

Light scattering by V4O7 film across the metal-insulator transition

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ABSTRACT

The experimental study of the angle-resolved hemispherical light scattering by V_4O_7 film within a broad temperature range across metal-insulator transition reveals complex structural reorganization of the film deposited on the c-cut sapphire crystal. The bidirectional scattering distribution function and the surface autocorrelation function were obtained from scattering data to visualize statistics of the spatially resolved contributions of optical inhomogeneities in normal and lateral directions to the surface. The measurements reveal an anisotropic surface roughness distribution due to the twinned domain structure, with significant anisotropy changes across the phase transition. The V_4O_7 film deposited on sapphire leads to a polydomain structure, minimizing elastic strain energy with distinct multiscale distributions of surface domains. Near T_c , the material shows the lowest roughness but the highest lateral disorder of the surface.

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INTRODUCTION

Optical scattering is one of the main processes accompanying the solid-to-solid transition in phase-change materials. Angle-resolved elastic light scattering is a powerful technique for the statistical analysis of surface irregularities of various sizes that should always be considered for the design and optimization of neuromorphic and optical devices based on complex phase-change materials such as V_4O_7 . For example, the light scattering within a plasmon resonance spectral range in vanadium oxide-based materials is a key factor for optimizing nonlinear optical media for various technological applications.^{1–3}

V_4O_7 demonstrates reversible *weakly* first-order metal-insulator transition (MIT) at a temperature point T_c of about 235–250 K,^{4–9} and the transition extends within a relatively broad temperature range of tens of Kelvins. V_4O_7 belongs to vanadium oxide Magnéli phases (V_nO_{2n-1} , $n = 3, 4, \dots, 9$)^{10,11} and shows MIT similar to other vanadium oxides, but with substantially different electronic, magnetic, optical, and thermal properties. Like other oxides, electron-electron correlations in V_4O_7 contribute to the exotic properties of this material, especially when the material undergoes a phase transformation.

The metrological precision of elastic light-scattering techniques can provide a deep understanding of structural dynamics across MIT, as it was already shown for VO_2 , V_3O_5 , and V_2O_3 oxides with different morphologies and concentrations of structural defects in quasi-equilibrium steady-state and ultrafast light-induced nonequilibrium regimes.^{12–18} The analysis of optical scattering across MIT in epitaxial VO_2 films showed that the internal elastic strain and structural defects influence thermal hysteresis and domain formation, providing crucial insights into the strain-dependent dynamics of the surface morphology and optical properties upon MIT. The light-scattering anomaly observed during the MIT in vanadium dioxide thin crystals was assigned to “transition opalescence,” caused by the coexistence of two phases with different optical properties.^{12,13} Optical scattering can contribute to noticeable losses during the phase transition of vanadium oxides, attributed to the formation of metallic domains.¹⁴ Here, we also note that the information regarding the scattering processes of V_4O_7 is very limited.

In this work, we apply angle-resolved hemispherical light scattering for the statistical analysis of surface irregularities of the V_4O_7 film across MIT. This method provides reciprocal-space imaging of

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surface roughness, offering robust statistical information about the distribution of optical inhomogeneities. Light-scattering measurements performed by a scatterometer equipped with a cryogenic system revealed specific changes in surface roughness statistics and its anisotropy across different spatial scales. Structural ordering of microcrystals on the mesoscale, such as self-organization, twinning, and polydomain formation, results in sharp diffraction peaks with 60° azimuthal symmetry. The evolution of the V_4O_7 surface morphology shows substantial differences compared to other vanadium oxides with first-order phase transition, where the V_4O_7 optical scattering decreases at T_c , indicating a transition without distinct phase boundaries.

EXPERIMENT

Thin films of V_4O_7 were deposited on Al_2O_3 (001) substrates via pulsed direct-current (DC) magnetron sputtering of a high-purity vanadium target (99.95%) in a reactive Ar/O_2 ambient. The background pressure was in the 10^{-7} Torr range, while the sputtering pressure was controlled at 10 mTorr using a combination of Ar/O_2 flow rates of 195/5.0 standard cubic centimeters per minute. The substrate temperature was maintained at $600^\circ C$ during deposition. The distance between the 2-in. diameter target and the substrate was 12 cm, and the pulsed DC sputtering power was set to 130 W.

