Optically-Triggered Nanoscale Memory Effect in a Hybrid Plasmonic-Phase Changing Nanostructure

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ABSTRACT: Nanoscale devices – such as all-optical modulators and electro-optical transducers – can be implemented in heterostructures that integrate plasmonic nanostructures with functional active materials. Here we demonstrate all-optical control of a nanoscale memory effect in such a heterostructure by coupling the localized surface plasmon resonance (LSPR) of gold nanodisk arrays to a phase-changing material (PCM), vanadium dioxide (VO₂). By latching the VO₂ in a distinct correlated metallic state during the insulator-to-metal transition (IMT), while concurrently exciting the hybrid nanostructure with one or more ultraviolet optical pulses, the entire phase space of this correlated state can be accessed optically to modulate the plasmon response. We find that the LSPR modulation depends strongly but linearly on the initial latched state, suggesting that the memory effect encoded in the plasmon resonance wavelength is linked to the strongly correlated electron states of the VO₂. The continuous, linear variation of the electronic and optical properties of these model heterostructures opens the way to multiple design strategies for hybrid devices with novel optoelectronic functionalities, which can be controlled by an applied electric or optical field, strain, injected charge or temperature.
Keywords: surface plasmons; metal nanoparticles; vanadium dioxides; plasmonic memory effect; phase transformation
Metallic nanostructures support localized surface plasmons (LSPs) and surface-plasmon polaritons (SPPs), coherent oscillations of conduction electrons at metal-dielectric interfaces. Surface plasmon excitations occur in ultrasmall mode volumes, generating high local electric fields in passive nanophotonic devices ranging from plasmon waveguides to plasmon lenses. The optical response of plasmonic structures can be controlled by electro-optical, thermal-optical or magneto-optical properties of functional materials in close proximity. All-optical modulation of SPP propagation in metal films and of LSPs in metal nanostructures is also well known. In all these schemes, however, the modulated plasmon response is necessarily transient; the metal nanostructures revert to their initial plasmonic states as soon as the external stimulus is removed. On the other hand, in many applications it would be desirable to retain functionality in a plasmonic device even after the triggering signal disappeared, allowing tunable, persistent modulation of plasmonic properties. Accessing multiple plasmonic states linearly and repeatedly by active control of plasmon modes could enhance functionality in optoelectronic devices and enable on-chip integration of optical and electronic functionalities.

Phase-change materials (PCMs) – already widely used in rewritable optical data storage and non-volatile electronic memory – offer unique possibilities for persistent, reversible tuning of surface plasmons by combining PCMs with metal nanostructures to create new or multiple plasmonic functionalities. A promising PCM for this purpose is vanadium dioxide (VO$_2$) with its strong correlations among orbital, spin and lattice degrees of freedom. A modest external stimulus can switch VO$_2$ through non-congruent electronic (from insulating to metallic) and structural (from M1 monoclinic to R rutile) phase transformations. When VO$_2$ is heated, this insulator-to-metal transition (IMT) occurs near a critical temperature ($T_c$) of 68 ºC in bulk crystals and thin films. The IMT can also be induced mechanically, electrically, optically
and by charge injection,\textsuperscript{27} enabling active modulation of LSPs and SPPs in metallic nanostructures integrated with VO\textsubscript{2}.\textsuperscript{28-31}

Although it is generally understood how plasmons are modulated in hybrid nanostructures switched between the fully insulating and fully metallic states,\textsuperscript{32} the potential for exploiting the entire strongly correlated phase space of a PCM has never been demonstrated \textit{at the nanoscale}. In this paper, we show how the plasmonic state of a model hybrid nanostructure can be controlled by sampling different initial states within the region of strong electron correlations. We demonstrate persistent optical modulation of localized surface plasmon resonance (LSPR) in a model heterostructure comprising Au nanodisks lithographically patterned on a thin VO\textsubscript{2} film. Throughout the temperature window of the IMT, the VO\textsubscript{2} can be latched thermally in the selected strongly correlated metallic state.\textsuperscript{33} Because of the hysteretic response of the first-order VO\textsubscript{2} phase transition, the LSPR can be persistently tuned by successive ultraviolet (UV) light pulses as long the system remains in the latched, initial state. This dynamic is highly repeatable, lasts for several hours and persists as long as the external stimulus is not varied.\textsuperscript{34-35} Hence the entire phase-space of this PCM can be optically addressed when the hybrid nanomaterial is latched at different starting temperatures during the IMT.

