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*Discussion of the Surface Science of Quasicrystals*

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**Abstract.** This paper contains a short review of four aspects the surface science of quasicrystals, together with a challenging list of open questions to be addressed by the scientific community in the near future. The first issue concerns the ability of surface science to shed light on bulk atomic structure. The second is the use of surfaces as quasiperiodic templates, particularly for films of periodic metals. Here, enforcing quasiperiodicity in the film may lead to unusual magnetic, tribological, or adsorption properties. The third concerns the effects of surface phasons and phonons on dynamical interactions with adsorbates, such as sticking coefficient, as well as on diffusion between the surface and near-surface region. The final area is tribology, where studies of quasicrystals have suggested that both adhesion and phononic friction may be important.

## Introduction.

At the recent conference “Quasicrystals—The Silver Jubilee”, a discussion panel consisting of Julian Ledieu, Vincent Fournée, Ronan McGrath, and Pat Thiel was asked (by the conference Chair, Ron Lifshitz) to consider the following questions:

1. In what ways can surfaces be more useful in testing the validity of structural models?
2. Using surfaces as quasiperiodic templates - just a toy or useful for something?
3. What are the potential effects of surface phonons/phasons on surface structure or properties?
4. Tribology - What have quasicrystals taught us that can be extrapolated to other materials?

This article is by no means an accurate record of the discussion that ensued, but is very much motivated by it. It reflects the author’s own views on the subject, and is provided here in the hope that it will inspire new research and lead to exciting discoveries.

### 1. Structural models.

In recent years, our community has been intrigued by the possibility that surface science may be able to help distinguish between different bulk structure models. This is because beautiful detail is provided by STM images, and this detail can be correlated with atomic arrangements in bulk structural models[1-4]. Both local motifs and longer-

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3 range tilings have been examined. However, there has been no attempt to use the STM  
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5 data to distinguish between different models. There have been several successful attempts  
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7 to correlate an individual set of data with an individual model, but in each case the  
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9 question remained unanswered: Would a different model have provided a better or worse  
10  
11 fit?  
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15 Another complication is illustrated by the well-studied 5-fold surface of  
16  
17 icosahedral Al-Pd-Mn. Within a given model, there are two main families of possible  
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19 surface terminations [5]. At this point, the identity of the family remains controversial.  
20  
21 Papadopolos et al. argue in favor of a family in which cut Bergmans are a dominant motif  
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23 [6]. Krajci et al. choose the other family, in which cut Mackays are a dominant motif,  
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25 based on DFT [7]. The problem illustrated here is that uncertainty about the structural  
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27 model is convoluted with uncertainty about the type of preferred termination. This  
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29 problem exists mainly for the icosahedral phases.  
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34 At the Conference, the opinion was expressed that surface science in fact will not  
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36 be able to distinguish between different models. This may be too pessimistic, since the  
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38 different models differ significantly in their chemical decorations. Focusing on the  
39  
40 chemical decorations of local motifs—local adsorption sites—may be a successful  
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42 strategy. (The results would, however, be subject to uncertainty from the possibility of  
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44 surface segregation or selective depletion by evaporation.) How can the chemical  
45  
46 decorations of local motifs be **measured? One approach** might be to use adsorbates that  
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48 are sensitive to chemical environment to probe the adsorption sites, which is a strategy  
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50 that has been used successfully at bimetallic surfaces of periodic crystals [8].  
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In considering data from experimental surface science, it must be kept in mind that there are two main types of techniques. First are those that provide information that is averaged over a macroscopic surface area, typically 1-4 mm<sup>2</sup>. Such an area includes many terraces. This category includes LEED, XPD, and LEIS. Second are those that provide microscopic information about individual features in real space. Chief among these is STM, providing information on the sub-nanometer scale. In STM, the main challenge is that of obtaining statistically-significant information about local motifs, and about characteristics of different terraces on a surface. While STM provides beautiful local detail at high magnification, that detail is lost at the low magnifications that are most useful for statistical averaging. It is important to know the degree to which surface features observed in STM are representative! Overcoming this problem is possible, but it is time-consuming and difficult, and there has been little attempt to do so to date. In the discussion at the Conference, it was noted that there may also be a strong bias introduced by the natural tendency of the experimentalists to choose the most beautiful STM images for publication; it is conceivable that some types of terraces do not image as well as others.

