**JoVE 53452\_R2: Reply to Editorial and Reviewers’ comments**

**Editorial comments:**  
  
Changes to be made by the Author(s):  
  
1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues. The JoVE editor will not copy-edit your manuscript and any errors in the submitted revision may be present in the published version.  
*→　We proofread the manuscript carefully and made several small amendments (changes are highlighted in the text).*

2. Please define the error bars in Figure 4 and 5 in the appropriate Figure Legend.

*→　We added to the Legends of Figures 4 and 5 a reference to the error bar calculation formalism, which is given as a Note in Procedure Step 6.8. We do not consider it helpful repeating the formalism itself in the Figure Legends, because defining all entering quantities would render those captions unnecessarily lengthy.*  
3. Please provide a complete reference for Reference 29, 31.

*→　Ref. 29 cites historically important fundamental work by Niels Bohr on electronic stopping of high energy ions in matter, published in a Danish journal on mathematical physics. This classic paper is frequently cited by the ion beam physics community in the exact form that I provided in the JoVE manuscript. Due to its age, no electronic version of the article (hence no DOI) exists to my knowledge. Therefore, please kindly understand that that Ref. 29 cannot further be ‘completed’.*

*→　Ref. 31 is the link to the website of the MALT accelerator facility, where application procedures are documented and application forms can be downloaded for potential users of the facility. Please explain what further ‘completion’ should be necessary here?*  
4. Please confirm that we will be able to access all relevant parts of the facility.

*→　I confirm that the filming team will have access to all the relevant sections of the MALT accelerator facility. That being said, it will be necessary to schedule the filming during a time when the accelerator facility has assigned machine time to our group (Fukutani-Wilde Laboratory) for NRA measurements. Only during such times (typically 4-5 times per year for about one week) our group has access to the beamlines and we are able to use the accelerator for NRA measurements. The next opportunity to do so is a machine time from* ***August 12 to August 21, 2015****. The next machine time cannot be expected before October or November 2015. To proceed publication of our video article, I therefore suggest scheduling the filming during the upcoming August machine time. Please kindly consider whether a JoVE filming set can be arranged in this period.*  
**Reviewers' comments:**  
  
**Reviewer #1:**   
The authors present a manuscript describing the protocol for nuclear reaction analysis at the MALT Tandem accelerator facility of the university of Tokyo. The procedure is known to be sensitive to the concentration of hydrogen atoms in the vicinity of surfaces. The potential of the method is demonstrated for a palladium surface and for SiO2 thin films deposited on silicon.  
  
The manuscript is well written and concise. The procedure is explained in detail and it is expected to reproduce the desired outcome. Nonetheless, some steps could benefit from a better explanation, in particular those concerning the participation of the assisting scientist:  
  
"Tighten flange bolts and evacuate UHV system following instructions from the assisting scientist." (line 275) "Obtain instructions from the assisting scientist to become familiar with the accelerator control system in the control room." (line 346)

*→　a) We gratefully appreciate the Reviewer’s careful reading and generally positive evaluation. We consider the particular instructions on the vacuum chamber evacuation (line 275) and operation of the accelerator control system (line 346) as highly facility-specific. Their description would require considerable additional text length but should be of little interest to most readers who may want to learn about the NRA technique in principle. We are prepared to support any collaborating scientist who may actually want to perform NRA measurements at our facility by making the additional instructions requested by the Reviewer available, but expect that this will only concern a tiny fraction of the article’s potential readership. With regard to manuscript length and readability, we would therefore prefer not to include also these details in the paper, although other (arguably equally technical) details have been given in the procedures (see also our reply to the last comment by Reviewer #1). The NOTE in lines 349-351 at least provides a brief principal description of the MALT control system. Actions performed with this control system will also be filmed and appear in the video that accompanies our article, so we believe that together with the video sufficient information is provided to form an adequately detailed impression of the protocol.*

Also, in section 5 (NRA Measurement at BL-2C), the acquisition time is not mentioned. From the introduction, I am lead to believe that the acquisition time should be shorter that at in the previous section (50 s).

*→　b) We thank the Reviewer for pointing out this significant shortcoming. We added the acquisition time setting for the NRA Measurement at BL-2C (for SiO2/Si) to the protocol (Step 5.2), and also provided a recommendation for the intensity of the 15N ion beam (50-100 nA on FC04) in Step 5.3. These parameters are determined by the requirement to obtain sufficient NRA -ray yield from the given H density in the target sample to achieve satisfactory counting statistics. In the present case, because the surface and interfacial H layer densities in the SiO2/Si targets are about one order of magnitude smaller than in the saturated surface H layer on Pd(110), the acquisition time in the described BL-2C measurement has actually been the same as in BL-1E, not shorter, despite higher sensitivity in BL-2C. We added an explanation of this situation to the discussion (lines 704-709 in the revised manuscript).*

From a physical point of view, the method is presented clearly and I find particularly refreshing that the authors are not trying to oversell it. It could nonetheless be beneficial to add a few more words concerning the spatial resolution of the technique, especially for metallic systems. In the text at lines 548-551, the H-uptake in the interfacial region is simply fitted to a sigmoid, whereas Figure 4 exhibits some corrugation. Is it possible to extract more information from this corrugation?

