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THE ELECTRONIC SPECTRUM OF THE THREE-DIMENSIONAL QUASICRYSTAL

D.V.Olenev[1], *E.I.Isaev, P.V.Slobodianiuk, Yu.Kh.Vekilov*

Moscow State Institute of Steel and Alloys, Theoretical Physics Department, 117936 Moscow, Russia

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The electronic spectrum of the icosahedral quasicrystal with a central atom decoration of Amman – Mackay network is investigated in the tight binding approximation. The quasicrystal is described as a structural limit of the optimal cubic approximants with increasing period. The electronic spectra for the first four optimal cubic approximants do not contain the hierarchical gap structure which is typical for Cantor set of the spectrum of one-dimensional quasicrystal. At the same time the spectrum with the increase of the order of approximant becomes singular in the whole energy scale.

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Quasicrystals are the objects which have non-crystallographic symmetry and positional long-range order (i. e. they coherently scatter electrons or X-ray) [1]. Quasicrystals reveal unusual physical properties. They have the linear temperature contribution to the electronic heat capacity which is a bit smaller than the one in the simple metals [2]. Recent experiments show that below $4K$ with increasing the structural order of quasicrystal the temperature dependence of $c_v(T)$ becomes exponential (this indicates the presense of a gap in the electronic spectrum) [3]. The resistivity of quasicrystals is anomalously high (it reaches nearly 2 Ω·cm at 0.5 K in the stable icosahedral phase of the system AlPdRe [4]). It strongly depends on the phase stochiometry. It decreases when the temperature increases and increases at increasing structural order as well as annealing of defects [2, 5]. The Hall constant of quasicrystals is large and strongly temperature dependent, the optical conductivity does not follow Drude's law, the thermoelectric power is also strongly temperature dependent [2, 6]. In the weak magnetic field the negative magnetoresistance is observed [2]. Most quasicrystalline phases (even alloys with transition elements) are weak diamagnetics in a wide temperature interval [6]. It is obviously that all the mentioned specific properties are tightly connected with electronic spectra of the quasicrystals.

The electronic spectra of quasiperiodic objects have been studied in detail for the cases of one- and two-dimensional quasicrystals [7 – 10]. The energy spectrum of one-dimensional quasilattice (two-fragment Fibonacci chain) consists of a Cantor set of Lebesque measure zero with self similar gap structure, and the wave functions are critical, being neither localized nor delocalized [7, 8]. In the two-dimensional case the energy spectrum contains a singular (non smooth) part and the most wave functions are critical [9, 10]. As a consequence of the exotic electronic structure of one- and two-dimensional quasicrystals there is a power law dependence of resistivity via object size [9]. The character of the electronic spectrum and the wave functions of three-dimensional quasicrystals with Amman – Mackay network structure have not been studied enough. Many explorers incline to think that this system does not contain any peculiarities in the behaviour

[1] e-mail: olenev@trf.misa.ac.ru

of the wave functions and energy spectrum (see, for instance [11]). For example, Marcus in his work on the electronic properties of the three-dimensional quasicrystal in the tight-binding approximation has not found any fundamental differences between those of quasicristals and crystals. He has concluded that the energy spectrum is smooth and the wave functions are delocalized [12]. On the other hand Niizeki and Akamatsu proposed that in the three-dimensional case "critical" wave functions do exist and that the energy spectrum has an exotic, singular continuous form [13]. The same results were obtained in the papers [14–16]. Recently the structural model of three-dimensional icosahedral quasicrystals has been proposed where the quasicrystal is made with the help of the set of four tetrahedra [11]. The electronic spectrum of the three-dimensional quasicrystal with such a structure reveales the strong oscillations of the density of states (DOS) in the whole energy range and the localization of selective states on the special topological configurations [11]. So the existing results are contradictory and new investigations are needed.

In the present work the electronic energy spectrum of icosahedral quasicristal is investigated in the tight-binding approximation. As quasicrystals do not possess translational symmetry and traditional methods for the calculations of the band structure of solids based on Bloch's theorem are no longer applicable, the objects of study are optimal cubic approximants of the icosahedral quasicrystal. The quasicrystal is regarded as the structural limit of a sequence of optimal cubic approximants with an increasing period and the electronic properties of approximants are investigated in the tight-binding approximation. The approximants are constructed by the projection method [17]. The Hamiltonian with constant transfer integrals for the nearest neighbours has been used to minimize the amount of free parameters (as the result for one- and two-dimensional quasicrystals show this type of Hamiltonian makes it possible to reproduce the characteristic features of the quasicrystalline object and to consider qualitatively the influence of quasiperiodicity on electronic structure of quasicrystals of the corresponding dimension [2, 7–10, 18]).

