

Study of the linear distribution of absorption spectra at the Ta L3 edge in $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ crystals using X-ray beam spectral-spatial modulation using adaptive X-ray optical elements

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Abstract

Based on previously proposed approaches for implementing monochromatic X-ray beam rapid spectra modulation method using adaptive X-ray optical elements, a new spatially resolved technique has been developed. This technique was applied to record the tantalum L3 edge absorption spectra linear distribution with 37 μm spatial resolution. This method accelerates the sample's structure study rate by increasing the illuminated area compared to the classical X-ray absorption spectroscopy method and increasing the volume of information obtained relative to X-ray fluorescence. This approach is promising for studying crystalline materials with complex structure used in the microelectronics industry.

Keywords

adaptive X-ray optics, X-ray spectroscopy, quick extended X-ray absorption spectroscopy (qEXAFS), synchrotron studies

1. Introduction

This work expands the adaptive X-ray optical elements (AXOE) application means in time-resolved X-ray spectroscopy methods. A bending piezoelectric actuator are based on a bidomain lithium niobate monocystal (LiNbO_3), viable materials for acoustoelectronics,

acousto-optics, and optoelectronics due to their high piezoelectric coefficients [1]. They are widely used in acoustic, optical, and piezoelectric sensor technologies, and using surface acoustic waves (SAW) they can be applied in acoustic filters, gas sensors, and chemical and biological sensors [2]. Here, the piezoelectric properties of LiNbO_3 are utilized in X-ray techniques making possible

for the X-ray diffraction element to adjust the angle at a frequency of ~ 100 Hz within a range of $\sim 0.3^\circ$ or higher [3–6]. Thus, AXOE can be integrated into classical X-ray optical schemes to increase the modulation speed of a monochromatic X-ray beam parameters and improve the efficiency of measurements.

Previously, the X-ray beam parameters rapid modulation method using adaptive X-ray optical elements (AXOE) has been successfully applied to time-resolved X-ray spectroscopy research implementation [7, 8]. In these researches, a simplified single-crystal scheme for X-ray beam monochromatization with one AXOE was used. In this geometry, the monochromatic beam spectral parameter rearrangement (energy or wavelength), leads to the spatial parameter of the beam (angle of radiation propagation) alteration as well. In classical time-resolved X-ray spectroscopy technique (quick Extended X-ray Absorption Spectroscopy, qEXAFS), the monochromatic beam spatial displacement was an undesirable effect. In the current work, the phenomenon of spatial beam rearrangement during spectra modulation using AXOE was utilized as the basis for a method of absorption spectra linear distribution registration.

A similar X-ray beam spatial modulation is typically used in the Energy Dispersive X-ray Absorption Spectroscopy (EDXAS) method. Using a bended crystal monochromator, the polychromatic X-ray beam is focused in one or two planes on the sample. In the work [9], the polychromatic beam is focused along the vertical axis while remaining parallel along the horizontal axis, enabling 1D scanning in the EDXAS technique. The advantage of this approach is the increased temporal resolution compared to the XAS method classical implementation, as the absorption spectra information is recorded in a single frame of a 1D/2D detector. The drawback of the EDXAS technique is that the spectral resolution of the recorded spectra is limited by the used detector pixel size.

Thus, this work advances the topic of using AXOE for time-resolved X-ray spectroscopy methods. Although the AXOE fast angular modulation capability is not utilized to achieve time resolution, the spatial-spectral beam modulation enabled by AXOE allows the implementation of an EDXAS-like time-resolved method without the need for specialized bended crystal-monochromators.

2. Experimental methodology

Developing the optical layout of 1D XAS method, a single-crystal monochromatization scheme with an AXOE as the radiation analyzer was chosen as the basis, similar to those used in [7, 8], implemented at the “PRO” station of the “KISI-Kurchatov” synchrotron source [10]. AdvaPIX TPX3 2D X-ray detector was used, and the synchrotron beam was collimated into a narrow horizontal line to create a linear illuminated area on the sample and the AXOE analyzer crystal (Fig. 1a).

With a static AXOE, a horizontal illumination area from the diffracted linear beam is observed on the sensor of the two-dimensional detector (Fig. 1a), propagating at a double Bragg angle θ_0 with the corresponding monochromatic energy E_0 . When AXOE oscillations are excited, the diffracted beam spectral-spatial modulation starts, leading to its displacement in the vertical plane. During integral recording by the two-dimensional detector, this results in vertical broadening of the illuminated area (Fig. 1b).

