Ordinary dielectric function of corundumlike $\alpha$-Ga$_2$O$_3$ from 40 meV to 20 eV

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The linear optical response of metastable $\alpha$-Ga$_2$O$_3$ is investigated by spectroscopic ellipsometry. We determine the ordinary dielectric function from lattice vibrations up to the vacuum ultraviolet spectral range at room temperature for a sample with a (0001) surface. Three out of four $E_u$ infrared-active phonon modes are unambiguously determined, and their frequencies are in good agreement with density functional theory calculations. The dispersion of the refractive index in the visible and ultraviolet part of the spectrum is determined. High-energy interband transitions are characterized up to 20 eV. By comparison with the optical response of $\alpha$-Al$_2$O$_3$ and with theoretical results, a tentative assignment of interband transitions is proposed.

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I. INTRODUCTION

The wide-band-gap semiconductor gallium oxide has a multitude of potential applications in spintronics [1], sensorics [2], solar-blind detectors [3], and transparent electronics [4,5]. It has long been known that Ga$_2$O$_3$ crystallizes in several different polytypes [6], each with distinct physical properties. The thermodynamically stable phase is the monoclinic $\beta$ phase which shows great potential for use as substrate material because it can be synthesized in thick boules [4,7–9]. Due to its high chemical and thermal stability matched with a large band gap and high breakdown voltage, this polytype has received increasing attention during the last decade. Specifically, the optical properties of monoclinic $\beta$-Ga$_2$O$_3$ were investigated in detail. Because of low crystal symmetry, phonon contributions [10,11] and interband transitions [11–13] are described by dielectric tensors containing four different nonzero tensor components. The Raman selection rules and tensor components are strongly influenced by the anisotropic properties of the material [14,15].

Additionally, other metastable polytypes of Ga$_2$O$_3$ are known, and their precise physical properties and even their crystal structures are still partly under debate [16,17]. These polytypes have not been studied yet in sufficient detail to unleash their obviously high potential. The corundumlike $\alpha$ phase of Ga$_2$O$_3$ [18] is especially attractive for technological and scientific purposes. Its most prominent advantage is the seamless integration with $\alpha$-Al$_2$O$_3$, which is available as high-quality large-area substrates at a relatively low price [19–21]. Moreover, several metal oxides that crystallize in the corundumlike structure are also available as bulk, e.g., Cr$_2$O$_3$ [22], opening the opportunity to investigate alternative substrates for $\alpha$-Ga$_2$O$_3$. The possibility to create alloys [23] with $\alpha$-In$_2$O$_3$ [24–28] or $\alpha$-Al$_2$O$_3$ is a further invaluable advantage. Consequently, the growth of phase pure alloys [29] and advanced functional heterostructures is possible [30].

$\alpha$-Cr$_2$O$_3$ is known for its interesting magnetic properties [22,31–33], and, similarly to Fe$_2$O$_3$, V$_2$O$_3$, or Rh$_2$O$_3$ in their corundum form, this magnetic oxide is nearly lattice matched to $\alpha$-Ga$_2$O$_3$ [34,35]. Alloys of these materials with $\alpha$-Ga$_2$O$_3$ [36] can be developed to realize new functionalities exploiting the semiconducting properties of $\alpha$-Ga$_2$O$_3$ [37,38] in conjunction with magnetic properties of corundumlike rare-earth oxides.

Experimental studies of the optical properties of $\alpha$-Ga$_2$O$_3$ are rather scarce. Phonon modes were investigated by Raman spectroscopy [39]. Electrical, optical, and magnetic properties of doped material were also studied [40]. Recently, we reported the properties of the fundamental absorption edge at 5.61 eV [41]. Some studies investigated $\alpha$-Ga$_2$O$_3$ by means of first-principles computations [17,42–44].
Here, we provide ellipsometry data for photon energies spanning the range from infrared lattice vibrations up to interband transitions of α-Ga2O3. We employ state-of-the-art spectroscopic ellipsometry experiments spanning an unusually large photon energy range [from ~40 meV (330 cm⁻¹) up to 20 eV] to obtain the dielectric function of the material. We compare the result with previous first-principles calculations and with ellipsometry results obtained on α-Al2O3. The optic axis of the sample used in this study was normal to the sample surface. Therefore, in normal-incidence experiments the electric field vector (\(\vec{E}\)) is perpendicular (\(\perp\)) to this axis (c or [0001] direction) and consequently, only the ordinary component of the dielectric tensor is accessible [41].

