A new imaging method using pulsed neutron sources for visualizing structural and dynamical information

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Abstract. Neutron imaging using pulsed neutron sources coupled with a 2-dimensional position sensitive detector applicable to the time-of-flight method can give information on the crystal texture of coherently scattering materials, dynamical information of incoherently scattering materials such as hydrogen, and magnetic field information. Bragg edges appeared at cold neutron region reflect the preferred orientation, crystallite size, and lattice spacing. To deduce such information from the neutron transmission data depending on the position we have developed a data analysis code, and applied this code to data of a welded iron sample. Furthermore, as examples of more realistic materials we have investigated quenched iron rods. The quenched region was clearly demonstrated by the lattice space distribution. Furthermore, difference in the bound state of water or hydrogen in wet and dry cement pastes have been observed by analyzing the gradient of the neutron transmission cross section at the cold neutron region. The magnetic field has been also measured by using the polarized neutrons, and the strength of the field was estimated easily by analyzing the wave length dependent data.

1. Introduction

Neutron transmission imaging based on steady neutron sources has been used in various fields as a method of non-destructive inspection. On the other hand, imaging using pulsed neutron sources has been developing a new field of the imaging with the aid of its energy resolving nature, since the time-of-flight method for energy analysis adopted in the pulsed neutron source allows us easily to obtain the neutron transmission depending on the neutron wavelength at each pixel of a two dimensional position sensitive detector. The transmission spectrum, in principle, depends on the total neutron cross section of the object material. The total neutron cross section generally consists of coherent elastic scattering, coherent inelastic scattering, incoherent elastic scattering, incoherent inelastic scattering, and absorption cross section. Furthermore, magnetic scattering should be included when the polarized neutron transmits a magnetic field. Coherent scattering is major cross section for almost all materials other than materials such as hydrogen, vanadium and so on. The neutron cross section exhibits change of its value at the cold neutron region; for the coherent scatterer there are Bragg edges corresponding to the Bragg scattering of each lattice plane, and for incoherent scatterer the cross section increases in proportion to the wavelength. Furthermore, absorption cross sections of some materials changes
drastically at a specific energy, for example, 0.4 eV for cadmium. At early stage of the time-of-flight imaging we tried to identify each alloy in an object[1], and then we got different contrast images at once by using the time-of-flight imaging[2]. Furthermore, a spatial depending image of crystal structural information was also obtained by analyzing the transmission spectra. Spatial distribution of strain, phase transition, texture, crystallite size were observed by using the pulsed neutron sources[3-11]. For deducing quantitative information on texture, crystallite size and strain, we have to analyze wavelength dependent transmission over wide wavelength region. Therefore, we have developed an analysis code ‘RITS’ [12] including similar functions of the analysis code of the neutron diffraction.

We have performed pulsed neutron transmission experiments at Hokkaido linac facility and also at J-PARC neutron source to study the crystallographic structure of a welded iron plate with a thickness of 6 mm and a quenched iron rod with a length of 20 mm. Furthermore, as an example of hydrogenous material we have studied water and hydrogen dynamical property in cement pastes to see difference between a dry cement paste and a wet cement paste. Finally, by using polarizing neutron we have observed image of a magnetic field. Here, we first explain the new analysis code ‘RITS’ and present the experiments performed at pulsed neutron source which indicating usefulness of the pulsed neutron imaging.

2. Crystallographic information imaging

2.1. Data analysis method

A Rietveld-type spectral analysis code for Bragg-edge transmission imaging, RITS (Rietveld Imaging of Transmission Spectra), was developed for quantitative evaluation of crystallographic information of a material, e.g., crystal lattice strain, preferred orientation, degree of crystallographic anisotropy and crystallite size [12-14]. This code calculates position-dependent transmission spectra based on an effective total cross section model, and then refines the structural parameters at each position by fitting the calculation data to the experimental data. It is very important to develop a theoretical model being able to analyze the experimental data. Hereafter, we present a new theoretical expression of a Bragg-edge transmission spectrum to deduce the crystallographic and metallographic information.

