

Dissipative hydride precipitates in superconducting niobium cavities

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(Dated: 17 October 2011)

We report the first direct observation of the microstructural features exhibiting RF losses at high surface magnetic fields of above 100 mT in field emission free superconducting niobium cavities. The lossy areas were identified by advanced thermometry. Surface investigations using different techniques were carried out on cutout samples from lossy areas and showed the presence of dendritic niobium hydrides. This finding has possible implications to the mechanisms of RF losses in superconducting niobium at all field levels.

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Superconducting radio frequency (SRF) cavities have become the primary particle accelerating structures in many modern accelerators (i.e. CEBAF, CESR, SNS) and are a technology of choice for future projects (i.e. XFEL, FRIB, Project X, ILC) due to the extremely low surface resistance in superconducting state and hence very high quality factors achievable in such structures. Bulk niobium is predominantly used to make accelerating superconducting cavities as a simple material having high enough T_c and critical fields. An intense research in recent years has been focused on overcoming limitations encountered in niobium cavities on the way to achieve higher accelerating gradients such as the high field Q-slope and quench (see¹ for review). An empirical recipe has been developed to achieve the highest gradients in SRF niobium cavities, which allows to eliminate the high field Q-slope via a combination of electropolishing and 120°C baking for up to 48 hours. Despite the recent effort, the physics behind the effect is still not understood, even though many possible explanations have been eliminated. In this context, the identification of any microstructural features on niobium surface, which exhibit RF losses at high surface magnetic fields and hence may be connected to any of the above-mentioned phenomena, is of a great value.

Among different experimental techniques used to gain understanding of the physics governing SRF technological breakthroughs, the most powerful approach is provided by using temperature mapping of the outer cavity walls during RF tests. Temperature mapping is realized by attaching several hundred of individual thermometers, typically based on carbon resistors, to the outer cavity surface. These thermometers are present during the RF measurements and the resulting temperature maps allow identifying areas of different dissipation, localizing the quench site, and obtaining a field dependence of losses at each thermometer location. Subsequent cutting of the areas of interest and subjecting them to extensive surface analytical and superconducting measurements makes it possible identifying the differences in near-surface properties leading to different RF dissipation mechanisms. Such studies have been performed on the high field Q-slope cutouts²⁻⁵ and low field quench sites^{6,7}. The source of the local dissipative areas found even after 120°C is not known, and is of the intense interest as mentioned above. From practical perspective these areas may be the cause of local quench and a quality factor degradation limiting the performance of niobium cavities.

To further study the origin of the high field RF losses, an elliptical single cell TESLA shape 1.3 GHz niobium cavity of 2.8 mm wall thickness and 50 μm grain size manufactured by

Advanced Energy Systems was subjected to $105 \mu\text{m}$ material removal from the inner surface by buffered chemical polishing (BCP) followed by $65 \mu\text{m}$ material removal by electropolishing (EP), 120°C ultra-high vacuum baking for 48 hours and a high pressure water rinsing (HPR) before RF tests. This sequence of processing steps is typically applied to state of the art niobium cavities in order to achieve the highest gradients, except for a high-temperature ($\sim 800\text{C}$) heat treatment, which was not applied to this cavity. The final RF test of the cavity was performed at Jefferson Lab with the temperature mapping system of 576 thermometers attached to the outside cavity walls measuring the local temperature increase with respect to the helium bath temperature sensor. The detailed design of the system is described in⁸. The measured quality factor dependence on the peak surface magnetic field at $T = 2 \text{ K}$ is shown in Fig. 1. The maximum surface magnetic field was limited by the localized quench

