Crystal monochromator with a resolution beyond 10^8

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Monochromatization with crystal diffraction has been achieved to a resolution ($\lambda/\Delta\lambda$) beyond 10^8. The monchromator is specifically designed for 23.880 keV synchrotron radiation (λ = 51.9 pm) for applications involving nuclear resonant scattering from ^{119}Sn. The design uses asymmetrically cut silicon (12 12 12) crystal reflections from two single-crystalline monoliths oriented in a dispersive geometry. A transmitted energy bandwidth of 140 $\pm$ 20 μeV was measured, corresponding to a resolution of 1.7 $\times$ 10^8. Methods of improving efficiency, wavelength stability and resolution are discussed.

Keywords: X-ray optics; crystal monochromators; nuclear resonant scattering; phonons; SnO.

1. Introduction

High-resolution monochromatization has important spectroscopic and metrological applications. At wavelengths corresponding to hard X-rays, applications include nuclear resonant scattering (Gerdau & de Waard, 2000), inelastic X-ray scattering (Burkel, 1991) and precise wavelength measurements (Shvyd'ko *et al.*, 2000). Improving the monochromator resolution has important implications for these measurements. Furthermore, there exists a range of resolutions that are inaccessible with X-rays that spans from present-day meV-resolution monochromators to the sub-μeV resolution of nuclear resonant scattering and radioactive sources. Ultra-high-resolution crystal monochromators can help to reduce this gap by producing bandwidths substantially below 1 meV.

Typically, a high-resolution monochromator improves the resolution of the incident beam from a pre-monochromated level of 10^4 to a resolution that is in the range 10^6–10^8 (Toellner, 2000). For 23.880 keV X-rays, previous high-resolution monochromators have achieved resolutions as high as a few times 10^7 (Mooney *et al.*, 1994; Chumakov *et al.*, 1998; Hu *et al.*, 1999). In the case of nuclear resonant scattering with synchrotron radiation, which is the specific purpose of the monochromator to be discussed here, there is a distinct advantage to improved resolution. For inelastic nuclear resonant scattering measurements, from which partial phonon density of states (PDOS) for the resonant isotope can be obtained, the resolution of the data is determined solely by the resolution of the incident radiation (Seto *et al.*, 1995; Sturhahn *et al.*, 1995). Therefore, the ability to resolve structure in the PDOS, as well as the ability to measure low-energy dynamical excitations that would normally lie too close to the elastic peak to be detected, are limited by the resolution of the monochromator. This motivates current attempts to improve the resolution of crystal monochromators that can be tuned at and around nuclear resonant energies.

This paper presents a widely tunable monochromator for 23.880 keV X-rays that is based upon the use of highly asymmetric ($\Theta_B - |\alpha| < 1^\circ$) Bragg diffraction from single crystals. Although the specific design focuses on ultra-high resolution ($>10^8$), the concept has applicability even for medium-to-high-resolution monochromatization over the entire range of wavelengths accessible with crystal diffraction. Achieving a desired resolution at a given wavelength requires one to make a judicious choice from accessible crystal-lattice reflections while considering efficiency and other practical issues. Even though the crystal monochromator presented here is for X-rays, the design principle and the related issues to be discussed are relevant to any quantum field amenable to crystal diffraction, *e.g.* neutrons.

2. Description of the optic

A simple multicrystal arrangement for achieving high resolution involves using two symmetric high-index crystal reflections that are arranged in a (+,+) geometry (Faigel *et al.*, 1987). This method has the flexibility to allow one to select the energy but has lower efficiency when compared with back reflections. To improve the efficiency and the resolution that one can obtain with this method, it is possible to use crystal reflections with extreme asymmetry angles (Toellner, 1996; Chumakov *et al.*, 1996). This method has been applied to the 14.413 keV nuclear resonance of ^{57}Fe to achieve sub-meV energy bandwidths using single-crystalline silicon (Toellner *et al.*, 1997; Chumakov & Rüffer, 1998).

