Two-dimensional imaging and modification of nanophotonic resonator modes using a focused ion beam

WILLIAM R. MCGEEHEE,1,† THOMAS MICHELS,1,2,† VLADIMIR AKSYUK1, AND JABEZ J. MCCLELLAND1,*

1Center for Nanoscale Science and Technology, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
2Department for Microelectronic and Nanoelectronic Systems, University of Technology Ilmenau, Germany
*Corresponding author: jabez.mcclelland@nist.gov

High-resolution imaging of optical resonator modes is a key step in the development and characterization of nanophotonic devices. Many sub-wavelength mode-imaging techniques have been developed using optical and electron beam excitation—each with its own limitations in spectral and spatial resolution. Here, we report a 2D imaging technique using a pulsed, low-energy focused ion beam of Li⁺ to probe the near-surface fields inside photonic resonators. The ion beam locally modifies the resonator structure, causing temporally varying spectroscopic shifts of the resonator. We demonstrate this imaging technique on several optical modes of silicon microdisk resonators by rastering the ion beam across the disk surface and extracting the maximum mode shift at the location of each ion pulse. A small shift caused by ion beam heating is also observed and is independently extracted to directly measure the thermal response of the device. This technique enables visualization of the splitting of degenerate modes into spatially-resolved standing waves and permits persistent optical mode editing. Ion beam probing enables minimally perturbative, in operando imaging of nanophotonic devices with high resolution and speed.

Nanophotonic resonators, such as whispering gallery mode resonators, are sensitive devices that have been used to measure temperature [1], mechanical motion [2–4], and a range of biological and molecular signals with both high resolution and speed [5,6]. Further development of microfabricated resonator-based sensors and transducers necessitates new techniques for device characterization and testing: spectroscopy of optical modes is often insufficient, and rapid, non-invasive techniques for spatial mapping of the mode structures are needed. Mode mapping is also important for calibration of interaction volumes and local field enhancements as well as for understanding the specific effects of fabrication imperfections. In one example, imperfections can break the spatial symmetry of circular resonators, resulting in spectroscopically observed resonance line splitting. However, spatial information about the imperfections can only be obtained by mapping the modes.

A number of diverse approaches have been developed for imaging nanoscale photonic and plasmonic resonators, including optical techniques such as near-field scanning optical microscopy (NSOM) and photo-modulation spectroscopy (PMS) as well as electron-based techniques including cathodoluminescence (CL), electron energy loss spectroscopy (EELS), and photoemission electron microscopy (PEEM). Each approach presents a distinct set of advantages and tradeoffs with regard to spatial, spectroscopic, and temporal resolution. They also vary in their ability to measure in operando, i.e., to determine the spatial distribution of a specific desired mode chosen for excitation by a single frequency tunable source. This is particularly important for photonic structures with many high quality factor (Q), spectrally dense states.

NSOM has been widely used to map nanophotonic devices [7–9] and encompasses a broad category of tip-based scattering techniques. Advancements in this technique have allowed for local, phase-sensitive measurement of evanescent field vectors [10–12]. NSOM is fundamentally based on an interaction between the mode’s evanescent fields and an externally-introduced physical scatterer and, while it allows in operando measurements, it requires a tradeoff between the scattered signal strength and the probe-induced perturbation to the mode. This probe-induced perturbation has been extensively studied and can be used as a signal for high-resolution imaging [13,14]. The spatial resolution of NSOM is practically limited by the attainable probe geometries. PMS [15,16], in which a focused laser beam creates a local perturbation to the refractive index and is scanned across the surface, also allows in operando measurement. The spatial resolution of PMS is limited by diffraction to the focal spot size of the probe laser.

Electron beam techniques such as CL, EELS and PEEM are attractive because they make use of the high spatial resolution of electron optics to extend imaging of plasmonic structures to the nanometer scale, and avoid the type of tip-induced perturbation of the resonator seen with NSOM. A tradeoff in this case is that the measurements are generally not in operando: mode excitation is typically broadband, either by an electron beam or by an ultrafast laser, with less ability to select specific modes to study, unless the resonator is very small with just a few modes.

