Review

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Posted Date: 4 March 2026

doi: 10.20944/preprints202603.0336.v1

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Reconciling the Adsorbed Water Layer with the Exclusion Zone: A Magnetic Field Model

Running title: Adsorbed Water Layer and the Exclusion Zone

Nygren J

Independent Scientist; johanngrn@gmail.com

Abstract

The relationship between the thin adsorbed water layer conventionally observed on hydrophilic surfaces and the much larger "exclusion zone" described in the literature has remained unclear. In this review, we survey the evidence for both phenomena and propose that they are intimately connected: the adsorbate constitutes a structurally distinct phase that generates a magnetic field, which in turn diamagnetically orders the surrounding water over much larger distances. This model reconciles the thin adsorbate with the much larger exclusion zone, and is consistent with available data, with broader implications for water's magnetic properties.

Keywords: exclusion zone; adsorbed water; fourth phase of water; magnetism

Introduction

It is well established in surface science that hydrophilic surfaces under ambient atmospheric conditions exhibit an adsorbed water layer of approximately 0.5–1 nm, consisting of a few molecular layers with partially ordered structure, as confirmed by techniques such as ellipsometry, AFM, and sum-frequency spectroscopy (Israelachvili, 2011; Asay and Kim, 2005; Thiel and Madey, 1987; Verdaguer et al., 2006; Xu and Salmeron, 2009). Beyond this thin adsorbed layer, a much larger zone of structured water that excludes solutes has been observed adjacent to hydrophilic surfaces. This phenomenon has deep historical roots: Szent-Györgyi (1957, 1972) proposed that water near biological surfaces was structurally organized, and Ling (1962, 1965, 2001) developed the polarized-oriented multilayer theory of cell water, describing intracellular water as "non-solvent water" that excludes solutes due to its altered dynamic structure. These ideas were later experimentally extended by Pollack and colleagues, who coined the term "exclusion zone" (EZ) for the solute-free region of up to hundreds of micrometers observed near hydrophilic surfaces (Zheng et al., 2006; Pollack, 2013), a phenomenon that has been independently reproduced by multiple groups, although Pollack's specific structural model for EZ water ($H_3O_2^-$ sheets) remains debated.

We propose a reconciliation between the conventional ~1 nm adsorbate and the much larger exclusion zone: only the adsorbate layer itself exists in the structurally distinct "fourth phase", while the bulk of the exclusion zone is diamagnetically ordered water — structurally ordered as confirmed by diffraction data (McGeoch and McGeoch, 2008), but not itself in the fourth phase. Rather, its ordering arises from diamagnetic alignment imposed by the magnetic field generated by the fourth-phase adsorbate. This is consistent with the observation that the EZ releases roughly 250,000 times fewer hydrogen ions than would be expected if the entire zone were in the fourth phase (Chai et al., 2009), corresponding to approximately 1 nm of fourth-phase water at the surface. The fourth-phase adsorbate, with its tightly packed, shifted ice-like lattice and expelled protons, generates a magnetic field due to the resulting charge separation and cooperative dipole alignment. This field then imposes long-range diamagnetic ordering on the surrounding water, generating the extensive solute-exclusion zone observed experimentally. Such a mechanism would also account for the observation that external magnetic fields induce exclusion zones in water (Shalatonin and Pollack, 2022).

doi:10.20944/preprints202603.0336.v1

The Adsorbed Water Layer and the Exclusion Zone

The adsorbed water layer on hydrophilic surfaces has been recognized for the better part of a century (Henniker, 1949) and has been increasingly well characterized with modern surface-sensitive techniques. Hydrophilic surfaces such as silica, mica, and metal oxides in contact with water exhibit a thin interfacial layer of approximately 0.5–1 nm, corresponding to two to three molecular layers, whose structure differs from that of bulk liquid water (Asay and Kim, 2005; Verdaguer et al., 2006). This phenomenon is observed both at solid–liquid interfaces and at surfaces exposed to water vapor, where the layer forms at moderate relative humidity and grows with increasing humidity while remaining on the order of a few nanometers even at near-saturation conditions (Asay and Kim, 2005; Thiel and Madey, 1987). Infrared and sum-frequency generation studies reveal a more ice-like hydrogen bonding network in the adsorbate, with reduced molecular mobility compared to the bulk (Asay and Kim, 2005; Thiel and Madey, 1987). Its presence is universal across hydrophilic surfaces and has been confirmed by a wide range of techniques including ellipsometry, ATR-IR spectroscopy, X-ray photoelectron spectroscopy, and AFM force measurements (Israelachvili, 2011; Xu and Salmeron, 2009; Verdaguer et al., 2006).

The exclusion zone, by contrast, extends over a dramatically larger length scale. Using microsphere suspensions, Pollack and colleagues demonstrated that hydrophilic surfaces such as Nafion immersed in water generate a particle-free zone extending up to several hundred micrometers from the surface (Zheng et al., 2006; Pollack, 2013). This zone also excludes dissolved solutes and dyes, and is accompanied by a measurable drop in pH in the region beyond the EZ, indicating a release of protons (Chai et al., 2009). The EZ grows in response to incident radiant energy, particularly infrared light (Chai et al., 2009), and can be induced by external magnetic fields in the absence of a hydrophilic surface (Shalatonin and Pollack, 2022). Diffraction measurements have confirmed that water within the EZ possesses structural order, with a lattice resembling ice but with each layer shifted by one oxygen position relative to the layers above and below (McGeoch and McGeoch, 2008).