We have considered a "central" decoration of the approximants: the atoms with one s-orbital per atom are located at the centers of rhombohedra. In this case one can write down the Hamiltonian of the system as follows

$$H = \sum_{j,j\neq i} |i\rangle t_{ij} \langle j|$$

where the transfer integrals are chosen equal to a nonzero constant ($t_{ij} = -1$) only for the nearest neighbors – for atoms belonging to rhombohedra which have a common face (taking into account that the next neighbors do not introduce anything qualitatively new and only complicate the calculations). Using the projection method the first four optimal cubic approximants of the icosahedral quasicrystals have been investigated, namely 1/1, 2/1, 3/2, 5/3 approximants. The approximant f_{n+1}/f_n (where f_n is the Fibonacci sequence with $f_0 = f_1 = 1$, $f_n = f_{n-1} + f_{n-2}$ and $\lim\limits_{n\to\infty} f_{n+1}/f_n = (1+\sqrt{5})/2 = \tau$) is connected with replacement by rational number f_{n+1}/f_n of the irrational number $\tau = (1+\sqrt{5})/2$ which describes the incommensurability of the icosahedral structure and is used for constructing the mentioned above approximants. With the increase of n the number of atoms in the basis the period of f_{n+1}/f_n approximant increases and the approximant "approximates" the icosahedral quasicrystal more accurate. The case $n \to \infty$ describes

the icosahedral quasicrystal. The unit cells of the considered approximants contained 32, 136, 576, and 2440 atoms in the basis, respectively.

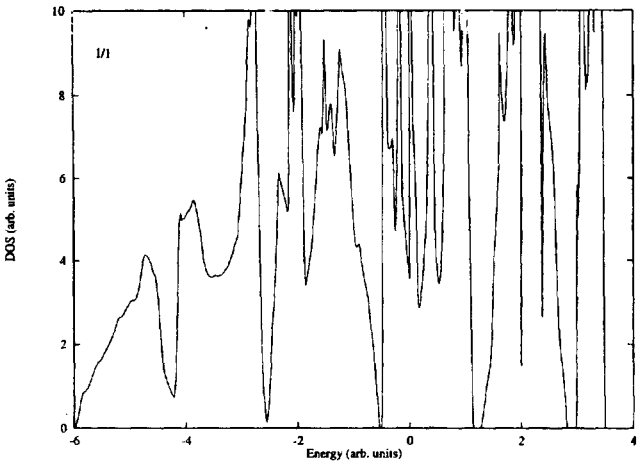

Fig.1

The results of DOS calculations are presented in Figures. The calculations for the first three approximants $(1/1, 2/1, 3/2)$ (Figs.1-3) have been done by the tetrahedra method using the values of energy levels in 40 **k**-points of the irreducible part of the Brillouin zone of the corresponding approximant. For $5/3$ (Fig.4) approximant we used only the 4 **k**-points in the irreducible part of the Brillouin zone. Because of a rather small volume of the Brillouin zone for $5/3$ approximant (with respect to the volume of the Brillouin zone for the first approximant) the increase of the number of **k**-points in calculation should weakly influence the calculated DOS. One can see from Figure that with the increase of the order of periodic optimal approximant the DOS curves become less smooth and more "spiky". This confirms the conclusion about the presence of the singular (non smooth) part in the energy spectrum of the icosahedral quasicrystal obtained by the level statistic method [14, 16]. In the opposite case (without singular part) the DOS is a smooth curve. Besides, one can see from Figure that the smoothness of the spectrum depends on the energy range: the spectrum is smoother in the range of small energies and the strong oscillations present mostly in high energy range. However, with the increase of the order of approximant the length of the smooth region of the energy spectrum decreases. For this reason one can believe that in the thermodynamic limit $(n \to \infty$, i. e. the case of quasicrystal) the strong DOS oscillations (a singular part of the spectrum) present in the whole energy range including the longwave part of the spectrum, that demonstrates that the energy zones in quasicrystal are flat and practically dispersionless (this obviously influences the optical properties of quasicrystals). The plots in Figure are in good agreement with DOS calculations for the first four periodic approximants carried out by Carlson in the nearly free electron approximation [19]: the gap is seen in the region of small energies of the spectrum of $5/3$ approximant, though in the energy spectrum of lower order approximants the like gaps are not exist.

Using the calculated DOS we determined the Lebesgue measure of the energy spectra of the investigated approximants. The Lebesgue measure of one-dimensional set S is defined as an exact low boundary of the length sum of the finite or countable set of intervals which