In CL measurements [17–24], a focused, scanning electron beam excites the resonator, and the spectrum of the cathodoluminescence provides a map of the local optical density of states. EELS experiments are similar, except the spectrum of the optical modes is observed in the energy loss of the electrons. [25] A related technique involves excitation with an ultrafast pulsed laser and observation of the spatial distribution of the modes by synchronized ultrafast electron microscopy [26,27]. This technique has the added benefit of excellent temporal resolution, in addition to having the spatial resolution of an electron microscope. PEEM also uses a pulsed laser to excite the optical mode, and either a UV
laser or a multiphoton process to generate photoelectrons from the surface of the resonator where mode intensity is present [28–30]. Imaging these electrons provides a map of the mode’s spatial distribution. In all these techniques, due to the broadband nature of electron or pulsed-laser excitation, it is challenging to study individual modes in spectrally dense resonators.

Recently, focused ion beams (FIB) have been introduced as a promising alternative for in operando measurement of nanophotonic resonator modes [31] with nanometer-scale resolution. As with NSOM and PMS, a specific mode is selected for imaging by tuning an external excitation laser, and the spectral resolution is limited only by shot noise in photodetection of the resonance excitation. Similar to PMS and in contrast to NSOM, the FIB-induced interaction is localized to just below the resonator surface, without perturbing the mode’s evanescent fields. However, like electron-based methods, advantage can be taken of the high resolution of charged particle optics, and the spatial resolution is limited only by the volume of the ion collisional cascade at the point of impact on the resonator, which can be of nanometer scale. With proper choice of ion species, beam energy, beam current and exposure time, high quality mode images can be obtained with minimal damage to the resonator. However, it should also be noted that focused ion beams have also been used to modify photonic devices for suppression of certain modes [32], and fine control over the location and strength of the FIB-induced modification of the resonator opens intriguing possibilities for fine-tuning of the optical modes with an unprecedented level of spatial control. For example, controllably perturbing resonators supporting multiple near-degenerate modes has been used to show phenomena such as mode coalescence and chirality at exceptional points [33,34].

Here we present an imaging technique that uses the time-varying response of a microdisk resonator to a pulsed ion beam of Li+. The temporal response of the resonator includes two primary effects: a spatially-dependent shift due to local modification of the device index and boundaries, and a rapid thermal shift in the resonance frequency from ion beam heating. We refer to these as the “optical” and “thermal” responses. These two effects occur over separate timescales and can be independently measured. Both responses are useful for device characterization, the optical shift for mapping the optical mode profile and the thermal shift for measuring thermal transport within the device. In our previous work [31], a modulated ion beam was used to make linear scans of the optical mode distribution. Here, we use detailed fitting of the time-varying response to increase the signal-to-noise ratio for a given ion dose, which allows quantitative separation of the two effects. These improvements enable increased speed of imaging, resulting in an ability to acquire two-dimensional images of the optical field without causing significant change in mode Q or resonant wavelength. For single resonances, we observe azimuthally-symmetric patterns as expected from the associated single-direction traveling wave fields. However, measuring on one component of a doublet line in the microdisk spectrum, we observe an azimuthally-periodic standing wave pattern, which is a result of the breaking of the rotational symmetry of the microdisk; presumably by a defect or fabrication imperfections. Notably, we show that significant persistent modification in the mode spectrum and the spatial pattern can be induced by extended application of the ion beam. As the first examples of such mode editing by the focused ion beam, we show permanent shifting of the resonance line by more than a linewidth, and conversion from spectral doublet to a singlet with the accompanying restoration of the azimuthally-symmetric traveling wave pattern.