Single-crystalline silicon is currently the best material for multicrystal high-resolution monochromators because high-quality silicon is commercially available in large quantities. Apart from monochromator design, fabrication and mounting issues that may affect long-range crystallinity through induced strain fields, crystal quality ultimately limits the maximum resolution that is attainable. Intrinsic crystal quality is determined largely by the growth process, which leads to impurities, vacancies *etc.* For a semiconductor, resistivity is a partial measure of crystal quality because it depends upon impurity concentration (Pawlik, 1988), which correlates with lattice constant variation (Windisch & Becker, 1990). Thus, to help avoid a crystal-quality-limited resolution, we used zone-refined very high resistivity single-crystalline silicon. Although the resistivity at the diffraction region was not measured, measurements performed by the manufacturer indicate that the resistivity should be in the range 60–130 kΩ·cm (Topsil, 2000).

Typically, one chooses the highest-order lattice reflection available, which for 23.880 keV X-rays (λ = 51.9 pm) corresponds to the silicon (17 11 5) crystal reflection. For our purposes though, we chose the silicon (12 12 12) crystal reflection, which is the second highest, because its larger X-ray scattering factor leads to better monochromator efficiency, albeit with a slight reduction in expected resolution. The Bragg angle for 23.880 keV X-rays diffracting from the silicon (12 12 12) crystal reflection at room temperature is 83.46°. Two monolithic crystals (25 mm $\times$ 25 mm $\times$ 60 mm) were given confirmed asymmetry angles of α = -83.0° (asymmetry factor $b \simeq 0.034$) for the first crystal and α = $+83.0^\circ$ (asymmetry factor $b \simeq 29$) for the second and arranged as shown in Fig. 1. The asymmetry angle on the first crystal results in an incidence angle of 0.46° with respect to the surface. This is well above the critical angle for total external reflection of 0.08°.

Non-uniformity of the asymmetry angle, *i.e.* slope error, can affect the transmitted bandwidth by inducing a distribution of refractive angular shifts between the crystals. In order to consider potential contributions to bandwidth broadening, we estimate the angular broadening due to asymmetry variation by calculating the distribution of refractive shifts and translate that into an energy spread

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(Toellner, 1996). Away from backscattering, the refractive angular shifts for the incident (Δ_-) and diffracted (Δ_+) beams in the asymmetric Bragg case are given by

$$\Delta_\pm = \frac{|\tilde{\psi}_{r0}|}{2\,\sin(2\Theta)}(1 + b^{\pm 1})\,, \qquad (1)$$

where $\tilde{\psi}_{r0}$ is the real part of the forward ($\vec{H} = 0$) Fourier component of the susceptibility, Θ is the kinematical Bragg angle, and $b = \sin(\Theta + \alpha)/\sin(\Theta - \alpha)$ is the asymmetry parameter. The variation of the refractive shift due to the variation of the asymmetry angle yields

$$\delta\Delta_\pm = \pm\left(\frac{|\tilde{\psi}_{r0}|}{2\,\sin^2(\Theta - \alpha)}\right)\left(\frac{b^{\pm 1}}{b}\right)\delta\alpha\,. \qquad (2)$$

From equation (2), one can obtain an estimate of the broadening of the transmitted bandwidth due to slope error from

$$\delta E \simeq E_0\,\frac{|\delta\Delta_+|_1 + |\delta\Delta_-|_2}{\tan\Theta_1 + \tan\Theta_2}, \qquad (3)$$

where 1 and 2 refer to the first and second crystal, respectively. Optical measurements of the surface flatness indicate a variation of asymmetry angle ($\delta\alpha$) over the diffraction region on both crystals of approximately 5 µrad. This amount of asymmetry variation leads to a broadening of approximately 0.2 µeV in the present case and therefore cannot contribute significantly to any measured broadening.

