Epitaxy of *M*/graphene/Ge (*M* = Fe, Sb) heterostructures: testing the limits of remote heteroepitaxy

<u>P. J. Strohbeen</u>,⁺¹ E. H. Shourov,¹ V. Saraswat,¹ D. Du,¹ M. S. Arnold,¹ and J. K. Kawasaki¹

¹Materials Science and Engineering Department, University of Wisconsin, Madison, WI, USA

It was recently demonstrated through the creation of GaAs/graphene/GaAs (001) heterostructures that monolayer graphene may act as a general platform for epitaxy through an atomic barrier[1]. However, the underlying mechanisms of "remote epitaxy" and its generalization to other material systems, e.g. transition metal compounds or oxides, remains unclear. Here, using M/graphene/Ge (M = transition metal or Sb) as a model system we (1) explore the limits of the remote epitaxy mechanism and (2) demonstrate that single layer graphene is also an excellent solid state diffusion barrier.

In systems containing more volatile species (M = Sb) we have found that carefully controlling growth kinetics both via substrate temperature and the cracked Sb species enables growth of nearly single oriented Sb/graphene/Ge (111) heterostructures. The resultant films are readily exfoliated using scotch tape (Fig. 1). In contrast, we find that when M = Fe, the films grown on graphene are polycrystalline regardless of substrate temperature and Ge orientation. Though we still show that the polycrystalline films are easily exfoliated. These results suggest that volatile adatom species may be a required ingredient for "remote epitaxy". With M = Fe we also show that graphene behaves as an excellent solid state diffusion barrier as supported by our in-situ x-ray photoemission spectroscopy (XPS) measurements as a function of annealing temperature. Our work suggests highly flux dependent growth mechanisms due to both the difficulty in wetting the graphene monolayer as well as the high

in-plane diffusivity on graphene. The effects of growth conditions as well as the effectiveness of graphene as a solid state diffusion barrier will be discussed.

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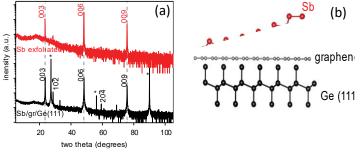


Figure 1: (a) Remote epitaxy and exfoliation of rhombohedral Sb films on graphene/Ge (111) substrates, (b) Schematic figure of how data in (a) was

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⁺ Author for correspondence: pstrohbeen@wisc.edu

(a) (b) Fe 2p_{3/21} Fe 2p_{3/2}I Reacted as-grown as-grown Fe 2p_{1/2} Fe 2p_{1/2} Fe 🖌 Fe Ge Ge 200 C 200 C Ge 250 C 250 C Ge reacted bulk 300 C Fe surface 300 C Ge Ge 725 705 700 725 720 715 710 705 700 720 715 710 Binding Energy (eV) Binding Energy (eV)

Figure 2. XPS measurements of Fe 2p core levels as a function of annealing temperature for (a) Fe/graphene/Ge (001) system and (b) Fe/Ge (001) system

Supplementary Information