

Koch, Hu, and Das Reply: Our study [1] demonstrates the important role of recrystallization processes for the stress evolution of polycrystalline Volmer-Weber films, which are not accounted for by the two mechanisms proposed by Friesen and Thompson [2] as well as Chason *et al.* [3]. In the following we try to resolve the dispute raised in the Comment of Friesen and Thomson [4]:

(1) We agree with Friesen and Thompson that film recrystallization definitely is not a reversible process. In the model of Abermann *et al.* [5] actually symmetric but irreversible processes are responsible for the seemingly reversible stress behavior observed during growth interruption and resumption. In Ref. [1] we correctly state that recrystallization processes, in particular, grain growth, give rise to tensile stress. Consequently, the tensile stress resulting from grain growth during growth interruptions [see Ref. [6]] is not reversible. Instead, the Laplace stress is transmitted immediately when growth is resumed, whereas recrystallization continues with a delay, which partly explains the initially steeper slope of the force curve. In the force curves of Volmer-Weber films typically a bend to smaller slopes is observed shortly after the maximum, where the films become continuous [visible also in Refs. [2,3]]. We interpret this bend as the onset of recrystallization and use it to estimate the corresponding tensile stress contribution. As illustrated in Fig. 1 [experimental data are from Fig. 4 of Ref. [1]], stress due to grain growth amounts about half of the overall stress change in the growth interruption. In Ref. [1] we explain the remaining, again seemingly reversible, stress contribution of Fig. 1 by another recrystallization process, namely, irreversible post-growth surface flattening. Here material moves via rapid surface diffusion from the compressively strained surface areas at the grain centers to the tensile grain boundary regions, thus leading to a fast tensile rise. Upon growth resumption the originally rougher surface configuration is recovered for kinetic reasons, by initially accumulating impinging atoms preferentially at the compressively strained regions. This mechanism is similar to the step-edge-barrier-induced kinetic roughening mechanism for epitaxial films described in Ref. [7]. Contrary to the epitaxial films, however, compressive stress arises not only due to the changing surface stress but also from the Laplace-strain field at the film surface and is therefore considerably larger.

(2) In contrast to the remarks by Friesen and Thompson [4], the driving force for the thermodynamically irreversible process of post-growth flattening is not the minimization of the of the elastic energy but of the several orders of magnitude larger surface energy ($\sim 1 \text{ J/m}^2$ versus $\sim 1 \text{ mJ/m}^2$ per monolayer).

(3) The last point concerns our use of surface tension instead of the surface stress. Of course, it would be desirable to calculate the Laplace stress of our stress model by the surface stress of the system Fe/SiO_x. Unfortunately

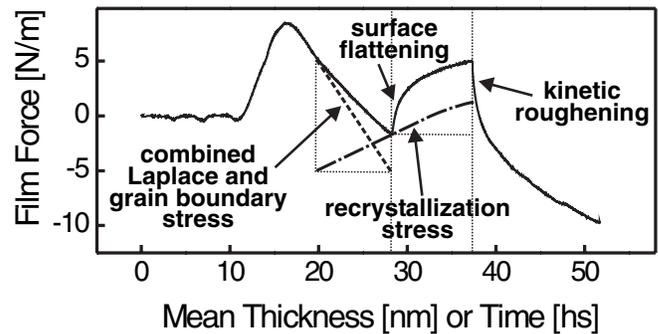


FIG. 1. Film forces evolving during deposition of Fe onto an oxidized Si(001) substrate at 520 K; growth was interrupted after deposition of 0.5, 1.6, 20, and 35 nm [from Ref. [1]].

neither experimental nor theoretical values are available. Therefore—as we state in Ref. [1]—we estimate the Laplace stress by extrapolating the Laplace pressure of an Fe droplet to the deposition temperatures of our experiments. Because of the crude assumptions we expect agreement with experiment within a factor of 2–3. Therefore, the nearly perfect agreement at 300 K is indeed coincidental; a larger deviation is observed at 520 K [as discussed in Ref. [1], the experimental stress value at 520 K includes the tensile stress contributions of grain boundary relaxation and recrystallization as well].

In view of our discussion we think that the tensile stress observed during growth interruptions indeed can be explained by recrystallization processes evidenced by our STM investigations. Accordingly, the seeming reversibility of the stress evolution after growth resumption is the result of symmetric but, in fact, irreversible processes.

R. Koch,¹ Dongzhi Hu,¹ and A. K. Das²
¹Paul-Drude-Institut für Festkörperelektronik
 Hausvogteiplatz 5-7
 D-10117 Berlin, Germany
²Department of Physics and Meteorology
 Indian Institute of Technology
 Kharagpur-721302, India

Received 30 August 2005; published 23 November 2005

DOI: [10.1103/PhysRevLett.95.229602](https://doi.org/10.1103/PhysRevLett.95.229602)

PACS numbers: 68.35.Gy

- [1] R. Koch, D. Hu, and A. K. Das, Phys. Rev. Lett. **94**, 146101 (2005).
- [2] C. Friesen and C. V. Thompson, Phys. Rev. Lett. **89**, 126103 (2002).
- [3] E. Chason, B. W. Sheldon, L. B. Freund, J. A. Floro, and S. J. Hearne, Phys. Rev. Lett. **88**, 156103 (2002).
- [4] C. A. Friesen and C. V. Thompson, preceding Comment, Phys. Rev. Lett. **95**, 229601 (2005).
- [5] R. Abermann, Vacuum **41**, 1279 (1990).
- [6] R. Koch and R. Abermann, Thin Solid Films **140**, 217 (1986).
- [7] C. Friesen and C. V. Thompson, Phys. Rev. Lett. **93**, 056104 (2004).