More interestingly, the subsequent temporal evolution of the LSPR depends strongly on the initial temperature from which the system is cooled, implying that the LSPR “memorizes” the initial state prepared in the hybridized plasmonic nanostructure and the strongly correlated electron state in the VO\textsubscript{2}. It is crucial to note that here the temperature is simply a proxy for the parameter that describes the initial, latched state of the VO\textsubscript{2} PCM. Known properties of VO\textsubscript{2} suggest, therefore, that the novel resonant memory effect described here should be observable.
regardless of whether the initial state is prepared by optically, electrically or mechanically initializing the hybrid plasmonic-phase-changing heterostructure.

**Mechanism of persistent tuning of a “memplasmon” resonance.**

Figure 1 shows the hybrid plasmonic-phase-change optical material, comprising periodic arrays of Au nanostructures on a thin VO$_2$ film (see Figure 1a), along with a schematic of the mechanism for persistently tuning the LSPR in Au nanostructures interacting with the strongly correlated metallic state of the VO$_2$. When the VO$_2$ layer is initialized in the strong-correlation regime (shaded area, Figure 1b) by setting the temperature, each successive UV-light pulse transforms more crystalline nanodomains of the VO$_2$ into the metallic state without recovering the initial state, manifested by a continuous change in optical transmission. This yields persistent tuning of the resonant plasmon response of Au nanostructures on top of the VO$_2$ film, unlike non-phase-changing, active plasmonic systems with transient resonance modulation (Figure 1c).
Figure 1: Model for persistent tuning of a “memplasmon” resonance. (a) Experimental schematic for the heating cycle of the phase transition: (a) Array of periodic Au nanodisks fabricated lithographically on a thin VO₂ film deposited on a SiO₂ substrate. Optical extinction measurements are conducted while successive UV-light pulses pump the VO₂ film into new states. The device is mounted on a temperature-controlled stage. (b) Hysteretic response in the optical transmission of VO₂, showing the region of strong electron-electron correlations during the heating cycle of the phase transition (shaded gray area). (c) Schematic comparison of plasmon resonance modulation schemes with conventional active media (blue) and the phase-transforming VO₂ (pink) when pumped by successive optical pump pulses (red). The strongly correlated electron state in the VO₂ is initialized and maintained by setting substrate temperature.

Sample fabrication and large modulation of LSPR via phase transformation.

Thin VO₂ films with nominal thickness of 50 nm were fabricated on glass substrates by pulsed laser ablation of a vanadium target in 250 mTorr oxygen. Subsequent annealing of the samples at 450 °C for 40 minutes in 10 mTorr of oxygen rendered the film stoichiometric, crystalline and switching. A complete description of the protocol for characterizing optical response and crystal structure can be found elsewhere.²³ Four different arrays of Au nanodisks were fabricated by electron-beam lithography on the VO₂ film; the nominal diameter D of the Au nanostructures was varied from 80 nm to 210 nm while keeping thickness constant at 25 nm. The Au nanodisks were arranged on a square lattice with a pitch of 500 nm, shorter than the wavelength of interest (above 700 nm) in order to avoid diffraction effects in the transmission measurements. Figure 2a shows typical SEM images of the Au nanostructures, exhibiting reproducibility over a large area.

Extinction spectra of the arrays, defined as (1 - Transmission), were acquired in an inverted optical microscope (Bruker Hyperion 2000) integrated with a Fourier-transform infrared spectrometer.³⁰ Extinction spectra were measured while a Peltier thermoelectric element mounted on the sample stage cycled the VO₂ film through its phase transition by heating and cooling.
through a set temperature program. The external temperature was maintained within ±0.1 °C over the entire measurement cycle in all experiments. Figure 2b shows the extinction spectra of three Au nanostructure arrays acquired below (T = 25 °C, black curves) and above (T = 75 °C, red curves) the insulator-to-metal transition of VO₂ and reveals that the LSPR of Au nanodisks experiences a substantial blue-shift due to the IMT.