The film stoichiometry and orientation of the V_4O_7 structure on the Al_2O_3 (C) substrate were verified by x-ray diffraction (XRD) using a Bruker D8 Discover diffractometer. In these measurements, x rays illuminated the $1 \times 5 \text{ mm}^2$ area of the sample, collecting data using θ - 2θ XRD geometry. The surface topography was probed by an atomic force microscope (AFM, Park Scientific Instruments, Autoprobe CP).

The light-scattering measurements were performed using a lab-built scatterometer equipped with a closed-cycle cryogenic system to control sample temperature. The main experimental geometry of the angle-resolved hemispherical elastic light scattering (ARHELS) used in this study is shown in Fig. 1. The scattering

apparatus allowed high-resolution angular measurements of far-field elastic light scattering within a full hemisphere. The sample was mounted at the cryofinger at the first focal point of the elliptical mirror that projected scattered light at the CCD (Charge-Coupled Device) detector. The cryogenic system controlled the sample temperature within the range of 164–290 K. A continuous wave semiconductor laser source with a wavelength of $\lambda = 450 \text{ nm}$, linear polarization, and an output power of $\sim 2 \text{ mW}$ was used in ARHELS measurements. The scattering indicatrix patterns were recorded by a 16-bit CCD camera with a 4096×4096 imaging pixel matrix (SBIG STX-16803) and recalculated to the bidirectional-scatter-distribution function,¹⁹

$$\text{BSDF}(\theta, \phi) = \left[\frac{dI_{\text{scatt}}(\theta, \phi)}{d\Omega} \right] \frac{1}{I_0 \cos \theta}, \quad (1)$$

where dI_{scatt} is the light scattered intensity within solid angle $d\Omega$, I_0 is the incident light intensity, and θ and ϕ are the polar and azimuthal scattering angles, respectively. This function was used to monitor the statistical contribution to the scattering field from surface inhomogeneities of different spatial frequencies f . The surface roughness component with a certain spatial frequency scatters light at a specific polar angle, where the relation between f , θ , and the lateral size d of surface irregularity is given by $f = 1/d = \sin \theta / \lambda$. Using a laser line of $\lambda = 450 \text{ nm}$ allows us to detect surface structures with spatial frequencies up to $2.2 \mu\text{m}^{-1}$.

Scattering data were used to calculate surface autocorrelation function ACF.²⁰ The procedure employs the Gerchberg–Saxton error-reduction (ER) combined with Fienup hybrid input–output (HIO) phase-retrieval algorithm.^{21–23} These algorithms are widely used in the fields of electron microscopy, crystallography, astronomical imaging, and x-ray diffraction imaging to reconstruct experimental data in the spatial domain (real space) from the frequency domain (reciprocal space). The algorithms involve looped Fourier transforms along with appropriate constraints in each domain. The HIO algorithm improves upon the ER algorithm through the modification of constraints. These constraints ensure that the algorithm can escape local minima, improving the accuracy of phase retrieval. In the context of our study, the ER and HIO algorithms are used to convert experimentally measured BSDF in reciprocal space into ACF in real space. The calculations incorporate direct (FFT) and inverse (IFFT) fast Fourier transform, using the parallel computing platform CUDA (Compute Unified Device Architecture Unit) for graphics processing unit (GPU, NVIDIA Tesla K80).²⁴

The ARHELS measurements were performed for 260-nm-thick and 40-nm-thick films. Both films showed nearly the same evolution of the light scattering across MIT. Therefore, here, we present data only for the 260-nm-thick film V_4O_7/Al_2O_3 (C) since the light scattering by the thicker film contains fewer artifacts related to the scattering at the film–substrate interface.

RESULTS AND DISCUSSION

The XRD measurements (Fig. 2) show that the deposition of vanadium oxide on c-cut sapphire crystal formed a single-phase stoichiometric V_4O_7 film with $(1\bar{1}\bar{1})_{V_4O_7}$ plane-parallel to the substrate. The electrical conductivity σ of the film (the inset in Fig. 2)