Despite the extensive characterization of both phenomena, they have largely been studied in isolation — with conventional surface scientists rarely acknowledging the exclusion zone literature, and exclusion zone researchers seldom engaging with the established body of work on adsorbed water layers. The adsorbed water layer is a topic of conventional surface science, studied in the context of adhesion, friction, and wetting. The exclusion zone, on the other hand, has been studied within a separate tradition — from Ling's work on non-solvent water and the polarized-oriented multilayer theory (Ling, 1962, 1965, 2001), through Szent-Györgyi's proposals of structured biological water (Szent-Györgyi, 1957, 1972), to Pollack's more recent characterization and fourth-phase model (Pollack, 2013). A key observation, however, is that the two phenomena differ by roughly five orders of magnitude in length scale — the adsorbate being ~1 nm and the exclusion zone extending up to ~300 μm — yet both involve structured water adjacent to hydrophilic surfaces. Furthermore, while the thin adsorbate is spectroscopically distinct from bulk water, the bulk of the EZ does not show the same degree of spectroscopic difference, suggesting that the ordering mechanism in the EZ may be fundamentally different from that of the adsorbate.

Proposed Model

A central challenge in understanding the exclusion zone is accounting for its long-range order. Neither van der Waals interactions nor hydrogen bonding networks are known to impose structural order on water beyond a few molecular layers from a surface. Ling's polarized-oriented multilayer theory, which invoked cooperative hydrogen bonding to extend the range of ordering, was met with skepticism on precisely these grounds. The exclusion zone, however, extends over five orders of magnitude further. This suggests that an additional mechanism is at work — one capable of long-range ordering. The observation that magnetic fields alone can generate exclusion zones (Shalatonin and Pollack, 2022) demonstrates that magnetic ordering is a viable mechanism.

We propose that the adsorbed water layer is magnetic. The precise mechanism by which it acquires magnetic properties is not yet established, but we note that a key feature of Pollack's proposed fourth-phase structure is the expulsion of interlayer protons, leaving adjacent hexagonal layers in direct contact — held together by van der Waals interactions rather than hydrogen bonding. This makes the fourth phase structurally analogous to van der Waals layered materials such as graphene. In such systems, a small rotational or translational displacement between hexagonal layers gives rise to flat electronic bands (Bistritzer and MacDonald, 2011), correlated electronic states (Cao et al., 2018), and emergent ferromagnetic order (Sharpe et al., 2019). Whether Pollack's proposed half-oxygen translational shift or a small rotational twist better describes the interlayer geometry remains to be determined, but either configuration could in principle support magnetic behavior.

This magnetic adsorbate behaves as a permanent magnet — amplifying ambient magnetic fields and retaining this amplification. The magnetic field it generates imposes diamagnetic ordering on the surrounding water molecules. This diamagnetically ordered water — structurally ordered as confirmed by diffraction data showing an ice-like lattice with each layer shifted by one full oxygen position (McGeoch and McGeoch, 2008), but distinct from the fourth phase itself — constitutes the exclusion zone.

This is supported quantitatively by Pollack's pH measurements, which show that the exclusion zone releases roughly five orders of magnitude fewer hydrogen ions than would be expected if the entire zone were in the fourth phase (Chai et al., 2009). The observed proton release corresponds to only approximately 1 nm of fourth-phase water — precisely the thickness of the adsorbed water layer established by conventional surface science (Israelachvili, 2011; Asay and Kim, 2005; Thiel and Madey, 1987).

Several further observations support this model. Infrared and ultraviolet radiation have been shown to expand the exclusion zone (Chai et al., 2009); within the proposed model, this can be understood as radiation interacting with the adsorbate layer to strengthen its magnetic amplification, thereby extending the range of diamagnetic ordering. Furthermore, while the thin adsorbate is spectroscopically distinct from bulk water, the bulk of the EZ is not, consistent with the EZ being diamagnetically ordered liquid water rather than a distinct thermodynamic phase. Additionally, if the entire exclusion zone were in the negatively charged fourth phase, as Pollack's model implies, the resulting net charge density would be enormous and difficult to sustain in a conducting medium such as liquid water (Elton et al., 2020; Segarra-Martí et al., 2014). Confining the fourth phase to the ~1 nm adsorbate reduces this charge by five orders of magnitude, yielding a far more physically plausible charge distribution.

Discussion

The model proposed here offers a reconciliation between two bodies of literature that have remained largely disconnected: the conventional surface science of adsorbed water layers and the exclusion zone research initiated by Ling, Szent-Györgyi, and Pollack. By identifying the ~1 nm adsorbate as the sole locus of the fourth phase, and the bulk of the exclusion zone as diamagnetically ordered water, several longstanding difficulties with Pollack's original model are resolved. The net charge density problem — that hundreds of micrometers of negatively charged $H_3O_2^-$ would be electrostatically implausible — is reduced by five orders of magnitude. The discrepancy between the predicted and observed hydrogen ion release disappears. And the need for a new long-range ordering mechanism is met by magnetism rather than by extending hydrogen bonding far beyond its known range.

The principal open question is the mechanism by which the adsorbate acquires its magnetic properties. The structural analogy with van der Waals layered materials, in which small displacements between hexagonal layers give rise to flat electronic bands and emergent ferromagnetism, provides a compelling candidate.

More broadly, if the adsorbed water layer is indeed magnetic, every hydrophilic surface in contact with water would be a source of local magnetic order — an observation of particular relevance to biology, where the density of hydrophilic surfaces is extraordinarily high.

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