Thus, using linear beam collimation and the AXOE, a two-dimensional spatial modulation of the beam is created in the detector sensor area. When recording the intensity of radiation passing through the sample, horizontal X-axis position of a point corresponds to the linear position of the illuminated point on the sample. During spectral-spatial modulation of the monochromatic beam

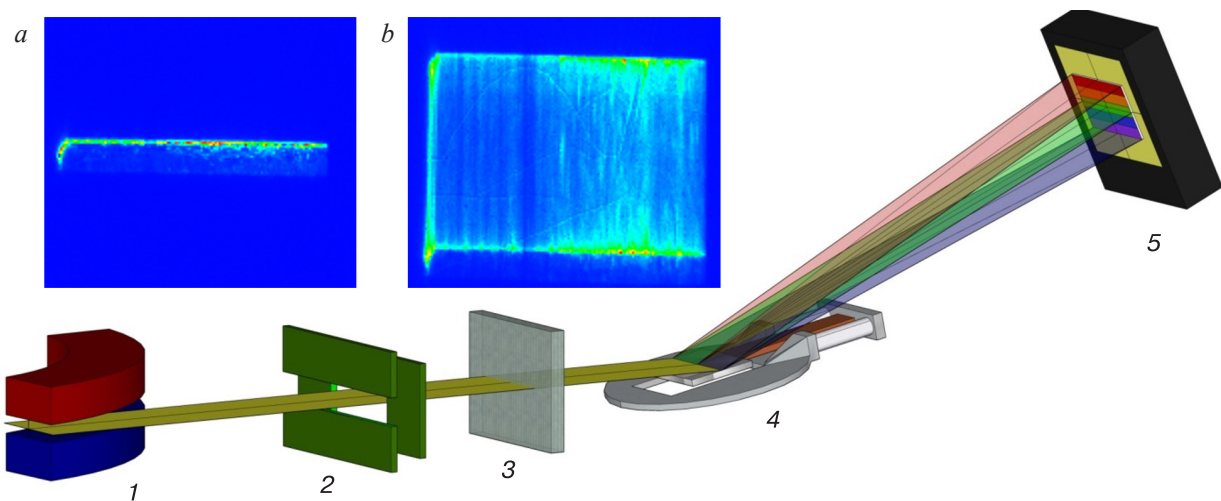


Figure 1. X-ray optical layout of the time-resolved 1D spectroscopy method: (1) synchrotron radiation source, (2) collimating slits, (3) sample, (4) adaptive X-ray optical element (AXOE), (5) 2D detector. Illuminated area on the detector for: (a) static AXOE and (b) active AXOE

using the AXOE, the Bragg diffraction condition is maintained, meaning there is a direct relationship between beam vertical angular deviation of the $\theta = \theta_0 + d\theta$ and the energy of the deviated beam $E = E_0 - dE$. Therefore, the vertical intensity distribution $I(y)$ in the detector illuminated area is converted into the radiation spectrum $I(E)$ for each horizontal coordinate x in the linear illuminated area of the sample, with this information $I(x,y)$ obtained in a single frame of the two-dimensional detector.

To record the absorption spectrum in the described method, it is necessary to record a reference frame of the intensity distribution $I_0(x,y)$ of the transmitted beam without the sample (instrumental function) to obtain the distribution $I_0(E,x)$ for fixing local spectral-spatial beam inhomogeneities arising during its generation, collimation, or diffraction. Based on the reference frame, the linear distribution of the absorption spectrum

$$\mu(E,x) = \frac{1}{d(x)} \ln \frac{I_0(E,x)}{I(E,x)},$$

is calculated, or

$$(E,x) = \ln \frac{I_0(E,x)}{I(E,x)},$$

in the case of a parallel-plane sample.

