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**Infrared absorption spectra at interfaces from first principles: Origin of LO and TO red shifts in ultrathin oxide films on silicon** FELICIANO GIUSTINO, ALFREDO PASQUARELLO, Ecole Polytechnique Fédérale de Lausanne (EPFL) — The interpretation of the vibrational spectra of thin $SiO_2$ films on Si(100) has recently been challenged by the observation of a pronounced thickness-dependent red shift of the asymmetric oxygen stretching mode in Si–O–Si intertetrahedral bridges. The origin of this red shift has variably been ascribed to compressive strain of the interfacial oxide, to void incorporation, or to the presence of substoichiometric silica. We here clarify the origin of this red shift by using a density functional approach. For this purpose, we first introduce a formalism to calculate both the longitudinal and the transverse infrared absorption spectra of thin films on transparent substrates. Then, we extend our formulation in order to spatially map the absorption intensity across interfaces. When applying this method to a realistic model of the Si(100)-$SiO_2$ interface, we find that that the red shift arises from a softening of the stretching modes in the substoichiometric interfacial layer. The cumulative effect of an interfacial layer with reduced vibrational frequencies and of a stoichiometric oxide with bulk-like modes is shown to consistently explain the thickness-dependent shift observed in experiments.

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