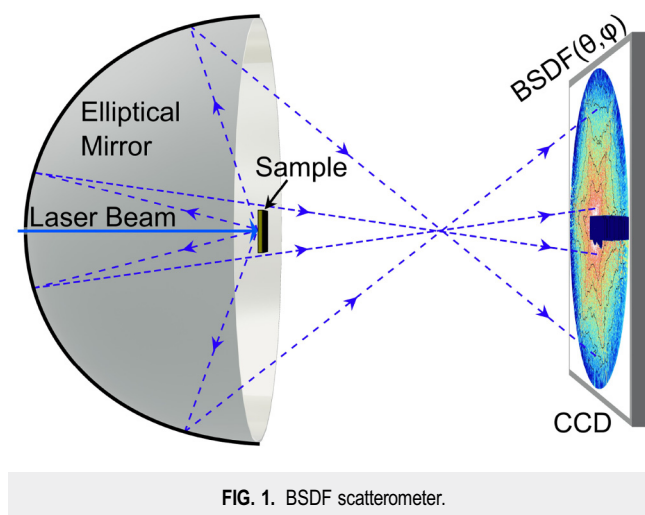


FIG. 1. BSDF scatterometer.

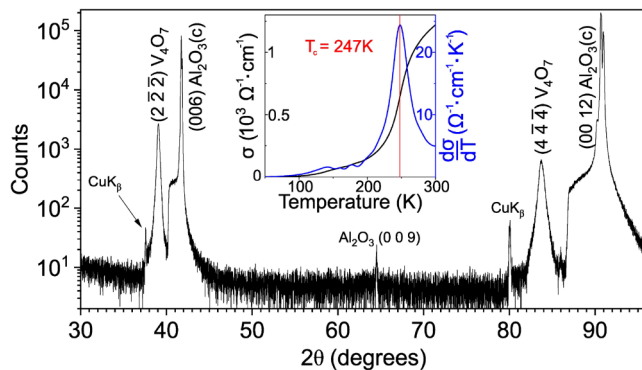


FIG. 2. XRD scan for the V₄O₇ thin film grown on Al₂O₃ (001). The inset shows the conductivity and its derivative as a function of temperature.

demonstrates a three-orders of magnitude change across MIT within a relatively broad temperature range: from 100 to 300 K. The derivative $d\sigma/dT$ defines the transition point $T_c = 247$ K. Thus, it is assumed that the low-temperature insulating phase of V₄O₇ belongs to the temperature range below ~ 200 K, while the high-temperature metallic phase is at room temperature.

Hemispherical light-scattering indicatrices of V₄O₇ were measured across the MIT region from 290 down to 164 K [Fig. 3(a), multimedia available online]. To obtain a spatially resolved contribution to the scattering signal from optical inhomogeneities, the BSDF (θ, ϕ) was recalculated vs surface spatial frequency [Fig. 3(b)]. For an optically smooth surface, the BSDF can be considered as a scaled function of surface power spectral density (PSD), a function that represents the surface roughness power per unit spatial frequency.¹⁵ This approximation is based on the Rayleigh smooth-surface criterion, where the film with *rms* roughness $\delta < \lambda/(4\pi)$ can be considered optically smooth.¹⁹ The AFM data yield $\delta = 5$ nm, which allows applying this criterion for the V₄O₇ surface. The rigorous computation of PSD from BSDF is a nontrivial task and requires material optical constants, that are unknown within the MIT region. Therefore, taking into account the proximity between BSDF and PSD, the quantitative surface analysis was performed using BSDF.

While PSD and BSDF provide statistical information in the spatial frequency domain, surface statistics in the real space are characterized by the surface autocorrelation function ACF(*r*). This function describes the correlation of surface irregularities concerning the translation length *r* in the surface plane (i.e., how the surface inhomogeneity at one point is related to the inhomogeneity at another point separated by a certain distance). The ACF(*r*) and PSD(*f*) are related through the Wiener-Khinchin theorem as a Fourier transform pair. Using the approximation of an optically smooth surface, the ACF(*r*) and BSDF(*f*) data can also be considered as the IFFT and FFT data set, respectively.¹⁵ The ER-HIO algorithm developed for the light-scattering metrology²⁰ provides significantly improved accuracy of ACF(*r*) computation [Figs. 3(c) and 3(d), multimedia available online], along with the reconstruction of the absent BSDF(*f*) data within the sample holder shadow. In this study, we employ normalized autocorrelation functions, ACF(*r*)/ACF(0).

