INTRODUCTION

Defects such as steps, kinks, and vacancies can have a significant effect on the reactivity of surfaces. The environments of atoms at defect sites are different than the environments of atoms on a perfect surface. These differences can alter the local charge distribution and create dipole moments that change the local chemical behavior of the surface. Electrons can be preferentially bound to point defects such as oxygen vacancies in oxides to maintain charge neutrality at the surface. Sites such as these can dictate adsorption and chemical behavior of heterogeneous surface reactions. Steps and kinks can also play an important role in crystal growth and have a strong influence on resulting crystal habits. Thus the type, density, and properties of defects is important information for understanding the overall chemical response of a surface toward any particular reactant.

The atomic and electronic structure of pyrite surfaces, and their chemical reactivity toward oxidation, is currently an active area of research. The importance of defects on these surfaces is becoming increasingly apparent. X-ray photoelectron spectroscopy (XPS) studies have indicated the presence of a variety of possible surface defects on cleaved pyrite (Buckley and Woods 1987; Birkholz et al. 1991; Karthe et al. 1993; Bronold et al. 1994a, 1994b; Nesbitt and Muir 1994; Nesbitt et al. 1998; Schaufuss et al. 1998). In-vacuum prepared growth surfaces are found to have a significant fraction of S deficiencies (Guevremont et al. 1997, 1998). Scanning tunneling microscopy (STM) studies have uniquely provided direct observations of various surface defects at the atomic scale (Eggleston and Hochella 1992a; Eggleston et al. 1996; Rosso et al. 1999a), but the data are not yet comprehensive. Surface defects created by cleavage are also poorly understood. There is an obvious need for more work in this area and this is the principal motivation behind the current study.

As a second motivation, we also investigate the stability of defects on this surface, a largely unexplored area on pyrite surfaces. In general, the atomic structure at surfaces at room temperature is in a constant state of dynamic change. Fluctuations due to the thermal energy create vacancies by promoting a fraction of relatively stable surface atoms to unstable adatoms (an isolated atom on a terrace). The highly undercoordinated adatoms perform a random walk across terraces until more stable attachment sites are again found such as at step edges or