![Fig. 1. Ion pulse imaging of microdisk resonator modes. (a) The silicon microdisk (10 μm diameter) is housed in a Li+ FIB vacuum chamber with optical fibers connecting the interrogating laser and photodiode to the device. The ion beam is scanned across the resonator mode (dashed line) and perturbs small volumes in the device near the disk surface. The resonator’s mode intensity (shown in cross-section) varies both radially and through the device thickness. (b) Spectroscopy of a 0th order transverse magnetic (TM) mode is shown with Q ≈ 20,000 at λ ≈ 1523 nm. The ion pulses incident on the microdisk shift the resonance to longer wavelength, and this shift is read out by tuning the laser (dashed line) to the low frequency side of the resonance and observing the time-varying change in optical transmission of the device. (c) The change is transmission (blue lines) is shown for a series of 0.5 ms ion pulses (blue bands) incident near the mode maximum, and the absolute magnitude of the spectral shift can be calculated from the transmission measurement in (b). Between each ion pulse, the laser is adjusted (orange band) to a nominal absorption value on the side of the resonance by turning on a slow feedback circuit.](image-url)
spectral shifts of the microdisk resonances are observed by positioning the probing laser at the low frequency side of a microdisk resonance and recording the transmitted power during ion dosing. The transmitted light is measured with a low noise photodetector, and the frequency response of the disk as shown in Fig. 1(b) is used to convert changes in optical absorption into a known spectral shift. This is calibrated by simultaneously recording the absorption spectrum from the microdisk and a Fabry-Perot cavity with a calibrated free spectral range of ≈ 1490 MHz. This side-of-line spectroscopic technique provides a direct measure of the resonances shifts, but is susceptible to drift arising from amplitude and frequency noise in the laser, polarization drift in the coupling fibers, and thermal drift of the microdisk resonances. Each of these effects is found to be negligible for the measurement duration described here. Laser frequency and photodetection shot noise are the dominant noise sources. Experiments are nominally performed at room temperature.

The focused Li⁺ ion pulses are positioned using the FIB as a microscope and registering the beam position using secondary electron images of fiducial marks on the device. The beam is then step-wise rastered across the structure with a series of ion pulses between 0.2 ms and 0.5 ms in duration. The ion pulses are spaced in time by ≈ 20 ms to allow time for recording the relaxation of the optical shift and for repositioning the laser to a nominal position on the side of the resonance feature where the sensitivity to spectral shifts is maximal as shown in Fig. 1(c). The tuning of the laser between ion pulses is accomplished using a sample-and-hold limited feedback circuit.

The response to a series of ion pulses across the edge of the microdisk structure is illustrated in Fig. 2. In this measurement, a sequence of ≈ 0.5 ms ion pulses containing ≈ 3000 Li⁺ ions each is incident near the edge of the microdisk. The magnitude of the response as the beam is scanned across the disk edge due to the spatial dependence of the optical mode. The ion-induced shift of the microdisk resonance is analyzed independently for each pulse using least-squares fitting to a two-component model. The two components represent a spatially-local shift arising from ion damage Δoptical and a position-independent shift arising from ion-beam heating Δthermal. The optical shift arises from modifications of the silicon lattice that change the local index and boundaries of the device. This modification of the device grows during the ion pulse (with length t_d) and relaxes afterward; we model this behavior through the piecewise-continuous function of time t expressed in Eq. (1).

The optical shift dynamics during the ion dose (0 < t ≤ t_d) are modeled as linear growth with exponential saturation to capture the finite bound on damage to the device. The relaxation of the shift (t_d < t) with magnitude Δrelax is modeled as exponential decay, similar to the model used to describe damage from 3 keV He⁺ ions in graphite [39] and is motivated by the linear response to ion damage and relaxation through interstitial-vacancy recombination in the dilute damage limit. Time scales for damage saturation and relaxation in this model are set by t_s and τ_s, respectively.

Fig. 2. Optical and thermal response of microdisk resonator to ion dose. (a) A cross section of the optical mode intensity (color scale) for the 0th radial order mode shown in Fig 1(b) is shown relative to the data in (b) and (c). (b) The ion beam is scanned radially across the edge of the microdisk with a series of ≈ 0.5 ms ion pulses, and the time-varying shift (blue lines) is recorded. Insets show the thermal response to the ion pulses (blue bands) off the disk (i) where no response is observed and on the disk (ii) where the overlap with the optical modes is minimal and only the thermal response of the resonator is observed. Each inset trace (blue points) is an average of 8 consecutive ion pulses with fitting for the thermal shift (dashed line) superposed. (c) The maximum optical shifts (closed circles) and thermal shifts (open circles) are plotted as a function of beam position. Each value is extracted from fitting the time-variant response to Eq. 1 as shown in inset (iii). The optical response is compared to numerical simulation of optical mode intensity at the disk surface (red line), and the thermal response is shown to be constant across the disk surface (blue line). Both responses account for the finite size of the ion beam. Inset (iv) shows the orientation of the ≈ 3 μm scan (red bar) using a FIB secondary electron image of the device. Error bars indicate the standard error of the mean; some are smaller than the data point.
surface swelling and modification of its optical index [31]. The ions
interact with the microdisk through a collisional cascade, damaging the
Si lattice as the ions scatter and slow in the device. The light mass of the
Li+ ion creates a disperse volume of damage with minimal material
sputtering [40]. Numerical simulation of the cascade in SRIM [41]
shows that 3.9 keV Li+ ions penetrate to a depth of ≈ 31 nm with an rms
deviation of ≈ 15 nm. The primary source of damage is due to knock-out
collisions that create ≈ 70 silicon interstitial-vacancy pairs per incident
ion. Surface sputtering is ≈ 0.3 Si per incident Li+, and ≈ 7 % of the ions
backscatter from the device while the rest remain as interstitials in the
crystalline silicon (c-Si) lattice. The lateral rms deviation in ion position
is ≈ 22 nm. Considering a single ion pulse with 3000 ions and a
conservative 100 nm estimation of the beam size, the expected ion flux
is ≈ 5 × 10^{12} cm^{-2} and the vacancy density is ≈ 0.2 % in the center of the
damaged volume.