The neutron transmission \( Tr(\lambda) \) as a function of wavelength \( \lambda \) is represented by

\[
Tr(\lambda) = \exp\left(-\sigma_{\text{tot}}(\lambda)\rho \lambda\right).
\]  

(1.1)

\( \sigma_{\text{tot}}(\lambda) \) is the neutron total cross section, \( \rho \) is the number density, and \( \lambda \) is the thickness of a material. In the low energy region, the total cross section consists of elastic coherent scattering, elastic incoherent scattering, inelastic coherent scattering, inelastic incoherent scattering and absorption parts, as follows:

\[
\sigma_{\text{tot}}(\lambda) = \sigma_{\text{coh}}(\lambda) + \sigma_{\text{ina}}(\lambda) + \sigma_{\text{ela}}(\lambda) + \sigma_{\text{abs}}(\lambda).
\]  

(1.2)

The elastic coherent scattering cross section is the most important for crystallographic information analysis because this component uniquely relates to Bragg-edges, and reflects the crystal structure. To describe an actual Bragg-edge transmission spectrum of a polycrystalline material, we proposed a modified expression of a Bragg-edge transmission cross section that is combined by the kinematical diffraction theory [15] with three new factors, \( R_{\text{hkl}}(\lambda - 2d_{\text{hkl}}) \), \( P_{\text{hkl}}(\lambda, 2d_{\text{hkl}}) \) and \( E_{\text{hkl}}(\lambda, 2d_{\text{hkl}}, F_{\text{hkl}}) \), as follows:

\[
\sigma_{\text{coh}}(\lambda) = \frac{\lambda^2}{2V_0} \sum_{\text{hkl}} \left| F_{\text{hkl}} \right|^2 d_{\text{hkl}}^2 R_{\text{hkl}}(\lambda - 2d_{\text{hkl}}) P_{\text{hkl}}(\lambda, 2d_{\text{hkl}}) E_{\text{hkl}}(\lambda, 2d_{\text{hkl}}, F_{\text{hkl}})
\]  

(1.3)

where \( V_0 \) is the unit cell volume, \( d_{\text{hkl}} \) is the crystal lattice plane spacing, and \( F_{\text{hkl}} \) is the crystal structure factor including the Debye-Waller factor.

The first new factor \( R_{\text{hkl}}(\lambda - 2d_{\text{hkl}}) \) is the edge profile function including the \( d_{\text{hkl}} \) information. This function describes the edge broadening due to the neutron pulse shape, the strain and the microstructure. The Dreele-Jorgensen-Windsor model [16] was implemented for strain imaging.
\[ R_{hkl}(\lambda - 2d_{hkl}) = \frac{1}{2} \text{erfc}(w) - \frac{\beta_{hkl} \exp(u) \text{erfc}(y) - \alpha_{hkl} \exp(v) \text{erfc}(z)}{2(\alpha_{hkl} + \beta_{hkl})}, \] (1.4)

where
\[ w = \frac{\lambda - 2d_{hkl}}{\sqrt{2}\sigma_{hkl}}, \] (1.5)
\[ u = \frac{\alpha_{hkl}}{2} \left[ \alpha_{hkl} \sigma_{hkl}^2 + 2(\lambda - 2d_{hkl}) \right], \] (1.6)
\[ v = \frac{\beta_{hkl}}{2} \left[ \beta_{hkl} \sigma_{hkl}^2 - 2(\lambda - 2d_{hkl}) \right], \] (1.7)
\[ y = \frac{\alpha_{hkl} \sigma_{hkl}^2 + (\lambda - 2d_{hkl})}{\sqrt{2}\sigma_{hkl}} \] and
\[ z = \frac{\beta_{hkl} \sigma_{hkl}^2 - (\lambda - 2d_{hkl})}{\sqrt{2}\sigma_{hkl}}. \] (1.8)

\( \sigma_{hkl}, \alpha_{hkl} \) and \( \beta_{hkl} \) are the broadening parameters of the Gaussian, the rise exponential and the decay exponential of the \( \{hkl\} \) edge, respectively.