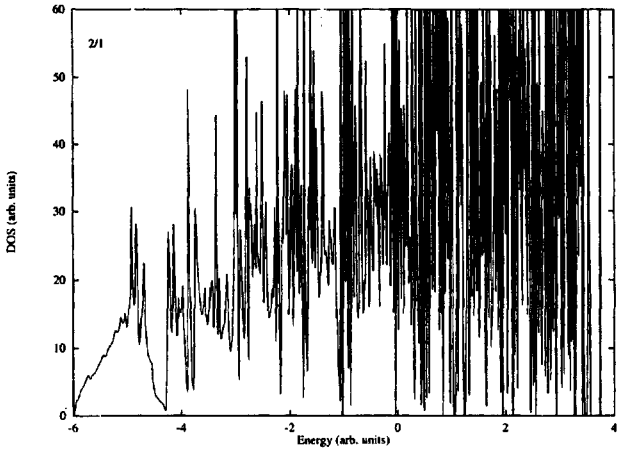

Fig.2

cover S. The Lebesgue measure of the energy spectrum was calculated as a total length of the permitable regions in the energy range [-6;6] (according to the well-known theorem on spectrum boundaries [20] the energy levels of the investigated system belong to the interval $-6 \leq E \leq 6$). The Lebesgue measures of the energy spectra for 1/1, 2/1, 3/2, 5/3 approximants normalized on the total band width B (because of the fast convergence of the integrated DOS for considered approximants the B value for 5/3 approximant has been choosed) turned out to be equal to 0.95, 0.98, 0.98 and 0.97, respectively. For this reason one may conclude that for the three-dimensional quasicrystal the dependence of the Lebesgue measure of the energy spectrum with respect to the order of approximant is small in contrast with the case of one-dimensional quasicrystal, where the Lebesgue measure of the energy spectrum of the periodic approximant decreases with system size according to a power law [7, 8]. A rather small difference of the Lebesgue measures of the energy spectra of the two latter approximants shows that the energy spectrum of three-dimensional quasicrystal occupies the region of a finite width on the energy scale. Besides that the values of the normalized Lebesgue measures of the energy spectra of considered approximants (which close to 1) and the tendency to the convergence of the results shows that the spectrum of the icosahedral quasicrystal does not have large energy gaps. So the energy spectrum of the three-dimensional quasicrystal has the "noncantor" (non self similar) character. This has to influence on the specimen length dependence of conductance and the related properties of three-dimensional quasicrystal.

In conclusion the results show that the electronic spectrum of the three-dimensional icosahedral quasicrystal with Amman – Mackay network structure significantly differ from the electronic spectrum of the one-dimensional quasicrystal. Like in the case of the one-dimensional quasicrystal the energy spectrum of the three-dimensional quasicrystal contains the singular part. However, due to the other topology the structure of the energy spectrum of icosahedral quasicrystal is nonself-similar and this is the reason of nonzero Lebesgue measure of the energy electronic spectrum of a three-dimensional quasiperiodic object. Besides that the energy spectrum of the investigated model of the icosahedral quasicrystal does not contain big gaps, and the quasiperiodicity of the structure is the reason of very strong oscillations of the DOS in the whole energy range including the long

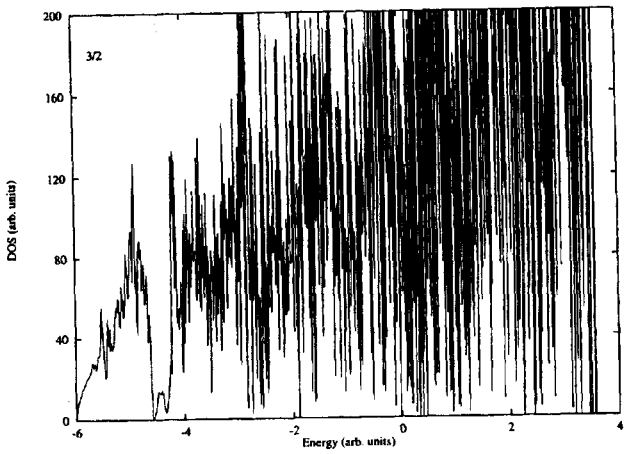

Fig.3

wave part of the spectrum (the same is obviously has to appear for the other excitation spectra of quasicrystal-phonons etc.). As a consequence the energy zones in quasicrystal are flat and practically dispersionless, the electrons are quasibound and have zero group velocity, and this is obviously the cause of anomalously low electric conductivity, low excitation energies of quasiparticles on the Fermi level, unusual magnetic properties (diamagnetizm) of quasicrystalline alloys [21]).

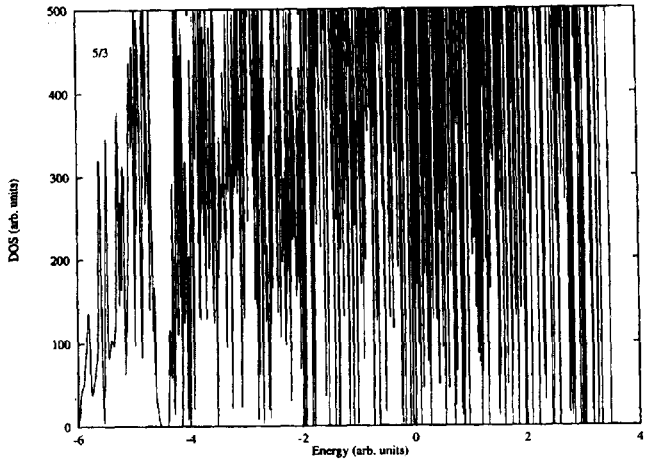

Fig.4

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