The energy resolution function, which gives the distribution of spectral flux transmitted as a function of energy, depends upon characteristics of the incident beam. In the present case, the diffraction plane is vertical, and the incident field has its polarization vector perpendicular to that plane (σ-polarization). The resolution function for the monochromator depends upon the distribution of incident radiation in vertical angle, as well as in horizontal angle. Fig. 2 shows a plot of the calculated transmission function of the monochromator for σ-polarized X-rays as a function of energy and vertical angle assuming zero horizontal divergence. Fig. 3 shows a plot of the calculated transmission function of the monochromator for σ-polarized X-rays as a function of energy and horizontal angle assuming zero vertical divergence. The horizontal angle–energy dispersion shown in Fig. 3 is significant for sources that are widely divergent in a plane that is perpendicular to the diffraction plane (Bortel *et al.*, 2000). For incident radiation that is highly collimated in the horizontal plane (42 µrad for the source used here), there is negligible change in the resolution function from the zero-horizontal-divergence limit. For σ-polarized X-rays, the theoretical energy bandwidth expected from this design in combination with an undulator source is 0.11 meV full width at half-maximum (FWHM) while its angular acceptance is 1.5 µrad FWHM. On the other hand, for a more horizontally divergent X-ray source, *e.g.* synchrotron radiation from a bending-magnet source, 1 mrad of horizontal divergence (rectangular distribution) would result in a resolution function that is broadened to 0.3 meV FWHM with a highly asymmetric distribution. Also, it is generally preferable for high-resolution monochromators to be used with the smallest beam size possible in order to maintain a small diffraction region. This minimizes resolution degradation due to a lack of ideal crystallinity that arises from thermal gradients, strain gradients and impurity gradients within the crystal. Consequently, this design is more suitable for small beams that are highly collimated along both transverse directions.

The tunability range of the monochromator depends strongly upon the chosen criteria. Due to the wavelength-dependent asymmetry factor, the transmitted bandwidth increases as the wavelength increases. As a result, the monochromator can be tuned 100 eV lower in X-ray energy if one can allow the transmitted bandwidth to broaden to 0.3 meV. On the other hand, if one restricts the bandwidth broadening to a maximum of 10%, then the tunability range is only 7 eV. Independent of the chosen criteria though, the tunability range

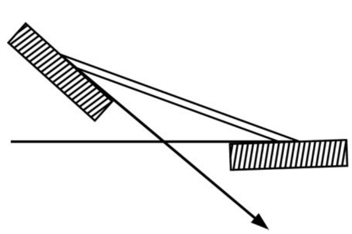

Figure 1
Design of the ultra-high-resolution monochromator. Two asymmetrically cut silicon (12 12 12) crystal reflections are oriented in a dispersive geometry.

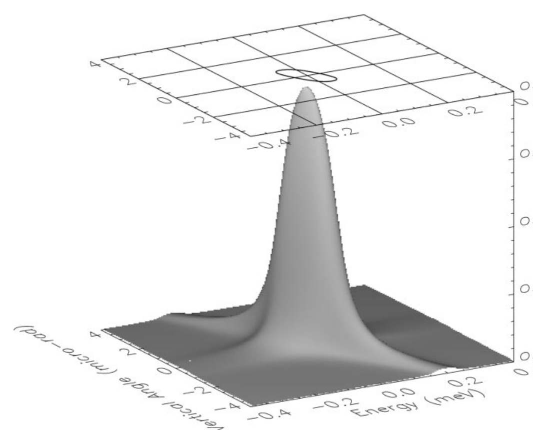

Figure 2
Calculated transmission function of the monochromator for σ-polarized X-rays as a function of energy and vertical angle assuming zero horizontal divergence. The contour is plotted at 50% of the peak transmission.

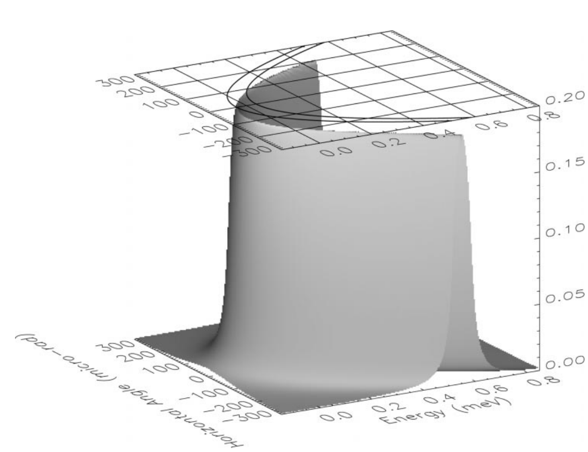

Figure 3
Calculated transmission function of the monochromator for σ-polarized X-rays as a function of energy and horizontal angle assuming zero vertical divergence. The contour is plotted at 50% of the peak transmission.

does not extend more than approximately 10 eV to higher energies due to the fact that the angle of the incident beam relative to the surface must remain above the angle for total external reflection.

