Poynor *et al.* Reply: Ocko, Dhinojwala, and Daillant agree with us about the role of methyl groups. Our unambiguous statement that [1] "…the depletion thickness reported in Fig. 2(b) must be regarded as an upper bound, although the existence of the depletion layer is unequivocal. This is because protons on the methyl-terminated hydrophobic monolayer and on water adjoining the monolayer are virtually invisible…" has been misquoted by Ocko, Dhinojwala, and Daillant, who assert [2] "(their) entire electron density depletion is attributed to a depletion of water."

Our Table I corrects additional factual mistakes in Table I of Ocko, Dhinojwala, and Daillant regarding data in the literature [1,3]. The product of the gap thickness and the electron density deficit is more consistent between the limits of the fitting range than Ocko, Dhinojwala, and Daillant have claimed. Table I also includes new experiments in which we tested experimentally our claim of a significant depletion by considering a counterexample, the interface of ethanol with octadecyltriethoxysiloxane (OTE) methyl-terminated monolayers. Measuring the reflectivity of this *wetted* structure under the same conditions as in our original work, we obtain the finding that methyl groups of the OTE monolayer explain all of the electron density gap for the wetting case but only half of that observed in our original [1] data.

Ocko, Dhinojwala, and Daillant suggest a second contribution to the observed electron density depletion—surface-induced orientation of water [2]. We note that this phenomenon, if substantiated, would be an intrinsic aspect of hydrophobicity. The large literature on this question suggests a certain statistical bias for one of the two hydrogen bonds of the average water molecule to point towards a hydrophobic surface, giving a broad orientational distribution [4,5]—not the distinct smecticlike layering proposed by Ocko, Dhinojwala, and Daillant. These simulations therefore do not support the contention that orientational ordering of water is a dominant source of electron density depletion when water meets a hydrophobic surface. Ocko, Dhinojwala, and Daillant also assert that low-density regions at the calcite-water interface observed by x-ray experiments are associated with water orientation [6]. The low-density layer observed in that system is *not*, however, from number density depletion. It is, instead, due to the intrinsic structure of the interfacial hydration layer coupled with high (1 Å) resolution in those measurements.

We are gratified that the frame of discourse has shifted from controversies of whether a zone of depleted electron density exists, which was the main point of our Letter, and on which we and Ocko, Dhinojwala, and Daillant agree. It is generally agreed that water is attracted at best only weakly to a hydrophobic surface. The matter of soft modes and ensuing *fluctuations* of interfacial density, which might have very long wavelength, has been emphasized on the theoretical side [7], inferred from measurements [8], and reported from one simulation [9]. To this fresh question, we encourage the community to turn its attention.

TABLE I. δD, ρ_{dep}, ρ_{avg}, and Γ as defined in Ocko, Dhinojwala, and Daillant [2].

System or interface	δD (Å)	ρ_{dep} ($e\text{Å}^{-3}$)	ρ_{avg} ($e\text{Å}^{-3}$)	Γ ($e\text{Å}^{-2}$)
Water/OTE [1] min	1.8	0	0.334	0.60
Water/OTE [1] max	4.0	0.167	0.334	0.67
Ethanol/OTE min	1.3	0	0.264	0.34
Ethanol/OTE max	1.8	0.09	0.264	0.30
Water/OTS [3]	3.8	0.237	0.334	0.37

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