The crystallographic anisotropy due to the preferred orientation in a polycrystalline material changes the whole shape of a Bragg-edge transmission spectrum. Therefore, we implemented the modified March-Dollase preferred orientation distribution function [17] as the second new factor \( P_{hkl}(\lambda, 2d_{hkl}) \) in the RITS code. The formulation is
\[ P_{hkl}(\lambda, 2d_{hkl}) = \frac{1}{2\pi} \int_0^{2\pi} \left( R^2 B_{hkl}^2 + \frac{1 - B_{hkl}^2}{R} \right)^{3/2} d\phi, \] (1.10)

where
\[ B_{hkl} = \cos A_{hkl} \sin \theta_{hkl} + \sin A_{hkl} \cos \theta_{hkl} \sin \phi. \] (1.11)

\( \theta_{hkl} \) is the scattering angle as a function of \( \lambda \) and \( 2d_{hkl} \) from the Bragg’s law. \( \phi \) represents the angle on a certain Debye-Scherrer ring of the scattering angle \( 2\theta \). \( A_{hkl} \) is the angle between the Bragg reflection vector \(<hkl>\) and the preferred orientation \(<HKL>\) that is parallel or perpendicular to the beam direction. The March-Dollase coefficient \( R \) provides the information of the degree of crystallographic anisotropy. If there is no anisotropy, both \( R \) and \( P_{hkl}(\lambda, 2d_{hkl}) \) are unity. When the texture grew, \( R \) departs from one. \( R = 0 \) or \( \infty \) means a single crystal specimen. Thus, we can get the information of the preferred orientation \(<HKL>\) and the degree of crystallographic anisotropy \( R \) from the shape change of a Bragg-edge transmission pattern.

The primary extinction effect is caused by the re-diffraction phenomenon inside one perfect crystal block (crystallite). This effect reduces the diffraction intensity by returning the diffracted neutrons in the direction of the transmitted beam. Hence, this phenomenon increases the transmitted neutron intensity, and decreases the Bragg-edge transmission cross section. Therefore, we implemented the Sabine’s primary extinction function [18] as the third new factor \( E_{hkl}(\lambda, 2d_{hkl}, F_{hkl}) \) in the RITS code. This formulation is expressed by combining the backward-scattering (Bragg) component \( E_B \) and the forward-scattering (Laue) component \( E_L \), as follows:
\[ E_{hkl}(\lambda, 2d_{hkl}, F_{hkl}) = E_B \sin^2 \theta_{hkl} + E_L \cos^2 \theta_{hkl}, \] (1.12)

where
\[ E_B = \frac{1}{\sqrt{1 + x}}, \] (1.13)
\[ E_L = 1 - \frac{x}{2} + \frac{x^2}{4} - \frac{5x^3}{48} + \cdots \] for \( x \leq 1, \) (1.14)
\begin{equation}
E_L = \sqrt{\frac{2}{\pi x} \left[ 1 - \frac{1}{8x} - \frac{3}{128x^3} - \frac{15}{1024x^5} - \cdots \right]}
\end{equation}

for \( x > 1 \),

and

\begin{equation}
x = S^2 \left( \frac{\lambda F_{hkl}}{V_0} \right)^2.
\end{equation}

\( S \) is the crystallite size along the beam direction. Thus, we can get the crystallite size information from the intensity increase of transmission data.

Figure 1 shows an example of the spectral fitting using the RITS code to Bragg-edge transmission data along the normal direction of an \( \alpha \)-iron plate of 5 mm thickness (cold rolled steel: 0.03%C, 0.04%Si, 0.5%Mn, 0.03%P and 0.03%S). The experiment was performed at the electron linac facility at Hokkaido University. The RITS model is more consistent with the experimental data than the traditional kinematical diffraction theory. Moreover, we can simultaneously obtain various crystallographic parameters. In the previous studies, it has been already indicated that the RITS code gave valid results of crystallographic information, by using various methods, e.g., the preferred orientation theories, the grains observations by an optical microscope, and the neutron diffraction experiments analyzed with their own Rietveld analysis code [12, 14].

![Figure 1](image_url)

**Figure 1.** Result of the Rietveld-type analysis of a Bragg-edge transmission spectrum of an \( \alpha \)-iron plate of 5 mm thickness. The crystal lattice parameter \( a \), the preferred orientation \( <HKL> \), the degree of crystallographic anisotropy \( R \), the crystallite size \( S \) and projected density \( \rho \times t \) can be simultaneously extracted by the RITS code.