Spectroscopic ellipsometry is considered to be an extremely sensitive tool to determine the linear optical response of materials. It can be easily implemented to work with uniaxial materials [45] (like corundum) and is sensitive to small anisotropies [46,47]. For our sample geometry, in which an oblique angle of incidence was used, only the ordinary dielectric function can be accessed with high sensitivity. The influence of the extraordinary tensor component can be modeled empirically in the infrared spectral region [48]. In the transparency regime, the effects of the extraordinary tensor element are negligible [49]. Only slight deviations of amplitudes may occur when describing interband transitions [50], while energy positions are correctly reproduced in the geometry used [51]. The influence of sample anisotropy becomes visible only in the case of materials where the extraordinary imaginary part of the dielectric tensor component yields a strong contribution while the ordinary one is close to zero. This occurs around the band edge of wurtzite AlN [52]. However, this is not the case for corundumlike α-Ga2O3, in which normal-incidence transmission and spectroscopic ellipsometry measurements yield similar absorption line shapes [41].

II. EXPERIMENTAL DETAILS

An α-Ga2O3 thin film grown by ultrasonic mist chemical vapor epitaxy on (0001) α-Al2O3 substrate was investigated experimentally. The substrate temperature was kept constant at 200 °C during the growth. The layer thickness was 473 nm, the root-mean-square surface roughness as measured by atomic force microscopy was 4.4 nm on a 1 × 1 μm scan area [53].

High-resolution x-ray diffraction of the sample provides an out-of-plane lattice parameter of 13.418 Å close to reported values [53,54]; therefore the layer is considered to be nearly or fully relaxed. The FWHM of a symmetric \(\omega\) scan of the (0006) reflex yielded 85 arcsec [53]. The sample was investigated by spectroscopic ellipsometry using different instruments in the different spectral ranges. In the infrared spectral range (300–6000 cm⁻¹) a Woollam IR-VASE based on Fourier transform spectroscopy was employed with resolution set to 4 cm⁻¹, while for photon energies between 0.5 and 6.5 eV, a Woollam VASE with autoretarder was used. There, the resolution was chosen to be better than 5 nm. The angle of incidence was varied between 46° (60°) and 74° in steps of 7° for the visible-ultraviolet (infrared) instrument. Ellipsometric parameters \(\Psi\) and \(\Delta\) were recorded for every photon energy and angle of incidence. In the higher energy range between 5 and 20 eV, an ellipsometer installed at the insertion device beamline of the Metrology Light Source (MLS) in Berlin, Germany was used [55]. The angle of incidence was set to 67° there. Spectra recorded at the synchrotron are not limited by resolution but by energy step width, which was set to 20 meV up to 10 eV and to 50 meV up to 20 eV. All studies were performed at room temperature.

The corundumlike crystal structure of α-Ga2O3 determines a dielectric tensor of the form

\[
\begin{pmatrix}
\varepsilon_{\perp,\perp} + i \varepsilon_{2,\perp} & 0 & 0 \\
0 & \varepsilon_{1,\perp} + i \varepsilon_{2,\perp} & 0 \\
0 & 0 & \varepsilon_{1,||} + i \varepsilon_{2,||}
\end{pmatrix}
\]

The (0001) surface of the samples allows sensitive experimental access to \(\varepsilon_{\perp}\) only, where \(\perp\) represents the electric field vector oriented perpendicular to (0001). \(\varepsilon_{||}\) is also called ordinary dielectric function. To measure the extraordinary dielectric function \(\varepsilon_{\perp}\), other sample surfaces than (0001) would be required.

