids and pyramids and for all the dot sizes considered.

We would like to emphasize that although the Schrödinger equation does not explicitly include the strain (that is, we do not diagonalize any 8×8 strain Hamiltonian matrix such as that in Ref. 12), its effects are nevertheless accounted for by the choice of strained values for effective masses and average confining potentials (for both electrons and heavy holes), relative to the pyramidal dot case.

The inclusion of a detailed treatment of the microscopic effects of the strain would, in fact, introduce excessive complexity in the model and would be beyond the scope of this work (which aims to provide the experimentalists with a simple, easy-to-code and quick-to-run method to analyze their spectra). In order to justify the choice of the parameters used in our calculations we, nevertheless, performed a set of calculations of the strain distribution in pyramidal structures with aspect ratio Q ranging from 1 to 4.5 using a method based on the Green's function technique,⁵ taking into ac-

count the anisotropy of the elastic properties as well.¹³ The carriers' strained confining potentials were then calculated as a function of position along the growth direction, in the framework of the eight-band $\mathbf{k} \cdot \mathbf{p}$ theory (see, for example Ref. 14). The results were in agreement with previously published data,^{11,15} and showed that the confining potentials of both electrons and holes are almost constant throughout the dot for $Q \geq 2$, with average values centered around 450 meV (for the electrons) and 266 meV (for the heavy holes), proving our constant-confining-potential approximation as a reasonable choice. The very good agreement obtained by our simple model with several experimental transition energies (see later) is nevertheless hard to explain. A compensation mechanism in which a positive difference in the confining potential [i.e., between our average strained value of 450 (266) meV and the average strained value calculated for the specific sample] is compensated by a negative difference in the value of the effective mass, (so that when the confining potential is < 450 meV, the effective mass is > 0.04 , and vice versa), could be responsible for this little difference in the transition energies.

Figure 1 presents the results for the electron and heavy-hole ground state energy levels of InAs square-based quantum pyramids and cubes, both with aspect ratio $Q = 1$ [for the cuboids, unlike the pyramids where $Q_p = b/(2h)$, define the aspect ratio $Q_c = b/h$], as a function of the cubic root of the volume, plotted relative to the unstrained GaAs conduction and valence band edge, respectively.

Despite the simplicity of the calculations, the ground state electronic and heavy-hole energies given by our method agree very well with previous, more sophisticated, theoretical studies of InAs self-assembled pyramidal dots^{3,9} [a comparison with those methods, as well as with experimental data, has been presented elsewhere:⁴ it is shown that the values ($m_{W, hh} = 0.59$, $V = 266$ meV) reported in Table II have proved to reproduce well the ground state energies of all the experimental spectra considered].

The model, as presented in Fig. 1, predicts no bound electron states for base lengths smaller than about 60 Å (i.e., $V^{1/3} < 33$ Å).

From Fig. 1 it is evident that for the ground state energy levels the relationship

$$E_{\text{gs}}^{\text{pyr}}(V) = E_{\text{gs}}^{\text{cub}} \left(\frac{V}{\alpha^3} \right) \quad (6)$$

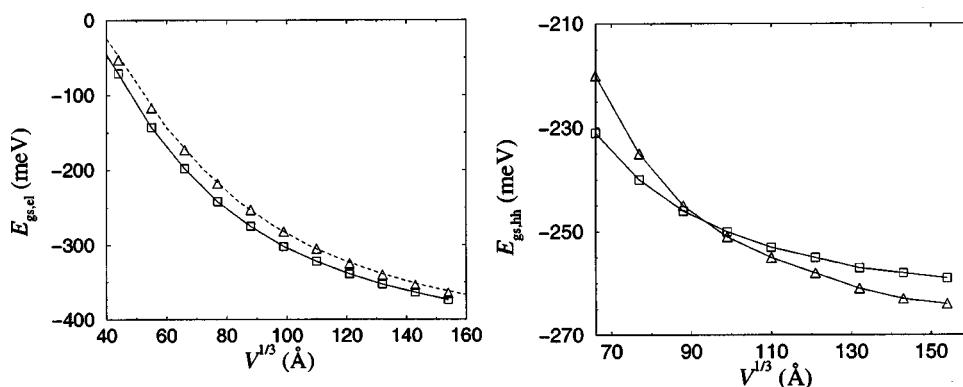


FIG. 1. Electron (left-hand side) and heavy-hole (right-hand side) ground state energy levels as a function of the cubic root of the volume, for InAs square-based pyramidal QDs (triangles) and cubes (squares), (both with aspect ratio $Q = 1$), with respect to the unstrained GaAs conduction and valence bands respectively. Fit for $\alpha = 1.091$ dashed line.

