Lin and Chiang Reply: Our Letter [1] reported a scanning tunneling microscopy (STM) observation of the recombinative desorption of H₂ from hydrogen-terminated Si(100) surfaces. The emphasis of our discussion was on the desorption pathway for dihydrides that are laterally confined by monohydride rows. Desorption from monohydrides at the initial stage was also described for completeness, although a similar study had been reported before [2]. The preceding Comment [3] raises an issue about monohydride desorption. The authors of the Comment argue that the desorption process could involve atomic diffusion, and, thus, STM observations might not reveal the whole picture. Specifically, they argue that an interdimer desorption process shown in Fig. 1 is much more important than the intradimer process. For dihydride desorption, they also question whether our experiment alone excludes the occurrence of H₂ desorption from a single dihydride. While it is true that STM is generally not well suited for studying details of fast surface dynamic processes, the specific arguments presented in the preceding Comment are not necessarily relevant or correct in the present context.

Desorption of H₂ from the monohydride phase has been discussed in the literature rather extensively. Quantum Monte Carlo and density functional calculations [4] show that the desorption barriers for the interdimer and intradimer processes are very similar, and, thus, one would expect that both processes can occur with significant rates [5]. For the interdimer process, the two Si dangling bonds created after desorption represent an unstable phase, and subsequent diffusion tends to join them into a Si dimer (see Fig. 1). It is possible that the entire process occurs in a concerted fashion. In any case, the end result is indistinguishable from the intradimer process based on STM observations.

The preceding Comment suggests that the interdimer process dominates based on their previous experiments involving pulsed laser heating [6]. The rapid quenching after the laser pulse results in frozen atomic configurations, thus yielding information relevant to transient states of the system. However, the high surface temperatures and high heating rates during pulsed laser irradiation can involve a host of other issues that are not necessarily simply related to quasistatic desorption at lower temperatures. For example, desorption can occur through photon-induced processes that are fundamentally different from thermal processes [7]. The high excitation level can lead to a high substrate carrier density, the build-up of a significant stress in the substrate, and even melting of the substrate surface at 1400 K [8], which can lead to very different desorption results.

A similar issue on the role of diffusive motion might be raised for the H₂ desorption from dihydride units. Figure 3(e) of our Letter shows a single row of dihydrides trapped within a (2 × 1) monohydride domain, which remains stable after all remaining dihydride rows have been converted into monohydrides. This is strong evidence against desorption from a single dihydride, and this conclusion is independent of the question of diffusion.

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