Ultrafast Structural Transformations in Fe: A Time-Resolved X-ray Diffraction Study

O. Liubchenko[i,*], J. Antonowicz[a], K. Sokolowski-Tinten[b,y], P. Zalden[c], R. Minikayev[i], I. Milov[d, e,f], C. Bressler[c,g,h], M. Chojnacki[h,$], P. Dłużewski[h], P. Dzięgielewski[a] A. Rodriguez-Fernandez[c], K. Fronc[i,$], W. Gawełda[#,z,x], K. Georgarakis[j], A.L. Greer[k], I. Jacyna[i], R.W.E. van de Kruijs[f], R. Kamiński[l], D. Khakhulin[c], D. Klinger[i], K. Kosyl[i], K. Kubicek[c,g,h], A. Olczak[a], N.T. Panagiotopoulos[k], M. Sikora[m], P. Sun[n], H. Yousef[c], W. Zajkowska-Pietrzak[i] and R. Sobierajski[i]

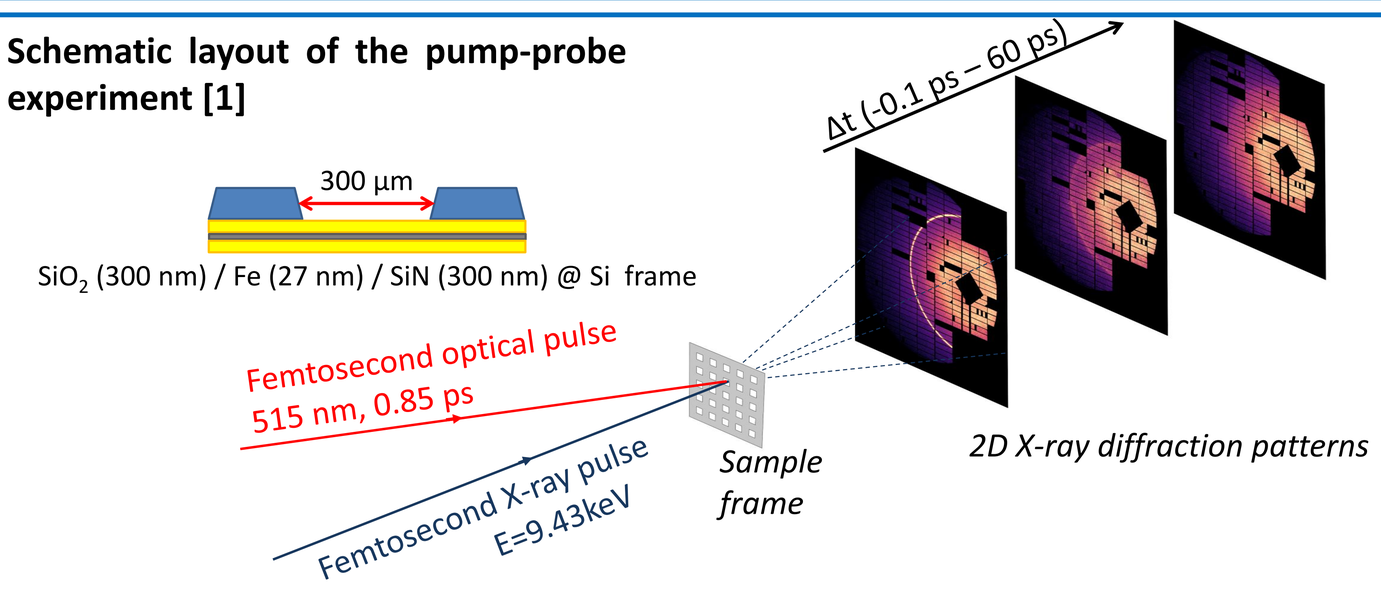
[a]Faculty of Physics, Warsaw University of Technology, Koszykowa 75, 00-662 Warsaw, Poland [b]Faculty of Physics, University of Duisburg-Essen, Lotharstrasse 1, 47048 Duisburg, Germany [y]Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Lotharstrasse 1, 47048 Duisburg, Germany [c]European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany [d]Advanced Research Center for Nanolithography (ARCNL), Science Park 106, 1098 XG Amsterdam, the Netherlands [e]Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany [f]Industrial Focus Group XUV Optics, MESA+Institute for Nanotechnology, University of Twente, Drienerlolaan 5, 7522 NB Enschede, the Netherlands [g]Department of Physics, Universitat Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany [h]The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany [i]Institute of Physics, Polish Academy of Sciences, Aleja Lotnikow 32/46, PL-02668 Warsaw, Poland [$]Research Centre MagTop, Institute of Physics, Polish Academy of Sciences, Aleja Lotnikow 32/46, PL-02668 Warsaw, Poland [j]School of Aerospace, Transport and Manufacturing, Cranfield University, Cranfield, MK43 0AL, UK [k]Department of Materials Science & Metallurgy, University of Cambridge, Cambridge, CB3 0FS, UK [l]Department of Chemistry, University of Warsaw, Żwirki i Wigury 101, 02-089 Warsaw, Poland [m]Academic Centre for Materials and Nanotechnology, AGH University of Krakow, Al. A. Mickiewicza 30, 30-059 Krakow, Poland [n]Dipartimento di Fisica e Astronomia "Galileo Galilei", Universita degli Studi di Padova, Padova 35131, Italy [#]Department of Chemistry, Universidad Autonoma de Madrid, Ciudad Universitaria de Cantoblanco 28049 Madrid, Spain [z]IMDEA Nanociencia, Calle Faraday 9, 28049 Madrid, Spain [x]Faculty of Physics, Adam Mickiewicz University, ul. Uniwersytetu Poznańskiego 2, 61-614 Poznan, Poland

