

How positively and negatively charged ions behave at interfaces

November 23 2021



Researchers from RUB and their cooperation partners have investigated what happens at the interface between an aqueous solution and a charged surface.
Credit: RUB, Kramer

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between a solid surface and an aqueous solution has been investigated by researchers from the Cluster of Excellence RESOLV at the Ruhr-Universität Bochum, its sister research network CALSOLV in Berkeley, and the University of Evry in Paris. At the SOLEIL synchrotron, they were able to use terahertz spectroscopy to observe exactly when and how the water shells around sodium and chloride ions are stripped away when voltages are applied in an electrolyte solution. They describe their results in the journal *Proceedings of the National Academy of Sciences*, or *PNAS*, published online on 15 November 2021.

Electrochemical double layer between electrolyte and solid interface

Electrolytes are chemical compounds in which separate ions occur. For example, when [sodium chloride](#) (NaCl) is dissolved in [water](#), the positively charged sodium ions and the negatively charged [chloride ions](#) separate and can move freely in the solution. Due to the electrical attraction between the ions and the [water molecules](#), a shell of water molecules forms around the individual ions—a so-called hydration shell that is stable in the solution. A [layer](#) of charge carriers forms in the immediate vicinity of the electrical boundary layer between the electrode and water. A positive and a negative charge layer are opposite each other, which is why this layer is also called an electrochemical double layer. According to chemistry textbooks, the following happens when a voltage is applied: the attraction between the electrode and the ions strips off the water shell and a charge transfer, a current, occurs.

This simple picture explains how a battery works. In the present work, the researchers from Bochum, Berkeley and Paris investigated whether it is also correct at the molecular level. They also checked whether the process is identical when negative or positive voltages are alternately applied.

Observation during the process is difficult

Observing chemical processes on a molecular level while a voltage is applied is a special experimental challenge. This is exactly what the scientists succeeded in doing in the current study with [terahertz spectroscopy](#), which they combined with simulations. For this purpose, the researchers investigated the electrochemical double layer that forms in a NaCl solution in the immediate vicinity of a gold surface at the SOLEIL synchrotron in Paris.

Terahertz spectroscopy makes it possible to follow the stripping of the hydration shell live. The researchers also showed for the first time how the water networks on the charged gold surface change. This is essential to understand how the total energy changes in the process. "It was astonishing for us to see that the process runs differently for positive and negative charges," sums up Professor Martina Havenith, spokesperson of RESOLV.

Asymmetric detachment of the hydration shell

The researchers found that the hydration shells of sodium and chloride ions behaved differently in the electrochemical double layer. The hydration shell of the positively charged ions was already detached at small applied voltages and the sodium ion was attracted to the electrode. For the negatively charged chloride ions, this only happened at a higher applied voltage. The team was able to attribute these differences to the behavior of the water networks at the interface. The scientists confirmed the results with the help of complex computer simulations.

"The method and the results can now be used to investigate the crucial role of water in other interfacial processes, for example at semiconductor/electrolyte interfaces," says Martina Havenith. The

results are important for understanding and optimizing electrochemical processes, for example for technological applications such as solar cell or fuel cell technologies.

More information: Serena R. Alfarano et al, Stripping away ion hydration shells in electrical double-layer formation: Water networks matter, *Proceedings of the National Academy of Sciences* (2021). [DOI: 10.1073/pnas.2108568118](https://doi.org/10.1073/pnas.2108568118)

Provided by Ruhr-Universitaet-Bochum

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