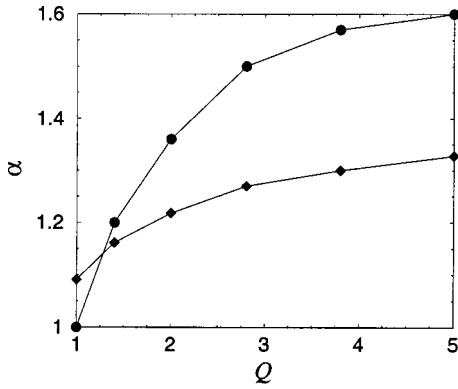


FIG. 2. Coefficient α of Eq. (7) as a function of the aspect ratio Q , for both electrons (diamonds) and heavy holes (circles).

holds, with $\alpha=1.091$ (dashed line in the figure) for the electrons (it is interesting to note that a similar value for the proportionality constant $\alpha_{el}(Q=1)=1.064$ has been found for the GaAs/Ga_{0.63}Al_{0.37}As system. This value constitutes an improvement to the one, calculated by means of a different approach, reported in a previous article¹⁶), and $\alpha=1$ for the heavy holes. In this particular case (i.e., $Q=1$), the heavy-holes ground state energies of the cubes are the same, to within 5%, as those of the pyramids, for $66 \text{ \AA} < V^{1/3} < 150 \text{ \AA}$, which is the region of interest, in which the typical (uniformly sized and distributed) experimental self-assembled pyramidal dot dimensions range.^{17–21} Nevertheless we have found that even for structures of a given shape and volume, E_{gs} varies depending on the particular aspect ratio Q of the dot. This variation is volume dependent in the sense that the range of Q within which $\Delta E_{gs}=[E_{gs}(Q) - E_{gs}(Q=1)]/E_{gs}(Q)$ (i.e., the percentual variation of the ground state energy of a structure with a given Q , relative to that of a structure with $Q=1$) is, say, 3%, is smaller for small volumes than it is for large volumes. In other words the variation of the ground state energy with Q is smaller the bigger the dot. This holds for both cuboids and pyramids, but the size of the variation is different for the two shapes. We have found that, by varying the aspect ratio Q of the square-based pyramids, the relationship in Eq. (6) remains valid (provided that a square-based cuboid with the same aspect ratio as the pyramid is used), but the proportionality constant α becomes a function of Q . Furthermore, two different $\alpha(Q)$ have to be used, one for the electrons and the other for the holes, as shown in Fig. 2, where every point has been obtained as the coefficient producing the best fit of the pyramidal dot energy curve to the cuboidal one [after the relationship in Eq. (6), and as illustrated in Fig. 1]. Equation (6) therefore becomes

$$E_{gs,carr}^{pyr}(V,Q) = E_{gs,carr}^{cub} \left[\frac{V}{\alpha_{carr}^3(Q)}, Q \right], \quad (7)$$

where carr (=carrier) implies electrons or heavy holes.

It is important to mention that by using the same parameters (i.e., strain induced effective masses and strained average confining potentials) for both cuboidal and pyramidal calculations we are not implicitly assuming the strain distribution in a quantum box to be the same as that in a pyramid

TABLE III. Comparison between the results of square-based pyramidal and cuboidal calculations [after the connection rule in Eq. (7)], and the peak positions of the experimental PL spectra, for the ground state transition energies of the samples of Refs. 19–21.

	E_{00}^{pyr} (eV)	E_{00}^{cub} (eV)	E_{00}^{exp} (eV)
Schmidt <i>et al.</i> ^a	1.017	1.019	1.01
Murray <i>et al.</i> ^b	0.962	0.964	0.961
Noda <i>et al.</i> ^c	1.199	1.195	1.220

^aSee Ref. 19.

^bSee Ref. 20.

^cSee Ref. 21.

(which is not the case, as shown, for example, in Ref. 5). We are proposing an alternative computational method for calculating the electronic ground state energy of self-assembled pyramidal quantum dots, that exploits the proportionality between the solutions of the pyramid and those of a much simpler and more symmetrical structure: the cuboid. In other words, given a pyramidal QD, whose electronic structure is determined by a characteristic set of parameters, its ground state energy can be obtained from a calculation where the pyramidal shape is replaced by a cuboidal one and use is made of the simple proportionality rule in Eq. (7). The cuboid therefore enters only as a computational expedient which allows us to greatly simplify the calculations (and not as a proper QD with its specific parameters which, due to a different strain distribution in that structure, are expected to be different from those of the pyramid). The advantage is that the integrals involved in the cuboidal calculations are much simpler than those relative to the pyramidal case (the mathematic involved extends over about 6 code lines compared to more than 600 in the latter), resulting in a substantially (about 20 times) lower computational time.

Utilizing Eq. (7) and $\alpha(Q)$ both for the electrons and for the heavy holes (from Fig. 2), the connection rule model has been applied in order to predict the position of the ground state transition peak in the experimental PL spectra of the samples grown by Schmidt *et al.*,¹⁹ Murray *et al.*,²⁰ and Noda *et al.*,²¹ which have aspect ratios Q of 1.428, 2.857, and 4.564, respectively, [for the latter, which was a rectangular-based pyramid, we defined the aspect ratio as $Q = \sqrt{b_x b_y} / (2h)$], and thus cover a large portion of the Q variation range. The results are presented in Table III together with the pyramidal calculation results and the experimental data.