### 2.2 Lattice space distribution of quenched steel rods

Here we refer one of examples of the spectroscopic imaging and its analysis. The lattice space analysis of quenched steel rods processed with the induction hardening. By the quenching process we obtain a toughened piece of steel. Such phenomenon is caused by the diffusionless transformation, and forms the martensitic microstructure. Its crystal structure expands a little from the original, and then the quenched piece has a strain inside and shows hardening. Normally, the hardening area is checked by method such as a hardness tester, but the information is limited in a local area. On the other hand, because the neutron has a large penetration depth, we can obtain the mean lattice space correlated with strain through the bulk depth on a wide area. For a realistic sample used in industry we need to measure thick sample. In this measurement feasibility for the thick bulk material is also tested. So we applied the Bragg edge lattice space analysis to the quenched steel measurement.
The experiment was carried at BL10 in J-PARC facility, Japan. It views the decoupled liquid hydrogen moderator, and the proton beam power was around 200 kW during the experimental period. The samples were cut out to 20 mm axial length from long quenched steel rods (JIS S45C, 20 mm diameter) which controlled the quenching depth as 3 mm, 5 mm and 7 mm, with 10 kHz induction hardening and tempering at 423 K. The used detector was the micro-channel plate (MCP) type, which has the 0.055 mm pixel resolution with 14 mm x 14 mm detecting area and is driven under the time event position recording [19]. The samples were set just before the detector window as their center axis parallel to the neutron beam. The obtained transmission spectra were analyzed with the RITS code and deduced the distributions of parameters such as the lattice space. Figures 2(a), 2(b) and 2(c) are the energy sliced radiographies for the quenching depth 3mm, 5mm and 7 mm. We can recognize the brightness difference between the sample center and their rim. The resultant lattice space images after the RITS code analysis are shown in Figures 3(a), 3(b) and 3(c). From these figures we understand that the rim areas are expanded in comparison with the sample centers, and that the effect is caused by the martensite phase formed in the quenching process. The hardening depth can be confirmed for each sample as is successively formed in the planned depth. Such information is important for industrial production, the RITS code analysis can offer the quantitative microstructural image piles upon the real image.

3. Imaging of dynamical information of hydrogen in cement pastes
Hydrogen is a typical incoherent scatterer and is included in many materials. Plant is one example of the typical material including water and the information of not only the water content but also the bound state of water will be sometime important. The freeze of water in a fuel cell is one of the issues for developing the fuel cell. Cement is also important as structure material, and the content of
hydrogen is observed by using the traditional neutron radiography. In the cement paste there exist various types of hydrogen: free water, crystal water and hydrides. It is useful to obtain the information on the hydrogen distribution in the cement paste with the information of its bound state relating to the dynamical motion of hydrogen. It is well known that the total neutron cross section of the water molecule increases with the neutron wavelength [20, 21] and at long wavelength region the total cross section is expressed by \[ \sigma = a\lambda + b \] [21]. Here, \( \sigma \) is the total neutron cross section, \( \lambda \) the neutron wavelength, \( a \) and \( b \) are coefficients. It was also indicated that ‘\( a' \)’, the gradient of the total cross section, become larger with increasing the water temperature [21]. The total cross section of bound water and bound hydrogen in a cement paste will be similar to that of ice since the quasi-elastic neutron scattering spectra of a cement paste were reproduced by superposition of free water and ice spectra [22]. The difference of the gradients of the total cross section between ice and water will be used to distinguish the free water and the bound water or the hydride in the cement paste. In order to study this feature, we performed experiments on two kinds of the cements pastes, dry and wet cement pastes, since in the wet sample free water exists in vacancies and in the dry cement the rate of the free water is much smaller than that of the wet sample. The experiments were performed at BL10 beam line in J-PARC/MLF, and the detector used was the MCP detector [19]. The flight path length was 14.5m from the moderator surface to the detector. We put the cement paste sample in front of the detector. The resolution and the active area was written in previous chapter. The cement pastes were made by gradually mixing water and Portland cement powder. The weight percent of the water and the cement powder was 50%, respectively. After the mixing the paste was casted in a mold with a diameter of 50 mm and length of 100 mm, and after about 72 hours several pieces of cement paste were cut out. The thickness of the samples was 0.5 cm and the diameter 50 mm. The thickness was almost the thinnest one we could make. To prepare the wet sample and the dry sample the pieces were cured in water and then dried.