The shift in LSPR for each array is plotted as a function of nanostructure diameter in Figure 2c, and shows that the larger Au nanostructures also exhibit larger resonance shifts because the dielectric contrast in VO₂ during its phase transformation increases with wavelength, further shifting the LSPR. In addition, the evanescent near-fields associated with the larger Au nanodisks sample a larger interaction volume in the phase-transforming VO₂ film. The ensemble of Au nanodisks is distributed over a VO₂ film having a broad range of grain sizes, each of which switches at a statistically determined temperature. Hence the average LSPR tuning should be the same for each nanostructure array and is expected to be a function of the effective or mean-field fraction of metallic phase in the underlying polycrystalline VO₂ layer.
**Figure 2: Modulation of localized surface plasmon resonances.** (a) Plan-view SEM image of Au nanostructures patterned on a 50-nm thick VO$_2$ film. Inset: Nanostructure array at higher magnification. Scale bars are 1 μm in each case. (b) Measured extinction spectra of Au nanostructures for three diameters $D$ at room- (black) and high-temperature (red). (c) Plasmon resonance peak shift due to the phase transformation of VO$_2$ film as a function of nanostructure diameter (black squares), with linear fit (black line).

**Hysteretic variation of localized surface plasmon resonance.**

We next investigate the changes in the LSPR in Au nanostructures during the phase transformation of VO$_2$ by measuring extinction spectra for the array of 130-nm diameter Au nanodisks while reversibly cycling the VO$_2$ film across the strong-correlation regime (30°C-80°C) with a resolution as small as 0.5°C. The measured extinction spectra in Figure 3a for the hybrid system show smooth transitions in both resonance wavelength and LSPR intensity. By fitting the extinction spectra, we can map the hysteresis in resonance wavelength of the Au LSPR as a function of substrate temperature (Figure 3b). The hystereses of the Au LSPR and the far-
field transmission of a bare VO$_2$ film (50 nm thick, Figure 3d) exhibit similar widths. The close similarity between the two hysteresis curves indicates that the evolution of the Au LSPR tracks the phase-transition properties of the underlying VO$_2$ film and can properly be used as an optical marker for the change in local optical properties in the phase-changing material. The slight reduction in thermal hysteresis width $\Delta T$ for the LSPR compared to that for the bare VO$_2$ film can be attributed to the presence of additional nucleation sites at the Au/VO$_2$ interface, which facilitate the phase-transformation of VO$_2$ by reducing the phase-transition barrier energy.$^{38}$

To further correlate the LSPR modulation with the phase changing dynamics of VO$_2$, Figures 3c and 3e show respectively the plasmon resonance wavelength of the Au nanodisks and the transmission intensity of the bare VO$_2$ film as a function of the VO$_2$ metallic fraction that is determined from the hysteretic parameters shown in Figures 3b and 3d by using the method described in Ref. 48. The results demonstrate a linear relationship between both plasmon resonance wavelength and transmission intensity, and the metallic fraction of VO$_2$, thereby indicating a continuous, linear variation of the electronic and optical properties of both systems.
Figure 3: Hysteretic response of localized surface plasmon resonance. (a) Temperature-dependent extinction spectra for Au nanostructures of 130-nm diameter on VO$_2$ through the heating cycle with a temperature increment of 0.5 °C around the phase-transition temperature. (b) Plasmon resonance hysteresis curve for the same structure extracted from (a). (d) Ensemble transmission hysteresis curve of a 50-nm VO$_2$ film. Here the sample was illuminated by a tungsten lamp and the integrated, unpolarized transmission intensity was recorded as a function of temperature using an InGaAs photodetector. In both (b) and (d), the black squares and red circles are the measured results for heating and cooling cycles, respectively, while the black and red solid lines are sigmoidal functions fits to the measured data. The green double-headed arrows define the thermal hysteresis width for each system. (c) and (e) Plasmon resonance wavelength from (b) and transmission intensity from (d) as a function of the VO$_2$ effective metallic fraction, respectively.

Persistent optical tuning of LSPR using successive UV-light pulses.
Apart from the external temperature, UV-light pulses can be used to trigger the insulator-to-metal transition from the prepared initial state because VO$_2$ has strong absorption in the UV range, as indicated by the large imaginary component of its dielectric function. Successive UV-illumination pulses also increase the areal density of the metallic phase in VO$_2$, gradually changing the average dielectric response of the film and consequently altering the persistent tuning characteristic of the LSPR. After a sufficiently long illumination time, the metallic nanopuddles connect percolatively and leave the VO$_2$ film in a pure metallic phase.