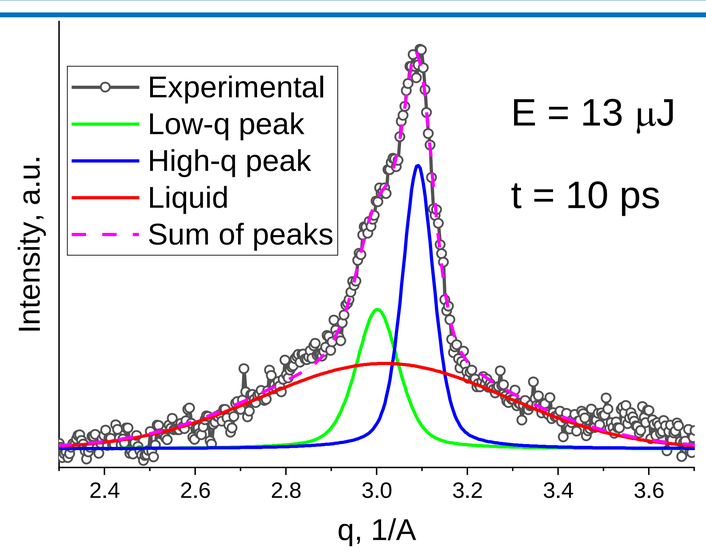
**Corresponding author: liubchenko@ifpan.edu.pl*

Abstract

The atomic structure of the thin Fe layer after sub-ps pulsed laser annealing has been studied by time-resolved X-ray diffraction. Due to very strong electron-phonon coupling, rapid heating of the lattice occurs on a 1 ps timescale leading to ultrafast structural transformations. Melting occurs on a timescale of 1-5 ps. At the highest excitation levels studied, ultrafast melting prevents any solid-solid transformations known from the equilibrium phase diagram. Below the threshold of complete melting, the body-centered tetragonal (bct) phase forms on a time scale of about 10 ps and remains stable for at least 60 ps. This experimental observation of structural changes supports, for the first time, the theoretical predictions of phonon softening in Fe upon heating.

