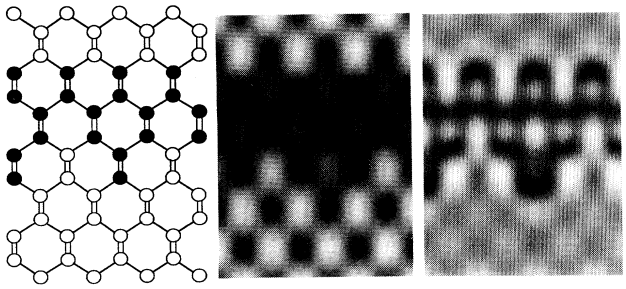
Ordered Structures at Si on Ge(001) Interfaces In a recent Letter [1], Ikarashi *et al.* claim to have determined the specific ordered structure existing at a Si on Ge(001) interface [Fig. 1(a)]. Although such ordering has now been appreciated for several years, these authors attribute a universal significance to their model by suggesting that all previously proposed [2] and determined [3–5] structures at similar interfaces are incorrect. The purpose of this Comment is to demonstrate that Ikarashi *et al.* have seriously overstated the significance of their structural model and that their notion of a universal ordered structure is a gross oversimplification of reality.

A Z-contrast image simulation of the Ikarashi *et al.* model structure [Fig. 1(a)] is shown in Fig. 1(b). The simulation reveals that the Z-contrast method has both sufficient resolution and compositional sensitivity to distinguish this model from other structures previously reported in the literature. In particular, the Ikarashi *et al.* model cannot explain the ordered phases observed at successive interfaces of an ultrathin $(Si_4Ge_8)_{24}$ superlattice (Fig. 1 of Ref. [3]), isolated Si on Ge interfaces (Fig. 5 of Ref. [4]), and antiphase boundaries of the ordered superlattice (Fig. 7 of Ref. [5]). Ikarashi *et al.* [1] are, therefore, incorrect in claiming that the Z-contrast imaging has insufficient resolution to determine the correct structures. Rather, their model is inconsistent with experiment showing that their simple notion of a universal ordered structure is unwarranted.

A disturbing feature of the study by Ikarashi *et al.* [1] is the lack of agreement between simulation and experiment. The multislice simulation in Fig. 1(c) reveals that the white ovals centered on the pure Si and Si-rich column positions, although of significant contrast, are not present in their experimental image. This is not surprising due to the combined influence of uncontrolled parameters such as tilt, strain, inelastic scattering, and absorption. The inability to experimentally reproduce the fine detail contained in the simulation means that in practice only gross statements can be made regarding column compositions.



a b c

FIG. 1. (a) Ordered structure proposed by Ikarashi *et al.* [1]. (b) ⟨110⟩ Z-contrast and (c) multislice phase contrast simulations of (a). The simulation in (c) corresponds to the experimental conditions used in Ref. [1]. Compositions are as in Ref. [1].

This insensitivity must be apparent to the authors since they assign 30% and 70% Ge contents to columns which are, respectively, indistinguishable from pure Si and Ge columns in their experimental image. We emphasize that these are lower limits to their errors in composition determination since an alloy column will in practice appear as pure Ge or pure Si at a composition determined by unknown parameters. In contrast, due to the dominant Z^2 dependence of the high-angle scattering cross section, Z-contrast imaging is extremely sensitive to small quantities of Ge in a column. This is why Z-contrast imaging has demonstrated that Ge segregation can occur to six monolayers [3–5], whereas Ikarashi *et al.* state only two monolayers [1].

In view of the compositional insensitivity of their imaging, the relevance of the authors' diffraction data is highly questionable. Since the ordered domain size is appreciably smaller than the coherence length, the diffraction data will average over a large number of phase variants and be sensitive to the relative translations between domains. Such translations depend on the growth conditions and would inevitably break the (2×2) symmetry in agreement with the authors' observations [1]. It is most likely that a combination of atom pump phases, incorporating translations between domains, would produce as good a fit to the data as the authors' "universal phase." However, this has not been considered by Ikarashi *et al.* [1] in their interpretation of the diffraction data.

In any event, the universal phase proposed by Ikarashi *et al.* [1] is clearly not universal [3–5].

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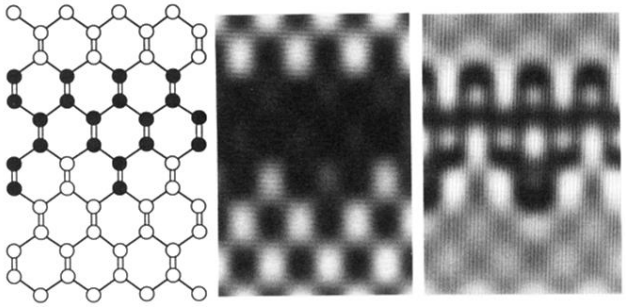

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