The hysteresis during the VO$_2$ phase transition allows for persistent tuning of the Au nanostructure LSPR when the external temperature is biased within the temperature range where strong electron-electron correlation exists. To demonstrate this effect, we prepared the VO$_2$ film in a strongly correlated metal state by maintaining the array and its substrate at a sequence of constant temperatures. The system was then illuminated by successive UV-light pulses either with a constant power density and varying pulse durations or with different power densities at constant pulse duration (varying fluence). After each such pulse sequence, we measured the extinction spectrum of the Au nanodisk array. Control experiments were also performed at two temperatures well below and above the critical temperature of VO$_2$, and the corresponding extinction spectra were recorded for comparison with those collected at temperatures within the strongly correlated range. To verify that the plasmon resonance shift observed in Au nanodisks was due to the altered dielectric response of VO$_2$, spectroscopic ellipsometry was used to characterize a bare VO$_2$ film of the same thickness (50 nm) and the change in dielectric response was measured using the same UV-illumination protocol used for the hybrid plasmonic system. The dielectric function of VO$_2$ was extracted by fitting the ellipsometric data with a sum of one Gaussian and one Lorentz oscillator under a global-minimization algorithm.
Figure 4a shows how the plasmon resonance is modulated by UV illumination for different pulse durations at constant power density (90 mW/cm²) while keeping the hybrid system at 64 °C and 68 °C, respectively. The LSPR wavelength extracted from each extinction spectrum as a function of total illumination time exhibits a substantial blue-shift after the initial few pulses, followed by a gradual saturation in the resonance wavelength for much longer pulse durations. Such behavior contrasts sharply with results from the control experiments performed at 27 °C (not shown) and 77 °C, in which the LSPR wavelength shows no sensitivity whatsoever to the UV illumination. Figure 4b shows similar results when the LSPR wavelength is modulated by UV-light pulses of constant duration (10 sec) but with various power densities: A large blue-shift and subsequent resonance saturation also occur when gradually increasing the power density of single UV-light pulses. The inset in Figure 4b plots the dispersive refractive index of the bare VO₂ film extracted from spectroscopic ellipsometry as a function of the UV-illumination power density at a constant temperature of 65 °C, which reveals a gradual decrease in the refractive index of VO₂ in the wavelength range of interest and accounts for the observed blue-shift in the LSPR of Au nanostructures.

In analogy to the general definition of modulation depth for silicon photonic modulators,⁴⁰ here we define the extinction ratio 10log(I₀/Iₚₜ) as the modulation depth for the Au/VO₂ hybrid heterostructure, where I₀ and Iₚₜ are the extinction intensities at a specific plasmonic wavelength for the system thermally latched within the phase transition region of VO₂ before and after a certain period of UV illumination, respectively. The inset of Figure 4a shows the measured extinction spectra for the Au nanodisks of 175-nm diameter on the VO₂ film before and after 120-sec, 90-mW/cm² UV illumination under the bias temperature of 64 °C, from which the modulation depth at the plasmonic wavelength 1029 nm is calculated to be 1.79 dB. Note that
this value might be underestimated because in the extinction calculation the transmission of each Au nanodisk array was normalized with respect to the transmitted light through a bare VO₂ area in close proximity to the array. This implies that the extinction contribution from the VO₂ itself has not been taken into account in the calculation of modulation depth.

**Figure 4: Persistent optical tuning of LSPR by UV-light pulses.** (a) Plasmon resonance wavelength of the Au nanodisks of 175-nm diameter on the VO₂ film as a function of the total UV illumination time with a constant power density of 90 mW/cm² while the whole sample was thermally latched at two different temperatures within the phase transition region, 64 °C (black squares) and 68 °C (red circles), and also above (77 °C, red empty circles). Each of the first three pulses has a duration of 10 sec, followed by another two pulses of 15 sec, respectively, and the last one of 45 sec. Inset shows the measured extinction spectra for the sample latched at 64 °C before (black) and after 120-sec UV illumination (red), and the modulation depth (i.e. extinction ratio) at a wavelength of 1029 nm is calculated to be 1.79 dB. (b) Resonance shift for the same structure as a function of the power density of each UV-light pulse of 10 sec duration. Inset shows the dispersive refractive index for a 50 nm bare VO₂ film illuminated at the same conditions as above.