The AFM topography shows that the V₄O₇/Al₂O₃(C) film consists of elongated sub-micrometer grains [Fig. 4(a)]. The ordering of the grains is traceable in the AFM image, and the ACF(*r*) calculated directly from AFM data by integrating the surface profile [Fig. 4(b)] shows a hexagonal pattern. This reveals 120° anisotropy of grain orientation within ~ 130 nm ACF length. However, at larger spatial scales, this ACF(*r*) does not show specific anisotropy owing to a lack of sufficient statistical information within the AFM-scanned area. In this regard, the light scattering measurements provide missing statistics of the surface at the mesoscale. The directional dependence of the light scattering in the BSDF indicatrices for High-*T* V₄O₇ [290 K in Figs. 3(a) and 3(b)] and preferable directions in corresponding ACF(*r*) [290 K in Figs. 3(c) and 3(d)] show 60° azimuthal anisotropy attributed to the anisotropic distribution of surface roughness owing to a twinned domain structure within, at least, several micrometer scale.

Taking into account XRD data (Fig. 2), the AFM and ARHELS measurements suggest that the V₄O₇/Al₂O₃(C) film experiences a domain formation with specific twinning that can be associated with the epitaxial nature of the film. This morphology can be understood in the context of minimizing elastic interaction energy between the film and the sapphire substrate via the formation of a polydomain structure. The system reduces the overall elastic strain energy compared to a monodomain structure through changes in the domain composition and orientation. The 120° anisotropy of polydomain morphology contributes to 60° azimuthal anisotropy of BSDF and ACF(*r*) for high-*T* V₄O₇. Increased light scattering along $\phi = 50^\circ, 110^\circ, 170^\circ, 230^\circ, 290^\circ$, and 350° at $T > T_c$ [290 K in Figs. 3(a) and 3(b)] corresponds to domain ordering along orthogonal azimuthal directions. As a consequence, this ordering contributes to increased ACF(*r*) values along $\phi = 20^\circ, 80^\circ, 140^\circ, 200^\circ, 260^\circ$, and 320° [290 K in Figs. 3(c) and 3(d)].

The origin of observed light-scattering anisotropy is the anisotropy of surface roughness at the nanoscale. Groups of V₄O₇ nanocrystals in the epitaxial film are organized in polysynthetic domains, building a typical “supercrystal”²⁵ with roughness anisotropy on mesoscale accessible by ARHELS. The structural mobility of nanocrystals in such a “supercrystal” is much higher, as compared to a single crystal. Therefore, relatively small alterations in the elastic energy, especially near triclinic-to-triclinic transition point T_c , can effectively change the domain pattern of the film. The phase separation during MIT produces an additional strain field, which is highly anisotropic for the epitaxial film. Despite the strain, the film remains continuous due to phase coherency. This coherency is provided by suppression of the strain field and net elastic energy via twinning of nanocrystals and reorganization of domain pattern.^{25,26} Beyond T_c , a temperature-dependent elastic deformation of the film can introduce further domain formation.

The PSD(*f*) function of the surface, calculated from AFM data, shows good match with BSDF(*f*) obtained at room temperature $T = 290$ K [Fig. 4(c)], supporting the approximation to consider BSDF(*f*) as a scaled PSD(*f*) function. While the *rms* roughness of V₄O₇ is $\delta = 5$ nm, the surface cross section shows the modulation of the surface height of about ~ 20 nm [the inset in Fig. 4(c)]. As the temperature drops to T_c , the BSDF(*f*) level diminishes, indicating a decrease of surface roughness. Moreover, the azimuthal anisotropy of BSDF(*f*) and ACF(*r*) significantly decreases