3. Experimental setup and equipment

The object of study was a $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ crystal – an important material in microelectronics and optoelectronics due to its unique properties, such as the piezoelectric effect, electro-optic effect, nonlinear optical characteristics, and high transparency in a wide spectral range [11, 12]. $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ combines the best properties as high piezoelectric coefficients of LiNbO_3 and high thermal stability of LiTaO_3 . Also, $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ material's phys-

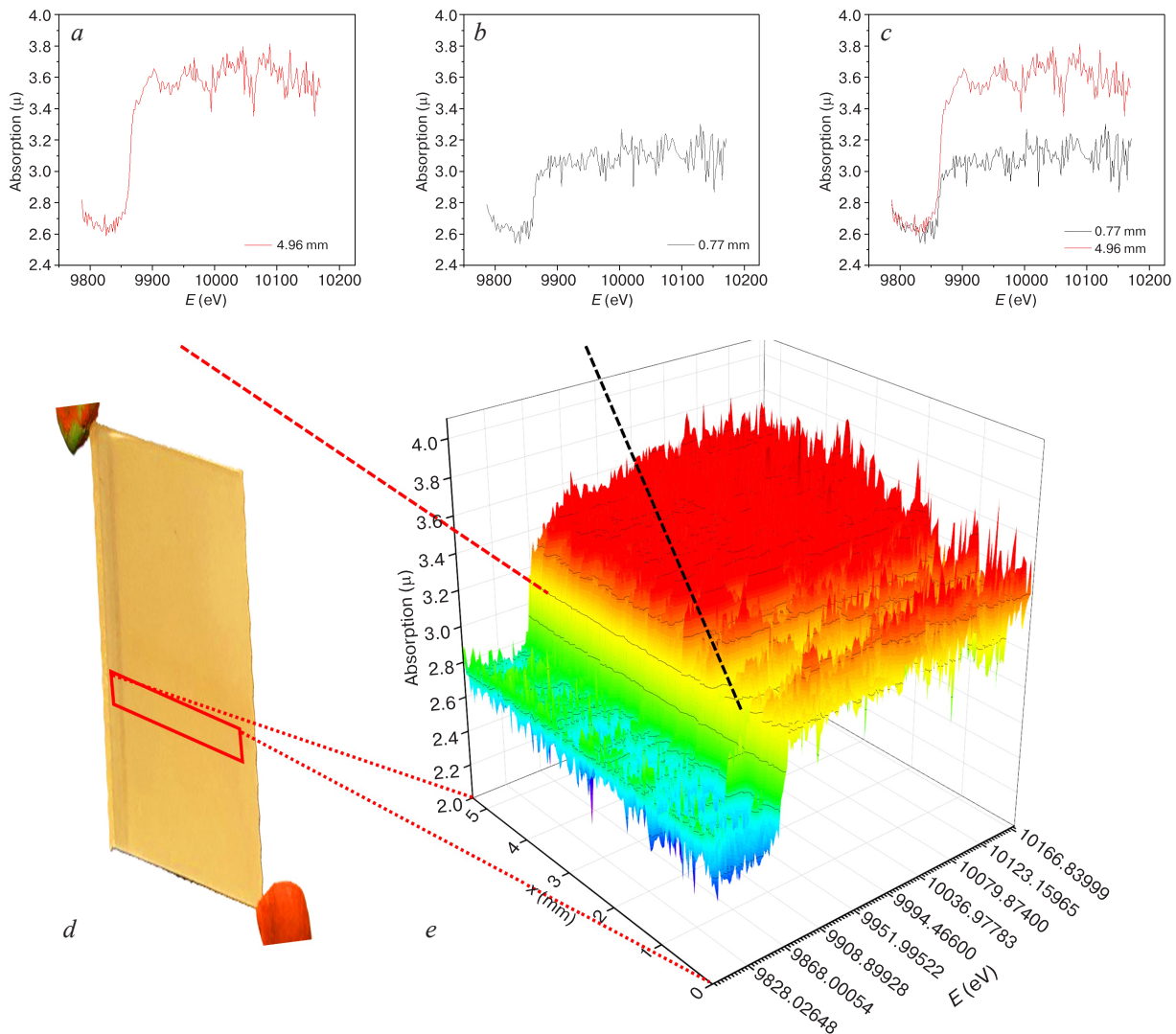


Figure 2. (a) Ta L3 edge at 4.96 mm position, (b) Ta L3 edge at 0.77 mm position, (c) Ta L3 edges at 0.77 mm and 4.96 mm comparison, (d) schematic linear X-ray illuminated area on $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ sample and (e) corresponding Ta L3 edge spectra linear distribution