**Evolution traces of LSPR registering the initial temperature states.**
In a temperature-controlled memristive device driven by the VO$_2$ phase transition, the resistance takes different paths for cooling and heating cycles; in particular, the time evolution of the resistance depends strongly on the bias temperature from which the cooling cycle starts.$^{41}$ This means that the initial temperature states or the associated information stored in the system can be retrieved by monitoring the evolving path of the system resistance. More generally, in a memristive metal/oxide/metal nanodevice, this corresponds to the hysteretic response of I-V curves in which the applied voltage and resultant current follow a highly nonlinear relationship and the evolutionary history of the current-voltage relationship depends on the initial bias voltage.$^{42,43}$

The optical-frequency, plasmon memory resonance described in this experiment exhibits features analogous to such memristive behavior. Figure 5a plots LSPR wavelength as a function of temperature during the cooling cycle starting from different bias temperatures. Each such evolutionary path is unique, yielding a characteristic parameter pair – thermal hysteresis width and resonance shift – for each bias temperature (Figure 5b). The unique evolution of the LSPR can also be observed by plotting the plasmon resonance wavelengths (extracted from Figure 5a for each different initial temperature and heating cycle) as a function of the effective metallic fraction of VO$_2$ (Figure 5c). The strong similarity between a memristive response$^{41-43}$ and the behavior of the hybrid plasmonic system further supports the idea that an optical-frequency plasmon resonance of the metallic nanostructure can store and read out the “memory” of the initial hybridized state in a broad range of phase-transforming materials like VO$_2$. 

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Figure 5: Temperature evolution of LSPR retracing the initial temperature states. (a) LSPR wavelength evolutions for 175-nm-diameter Au nanodisks on VO$_2$ through cooling cycles starting from different initial temperatures $T_0$ (red circles to dark-blue left-triangles). The black squares are the resonance positions through the heating cycle. The quantities $\Delta T$ and $\Delta \lambda$ represent the thermal hysteresis width and the resonance shift amplitude for each initial state of the film, with results shown in (b) as a function of initial bias temperature $T_0$. Solid lines in (a) are sigmoidal fits to the measured data points while black and blue solid lines in (b) are guides to the eye. The plasmon resonance wavelength extracted from (a) for different initial temperatures and heating cycles are re-plotted in (c) as a function of the effective metallic fraction of the VO$_2$.

Discussion

The hysteretic nature of the first-order phase transition in VO$_2$ is the key to realizing the persistent plasmon resonance tuning in practice. In this experimental configuration a bias temperature somewhere in the transition region from insulator to metal, that is, between 60°C and 80°C (Figure 4), initializes the VO$_2$ in a strongly correlated state. The UV-modulation experiments reveal that the LSPR of Au nanostructures is “pinned” at a wavelength defined by the initial state of the VO$_2$ film and the duration of illumination. Successive UV-light pulses drive the LSPR to a new wavelength as more VO$_2$ domains are switched from insulating to
metallic. The hysteretic response of this hybridized heterostructure after the triggering impulse fades away (as demonstrated in Figure 5) is then essential for the long-lasting memory of the UV-light induced change which is “imprinted” into the LSPR frequency.