Schematic layout of the pump-probe experiment [1]

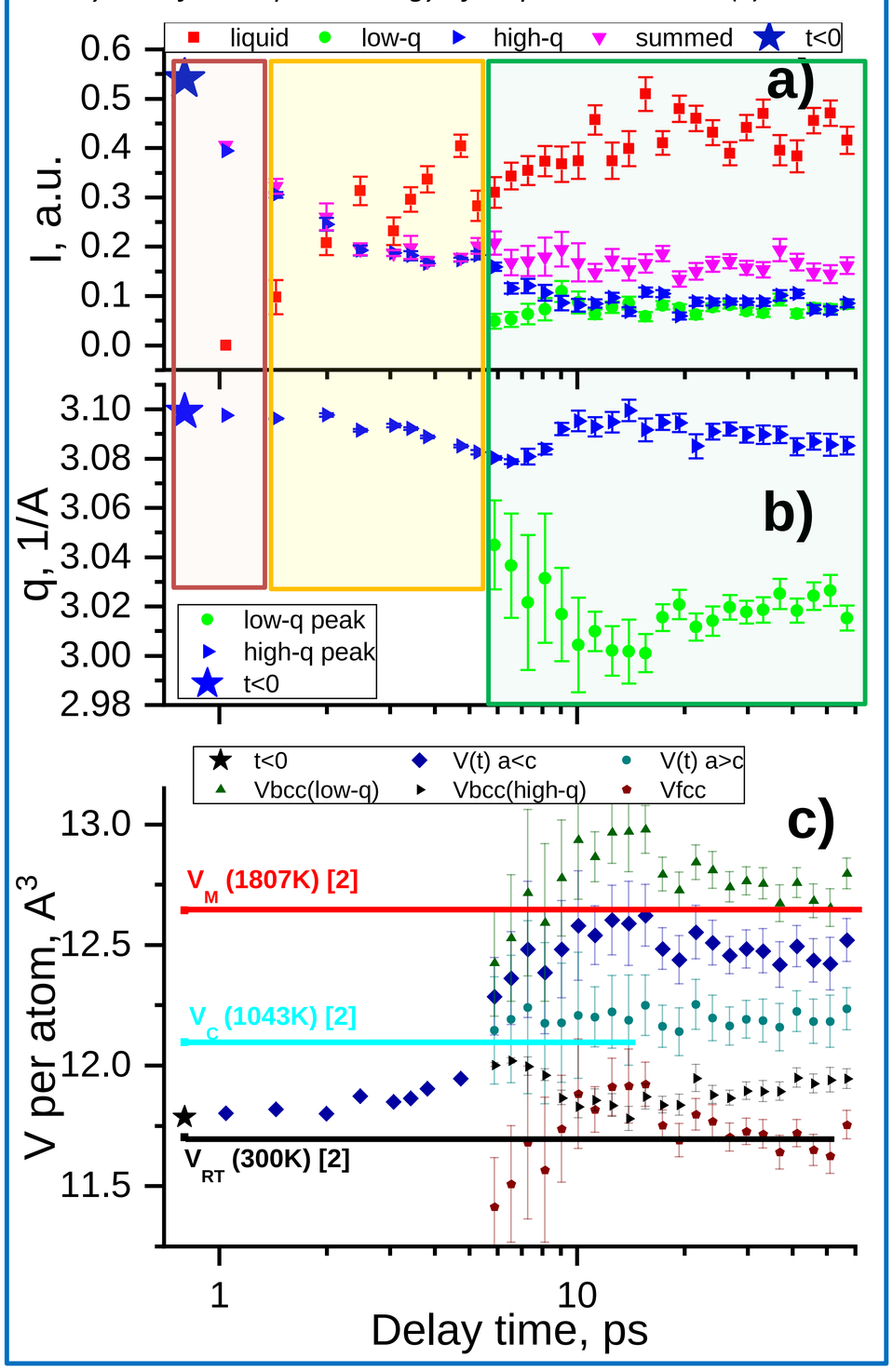




The integrated XRD pattern and peak deconvolution.

Experimental results

Changes of integrated intensities (a) and Bragg peaks positions (b) with delay time. Changes in volume per atom for different interpretations of origins of low-q peak as a function of the delay time for the pulse energy of 13 µJ are shown in (c).