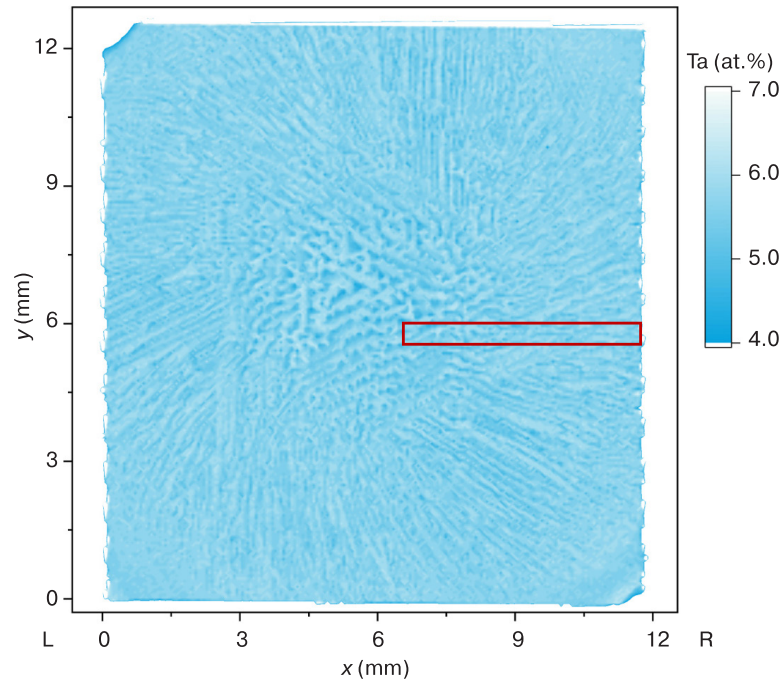


Figure 3. Ta concentration distribution in the $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ sample, recorded by fluorescence spectroscopy (XRF) method. The illuminated sample area during 1D spectroscopy is highlighted

ical properties can be significantly adjusted by changing its composition. In addition to general acoustoelectronics, acousto-optics, and optoelectronics applications, ferroelectric $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ crystals can be used to create layered structures on semiconductor crystals of Si or Ge [13].

The sample was grown from a charge composition of 95 mol.% LiNbO_3 and 5 mol.% LiTaO_3 using the Czochralski method with parameters adapted from those used for pure LiNbO_3 : reduced pulling rate, increased

rotation speed, and higher temperature gradient at the crystallization front. A thin $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ monocrystal was placed on a “white” beam collimated to linear area 5.5×0.1 mm.

The AXOE used as X-ray analyzer crystal and had an oscillation frequency of 90 Hz and an angular readjustment range of 780 arcseconds, and was set at the Bragg angle for the Ta L3 absorption edge (9.88 keV). The two-dimensional AdvaPIX TPX3 detector with a sensor size of 13×13 mm was placed at a 400 mm distance from

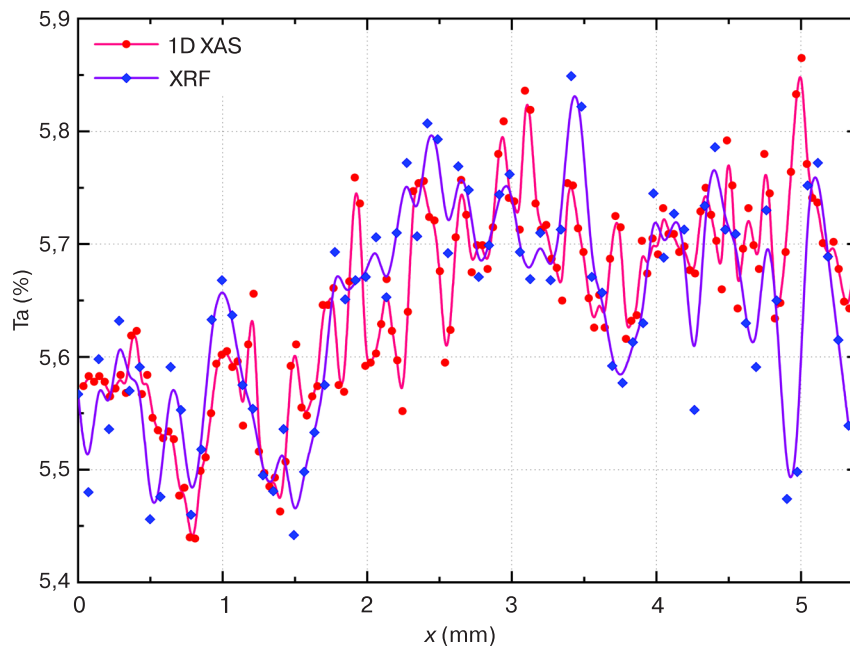


Figure 4. Ta concentration linear distribution obtained by: 1D spectroscopy with AXOE (red line) and X-ray fluorescence spectroscopy (blue line)