As noted in the discussion of Figure 5, this LSPR memory effect in metal nanostructures resembles in some ways the memory capacitance and memristive responses of VO₂ in memory metamaterials⁴⁴ and phase-transition-based memory devices.⁴¹ However, the plasmon memory resonance observed here at optical to near-infrared frequencies is based on the hybrid response of the Au nanodisks and the hysteretic response of VO₂. This is distinctly different from the metamaterial memristive system⁴⁴ that exploits the hysteresis in VO₂ dielectric constants at terahertz or microwave frequencies. Finally, it should be noted that although UV pulses of a few tens of seconds were used in this study, the phase transition dynamics of VO₂ is not intrinsically restricted to slow timescales. The absorbed UV light in the present experiment causes both transient local heating and electronic excitation, leading to the formation of new metallic nanopuddles in the insulating VO₂ host and driving the film toward completed formation of the metallic, rutile phase. If the UV energy density is high enough to cause the transition to occur by local heating, the reversion from the rutile metallic state to the insulating state could require nanoseconds to microseconds. If, on the other hand, the UV light pulse is ultrashort²⁶ and the transition proceeds primarily through electronic excitation to the recently observed metallic monoclinic state,⁴⁵-⁴⁷ the complete transition cycle could then occur on a picosecond time scale without a structural phase transition, making it possible to consider practical applications. Another potential route to an ultrafast device using the hybrid material system demonstrated here would be by electron injection initiated by resonant excitation of a noble-metal nanoparticle.²⁷
The plasmon memory resonance effect can be generalized in a number of ways. Because the critical temperature and the slope of the hysteresis curve for a VO$_2$ film can be substantially altered by impurity doping, electrical bias, charge injection or substrate-induced strain, even for the ultrafast laser-driven phase transition, a plasmon memory device can in principle be operated at lower temperatures, and over a broader temperature range, than what is demonstrated here. Conversely, since the initial strongly correlated metal state can be prepared by applied bias voltage, charge injection, strain or laser illumination, a plasmon memory device can be designed to operate at a constant temperature. The thermal hysteresis width of the IMT in VO$_2$ can be altered by changing fabrication parameters to yield differing grain sizes in a film, ordered arrays of nanostructures or strained one-dimensional nanostructures. Flexible control over both the transition temperature and hysteresis width of the VO$_2$ also allows for stable modulation of the plasmon resonance wavelength.

Other nanostructure configurations can easily be imagined: For example, one could imagine a double-layer lithography procedure in which one would remove all the VO$_2$ outside the range of the plasmon near-field, leaving a double-layer Au/VO$_2$ heterostructure array with a defined pitch. Finally, since the phase transformation in VO$_2$ is associated with strong electron-electron correlations, the plasmon memory resonance offers a unique opportunity to monitor the interaction between the collective oscillation of free electrons in the surface plasmons and strongly correlated electrons during the interband transitions in VO$_2$.

Although these experiments employed Au nanostructure arrays, the behavior observed here should also occur in individual metal nanostructures, and be observable using single-particle spectroscopy to monitor the plasmon resonance shift. In fact, single-particle studies could explore the metal-to-insulator phase-transition on the nanoscale by monitoring electromagnetic
near fields associated with localized surface plasmons. For example, an individual metal nanostructure used as a nanoprobe could detect coexisting insulating and metallic phases in VO$_2$ and the divergence of the real dielectric function near the onset of the insulator-to-metal transition.

**Conclusion**

We have demonstrated a novel plasmon memory resonance by hybridizing the surface plasmon resonance in metallic nanostructures with the strong electron-electron correlation effects in a phase-changing VO$_2$ thin film. Analogous to the memristive response of VO$_2$, this plasmon memory resonance exhibits persistent tuning of the resonance frequency when illuminated by ultraviolet light, in contrast to plasmon modulation schemes using conventional active media. The temperature and temporal evolution of the observed memplasmon resonance depends critically on the correlated initial state of the VO$_2$, and thus enables reversible operation of the device to re-initialize the hybrid plasmonic memory material.

This memory effect on the resonance frequency of a plasmonic nanostructure due to the strong electron-electron correlations in VO$_2$ exhibits an optical analog of memory circuit devices, the plasmon memory resonance, but now in the optical to near-infrared range rather than the microwave and THz range.$^{41-43}$ Nanoscale integration of plasmonic structures with phase-changing materials such as VO$_2$ could enable further miniaturization of an entire class of memory devices for next-generation nanophotonic circuitry – simply because such devices would work at shorter wavelengths than THz or microwave-based devices. The dual-excitation scheme presented in this work could potentially be extended to other hybrid plasmonic/SECM systems, incorporating the controlled metal-to-insulator phase transition in NdNiO$_3$, colossal magnetoresistance in perovskite materials like LaMnO$_3$ and Pr$_{0.7}$Ca$_{0.3}$MnO$_3$.55
Combined with novel nanostructure architectures and excitation schemes, this proof-of-concept demonstration has a significant impact on the possibilities for transferring the basic concepts in conventional circuits — such as memristive behaviour and memory capacitance — to nanoscale optical devices operating in the visible and near-infrared regimes.

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References