Ion damage shifts the microdisk resonances due to local expansion of
the structure and modification of its refractive index. The magnitude of the
observed shift in Fig. 2 is ≈ 1.5 pm for ≈ 3000 ions — this is a fractional
shift of the resonant wavelength of ≈ 10 %. The contribution of total shift
due to modification of the refractive index can be estimated from the
amorphization fraction, the known ≈ 10 % index difference between
crystalline and amorphous Si [42], and the fraction of the mode volume
that is damaged. The estimated shift is ≈ 0.15 pm, which is in the same
direction as the signal but constitutes only a tenth of the observed
value [31,43]. Calculation of the contribution from surface swelling can
be done through optical eigenmode perturbation theory [31], and an
expansion of ≈ 0.6 nm would accommodate the observed 1.5 pm shift.
The low-energy-Li+-induced expansion of c-Si is unknown and
can be approximated by expansion in ion species, dose, and
accelerating voltage. Measurements with 80 keV He+ implanted in c-Si
show expansion at the percent level and suggest that surface swelling is
likely the explanation for the presently observed signal [44]. Generally,
the amplitude of the optical response is expected to differ for fields
normal and perpendicular to the disk surface. For the modes explored in
this work, the electric field normal to the microdisk surface is
negligible, and the expected response can be approximated using the
mode intensity. The proximity of the ion damage to the disk surface may
also play a role in the magnitude of this effect as self-interstitial silicon
has been observed to migrate to the surfaces instead of remaining as
lattice interstitials [45].

In addition to the spatially varying optical response, there is also the
rapid thermal shift of the resonance, as shown in Fig. 2 (ii), caused by
ion-beam heating of the microdisk. This response appears independently
of beam location on the microdisk due to rapid thermalization across the structure. The thermalization time constant of the microdisk is estimated as \( \tau = \frac{H}{W} = 300 \, \text{ns} \), where \( D \) is the thermal
diffusivity of c-Si (≈ 0.8 cm²/s) and \( L \) is the diameter of the disk.
Thermalization with the substrate is limited by conduction through the
silicon nitride support structure, and the temperature rise of the disk
can be estimated through the balance of the ion heating (≈ 4 nW) and
thermal conduction of the nitride column \( \Gamma \text{SiN} / \text{Si} / H \), where \( \Gamma \) is the
increase in temperature of the microdisk, \( A \) is the cross-sectional area of the
support (≈ 1.4 μm²), and \( H \) is the height of the support (≈ 2 μm). The
thermal conductivity of the nanofabricated silicon nitride structures \( C_{\text{SiN}} \)
is estimated as ≈ 5 Wm⁻¹K⁻¹ [46]. Using these values, we estimate the
temperature rise of the disk at \( \Delta T \approx 0.5 \, \text{mK} \).