the AXOE, with the modulated beam vertical displacement on the detector sensor reaching 4.95 mm.

The spectral modulation range of X-ray monochromatic beam reached 130 eV when operating at 9.88 keV. To increase the range of recorded absorption edge spectra, a series of 5 frames was recorded with a stationary detector and sequential adjustment of the AXOE angular position, ensuring the illuminated area covered a larger vertical range, and the spectral range reached 383 eV. Similarly, reference frames without the absorber were recorded. The accumulation time for a single frame was 60 s.

Based on the recorded series of beam intensity distribution passing through the sample and the reference frames, the linear distribution of the absorption spectra at the Ta L3 absorption edge was calculated (Fig. 2).

In the obtained experimental data, the signal-to-noise ratio does not allow the absorption spectrum fine structure identification, presumably due to the low Ta content in the sample. However, the position of the absorption edge and the local absorption step, corresponding to the local Ta concentration distribution in the illuminated area, can be determined with high accuracy.

The distribution of tantalum content in the $\text{LiNb}_{(1-x)}\text{Ta}_x\text{O}_3$ sample was also investigated using X-ray fluorescence spectrometry, which allows for the study of element distribution on the surface or in the volume of the sample by recording the excited characteristic X-ray radiation. The results of the distribution obtained by X-ray fluorescence spectrometry [14] are shown in Fig. 3.

Based on the Ta two-dimensional distribution in the sample obtained by fluorescence spectroscopy (XRF), the Ta linear distribution in the area studied by the 1D spectroscopy method using the AXOE was extracted. The obtained relations are shown in Fig. 4.

The obtained relations correspond to each other, with discrepancies in individual peaks possibly due to local mismatches between the actual illuminated area in the 1D spectroscopy method and the calculated illuminated area selected from the fluorescence image.

The presented curves show similar trends, such as reduced Ta concentration in the 0–1.7 mm region and the repetition of several oscillations in Ta concentration that arose during the sample growth process. Local discrepancies in the dependencies may result from the mismatch between the sampling area of the spectroscopy (see Fig. 3) and the actual illuminated area of the sample. It should be noted that the fluorescence spectroscopy method spatial resolution is 71 μm , while the 1D spectroscopy method resolution is 37 μm .

A comparison of the obtained Ta concentration linear distribution in the sample demonstrates the legitimacy of the absorption spectra processing results obtained by the 1D spectroscopy method using AXOE. Thus, the Ta L3 absorption edge spectra linear distribution obtained by the 1D spectroscopy method contains information about the absorption edge local position and the absorption step, sufficient to obtain information about the element concentration linear distribution and chemical state of the selected element in the studied sample.

Conclusions

The implemented method of 1D absorption spectroscopy using adaptive X-ray optical elements (AXOE) allows of absorption spectra linear distribution recording with high spatial and spectral resolution in a single detector frame.

This methodology is promising for studying samples with a gradient in chemical composition with temporal resolution. The temporal resolution achieved in this study – 60 s per frame – exceeds the speed of linear imaging by XRF or XAS methods and can be significantly improved with the use of a more intense X-ray source. The spatial and spectral resolution of the method can be enhanced by using a detector with higher resolution.

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Author contributions

Eliovich Ya.A. – development of the topic and experimental technique, supervising, text review; **Protsenko A.I.** – development of experimental technique and conducting the experiment, text and data preparation; **Kulikov A.G.** – fluorescence spectroscopy realization, **Mololkin A.A.** and **Fakhrtdinov R.R.** – sample synthesis and preparation; **Korzhov V.A.** – conducting the experiment, data preparation; **Pisarevsky Yu.V.** – development of experimental technique; **Kovalchuk M.V.** – supervising.

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