The ground state transition energies calculated with the cuboidal approximation, following the connection rule in Eq. (7), agree very well both with those obtained by means of the full pyramidal calculation and with the experimental values of such transitions. This agreement not only proves the validity of the connection rule in Eq. (7), but the accuracy of the $\alpha_{carr}(Q)$ curves (in Fig. 2) as well.

It has now been proved that by knowing the dimensions of the pyramidal dot it is possible to deduce easily and without much computational effort, the ground state transition energy and therefore the position of the first PL spectrum peak, using the energy of an equivalent square-based cuboid with the same aspect ratio (and the same characteristic pa-

TABLE IV. Theoretically predicted [after the connection rule in Eq. (7), and the $\alpha_{\text{carr}}(Q)$ curves of Fig. 2], dimensions of a pyramidal dot with a ground state transition energy $E_{\text{trans}}^{\text{gs}}=961$ meV, and Q ranging from 2.5 to 3. The actual dimensions from Ref. 20 are also displayed for comparison.

Q	h (Å)	b (Å)
2.5 (theory)	72	360
2.857(exp) ^a	70	400
3.0 (theory)	68	409

^aSee Ref. 20.

rameters) as the pyramid, whose volume is determined by Eq. (7). In addition the versatility of the model is such that, starting from the PL ground state peak position, it can be applied in reverse, to deduce the dimensions of the pyramidal dot, allowing those experimentalists who do not have access to microscopic analysis equipment to estimate the size of the dots they have grown (though they do need to have an estimate of the aspect ratio Q). The latter does represent a limitation to the applicability of the model, but the aspect ratio can often be estimated from supporting microscopy of similarly grown samples.

As an example of this (reverse) application of the model, we have deduced the dimensions of the dots whose ground state energy transition is 0.961 eV and whose aspect ratio ranges between 2.5 and 3 (i.e., the pyramids grown by Murray *et al.*,²⁰ which have $h=70$, $b=400$, and $Q=2.857$). The procedure is to draw two (one for the electrons and the other for the heavy holes) graphs similar to those reported in Fig. 1, for each value of the aspect ratio. Since the cuboidal equivalent volumes for the electrons $V_{\text{el}}^c=V_p/(\alpha_{\text{el}}^3)$ are different from those for the heavy holes $V_{\text{hh}}^c=V_p/(\alpha_{\text{hh}}^3)$ (the constant α being different), one of the graphs, say the electron one, needs to be rescaled by the factor $\alpha_{\text{el}}/\alpha_{\text{hh}}$ ($\alpha_{\text{hh}}/\alpha_{\text{el}}$ has to be used for the heavy-hole curve), before direct comparison to the other. Now in each pair of graphs, for the same Q , there is now only one value for the volume (of the equivalent cuboid), for which $E_{\text{gs,el}}$ and $E_{\text{gs,hh}}$ satisfy

$$E_{\text{gs,el}}+E_{\text{gs,hh}}=E_{\text{gs,tr}}-E_{\text{GaAs}}, \quad (8)$$

where $E_{\text{gs,tr}}$ is the experimental ground state transition energy and E_{GaAs} is the GaAs band gap. The volumes of the pyramids are finally obtained by using Eq. (7). The results are displayed in Table IV together with the experimental values.

The accuracy of the prediction depends on the accuracy to which Q is known. Otherwise by knowing one of the dot dimensions, say h , (which, as the case of Sauvage *et al.*² shows, is a condition more likely to happen than to have an estimate of its aspect ratio) and the ground state transition energy, our model allows the determination of the aspect ratio and thus the other dimension, b . Unfortunately in this case the connection rule is not applicable and we have to resort to a full pyramidal calculation.

IV. CONCLUSIONS

A theoretical investigation of the ground state electronic structure of InAs/GaAs quantum confined structures has been presented. Energy levels of cuboids and pyramidal shaped dots have been calculated using a single-band, constant-confining-potential model that has proved⁴ to well reproduce both the prediction of more sophisticated treatments and several features of the experimental PL spectra.

Aiming to provide experimentalists with a simple method, easy-to-code and quick-to-run, with very limited computational demands, to analyze their spectra, a connection rule has been found which allows calculation of the energy levels of pyramidal dots starting from those of cuboids of the same material (that is, with the same effective masses and confining potentials as the pyramid), aspect ratio and of suitably chosen dimensions. As an example, this rule has then been applied to successfully reproduce the position of the ground state transition peaks of some experimental PL spectra relative to self-assembled pyramidal dots. Furthermore, starting from the knowledge of the ground state transition energy and an estimate for the aspect ratio Q of a pyramid, the rule has been used to predict its dimensions.

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