The ordinary dielectric function was obtained by fitting \(\Psi\) and \(\Delta\) data from all angles of incidence together in a multilayer model [56], which also accounts for the surface roughness via the Bruggeman effective medium approximation [57] and for the (0001)-oriented α-Al2O3 substrate. The dielectric functions published by Malitson and Tomiki et al. [58,59] for the visible-ultraviolet spectral range were used to describe the sapphire substrate. In the infrared spectral range, we determined experimentally the dielectric response of...
sapphire on polished sapphire substrates with different surface orientations, i.e., (0001), (1100), and (1120) orientations.

After modeling the experimental data by superposition of different Kramers-Kronig consistent model functions optimized to match the experimental line shapes, a numerical point-by-point fit was performed without a priori assumptions on the line shape of $\tilde{\varepsilon}_{\perp}$. The excellent agreement of this point-by-point fitted model with the experimental data is displayed in Figs. 1, 2, and 3 for the different spectral ranges.

### III. AB INITIO CALCULATION OF THE INFRARED-ACTIVE MODES

The frequencies of the infrared-active modes of $\alpha$-Ga$_2$O$_3$ were calculated using a plane-wave pseudopotential approach to density functional theory (DFT) as implemented in the ABINIT code [60]. The calculations were performed in the local density approximation (LDA) using the Perdew-Wang parametrization of the exchange interaction and Troullier-Martins pseudopotentials. The 3$d$ electrons of Ga were included as valence states. A $4 \times 4 \times 4$ Monkhorst-pack $k$-point sampling and a plane-wave basis set with an energy cutoff of 95 Ha were used. A full structural relaxation of the $\alpha$-Ga$_2$O$_3$ unit cell was carried out, and the hexagonal lattice parameters $a = 5.004$ Å and $c = 13.410$ Å were obtained, in excellent agreement with the reported high-resolution x-ray diffraction values ($a = 4.983$ Å and $c = 13.433$ Å) [54]. Phonon frequencies were determined by diagonalizing the dynamical matrix derived from the perturbation theory linear response approach [60]. The calculated frequencies of the zone-center infrared-active phonons are listed in the first column of Table I.

### IV. RESULTS AND DISCUSSION

#### A. Infrared dielectric function

For the infrared dielectric function, we use a sum of Lorentzian oscillators and the dielectric limit to describe the point-by-point fitted experimental result. Therefore,

$$\varepsilon_{\text{IR}}(\omega) = \varepsilon_{\infty} + \sum_i \frac{S_i \omega_{\text{TO},i}^2}{\omega_{\text{TO},i}^2 - \omega^2 - i \omega \gamma_{\text{TO},i}}$$

is used for the ordinary and extraordinary dielectric functions, separately. The space group of corundum crystals is $D_{3d}^4 (R3c)$ expressing a ditrigonal-scalenohedral symmetry. At $\Gamma$, there are two infrared-active phonon modes with $A_2u$ symmetry for the extraordinary dielectric function and four modes with $E_u$ symmetry for the ordinary one.

The $\alpha$-Al$_2$O$_3$ substrate has the same symmetry, and the corresponding experimental phonon modes are listed in Table I. Its dielectric limits are found to be $\varepsilon_{\infty,\perp} = 3.08$ and $\varepsilon_{\infty,||} = 3.05$ from model fits. We note that synthetic sapphire was extensively investigated earlier with nearly identical results [61]. However, a more complicated anharmonic product description of the infrared part of the dielectric function was used there [62] which is not required in the present study.

For $\alpha$-Ga$_2$O$_3$ our experimental geometry is only sensitive to the ordinary component of the dielectric tensor. For a proper data modeling routine, we therefore have to make an assumption about the extraordinary part of the dielectric tensor, i.e., the characteristic energy $\omega_{\text{TO},i}$, broadening factor $\gamma_{\text{TO},i}$, and amplitudes $S_i$ of the two $A_{2u}$ modes. For this purpose, we carefully compared the energy positions of Raman active phonon modes of $\alpha$-Al$_2$O$_3$ and $\alpha$-Ga$_2$O$_3$, which are fully explored [39,63]. Next, the results for the ordinary dielectric functions of $\alpha$-Al$_2$O$_3$ and $\alpha$-Ga$_2$O$_3$ were compared.