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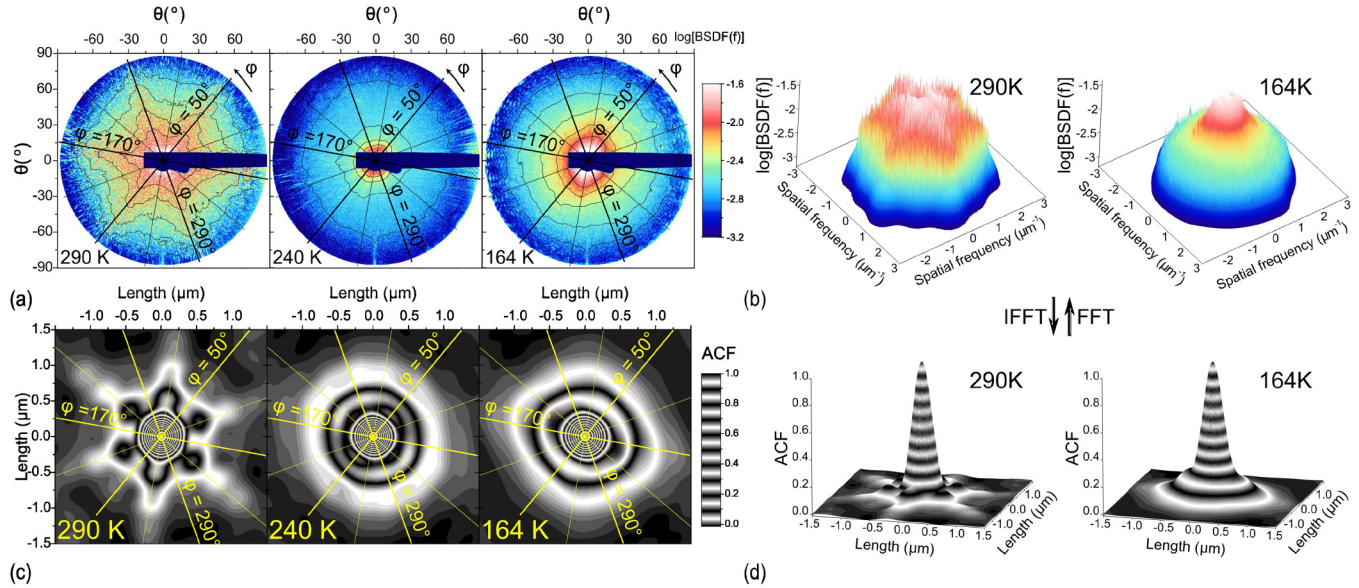


FIG. 3. (a) Evolution of BSDF(θ, ϕ) scattering indicatrix for $\text{V}_4\text{O}_7/\text{Al}_2\text{O}_3(\text{C})$ vs temperature. The full set of BSDF(θ, ϕ) indicatrices is shown in Fig. S1 in the [supplementary material](#). (b) 3D-plots of the reconstructed BSDF(f) indicatrix for the high- T and the low- T phase of V_4O_7 . (c) ACF(r) computed from the scattering indicatrix. The full set of ACF(r) data is shown in Fig. S2 in the [supplementary material](#). The repeating gray scale pattern is used to highlight small modulations of spatial anisotropy. (d) 3D plots of the ACF(r). IFFT and FFT are inverse and direct Fourier transforms, respectively, applied for the ACF-BSDF data computation in the ER-HIO algorithm. Multimedia available online.

[240 K in Figs. 3(a) and 3(c)] due to the reconstruction of the surface near T_c , where the *rms* roughness becomes minimal. As the temperature drops below T_c and the MIT completes, the scattering level and anisotropy of BSDF(f) and ACF(r) increase again. It was found that the level of BSDF(f) for low- T V_4O_7 at $T = 164$ K approaches similar values as it was in the High- T phase at $T = 290$ K. This indicates that the surface *rms* roughness in low- T and high- T phases is nearly the same.

The BSDF(f) and PSD(f) of surface roughness [Fig. 4(c)] reveal several regions with power-law dependence indicating multi-scale distributions of the surface domains. The mapping of these functions in a log-log graph vs spatial frequency f falls into several distinct linear regions: $3.3\text{--}10\mu\text{m}^{-1}$ and above [Fig. 4(c)], and $0.3\text{--}0.7\mu\text{m}^{-1}$, $0.7\text{--}1.8\mu\text{m}^{-1}$, and $1.8\text{--}2.2\mu\text{m}^{-1}$ [Fig. 5(a)]. These regions correspond to spatial frequencies with different statistics of surface roughness. Because of the 120° -fold twinning of the $\text{V}_4\text{O}_7/\text{Al}_2\text{O}_3(\text{C})$ film, the surface is not considered isotropically rough, and, therefore, should be treated rather in terms of a 1D surface. The attempt to analyze the light scattering in terms of a 2D isotropically rough surface resulted in a noticeable error. Therefore, BSDF(f) and also the power spectrum were approximated by the inverse power law of a 1D surface,

$$\text{BSDF}(f) = \frac{\tilde{K}_n}{f^n}, \quad (2)$$

where $\tilde{K}_n$ and n are fractal parameters.^{27,28}

The parameter n defines the tilt of the function and the surface can be considered in terms of the fractal approach when $1 \leq n \leq 3$. The higher fractal parameter n corresponds to a smoother surface, with the *marginal fractal* surface at $n = 3$, while at $n = 1$, close-packed grains of a rugged surface are associated with *extreme fractal*.²⁹ Temperature dependence of n values was derived from BSDF data for three regions of spatial frequencies and summarized in Fig. 5(b).