We could obtain the data up to about 12 Å due to frame overlap of the neutron beam. Figure 6 shows the transmission images obtained by whole energy range of neutrons measured. There is difference of the contrast in each sample corresponding to mainly hydrogen content at each pixel. It is also recognized that the wet sample includes the larger number of hydrogen atoms compared with the dry one. At each pixel we got a wavelength dependent transmission and deduced a micro-cross section.

![Figure 6. Transmission images of a dry cement paste (left) and a wet cement paste (right). The darker image of the wet sample indicates higher content of hydrogen.](image)

By fitting a linear function to the transmission cross section in a range of 7-11.3 Å, the gradient \( a \) [barns/Å/hydrogen] was determined. In this process we summed up 16x16 pixel data to improve the statistics. Figure 7 shows spatial distributions of the gradient of the total cross section obtained by the transmission data. In the results, the gradient changes dependent on position in the same sample. In the dry cement case the gradients are generally larger than those of the wet one. This is inconsistent with the expected result that bound water would give smaller gradient. This is considered to be due to a reason that since in the wet sample there are many free water molecules and sample thickness was not sufficiently thin to avoid the up-scattering of the long wavelength neutrons. The temperature of the sample was much higher than the neutron energy used for analysis. Therefore, the long wavelength neutrons incident on the sample have high possibility for scattering obtaining the energy, namely up-
scattering. The energy transferred to the neutron is much higher than the incident neutron energy. Therefore, the energy spectrum of the up-scattered neutrons will not depend very much on the incident energy. As a result the transmitted neutron has similar energy spectrum. This trend makes the total cross section gentler dependence of the wavelength. Therefore, the effective cross section obtained by the transmission data was in some sense an averaged cross section over the thermal energy. On the other hand, in the case of the dry cement paste the scattering was mainly elastic by the bound hydrogen. Therefore, the energy of the neutron changed not so much, and the effective cross section obtained by the transmission data reflects the incident energy. We have confirmed this effect by simulation calculation. The result indicate that for deducing the gradient we should use thinner sample or use a collimator to remove the scattered neutrons. However, by adopting suitable experimental method we will get information on the bound state of hydrogen in materials. Anyway, we need more complicated analysis for getting the quantitative information.

4. Magnetic field imaging using polarized neutrons

The neutron imaging technique using neutron polarization analysis method has been developed to visualize the spatial distribution of magnetic field [23]. This technique is based on the motion of a neutron spin passing through a magnetic field, and the rotation of a neutron spin is measured as the change in the neutron polarization with spatial resolution using a two-dimensional neutron detector.

The neutron spin rotation angle $\phi$ due to the Larmor precession in a magnetic field $B$ is expressed by following equation,

$$\phi = \gamma \int \text{path} B ds \tag{4.1}$$

where $\gamma$ is the gyromagnetic ratio of the neutron and $v$ the neutron velocity. Therefore, we can get the cumulated magnetic field strength along the neutron beam path from the rotation angle using polarized monochromatic neutron beam. However, as measurements of neutron polarization with a monochromatic neutron beam results in an uncertainty of the rotation angle due to the periodicity of $2\pi$, the study of wavelength dependence of the polarization is necessary to determine the rotation angle. Then, integral of magnetic field along the flight path of polarized neutrons can be quantified.

The usage of the pulsed neutron beam, whose energy can be recorded precisely by the time-of-flight (TOF) method, possesses an advantage on the measurements of energy-dependent phenomena, so the polarized pulsed neutron beam is suitable for the magnetic field imaging experiments. Recently, we
have performed the first proof-of-principle experiment of polarized pulsed neutron imaging and successfully confirmed the possibility of quantification of magnetic field strength inside the solenoid coil and the direction from the quantized axis of the incident neutron spin by analysing the wavelength dependence of polarization after transmission of the sample [24].

On the other hand, to analyse the magnetic field vector, 3D polarization analysis is required, for which each polarization vector of incident and transmitted neutron beams was controlled three dimensionally [25]. The polarization change due to the spin rotation in a magnetic field is described by a $(3 \times 3)$ matrix $D$, and its elements are expressed by direction cosines of the magnetic field vector [26]. Thus, measuring and analysing the wavelength dependence of each element of matrix $D$ at each position allow determination of the spatial distribution of the magnetic field vector. Preliminary imaging experiments using 3D polarization analysis were performed at the beam line of BL10 of MLF at J-PARC. The schematic illustration of our experimental setup was shown in Figure 8. Electric currents, which changed synchronized with the flight time of the pulsed neutron to give the same spin rotation angle to wide wavelength range neutron beam, were applied to the spin rotators.

