Iron Equilibrium Fractionation Factors from Ab initio, NRIXS, and Lab Experiments: The Path Forward

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Iron isotope variations have been found in a variety of samples and settings, ranging from hemeproteins to extraterrestrial igneous rocks. The strongest impediment to interpreting iron isotope variations in natural samples is the lack of a robust set of iron equilibrium fractionation factors between phases. Three approaches can be used to estimate equilibrium fractionation factors: ab initio calculations [1], NRIXS measurements [1,2], and laboratory experiments [3,4]. Each method has its weaknesses and strengths but evaluating their accuracies can be difficult. Consistency between equilibrium fractionation factors estimated by several independent approaches is a necessary condition to gain confidence in these methods; keeping in mind that natural and synthetic minerals can depart from ideal stoichiometry. Few phases have been studied by the three approaches. Blanchard et al. [1] recently estimated the β-factor of goethite using ab initio and NRIXS. The β-factor estimated by NRIXS is slightly higher than that predicted by the ab initio approach, which itself predicts a higher equilibrium fractionation between goethite and Fe(II)aq than is measured [4]. In order to assess the reliability of the equilibrium fractionation factors estimated using the various techniques, we have measured by NRIXS at the APS synchrotron the β-factors for synthetic ferrihydrite, natural goethite, hematite, pyrite, and siderite. Ab initio calculations are available for all these phases. We will provide a side-by-side comparison of the NRIXS and ab initio approaches with a perspective on what it tells us about laboratory experiments.

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