The evolution of BSDF(f) across MIT along two representative directions with higher ($\phi = 110^\circ$) and lower ($\phi = 80^\circ$) scattering levels [Fig. 5(a)] shows quite different trends for three different regions of spatial frequencies associated with different statistics of surface roughness. At the same time, for each specific region, the trends are similar for different azimuthal directions at $f < 1.8\mu\text{m}^{-1}$ indicating similar surface reconstruction. Moreover, positions of majority diffraction peaks do not noticeably shift with temperature, evidencing that the domain pattern remains nearly constant across MIT at $f < 1.8\mu\text{m}^{-1}$. However, at $f > 1.8\mu\text{m}^{-1}$, a higher number of diffraction peaks demonstrates a shift during MIT, indicating the formation of new domain patterns. Within this region, n -curves in Fig. 5(b) show different slopes for $\phi = 110^\circ$ and 80° at $T > T_c$. This behavior indicates that while along one direction, the surface becomes smoother, along another direction, it becomes rougher. Such a surface reconstruction occurs within the range of spatial frequencies corresponding to typical sizes of V_4O_7 grains and, therefore, is associated with MIT and the evolution of optical inhomogeneity within or at the boundary of individual crystallites.

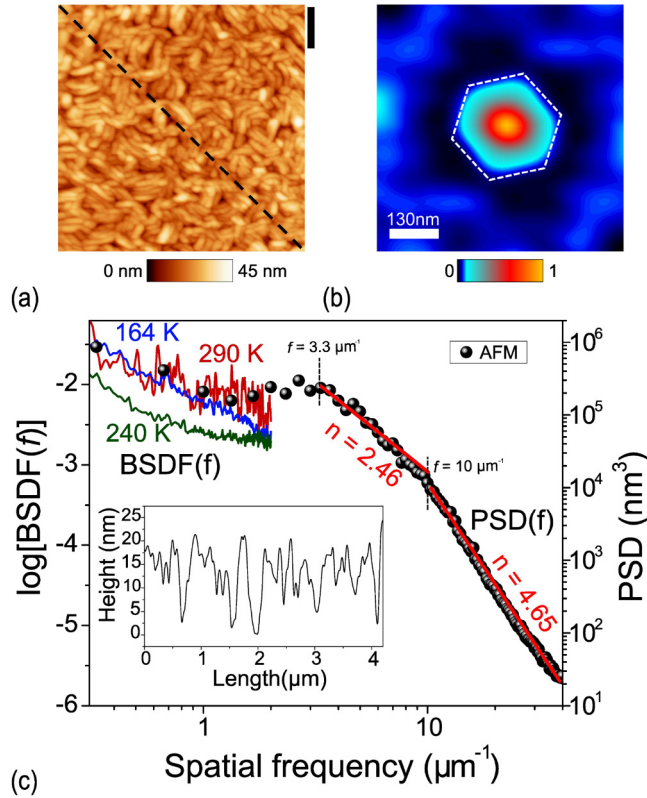


FIG. 4. (a) $3 \times 3 \mu\text{m}^2$ AFM topography scan. The scale bar represents the 500 nm reference length. (b) Surface autocorrelation function (ACF) computed from AFM topography. The hexagonal pattern is outlined by a dashed line. (c) PSD(f) obtained from AFM topography and log[BSDF(f)] cross section of the scattering indicatrix at $\varphi = 110^\circ$. The inset shows the cross section of the AFM image along the dashed line in (a).