## **2. Quasiperiodic templates.**

One intriguing possibility for using quasicrystals as templates, is in growth of magnetic films. Theoretical work has predicted unusual magnetic structures associated with quasiperiodicity [9-13]. To date, the search for quasiperiodic structure in films of magnetic metals has been intense, but unsuccessful. Studies of Fe, Ni, and Co deposited on d-Al-Ni-Co and i-Al-Pd-Mn have shown that the deposited metal either intermixes

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3 with the substrate [14-16] [17], or otherwise fails to adopt quasiperiodic structure on the  
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5 atomic scale as would be necessary to test the theoretical predictions [18-21] [17]. This  
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7 has been disappointing, given that a number of other studies had reported pseudomorphic  
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9 growth, either short-range or long-range, of non-magnetic adsorbates on quasicrystals  
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11 [22-25] [17, 26], and it was naturally expected that pseudomorphism would be found in at  
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13 least some magnetic adsorbates as well.  
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17 However, it is possible that pseudomorphic growth can still be achieved by tuning  
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19 growth variables, namely flux and deposition temperature, along with their temporal  
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21 variation. These control the kinetics of formation of local structures, and they have not  
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23 yet been exploited in magnetic film growth. An especially interesting aspect of the  
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25 kinetics of film growth is the influence of specific adsorption sites acting as trap sites  
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27 [27], which may especially favor local pseudomorphic nucleation. Another possibility  
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29 for controlling growth is the use of surfactants. As an example, fluorine suppresses  
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31 interdiffusion when Fe films are grown on Cu, and the F can be removed cleanly by  
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33 electron stimulated desorption after growth [28]. Some work exploring the effect of In as  
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35 a surfactant in Au and Pt films on d-Al-Ni-Co quasicrystals has been reported. There, the  
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37 system is apparently dominated by the formation of Au-Al or Pt-Al surface alloys, and  
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39 the In surfactant did not prevent this, [29-33] although one can speculate that other  
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41 surfactants might be more effective.  
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48 Quasicrystals may also be useful as templates for non-magnetic pseudomorphic  
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50 films, in order to test whether basic properties are adopted by the pseudomorphic film.  
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52 Recent results show that a pseudomorphic monolayer of Pb on five-fold i-Al-Pd-Mn  
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54 exhibits an electronic pseudogap, which is an important result [25]. It would also be very  
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3 interesting to test friction forces on a pseudomorphic quasicrystalline film, to determine  
4 whether the low friction associated with the substrate propagates to the film. Finally, the  
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6 adsorption properties of pseudomorphic films would be of significant interest; one report  
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8 has appeared recently on this subject [17], for C<sub>60</sub> adsorbed on a pseudomorphic Pb film  
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10 on d-Al-Ni-Co. Here, the adsorbate formed a hexagonal lattice, indicating that the  
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12 corrugation of the potential energy surface is sufficiently weak that adsorbate-adsorbate  
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14 interactions dominate. However, much remains to be explored, including not just the  
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16 static structure but also the adsorption dynamics as mentioned in the following section.  
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### 25 **3. Phonons and phasons.**

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27 The effect of phonons and phasons at surfaces of quasicrystals has received some  
28 attention [34-38], but relative to atomic or electronic structure the level of scrutiny has  
29 been small. There are at least two surface phenomena that could be affected by these  
30 types of dynamics: Mass transport between the surface and the bulk; and energy  
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32 dissipation at surfaces.  
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39 Phasons may facilitate mass transport between the surface and the bulk. Phasons  
40 play a role in bulk self-diffusion, since measurements of diffusivity in i-Al-Pd-Mn have  
41 been interpreted to mean that for some metals (Pd and Au), diffusion is assisted by  
42 collective phason flips involving many (20 to 30) atoms [39]. This was based upon an  
43 Arrhenius analysis of the diffusivity for these elements. The Arrhenius representation of  
44 the data consisted of two straight lines intersecting at 720 K, indicating different  
45 processes, with the low-temperature process (assigned as phason-assisted diffusion)  
46 having a lower activation barrier and an extremely low pre-exponential factor. Very  
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3 similar behavior was observed, and a similar interpretation was given, in a study of Al  
4 migration from the bulk to the surface of i-Al-Cu-Fe [34]. In the latter work, migration  
5 of Al to the surface was triggered by exposure to oxygen, which caused selective  
6 oxidation of Al at the surface. The Arrhenius plot of the rate of Al diffusion showed two  
7 straight lines intersecting at 770 K. From this, an activation barrier of 0.6 eV was  
8 estimated for phason-flip transport of Al below 770 K. This barrier was at least a factor  
9 of 2 lower than the barrier for normal vacancy-mediated self-diffusion of Al. Recently,  
10 another study showed that when Pd and Mn were deposited on the surface of i-Al-Pd-Mn  
11 and then were allowed to diffuse into the bulk by heating above room temperature,  
12 diffusion into the bulk was characterized by anomalously low activation barriers. These  
13 low barriers were attributed to phasons as well [38]. Overall, the evidence for phason-  
14 assisted diffusion between surfaces of quasicrystals and the underlying bulk is tantalizing.