### TABLE I. Calculated (first column) and experimentally obtained IR parameters (other columns). Values in italic were fixed and not fitted. $\omega_{\text{TO}}$ is the phonon mode resonance energy, $\gamma_{\text{TO}}$ its broadening parameter, and $S$ the corresponding amplitude factor. $l$ is used for indexing the different $E_u$ and $A_{2u}$ symmetry phonon modes in both materials.

<table>
<thead>
<tr>
<th></th>
<th>$\alpha$-Ga$_2$O$_3$</th>
<th>$\alpha$-Al$_2$O$_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\omega_{\text{TO},i}$ (cm$^{-1}$)</td>
<td>$\gamma_{\text{TO},i}$ (cm$^{-1}$)</td>
</tr>
<tr>
<td>$E_u$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>220</td>
<td>333.4</td>
</tr>
<tr>
<td>2</td>
<td>326</td>
<td>333.4</td>
</tr>
<tr>
<td>3</td>
<td>462</td>
<td>469.9</td>
</tr>
<tr>
<td>4</td>
<td>551</td>
<td>562.7</td>
</tr>
</tbody>
</table>

| | $A_{2u}$ | | | | | | |
|---|---|---|---|---|---|---|
| 1 | 269 | 280 | 20 | 12 | 396.7 | 3.16 | 6.73 |
| 2 | 531 | 544 | 20 | 1 | 582.7 | 2.95 | 1.71 |
Finally, the phonon frequencies computed by first-principles calculations (see Sec. III) were also taken into account.

In Fig. 4, we compare the characteristic phonon energies of $\alpha$-Al$_2$O$_3$ and $\alpha$-Ga$_2$O$_3$ and a linear relationship is fitted to the data. From this linear dependence, the three missing infrared-active phonon modes are determined to be around 231 cm$^{-1}$ ($E_u^1$), 247 cm$^{-1}$ ($A_{2u}^1$), and 480 cm$^{-1}$ ($A_{2u}^2$). Note that the slope of the interpolation line (1.26) matches approximately the square root of the reduced mass ratio of Ga$_2$O$_3$ and Al$_2$O$_3$ (1.19). However, for use in our model fits, we iteratively changed the extracted $A_{2u}$ phonon energy positions to improve the agreement (Fig. 1). We finally assumed the values of 280 and 544 cm$^{-1}$, as reported in Table I in italic. These values are also in good agreement with computational results (Sec. III and Table I) which systematically underestimate the experimentally obtained room temperature phonon mode frequencies. The order of phonon mode amplitudes is basically maintained when going from $\alpha$-Al$_2$O$_3$ to $\alpha$-Ga$_2$O$_3$. The broadening factors are substantially higher for the thin epitaxial $\alpha$-Ga$_2$O$_3$ film, because it is expected to contain a density of point and structural defects larger than that of the high-quality bulk sapphire substrates investigated. For the remaining $E_u$ phonon mode of $\alpha$-Ga$_2$O$_3$, a reliable experimental estimate cannot be given because this phonon is found far from the spectral range of our measurements.

Our analysis of the dielectric function for the $E_u$ phonon modes of $\alpha$-Ga$_2$O$_3$ allows us to unambiguously assign three of the four $E_u$ modes. The missing undetectable mode is expected to have low amplitude $S \approx 0.1$ and a characteristic energy $\omega_{\ell} \approx 275$ cm$^{-1}$, which is in agreement with the computational result of 220 cm$^{-1}$. Figure 5 presents a comparison of the point-by-point fitted experimental dielectric function of $\alpha$-Ga$_2$O$_3$ to the model given by Eq. (2). No significant residual contributions from neither substrate modes nor the extraordinary phonon modes are detectable, which corroborates the validity of the assumptions we made for the $A_{2u}$ modes. The ordinary dielectric limit of $\alpha$-Ga$_2$O$_3$ is found to be $\varepsilon_{\infty,\perp} = 3.75$ from our model fits. Maintaining the ratio of ordinary and extraordinary dielectric limits from the theoretical calculations published in Ref. [17] of $\approx 1.03$, we obtain $\varepsilon_{\infty,\parallel} = 3.64$. Please note that for both corundum materials, $\varepsilon_{\infty,\perp} > \varepsilon_{\infty,\parallel}$, which implies that these materials have a negative birefringence.