The thermo-optic coefficient of c-Si \( \beta_{\text{Si}} \approx 2 \times 10^{-6} \, \text{K}^{-1} \) at 1550 nm and \( T = 300 \, \text{K} \) [47] predicts an
expected thermal shift of ≈ 0.15 pm given this temperature rise and is
consistent with observed values of ≈ 0.12 pm seen in Fig. 2. The role of
thermal expansion is minimal given the low thermal expansion coefficient
of c-Si at ≈ 2.6 × 10^{-6} K⁻¹, two orders of magnitude smaller than the
effect from the change in index. In general, microdisk resonators are
extraordinarily sensitive thermometers, and the data in Fig. 2 (ii) show
similar capability to state-of-the-art microdisk thermometers [48,49].

![Fig. 3. Two-dimensional imaging of nanophotonic modes. (a) Image of the 0th order radial mode showing the spatially-dependent optical shift. (b) Data from (a) are compared with a 2D model of the mode intensity (inset). The optical shift (closed circles) is compared with the numerically calculated mode intensity (red line) averaged by radial position on the microdisk after fitting. The thermal shift (open circles) shows effects of non-linear driving of the resonance at the mode position. (i) The thermal response away from the optical mode position shows a hysteresis response in which the original temperature is not restored after the ion pulse. This resonance is the same as described in Fig. 2 and was interrogated using 250 μs, 1 pA pulses (≈ 1500 ions). (c) Image of a 1st order radial mode showing the optical response to 300 μs ions on a separate device with \( Q \approx 5000 \) and \( \lambda = 15.38 \, \text{nm} \). (d) The optical response (closed circles) is compared to the numerically calculated mode profile (red line, inset) and the thermal response (open circles) again shows a hysteretic response that is uniform across the disk except at the mode maximum.

Using the understanding of the time varying response of the
microdisk to ion pulses, images of the optical mode intensity were
acquired with high signal to noise ratio. Two-dimensional images of 0th
and 1st order radial TM modes on two separate devices are shown in Fig.
3 to demonstrate this ability. For each of these images, an array of ion
pulses was positioned near the microdisk edge and the two-component
fitting procedure described in Eq. 1 was used to extract the optical and
thermal response from each ion pulse. The images are composed of 864
and 900 ion pulses lasting 250 μs and 300 μs, respectively (total of \( \approx 2 
\text{million ions} \); total image acquisition time is less than 20 s. The
measured optical shifts are compared to numerical simulations of the
mode intensity. The finite beam width and technical noise are again
modelled as a Gaussian blurring of the expected mode profile. The
comparison to the predicted mode shows strong agreement that is most
visible in the azimuthally averaged data shown in Fig. 3 (b, d). Notably,
the agreement indicates that non-linear effects from ion damage are
minimal and the ion dose can be considered in the linear-response
regime. Higher ion dose leading to significant amorphization is known
to have a non-linear scaling and would limit this technique at fluences in
the regime of \( 10^{16} \text{cm}^{-2} \) [50].

The thermal shifts shown in Fig. 3 are observed to be constant across
the disk surface except near the modal maxima. The resonances used
for these images were excited in a quasi-linear regime in which the
optical mode spectrum is skewed by a thermal shift due to the absorbed
optical power, increasing the apparent slope on the low frequency
shoulder of the mode’s absorption line. While this non-linear driving is
useful for increasing sensitivity to small frequency shifts, it induces
hysteresis behavior in the thermal response of the system. This
hysteresis is due to interplay between ion heating and rapid
modification of the resonance absorption and is evident in the
modification of the shape of the thermal response shown in Fig. 3(i) away from a step response and in the shift’s partial recovery after the ion pulse. We model this behavior by fitting to a heuristic function, and the plotted values correspond to the maximum of that function.

Repeated imaging of a disk mode is ultimately limited by broadening of the optical resonances, attributed to increased optical losses due to ion damage. Nevertheless, it is possible to image large areas with minimal device degradation. The imaging in Fig. 3(c) was repeated 6 separate times with an estimated total dose of ≈ 2 pC (≈ 0.6 pC in the mode volume). During this process the mode Q decreased by approximately a factor of 2. The loss in sensitivity can be partially offset through increasing the optical power and thereby decreasing the detection shot noise. A careful measurement of the mode spectrum changes in a doublet resonance during ion imaging is shown in Fig. 4(a). Starting from the pristine resonator with the mode used in Fig 3(c), we measure the spectrum before, in the middle, and after two imaging sequences. The spectrum is observed to globally shift to longer wavelength by ≈ 100 pm per image with ≈ 750 000 ions incident on the portion of the microdisk occupied by the mode. At the same time, the lower frequency peak was broadened by ≈ 75 pm per image. The process of adjusting the interrogation laser frequency to follow the resonance is shown in Fig. 4(b) during one of the imaging sequences. Here the stepwise shifting of the mode follows the rastering of the ion beam across the disk surface, demonstrating a gradual and controlled shift of the mode line by more than a linewidth. We note that associated with this shift is an inevitable, but relatively minor, change in Q. Understanding and limiting this change in Q is the subject of a future study.