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Phason and phonon excitations represent channels of energy dissipation, both in  
the bulk and at the surface. Energy dissipation at surfaces affects phenomena as diverse  
as spectroscopic linewidths, adsorption probabilities (sticking coefficients), ballistic  
diffusion, and tribological friction [40]. Spectroscopic linewidths of adsorbates on  
surfaces are determined by the lifetimes of excited states, which in turn are determined by  
the surface's efficiency in quenching the excited states. Sticking coefficients depend, in  
some circumstances, on the ability of the surface to absorb the translational or other  
energy components of the incoming gas particle. Ballistic diffusion occurs when an  
adsorbate temporarily retains the energy released by the adsorption event, coming to rest  
when that energy is absorbed by the substrate. In friction, also, energy dissipation is key.  
It is discussed more fully in the following section. Other examples abound [40]. The main

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3 point is that many phenomena in surface chemical dynamics share a common basis in  
4 energy dissipation. Hence, any of these phenomena could reflect dynamic peculiarities of  
5 quasicrystals that would be important in defining the surface properties of quasicrystals—  
6 particularly in terms of their dynamic interactions with adsorbates—yet none has been  
7 investigated. Hence, this is an interesting avenue of possible future research.

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15 It is difficult to predict whether quasicrystals will be relatively more or less  
16 efficient in surface energy dissipation. On one hand, phasons represent a channel for  
17 energy dissipation that is not available in periodic crystals. On the other hand, there is  
18 some expectation that cross sections for phonon excitations at surfaces may be relatively  
19 small in quasicrystals [37].

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27 Phasons themselves present interesting challenges to our understanding of  
28 quasicrystals, as discussed by Widom elsewhere in this issue [41].  
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#### 34 **4. Tribology.**

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36 Classically, the friction coefficient,  $\mu$ , is cast as the sum of two terms: One due to  
37 adhesion between the sliding interfaces, and one due to mechanical resistance to  
38 formation of the wear track, called ploughing [42]. These two terms alone are not  
39 sufficient to account for measured values of  $\mu$  between unlubricated metals, which leads  
40 other factors to be invoked, such as junction growth, and phononic friction (excitation of  
41 lattice phonons at the interface). Electronic friction (excitation of electron-hole pairs at  
42 the interface) can also play a role, but its contribution is thought to be generally small  
43 compared with phononic friction [43-45].  
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Quasicrystals exhibit low coefficients of friction in dry sliding, relative to other materials of similar composition [46]. Because quasicrystals are hard, it has sometimes been suggested that their hardness alone may account for their low coefficients of friction [47]. However, over the past several years, there have been two major sets of studies focusing on the fundamental origin of low friction in quasicrystals. Both have pointed to factors other than hardness as being very important. In one set, comparisons were made between quasicrystals and a large group of related metals and oxides [48-55]. The friction coefficient was measured in high vacuum with a pin-on-disc tribometer under carefully standardized conditions. This work showed that the low friction of quasicrystals did not correlate with their hardness, but rather with their reversible work of adhesion when placed in contact with polar liquids. Both the adhesion and the friction coefficient, in turn, showed good correlation with the electronic structure, indicating this as a basic feature responsible for low friction in quasicrystals.

The other set of studies exploited atomic force microscopy to measure friction forces in a single-asperity, elastic contact in ultrahigh vacuum [56-63]. A quasicrystal sample was chosen that was quasiperiodic along one axis in the surface plane, and periodic along the orthogonal axis. Under these experimental conditions, neither adhesion nor hardness could play a role in the difference in friction forces measured in the two directions. On the atomically-clean surface, those friction forces were higher by a factor of eight when sliding occurred parallel to the periodic axis. Consideration of various possible explanations led to the conclusion that phononic or electronic friction were most probably lower in the quasiperiodic direction than the periodic direction. Because

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3 phononic friction is generally thought to outweigh electronic friction, phononic friction  
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5 was concluded to be most viable [44, 45].  
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8 Have these two sets of experiments yielded incompatible results? The answer is  
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10 no, but they have certainly yielded different results. They are not incompatible, because  
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12 adhesion and phonon excitation both can contribute to low friction on quasicrystal  
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14 surfaces—they are not mutually exclusive. Their relative importance probably depends  
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16 upon the conditions of sliding, e.g. elastic vs. inelastic contacts [63]. Both studies  
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18 converge in the conclusion that hardness is not the root cause of low friction in  
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20 quasicrystals.  
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24 Regarding future directions in the tribology of quasicrystals, one opportunity  
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26 stems from the lack of solid evidence from which to decide whether the friction  
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28 anisotropy is due to electronic or phononic friction. This calls for further exploration.  
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30 That may come from molecular dynamics simulations of sliding interfaces [64].  
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32 Molecular dynamics models typically do not include electronic excitations. Thus, if such  
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34 a model can reproduce the friction anisotropy, it will serve as strong evidence for  
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36 phononic friction. This, in turn, will enable predictions of whether materials exhibit low  
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38 friction, from their fundamental properties.  
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45 In summary, many possibilities still exist for exciting fundamental research on the  
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47 surface physics and chemistry of quasicrystals.  
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