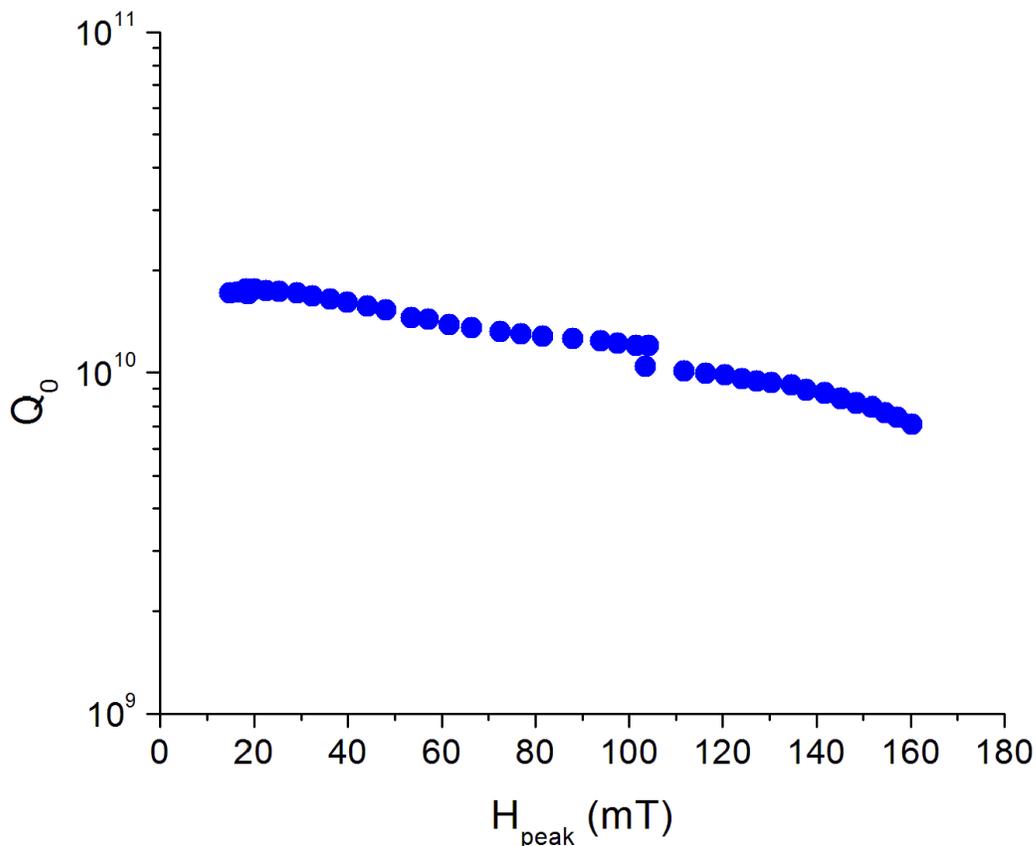


FIG. 1. The quality factor Q_0 of the cavity plotted against the peak surface magnetic field H_{peak} as measured during the RF test.