### B. Optical properties in the visible-to-ultraviolet spectral range

The complex ordinary dielectric function of $\alpha$-Ga$_2$O$_3$ is shown in Fig. 6 for the range of photon energies from 0.5 up
to 20 eV. Experimental point-by-point fits were performed on data obtained by using two ellipsometers as described above. First of all, we note that the imaginary part of the dielectric function $\varepsilon_2$ is not zero for photon energies higher than $\sim 5.6$ eV. This is the onset of strong interband absorption of the material associated with the direct band gap. The shape, temperature dependence, and derivation of the exact energy value of the direct band gap was discussed in detail recently [41]. The major findings were the determination of a fundamental direct band gap at 5.61 eV and a higher energy transition at 6.44 eV. An exciton binding energy of about 110 meV was deduced. Both absorption steps are clearly visible in Fig. 6 in the $\varepsilon_2$ spectrum and in the corresponding Kramers-Kronig maxima in the $\varepsilon_1$ spectrum.

The refractive index $n$ is equal to $\sqrt{\varepsilon_1}$ in the energy range where $\varepsilon_2 = 0$, i.e., for photon energies below the band gap. An analytical description of the point-by-point fitted $\varepsilon_{1,2}(\hbar\omega)$ can be made using the model proposed by Shokhovets et al. [64]:

$$
\varepsilon_1(\hbar\omega) = 1 + \frac{2}{\pi} \frac{A_G}{2} \ln \frac{E_{1G} - (\hbar\omega)^2}{E_{1G} - (\hbar\omega)^2} + \frac{A_H E_{1H}}{E_{1H} - (\hbar\omega)^2},
$$

It is derived by a Kramers-Kronig transformation of a representative simplified model for $\varepsilon_2(\hbar\omega)$ consisting of a constant $A_G$ for $\varepsilon_2$ above the effective absorption onset $E_{1G}$ and an upper energy contribution given by a $\delta$ function at $E_{1H}$ weighted by $A_H$ representing effective high-energy contributions. By fitting Eq. (3) to $\varepsilon_1(\hbar\omega < 4.8$ eV), we obtain the parameters given in Table II. In this model, the dielectric limit $\varepsilon_{\infty}$ can be found as the asymptotic limit at $\hbar\omega \to 0$ of Eq. (3), and we obtain $\varepsilon_{\infty} = 3.75$. This value is in perfect agreement with $\varepsilon_{\infty,1}$ derived from the infrared analysis (see above).

The high-energy interband excitonic transitions are visible as peaks in $\varepsilon_2(\hbar\omega)$. We expect a certain similarity between the observed features and those of $\alpha$-Al$_2$O$_3$ because the crystal symmetry plays a major role in shaping the band structure. Substitution of Ga for Al in the corundum sesquioxide is expected to decrease transition energies. Similar behavior is reported for $\beta$ sesquioxides [20] or when comparing AlN and GaN, both in the zinc-blende [65] and in the wurtzite polytype [66]. Also the classical III-V zinc-blende semiconductors AlAs and GaAs show a similar behavior [67].

The high-energy interband transition bands of sapphire were reported for a very wide spectral range [59,69,70]. Here, we use the experimental results by Tomiki and colleagues [59] and follow a later analysis by Harmann et al. [68], where six characteristic transition energies were identified in the spectral range up to 18 eV for the ordinary dielectric function (Table III). Visual inspection of $\varepsilon_2$ for $\alpha$-Ga$_2$O$_3$ shows that the overall line shape is quite similar (Fig. 7). We therefore assign high-energy transition peaks following the identification performed for sapphire [68]. The first six characteristic transition energies are summarized in Table III.