A crystal monochromator with this degree of resolution places enormous demands on thermomechanical stability as well as angular control. Due to the room-temperature coefficient of thermal expansion for silicon, a temperature change on both crystals of 1 mK results in an energy shift of the transmitted X-rays of 0.06 meV. Thus, temperature monitoring is necessary. Thermistors are mounted approximately 1 cm from the diffraction region on both crystals to determine changes in temperature. To reduce thermal influences, additional measures are taken to minimize both thermal fluctuations and thermal drifts. Crystal strains introduced from mounting need to be minimized to reach the ideal energy width. The crystals are mounted on aluminium holders with a thin film of grease to mitigate mounting strains and are held in place with spring-loaded nylon pins. An angle-to-energy ratio of 0.73 μrad meV^{-1} implies that the calculated angular width of the theoretical resolution function is 0.082 μrad FWHM. To effect the necessary angular motions, the crystal assemblies are mounted on Kohzu high-resolution rotation stages, which have a minimum step size of 0.025 μrad. Calibration of the angular scale is performed with the use of Heidenhain ROD800C angle encoders with 4096 interpolation, which results in an encoder resolution of 0.043 μrad. Experience has shown that the angular motions necessary to scan the monochromator in energy are so small that vibrations can produce significant broadening in a measurement of the resolution function. Consequently, we mitigated vibrational influences by placing the rotation stages on a massive (1300 kg) granite slab supported by four 50 cm-tall sand columns.

3. Results

All measurements were carried out at the 3-ID undulator beamline at the SRI-CAT of the Advanced Photon Source. Synchrotron radiation produced from an undulator was pre-monochromated at 23.880 keV to a bandwidth of approximately 1.8 eV using a water-cooled diamond (1 1 1) premonochromator. Due to the many factors that might lead to disagreement between theory and experiment in the ultra-high-resolution limit, it is important to know the X-ray scattering factor in order to accept the theoretical results using the theory of X-ray dynamical diffraction. To verify the crystal scattering factor of the silicon (12 12 12) lattice reflection, we first measured the energy-resolution function of a monochromator composed of two silicon (12 12 12) crystals with a very modest asymmetry of 43° (asymmetry factor $b \simeq 1.2$) oriented in a (+,+) geometry. The energy-resolution function is measured by energy scanning the monochromator through the 23.880 keV nuclear resonance of ^{119}Sn while monitoring the delayed flux that is scattered into the forward direction. This measurement produces the resolution function of the monochromator because of the delta-function-like energy response of the nuclear resonance in this scattering geometry. The result is shown in Fig. 4 and gives an energy bandwidth of 0.47 meV FWHM. This measurement is important because it independently verifies the magnitude of the scattering factor of the silicon (12 12 12) lattice reflection including the room-temperature Debye–Waller factor in a way that avoids the experimental difficulties associated with ultra-high resolution, such as thermal stability, crystal-quality-limited resolution *etc*. To simulate the data we use an X-ray scattering factor (including Debye–Waller factor) of $(1.94 + 0.053i)$ electrons/unit cell, while the room-temperature Debye–Waller factor is 0.18.

To characterize the ultra-high-resolution monochromator we worked at a distance of 63 m from the source. At this distance, and given the vertical source size of this beamline of 0.05 mm, the vertical spatial acceptance of the monochromator is 0.14 mm for this source. A measurement of the resolution function of the monochromator is shown in Fig. 5. The measured energy bandwidth that is transmitted is 0.14 meV FWHM, corresponding to a resolution of $\lambda/\Delta\lambda = E/\Delta E = 1.7 \times 10^8$, which implies a longitudinal coherence length of 8.8 mm. Currently, the measured transmitted flux in this bandwidth is approximately 4×10^6 photons s^{-1} at 100 mA of storage-ring current. In order to achieve this resolution, it was necessary to reduce the incident vertical beam size to 0.05 mm. The resulting size of the diffraction region was 6 mm along the diffraction plane and 4 mm transverse to the diffraction plane, while the extinction depths for each crystal are in the range 8–25 μm. The flux incident on the monochromator within the 0.05 × 4.0 mm beamsize was 7×10^{11} photons s^{-1}.