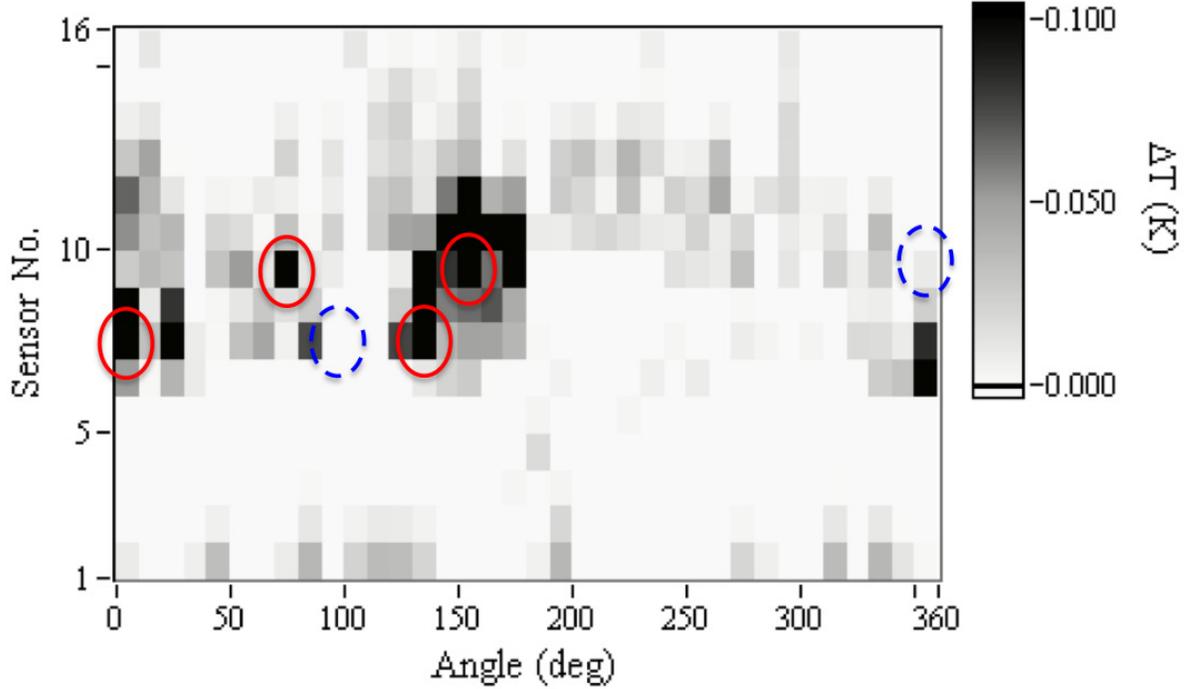


FIG. 2. The temperature map at the highest achieved field of $H_{peak} = 160 \text{ mT}$. Locations from where the samples were extracted are marked with red solid circles for hot spots and blue dashed line circles for cold spots with negligible RF losses.

at about 160 mT. The temperature map obtained at the highest field before quench is shown in Fig. 2. Based on the temperature map, three different kinds of locations were identified: (i) exhibiting strong RF losses (black circles in Fig. 2a), (ii) exhibiting weak losses (brown circles), and (iii) the quench site. Typical individual temperature sensor readings at hot and cold spots are shown in Fig. 3.

Circular samples of about 1 cm in diameter were extracted from the selected locations using the automated milling machine with no lubricant to prevent possible contamination. In addition, the rotation speed of the milling tool was kept at 375 rpm to prevent heating of the samples whose temperature did not exceed a few degrees above the room temperature. A series of investigations was performed on each of the samples to elucidate possible surface structure differences leading to the different RF losses. SEM investigation at different electron accelerating voltages uncovered dendritic structures ranging in size from 1 to 15 μm in two hot spots exhibiting the strongest heating. SEM images for the hot spot 150-10 are shown in Fig. 4. Similar imaging of the cold spots did not indicate any of the dendritic

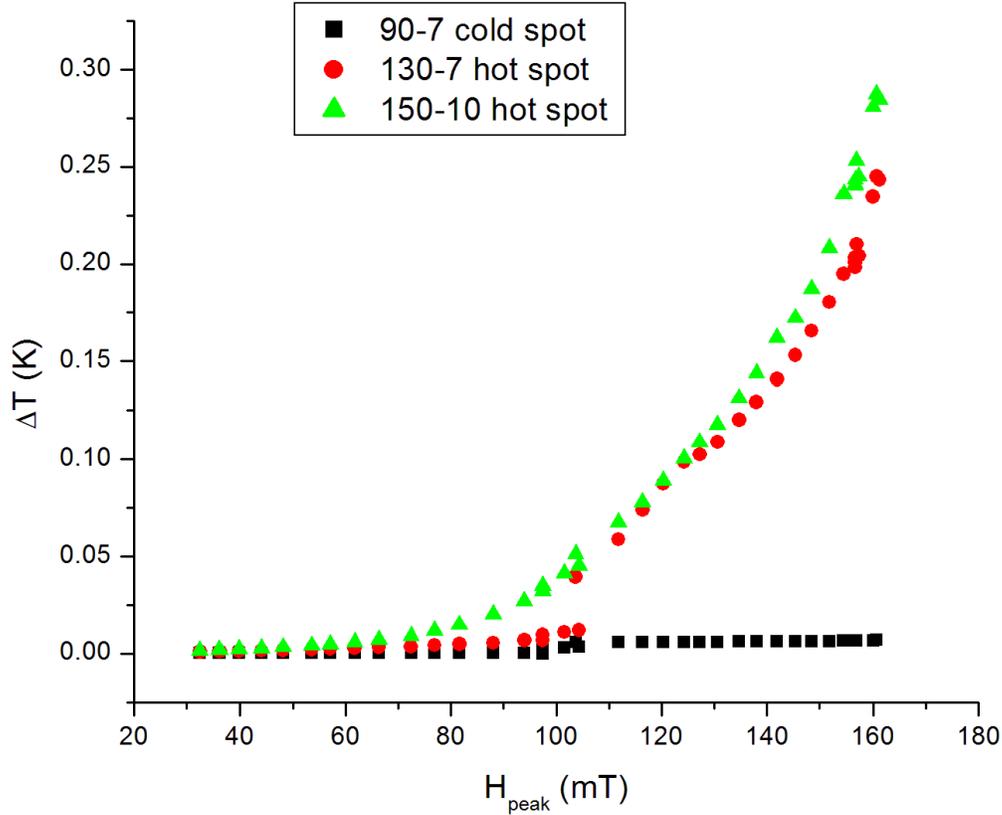


FIG. 3. Temperature sensor readings versus peak magnetic field for two hot spots and one cold spot.

objects present.