While the BSDF(f) provides information about the surface inhomogeneity in the direction perpendicular to the surface, the ACF(r) shows the correlations in the lateral plane. In this regard, the evolution of surface morphology in this plane can be characterized by surface autocorrelation length (ACL). The ACL is defined as a *lag length* at which the ACF(r) falls to a $1/e$ fraction of its maximum value. Figure 6(a) shows the angular distribution ACL in the surface plane for a set of temperatures. The azimuthal map shows the elongation of ACL along the preferred direction $\phi = 85^\circ$ in the high- T phase at $T = 290$ K. After the MIT completes, this direction changes to $\phi = 140^\circ$ at $T = 164$ K for the low- T phase. This behavior indicates significant anisotropy of the surface reconstruction during MIT. Moreover, as the temperature approaches the T_c point, the ACL acquires minimal values [Fig. 6(b)]. This is the result of the reduced correlations between optical inhomogeneities of the surface since the system passes a nonequilibrium region of structural transformation. Here, we note that the V_4O_7 shows similar behavior to VO_2 ,¹³ but quite different from V_3O_5 ,³⁰ where the ACL monotonically decreases as the temperature drops.

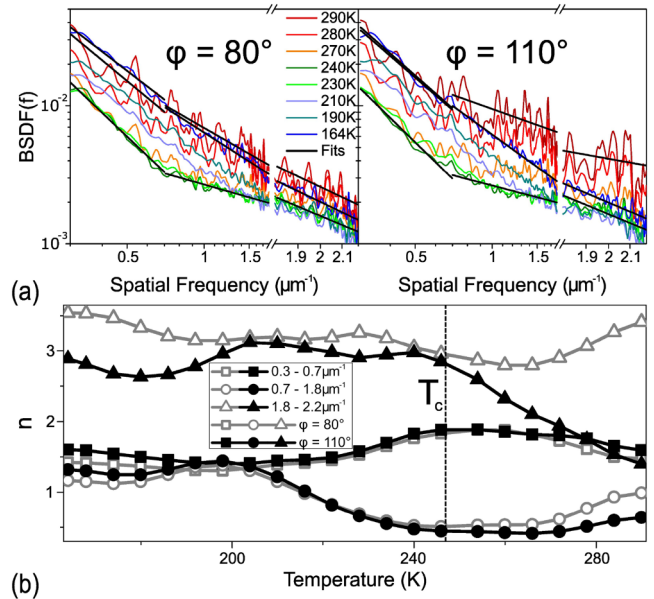


FIG. 5. (a) Cross sections of BSDF(f) indicatrix at $\varphi = 80^\circ$ and 110° . Straight lines are fits to Eq. (2) at 290, 240, and 164 K. (b) Temperature-dependent dynamics of the fractal surface parameter n for three ranges of spatial frequencies.

We also note that some nonmonotonic behavior of ACL appears below ~ 205 K [Fig. 6(b)], and the BSDF acquires nearly the same fractal parameter n within two regions $f = 0.3\text{--}0.7 \mu\text{m}^{-1}$, $f = 0.7\text{--}1.8 \mu\text{m}^{-1}$ [Fig. 5(b)]. This behavior can be associated with the final stage of the MIT with temperature decrease.

One of the straightforward approaches to monitor MIT is the measurement of the integrated scattering signal I_s within the hemisphere [Fig. 6(b)]. Here, I_s is the total scattering intensity within the hemisphere normalized to incident light intensity I_0 . In many cases, when the temperature approaches the T_c point, the system exhibits a “transition opalescence” via a noticeable increase in the scattering signal, as was observed for the first-order transition in VO_2 .^{12,13} In VO_2 and also V_3O_5 ,³⁰ the light scattering monotonically increases across MIT with the temperature decrease. However, the 260-nm-thick $\text{V}_4\text{O}_7/\text{Al}_2\text{O}_3(\text{C})$ film shows a different behavior of I_s : the light scattering reaches its minimal value within the MIT region. This indicates that the system does not form a mixture of low- T and high- T phases with sharp phase boundaries and strong spatial modulation of optical constants. Instead, the whole film experiences a uniform triclinic-to-triclinic MIT across all spatial scales. The MIT in V_4O_7 appears in the ARHELS signal as a continuous transition, where the coexistence of different triclinic phases near T_c results in higher optical homogeneity and lower surface roughness. As a result, $\text{V}_4\text{O}_7/\text{Al}_2\text{O}_3(\text{C})$ demonstrates a quite unusual state near T_c with a minimal spatial correlation of the surface roughness (i.e., highest lateral disorder with lowest ACL) and, at the same time, a minimal level of light scattering due to lowest *rms* roughness.