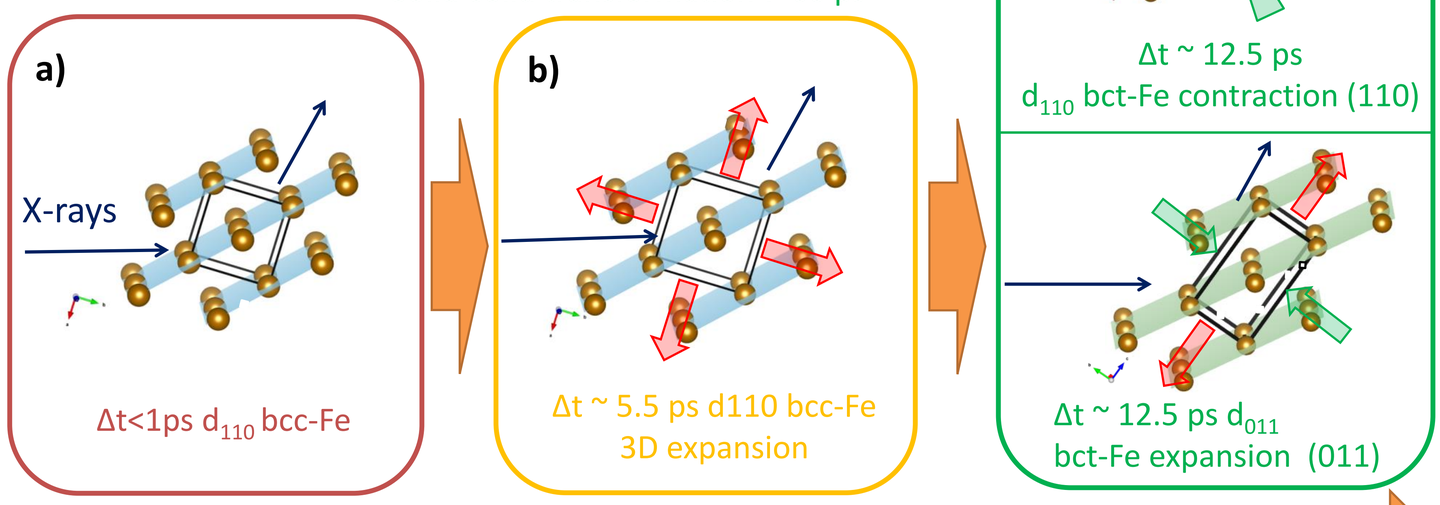
Interpretation of the results

Regimes of structural transformations

Heating 0-1 ps

Melting & Thermal expansion 1-5 ps

Solid-solid transformation 7-60 ps



Sketch of changes in interplanar spacing and unit cell distortion for a selected delay time. a) "cold" lattice just before laser irradiation and in first 1-2ps after irradiation. b) 3D thermal expansion of bcc lattice. c) bct lattice distortion and lattice expansion.

2 Bragg peaks interpretation:

1) Coexistence of two BCC-Fe phases with different lattice parameters

2) Tetragonal distortion of BCC-Fe unit cell, case a<c (a=b)

3)Tetragonal distortion of BCC-Fe unit cell, case a>c (a=b)

4) Coexistence of FCC-Fe and BCC-Fe phases

Phonon softening as a reason of bct distortion

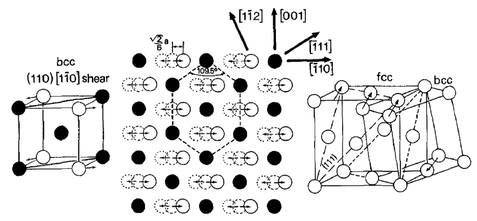


Fig. 7 Scheme of the bcc→fcc transition.

BCC-FCC phase transition pathway [3]

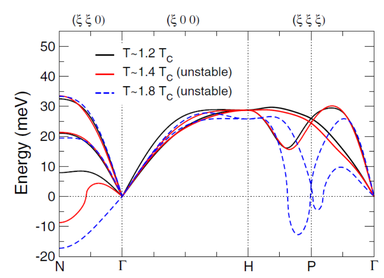


Figure 2 | Calculated phonon dispersions of paramagnetic bcc iron near the α-to-γ and γ-to-δ phase transitions for different temperatures.

Changes in dispersion curves in Fe with overheating over Curie temperature [4]

Acknowledgments

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Summary

- Ultrafast heating occurs at timescale approx. 1 ps, which indicates on strong electron-phonon coupling in Fe.
- Ultrafast melting is observed up to 5 ps.
- Bct distortion occurs after melting due to phonon softening at timescale approx. 7ps.
- Ultrafast structural changes caused by phonon softening were observed experimentally for the first time