*→　We have very good reason to believe that the corrugation in Figure 4 results from background fluctuations in our -detection system rather than reflecting real concentration gradients in the H depth distribution in the Pd single crystal. We added these reasons and further explanations of the near-surface depth resolution to the discussion in lines 552-562 (revised manuscript) and thereby explained to the reader why our use of the sigmoid fit function is justified:*

*“Figure 4 also shows that the depth resolution of the NRA H profile in the near-surface region is limited by the width of the surface resonance peak to about 2-3 nm (≈FWHM/S). Therefore, any abrupt features in the H profile such as the point-to-point -yield variations around 16 nm depth cannot correspond to actually existing steep H concentration gradients, because such would be smeared out by the surface peak width and additional energy broadening due to 15N ion straggling3. Hence, the -yield corrugations in the plateau region of the H profile (5 to 22 nm depth) reflect fluctuations of the BGO background count rate (separate background measurements confirm that such random fluctuations occur) and do not contain physical information on the depth distribution of the Pd-absorbed hydrogen. The latter distribution is expected to be rather smooth in the homogenous single crystal, where H diffusion is rapid (several 100 nm/s even at 145 K)3, 13, 15. Thus, the approximately constant concentration of the bulk-absorbed hydrogen in the near-surface region of the Pd crystal after exposure to 2000 L H2 at 145 K can be evaluated by fitting the plateau data in Figure 4 to a sigmoid function that rises to its half-height at Eres with the same width as the Gaussian surface peak. …”*

Also, a discussion of the time resolution of the procedure could be included. Is it possible to investigate the transient evolution of a system and, if so, on which timescales?

*→　Yes, it is possible to observe transient evolutions of H densities with NRA, such as upon adsorption, desorption, absorption, or diffusion of hydrogen in target materials, on the time scale defined by the acquisition time. We added a brief description of these circumstances to the discussion in lines 709-711.*  
The main critique to the manuscript is that the procedure appears to be valid only in a single facility. Then again, this is probably a necessary requirement for describing the methodology used in such detail.

*→　In this and also his first comment, Reviewer #1 points out a ‘dilemma’ that also we felt during authoring the article. The unique JoVE format puts much focus on the experimental procedure. Thus, the methodology has to be described in quite some technical detail to enable reproduction of the results on one hand, yet of course the article should still remain readable and generally informative to a wider audience, on the other. Especially because our particular method involves an accelerator and custom design experimental stations, many procedure steps are necessarily facility-specific. We are afraid that this cannot be helped, but hope nonetheless that our descriptions will enable the reader to extract the experimental principles of the NRA technique and to potentially extrapolate from these to situations in other laboratories. We have strived to maintain a reasonable balance between facility-specific detail and explanation of the principle ways of the experiment in the article, and our deliberate omission of, e.g., evacuation steps of a specific vacuum system and detailed descriptions of the controls at MALT were meant as ways to preserve this balance.*

**Reviewer #2:**   
1. One point that is not mentioned that can be important is the beam spot size (or beam intensity in (beam current)/cm2). Unlike most other elements undergoing MeV ion beam analysis, H can be lost during analysis limiting the application of this method to materials in which the H is only weakly bound. With the same beam current, if the beam spot is 1 mm by 1 mm, the same beam current with a 1 cm by 1 cm beam spot has 100 time less beam intensity, making analysis of even very delicate materials possible.  
*→　We thank the Reviewer for pointing out an important point. Following his suggestion, we have provided a more detailed explanation of possible beam-induced H loss from the target material and included mentioning of the possibility to reduce such effects by defocusing the ion beam in the discussion (lines 666-676 of the revised manuscript):*

*“The tendency of H to desorb from the sample or to redistribute inside the target by diffusion under the ion beam varies strongly between different target materials and should be evaluated as part of any given analysis by monitoring the -yield at the probing depth of interest as a function of the ion beam dose. Without a compensating H2 background as applied here to Pd(110) where H readsorbs readily, in many cases a more or less pronounced exponential decay of the H-signal can be observed. Measuring and extrapolating such H-loss functions to zero 15N exposure allows reproducing the original H density on or inside the target prior to perturbation by the ion beam (for details, see Ref. 3). If the target size permits, reducing the current density (nA/cm2) in the beam-irradiated surface spot by defocusing the ion beam with aid of the MQ04 magnetic lenses (Protocol Steps 2.3.5 and 3.10) may alleviate H losses during the analysis.”*

2. The authors don't seem to say how they established the absolute calibration of the method. For anyone wanting to establish this method in their own laboratory, this is a key step usually accomplished by measuring samples with well known H content (either bulk of ion implanted).

*→　We actually did mention the sensitivity calibration using a Kapton foil concentration standard in line 542 (original manuscript), but this admittedly very brief and embedded text may have failed to catch the reviewer’s attention. To describe the calibration procedure more explicitly, we added a few more details in lines 545-546 and 568-569 of the revised manuscript.*

3. I don't think a viewer (reader) needs to know all the local accelerator/data collection protocols. They are specific to that particular institution.

*→　We would agree with the Reviewer’s assertion, but are afraid that the JoVE’s focus on the experimental procedure makes it inevitable to include a fair amount of technical detail that (especially in case of our particular accelerator-based method) is facility-specific. Please also refer to our reply to a similar remark by Reviewer #1.*

4. I have no doubt that the examples shown to demonstrate the method are correct. The examples illustrate some of the important types of application of 15N hydrogen profiling.

*→　We thank the reviewer for this positive assessment.*