Energy dispersive X-ray spectroscopy (EDS) was performed at different spots on the precipitate and in the surrounding areas. Individual EDS energy spectra did not reveal any contamination, while EDS mapping showed a slightly higher oxygen concentration in the “border” area of the stars not apparent from individual spectra. It should be noted that EDS is not sensitive to any elements below atomic number of 12 (carbon), and in particular hydrogen is not identified.

In order to investigate the nanoscale structure, Zeiss 1540XB FIB-SEM system was used to prepare site-specific TEM samples from the areas containing branches of the precipitates. The area around the precipitate was capped with a protective carbon layer of about $2 \mu\text{m}$ thickness to preserve the near-surface structure throughout milling and thinning processes.

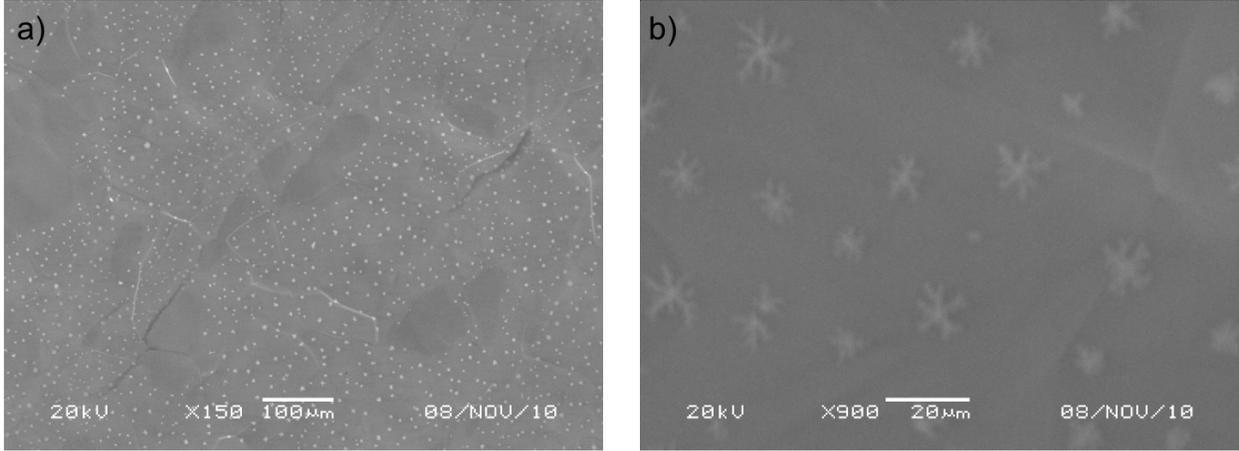


FIG. 4. SEM secondary electron images of the dendritic objects found on the 150-10 hot spot cutout.

The analytical transmission electron microscope FEI CM30T was used for imaging. The cross-sectional bright field TEM image of one of the areas containing the precipitate is shown in Fig 5. Selected area diffraction patterns have been obtained from the precipitate and from the surrounding niobium showing the difference in the crystalline structure. The precipitate is extending about 100 nm into niobium.

In addition to SEM/EDS and FIB/TEM studies, electron backscattered diffraction (EBSD) studies are being done on the cutout samples to elucidate possible connections with the dislocation substructure and will be reported in a future publication. Furthermore, comparative elastic recoil detection (ERD) measurements of the near-surface hydrogen distribution were performed on one of the cold spots and are reported in⁹.