We used the ultra-high-resolution monochromator to measure the vibrational excitations in a ^{119}SnO powder using the method of inelastic nuclear resonant scattering (Seto *et al.*, 1995; Sturhahn *et al.*, 1995). The result of approximately seven hours of data collection is shown in Fig. 6.

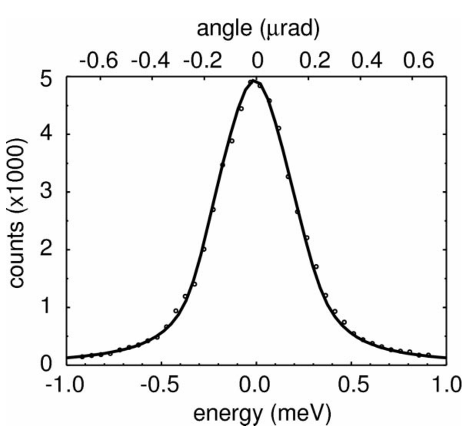

Figure 4
Energy-resolution function for two silicon (12 12 12) crystals with a very modest asymmetry of 43° (asymmetry factor $b \simeq 1.2$) oriented in a (+,+) geometry. The solid line is a fit using the theory of dynamical diffraction and is used to determine the silicon (12 12 12) structure factor.

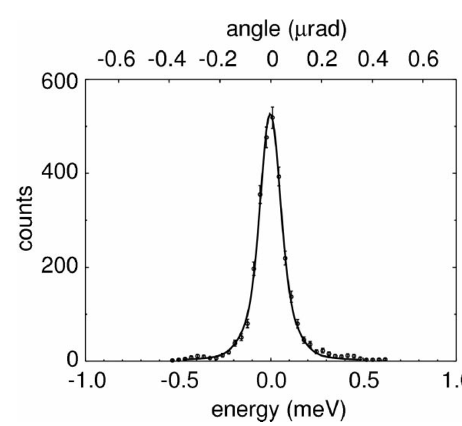

Figure 5
Energy-resolution function for the ultra-high-resolution monochromator. The solid line is a simulation using the theory of dynamical diffraction with the structure factor determined from the result of Fig. 4 and its energy scale stretched by 27%. The FWHM is 140 μeV.

4. Discussion

The discrepancy between the measured bandwidth of 0.14 meV and the expected value of 0.11 meV may be due to thermal effects, intrinsic crystal quality or mechanical strain. Thermal effects, such as gradients, fluctuations and drifting, affect the resolution function as well as the wavelength stability of the transmitted X-rays. Thermal fluctuations at the diffraction region are due to heat transfer at the convective-air–silicon interface, motion of the incident X-ray beam, which produces a local variation of the thermal load, as well as variation of incident-beam intensity. A direct temperature measurement at the diffraction region is difficult, but we did observe thermal fluctuations of 0.15 mK approximately 1 cm from the diffraction region. If this much thermal fluctuation exists at the diffraction region, then it would account for 0.01 meV in broadening of the resolution function. Another possible source for the bandwidth discrepancy may be related to a noted degradation of the measured bandwidth as the diffraction region was moved away from the central region of the crystals. This trend may be due to impurity gradients that can be inferred from the radial resistivity profile of the silicon ingot from which the crystals were cut. Also, thermal gradients at the diffraction region may cause bandwidth broadening, and simulations suggest that thermal gradients generally increase as one moves from the center of the crystal toward the edges. Crystal strain induced by the cutting and polishing procedure and possibly the residual strain of mounting may also contribute to the broadening despite precautionary measures taken to avoid such effects. Consequently, non-vanishing impurity concentration, which leads to an intrinsic lattice-spacing variation, thermal gradients, thermal fluctuations, and perhaps mechanical strain, may all contribute to the measured broadening of the transmitted bandwidth.

Beyond the issue of broadening, the data of Fig. 6 clearly demonstrate that inelastic nuclear resonant scattering with this degree of resolution is obtainable at current third-generation synchrotron sources. Despite this, ultra-high-resolution monochromatization can be significantly improved over the current implementation in terms of its efficiency, wavelength stability and resolution. The low efficiency has its origin in a mismatch between source divergence and the angular acceptance of the monochromator, as well as the low reflectivity of each diffracting crystal (*cf*. Fig. 2). The stability of the transmitted wavelength is directly related to the thermal sensitivity of the monochromator. Thermal influences affect the transmitted wavelength by altering the atomic interplanar spacing through thermal expansion and contraction of the lattice. The resolution is limited by crystal quality and thermal effects.