![Figure 8](image)

**Figure 8.** Schematic illustration of the experimental setup. The distance written in the bottom of the figure is from the source of pulsed neutron beam. Both polarizer and analyser consist of magnetic super mirrors. 2D detector was composed of a position-sensitive photomultiplier and a ZnS scintillator [27]. The beam size was 12 mm x 12 mm and the channel width of this detector was 0.25 mm.

After deciding the applied current condition of each spin rotator, test measurements were performed by superposing a small guide field at the centre of two spin rotators to check whether they work properly. Results of the wavelength dependence of matrix elements $D_{ij}$ ($i, j = x, y, z$, where $z$ axis is taken parallel to the beam direction) were shown in Figure 9. In this figure, matrix elements regarding $x$ and $z$ axis showed oscillating behaviour apparently. The diagonal elements $D_{xx}$ and $D_{zz}$ could be fitted with cosine curves though the off-diagonal elements $D_{xz}$ and $D_{zx}$ could be done with sine curves. This indicates that the magnetic field was along $y$ axis and its orientation was a positive direction. In addition, the averaged magnetic field strength was evaluated to be 0.175 mT from the oscillation frequency. These results were in good agreement with the results measured by a magnetic field detector.

![Figure 9](image)

**Figure 9.** Wavelength dependence of matrix element $D_{ij}$ ($i, j = x, y, z$). Incident neutron beam polarization was parallel to (a) $x$ axis, (b) $y$ axis and (c) $z$ axis. Solid lines indicated the results of fitting using a sinusoidal function.

Then, we placed a soft magnetic metal foil sample with a thickness of 0.03 mm, in the midway of the spin rotators and performed imaging measurements. This soft magnetic foil was a commercial...
amorphous Fe alloy used for the core of an electric transformer, with almost isotropic magnetic properties. The sample size was 50 mm in height, 30 mm in width and the thickness of 0.03 mm. The neutron beam passed across the central 12 mm by 12 mm region. The obtained images of diagonal elements were shown in Figure 10. The images of $D_{xx}$ and $D_{zz}$ showed stripe patterns, whose width were 2 ~ 3 mm, while that of $D_{yy}$ was almost homogeneous. These results suggest that the component of magnetic field vector along $y$ axis is homogeneous inside the foil, but $x$ and $z$ axis components were different at each position. The stripe structures shown in the images of $D_{xx}$ and $D_{zz}$ can be regarded as the evidences of the magnetic domain structure in the soft magnetic foil. Moreover, the features of polarization distribution images shown in Fig. 10 can be explained by a picture, that the small magnetic field applied along $y$ axis magnetized the sample but partially there remained some magnetization component which could not align with the applied field. In other words, the magnetization vector inside the foil almost directed along the $y$ axis, but it should canted toward $x$ or $z$ direction.

Because of insufficiently optimized set up, quantifying each component of the magnetic field vector inside the sample using off-diagonal matrix elements was difficult. By removing the stray field and tuning the spin rotators more precisely, polarized pulsed neutron imaging using 3D polarization analysis method would enable the quantitative imaging of arbitral magnetic field vector.

![Figure 10. Images of matrix element $D_{ii}$ (i = x, y, z) of soft magnetic metal foil sample. The incident neutron wavelength was 4.26 Å. The right figure is the photograph of the sample. FOV means field of View.](image)

5. Conclusions
It has been demonstrated that the pulsed neutron imaging can give image of the crystallographic information by one measurement. Such information for thick materials has not been obtained by other method such as X-ray and electron beam scattering. The imaging of the hydrogen dynamical (bound state) information was tried. The difference of the gradient of the total neutron cross section was indicated in two sample, which suggests the possibility to obtain the information on the hydrogen bound state. However, for the detailed data analysis we need a simulation calculation using a precise neutron cross section. Furthermore, it has been indicated that the pulsed neutron measurement can give easily an image of the strength of the magnetic field. The pulsed neutron imaging is still at developing stage and further improvement is required to deduce more precise quantitative information and also to deduce the new information that has not been obtained so far.

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