**Fig. 4.** Persistent spectroscopic shifts of microdisk modes due to ion imaging. (a) A doublet mode (same as Fig 3c) spectrum is measured interleaved around two separate 2D imaging sequences. The curves correspond to the initial spectrum (black), and the spectrum after the first (blue) and second (red) 2D image. The spectra show that the modes are red-shifted and broadened during imaging, and that coupling to the lower-wavelength mode becomes dramatically reduced. (b) The wavelength of the laser is monitored using an optical wavemeter during the second imaging sequence (shaded area) and shows the stepwise shift of the disk resonance as the pulsed ion beam is rastered across the microdisk surface. The inset shows the resulting map of the optical mode.

Ion imaging can also be used to explore mode doublets. For perfectly circular disks, clockwise (CW) and counter-clockwise (CCW) whispering gallery modes are degenerate due to rotational symmetry of the disk. However, small fabrication imperfections and defects are in some cases sufficient to break the symmetry, leading to doublets in the mode spectrum. These doublets correspond to pairs of standing wave modes with slightly different energies, arising from mixing between CW and CCW propagating waves. The nodal position of the lower- and higher-energy standing waves is dictated by the details of the spatial symmetry breaking disorder. Modifying the symmetry-breaking disorder and/or modifying the photon lifetime (linewidth) can cause the relative coupling of the two modes to change as is shown in Fig. 4(a). In this data, repeated ion dosing dramatically reduces the contrast of the high-frequency mode. Additionally, distinct standing wave modes can be individually measured using this technique as shown in Fig. 5(a) where a TM, 0th radial order mode is imaged. Given sufficient spectral separation between the modes, independent addressing and imaging can be achieved. Here we image the lower frequency mode of a doublet and observe five peaks on the standing wave pattern. The
interferometric visibility of the observed standing wave is $\approx 0.28$. The maximum theoretically expected value for the visibility is $\approx 0.7$ due to the $90^\circ$ phase shift of the azimuthal electric field relative to the radial and normal components of the field. The finite resolution of the ion beam and finite spectral mode overlap shown in Fig 5(d) also contribute to a reduction in the standing wave visibility. Assuming Gaussian blurring with an rms radius of 100 nm, one expects a visibility of $\approx 0.29$ in the mode response, in strong agreement with the data shown in Fig. 5(b). Additionally, the mode order is found to be $\ell = 35$ in agreement with spectroscopy of the disk modes. Further imaging of this microdisk leads to permanently broadening of the mode's spectrum. When the photon lifetime becomes shorter than the CW-CCW scattering rate, the mode splitting and the resulting standing wave are no longer resolved. Imaging of the mode distribution after this broadening in Fig 5(e) shows no visible standing wave and demonstrates the ability to spatially image and permanently edit high quality factor optical modes.

We have imaged the optical and thermal responses of microdisk resonators to pulses of low energy ions from a focused beam. This has enabled us to form images of the device's optical mode intensity in the linear response regime with both higher sensitivity and speed. With higher cumulative dose, we demonstrate persistent editing of mode spectra and spatial patterns. Future extensions of this technique using focused ion beams to locally perturb optical structures at higher spatial and temporal resolution will allow for improved imaging of sub-wavelength structures. Fundamentally, the technique is only limited by the volume of the ion cascade in the material, and this volume can be minimized using high resolution, low energy beams that are currently being developed using MOTIS and similar sources [51]. There is also an opportunity to use high energy ions that will penetrate deeper into nanophotonic devices to allow for mapping of optical structures using He+ and other forms of ion microscopy. Additionally, detailed understanding of the time varying optical response will provide insight into the process of thermal annealing of ion damage, which is of relevance to a broad range of electronic and optical materials.

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† These authors contributed equally to this work

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