Our findings of dendritic precipitates in two hottest spots represent to our knowledge the first direct correlation of high field (> 100 mT) RF losses with this particular microstructural feature. Based on SEM/EDS results along with TEM imaging and diffraction we believe that the observed dendritic precipitates are most likely niobium hydrides similar to the ones reported in^{10,11}, which represent β -phase formed upon rapid cooling of the Nb-H solution. Magnetic properties of similar objects were investigated by Vinnikov and Golubok¹², and these precipitates were found to be either superconducting even at 4.2 K or lying more than 50 nm under the surface while possessing a significant flux pinning strength. In our case, these precipitates appear to be right next to the surface and hence are probably weakly superconducting at the RF test temperature of 2 K resulting in the increased RF losses as

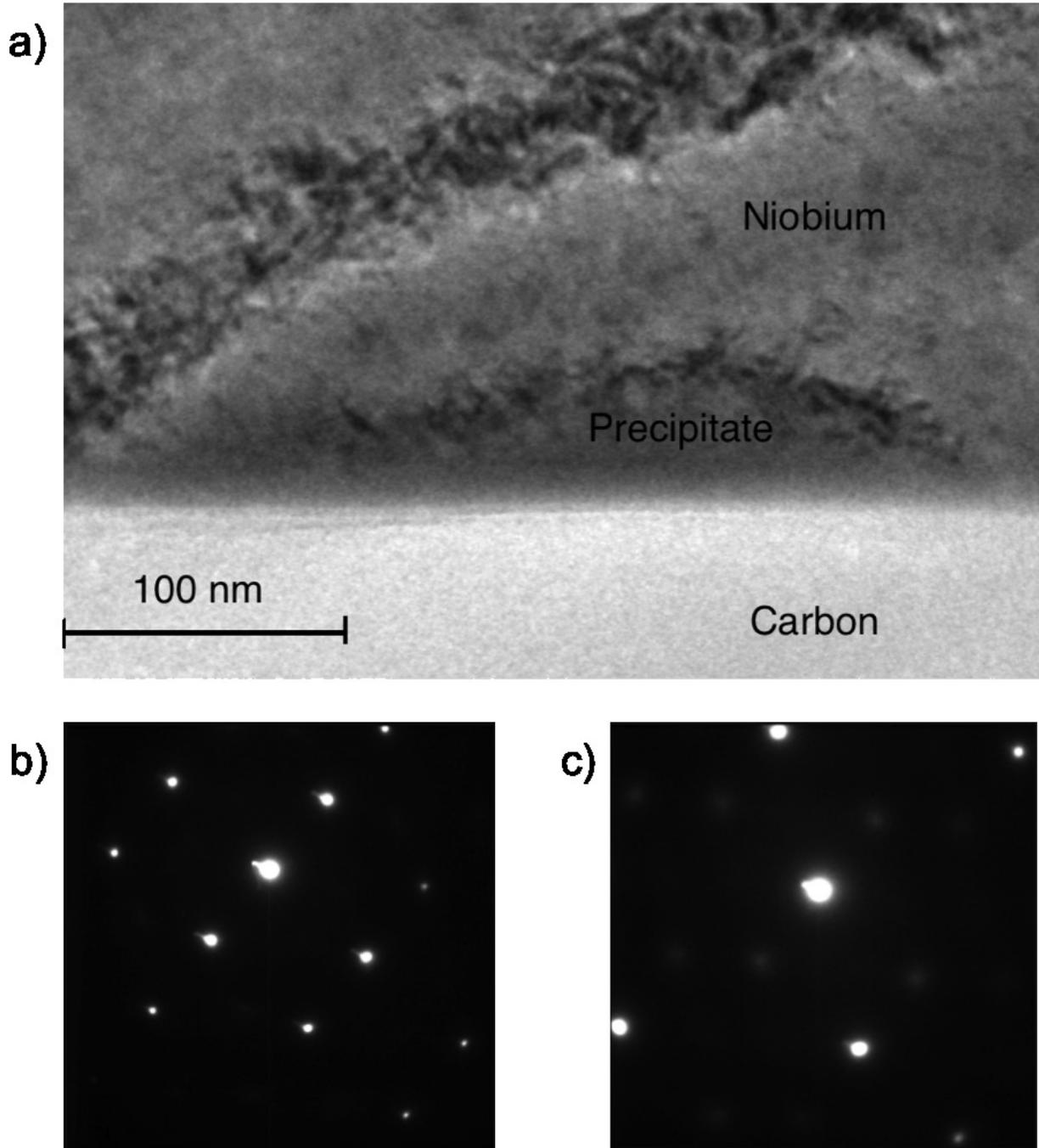


FIG. 5. A cross-sectional TEM image of the area containing the precipitate (a) and the selected area diffraction (SAD) patterns showing different crystalline structures in the precipitate area (b) and the surrounding niobium (c).

registered at the corresponding locations.