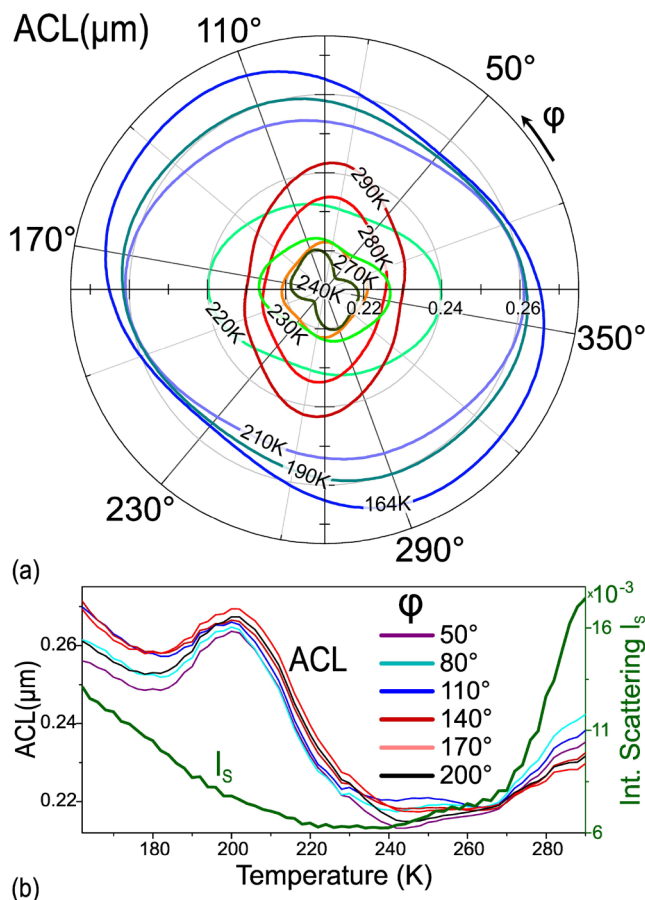


FIG. 6. (a) Azimuthal dependence of the autocorrelation length (ACL). (b) Temperature dependence of the ACL and integrated scattering I_s .

CONCLUSION

In this paper, we discussed the hemispherical light scattering and structural properties of V_4O_7 film deposited on c-cut sapphire crystal across its metal-insulator transition. The 60°-symmetry of scattering indicatrices reveals 120° azimuthal anisotropy of V_4O_7 polydomain morphology, indicating the epitaxial nature of the $V_4O_7/Al_2O_3(C)$ film. The temperature-dependent evolution of the scattering indicatrix and surface autocorrelation function reveals significant changes in surface roughness and its anisotropy across the MIT. The optical inhomogeneity of the material decreases to a minimum at T_c , indicating a uniform triclinic-to-triclinic transition without sharp phase boundaries or phase separation.

The study emphasizes the distinct multiscale distributions of surface domains. The analysis of fractal parameters indicates different roughness statistics at varying spatial frequencies, highlighting the complex nature of surface reconstruction upon phase transition. The autocorrelation length demonstrates significant sensitivity to the MIT, providing a reliable relationship with electrical conductivity, integrated scattering, and fractal parameters. For the

$V_4O_7/Al_2O_3(C)$ system, the autocorrelation length reaches its minimal values near T_c , indicating maximal lateral surface disorder, while the optical inhomogeneity in the normal direction to the surface, material roughness, also drops to the minima, showing opposite behavior to the “transition opalescence.” This can be attributed to the specific nature of the weakly first-order transition in V_4O_7 , which appears in angle-resolved scattering as a continuous transition exhibiting properties of both first- and second-order MITs.

Overall, these results contribute to a deeper understanding of the surface morphology and optical properties of V_4O_7 during the structural phase transition, showing the contribution of surface inhomogeneities in light scattering. This knowledge is crucial for advancing the application of V_4O_7 in optoelectronic devices and improving the design of materials with adjustable optical properties.

SUPPLEMENTARY MATERIAL

Additional results of bidirectional-scatter-distribution function measurements and surface autocorrelation function of the V_4O_7 film are included in the [supplementary material](#).

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Alexander Bartenev: Investigation (equal); Visualization (equal); Writing – original draft (equal). **Camilo Verbel:** Investigation (equal). **Fernando Camino:** Investigation (equal). **Armando Rua:** Funding acquisition (equal); Investigation (equal); Project administration (equal); Supervision (equal); Writing – original draft (equal). **Sergiy Lysenko:** Investigation (equal); Supervision (equal); Writing – original draft (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request and its [supplementary material](#).

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