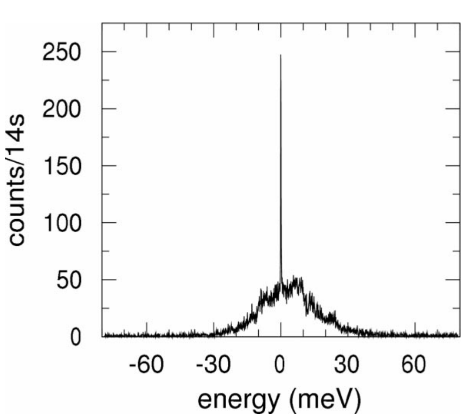

Figure 6
Inelastic nuclear resonant absorption spectrum of ^{119}SnO powder. The data took approximately 7 h to collect.

The efficiency can be improved over the current measurements by operating the monochromator as close as possible to the source, as well as through the use of collimating optics to reduce the divergence of the incident beam (Baron *et al.*, 1999). Beyond this, the low efficiency and severe thermal sensitivity can be mitigated by cryogenically cooling the crystals (Toellner, 2000). Cooling the crystals has a number of positive effects. First, at lower temperatures the Debye–Waller factor increases, which results in greater reflectivity for high-index lattice reflections. Second, the angular acceptance is proportional to the Debye–Waller factor and so also experiences an increase. Third, at lower temperatures the coefficient of thermal expansion decreases, resulting in reduced thermal sensitivity and therefore greater wavelength stability. Furthermore, in the special case of silicon, the coefficient of thermal expansion vanishes around 123 K before going negative at lower temperatures and finally vanishing at absolute zero. Thus, whether at 123 K or at very low temperatures, the sensitivity to thermal gradients, thermal fluctuations and thermal drifts vanishes whether produced by the incident beam or otherwise. Finally, maintaining the diffracting crystals at such cryogenic temperatures allows wavelength stability even in the presence of more intense radiation sources. It should be noted that it may not be sufficient to compensate for the low efficiency of the monochromator by increasing the strength of the radiation source alone, because this will increase the thermal load on the crystals and may degrade the monochromator's performance.

A side-effect of cryogenically cooling the monochromator is a slightly decreased resolution. The trade-off between efficiency and resolution is heavily favored for efficiency though, because the ratio of integrated transmission to transmitted bandwidth can improve by an order of magnitude or more, by cooling the monochromator. Another issue is the technical difficulty of cryogenically cooling the silicon crystals while maintaining the requisite angular stability.

In order to improve the resolution significantly beyond what was obtained here, it may be necessary to improve crystal quality to avoid a crystal-quality-limited resolution. Better crystal quality is achieved through reduced impurity concentrations, which includes isotopic impurities. Naturally abundant silicon contains 92.23% ^{28}Si, 4.67% ^{29}Si and 3.10% ^{30}Si (Lederer & Shirley, 1978). Due to the isotopic dependence of the lattice constant, an isotopic distribution induces a variation in interplanar spacing that can only be removed by manufacturing the crystal with a single isotope. If crystal quality is not a limiting factor and if thermal influences can be mitigated, then further improvements in resolution may be possible.

5. Conclusion

We have presented a 140 μeV-bandwidth monochromator for 23.880 keV X-rays. We have also used the monochromator for a measurement of the vibrational excitations in ^{119}SnO using the technique of inelastic nuclear resonant scattering. The data clearly demonstrate that inelastic nuclear resonant scattering with this degree of resolution is obtainable at current third-generation synchrotron sources – but also shows the need for increased flux. We have suggested ways to improve the flux by decreasing the size and divergence of the incident beam, as well as by improving monochromator efficiency through the use of cryogenically cooled crystals. Furthermore, wavelength stability is extremely important in metrological applications where unpredictable changes in wavelength can be a significant source of uncertainty. The use of cryogenically cooled crystals would also significantly enhance the stability of the transmitted wavelength as long as the necessary angular stability can be maintained. In order to improve the resolution significantly beyond what was obtained here, we have suggested the need for improved crystal quality. Improved crystal quality may require the use of isotopically pure single crystals.

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