In summary, we directly observe for the first time dendritic hydride precipitates in superconducting niobium cavities. These precipitates, most likely representing the β -phase of

niobium hydride, exhibit strong RF losses when exposed to high magnetic fields on the cavity surface. Our results suggest that the state of hydrogen precipitation may be of importance for the performance of niobium cavities not only in the lower field range as thought before in the context of a so-called “hydrogen disease”, but also at highest gradients. A correlation between increased cavity quality factor at 100 mT and reduced hydrogen concentration has already been reported¹³. As a hypothesis, the presence of dislocations and vacancies as hydride nucleation centers upon cooling down to 2 K may be a necessary condition for the hydrides to form and RF losses to appear. It is unclear at this point if hydrogen precipitation is connected to the high field Q-slope as well, and it is the subject of ongoing research.

The study was accomplished at Fermilab operated by Fermi Research Alliance, LLC under Contract No. DE-AC02-07CH11359 with the United States Department of Energy. The authors would like to acknowledge Michael Morrone from Jefferson Lab for the help with the cavity test. The transmission electron microscopy and focused ion beam preparation was accomplished at the Electron Microscopy Center for Materials Research at Argonne National Laboratory, a U.S. Department of Energy Office of Science Laboratory operated under Contract No. DE-AC02-06CH11357 by UChicago Argonne, LLC.

REFERENCES

- ¹H. Padamsee, *RF Superconductivity: Volume II: Science, Technology and Applications* (Wiley-VCH, 2009).
- ²A. Grassellino, “Muon spin rotation/relaxation studies of niobium for SRF applications,” in *Proceedings of the 15th International Conference on RF Superconductivity* (2011).
- ³A. Romanenko and H. Padamsee, “The role of near-surface dislocations in the high magnetic field performance of superconducting niobium cavities,” *Supercond. Sci. Technology*, **23**, 045008 (2010).
- ⁴A. Romanenko, “Crystalline microstructure role in the high-field Q-slope,” in *Proceedings of 14th International Conference on RF Superconductivity* (2009).
- ⁵A. Romanenko, H. Padamsee, G. Ereemeev, and J. Shu, “Comparative surface studies on fine-grain and single crystal niobium using XPS, AES, EBSD and profilometry,” in *Proceedings of PAC2007* (2007).
- ⁶W. Singer, X. Singer, S. Aderhold, A. Ermakov, K. Twarowski, R. Crooks, M. Hoss,

- F. Schölz, and B. Spaniol, “Surface investigation on prototype cavities for the European X-ray Free Electron Laser,” *Phys. Rev. ST Accel. Beams*, **14**, 050702 (2011).
- ⁷X. Singer, S. Aderhold, A. Ermakov, W. Singer, K. Twarowski, M. Hoss, F. Schölz, and B. Spaniol, “Surface investigation on prototype cavities for the European XFEL,” in *Proceedings of the 1st International Particle Accelerator Conference* (2010).
- ⁸G. Ciovati, *Investigation of the superconducting properties of niobium radio-frequency cavities*, Ph.D. thesis, Old Dominion University (2005).
- ⁹A. Romanenko and L. V. Goncharova, “Elastic recoil detection studies of near-surface hydrogen in cavity-grade niobium,” *Supercond. Sci. Tech.*, **24**, 105017 (2011).
- ¹⁰M. L. Grossbeck and H. K. Birnbaum, “Low temperature hydrogen embrittlement of niobium II - microscopic observations,” *Acta Metallurgica*, **25**, 135 (1977).
- ¹¹T. Schober and U. Linke, “A metallographic study of the niobium-hydrogen system. Part I. Beta-phase room temperature morphologies,” *J. of the Less-Com. Met.*, **44**, 63 (1976).
- ¹²L. Y. Vinnikov and A. O. Golubok, “Direct observation of magnetic structure in niobium single crystals with hydride precipitate pinning centers,” *Phys. Stat. Sol.*, **69**, 631 (1982).
- ¹³G. Ciovati, G. Myneni, F. Stevie, P. Maheshwari, and D. Griffis, “High field Q slope and the baking effect: review of recent experimental results and new data on Nb heat treatments,” *Phys. Rev. ST Accel. Beams*, **13**, 022002 (2010).