The similarities in shapes and amplitudes (Fig. 7) of the identified high-energy features lead us to a tentative one-to-one correlation of the related features for both materials. Accordingly, the arrows shown in Fig. 7 correlate the characteristic transition bands of both materials. Several attempts were made to assign the observed bands of the dielectric function of sapphire to different regions of the band structure in a simplified atom orbital picture [70,71]. The dominant contributions in $\varepsilon_2$ of $\alpha$-Al$_2$O$_3$ are believed to originate mainly from transitions between $2p$ valence bands and $13p$ conduction bands [70]. Recent first-principles results of different Ga$_2$O$_3$ polytypes [17] yield a similar assignment for the two main absorption contributions in $\alpha$-Ga$_2$O$_3$. The transitions $k = 3$ and 4 (Table III) are therefore tentatively assigned mainly to transitions between $2p$ valence bands and Ga$_4s$/$4p$ conduction bands.

These assignments are further corroborated by comparing the theoretically calculated dielectric function including Coulomb interaction [17] of $\alpha$-Ga$_2$O$_3$ with our experimental result (Fig. 8). The overall line shape of our experimental result is nicely reproduced by the computational spectrum. However, the theoretical curve is found at lower energies compared to the experimental spectrum with higher discrepancies at higher

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**Table II.** Parameter set for the empirical description of $\varepsilon_{1,2}$ below 4.8 eV at $T = 295$ K.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{1G}$ (eV)</td>
<td>5.8</td>
</tr>
<tr>
<td>$E_{1H}$ (eV)</td>
<td>11.1</td>
</tr>
<tr>
<td>$A_G$</td>
<td>1.3</td>
</tr>
<tr>
<td>$A_H$ (eV)</td>
<td>38.5</td>
</tr>
</tbody>
</table>

**Table III.** Characteristic energies of high-energy transitions of the ordinary dielectric functions of $\alpha$-Ga$_2$O$_3$ and $\alpha$-Al$_2$O$_3$ [68].

<table>
<thead>
<tr>
<th>$k$</th>
<th>$E_k$ (eV)</th>
<th>$\varepsilon_{Ga_2O_3}$ (eV)</th>
<th>$\varepsilon_{Al_2O_3}$ (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5.60</td>
<td>9.25</td>
<td>9.25</td>
</tr>
<tr>
<td>2</td>
<td>6.44</td>
<td>10.88</td>
<td>10.88</td>
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<tr>
<td>3</td>
<td>9.70</td>
<td>12.57</td>
<td>12.57</td>
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<tr>
<td>4</td>
<td>11.18</td>
<td>13.32</td>
<td>13.32</td>
</tr>
<tr>
<td>5</td>
<td>12.79</td>
<td>14.91</td>
<td>14.91</td>
</tr>
<tr>
<td>6</td>
<td>15.37</td>
<td>17.40</td>
<td>17.40</td>
</tr>
</tbody>
</table>
energies. Most likely, this fact originates from a slight underestimation of the quasiparticle corrections and/or screening effects. Nevertheless, the overall agreement visible in Fig. 8 allows one to conclude that the general findings of Ref. [17] can describe correctly the optical spectra of α-Ga$_2$O$_3$.

V. SUMMARY AND CONCLUSIONS

Spectroscopic ellipsometry measurements on the metastable α phase of Ga$_2$O$_3$ have been performed over an extended energy range up to 20 eV. The ordinary dielectric function $\varepsilon_\parallel$ has been determined by point-by-point fitting of the ellipsometry data without a priori assumptions on the $\varepsilon_\perp$ line shape. Excellent agreement with the experimental data is achieved. The infrared dielectric function is accurately described with a sum of Lorentzian oscillators, which has allowed us to clearly identify three out of the four expected $E_p$ infrared-active modes at frequencies of 333.4, 469.9, and 562.7 cm$^{-1}$. These frequencies are in good agreement with theoretical values of infrared-active modes that we have calculated using a DFT approach.

The dielectric function in the ultraviolet below the absorption onset was analyzed using Shokhovets’ model, and the dielectric limit $\varepsilon_\infty = 3.75$ found is in perfect agreement with the value derived from the infrared data. The high-energy interband excitonic transitions are clearly visible as peaks in $\varepsilon_2(h\omega)$. These have been tentatively identified by analogy with the well-characterized α-Al$_2$O$_3$ interband transitions by establishing a one-to-one correspondence between the peaks observed in their respective absorption spectra. Our analysis suggests that the observed absorption peaks originate mainly from O2$p$ valence band to Ga4$s$/4$p$ conduction-band transitions.

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