

Supplemental graphs

Calculation of spectral optical constants using combined ellipsometric and reflectance methods for smooth and rough bulk samples

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Other glass sample measured with another ellipsometer

In Figures S.1 to S.7, the fully opaque curves are supplemental results obtained from measurements performed with a different ellipsometer (SOPRA company, IRSE-5E model, silicon carbide source, rotating-analyzer configuration, polarizer at mirror angles of $+45^\circ$ and -45° , FTIR spectrometer, DTGS thermoelectrically cooled detector) and a different glass sample (slide from another lot, black electrical tape attached to its back side), at each angle of observation $\theta_o \in \{45^\circ, 75^\circ\}$, with a spectral resolution of 8 cm^{-1} and with 128 averaged scans. For comparison purposes, corresponding data from the main paper are kept in the background, as semi-transparent curves. As can be observed, when an ellipsometer from a different company is used to investigate a different glass sample, the main trends remain consistent with those discussed in the paper, for all presented parameters. The observed undesirable discrepancies are worse (see Fig. S.3), and they are still well corrected (see Fig. S.7) using the method presented in the paper. Therefore, the undesirable spectral phenomena discussed for glass in the main paper and the improvements obtained with the presented method are not specific to the optical instrument used to take the measurements or to the sample analyzed.

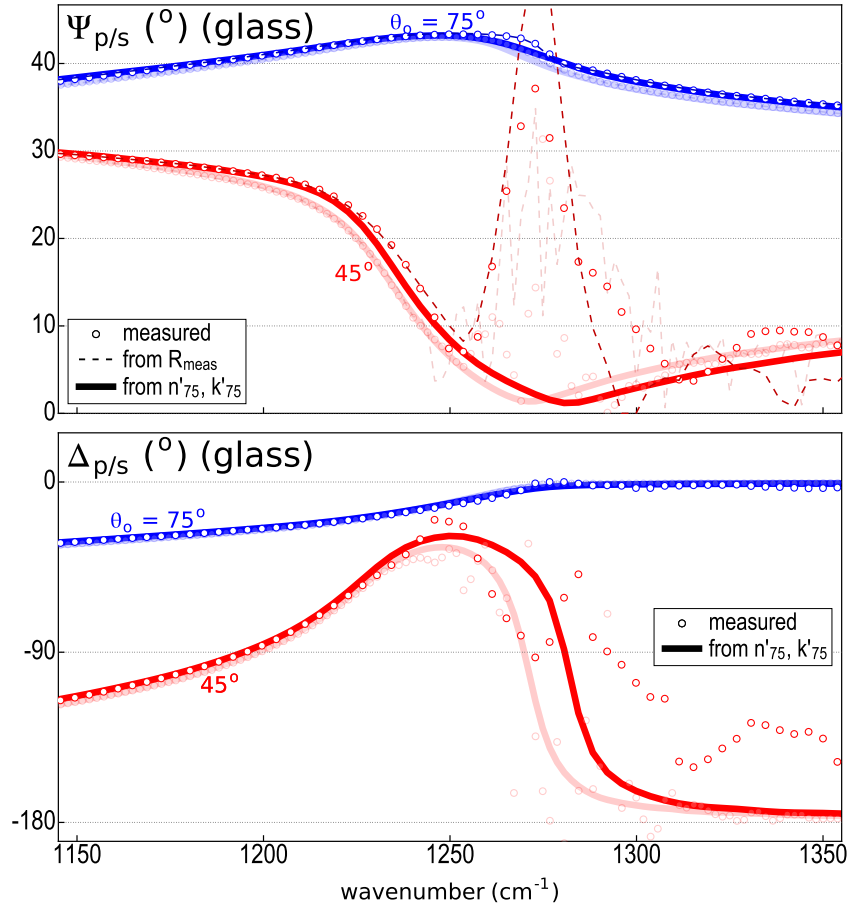


Figure S.1: Ellipsometric angles $\Psi_{p/s}$ and $\Delta_{p/s}$ (other glass sample, other ellipsometer).

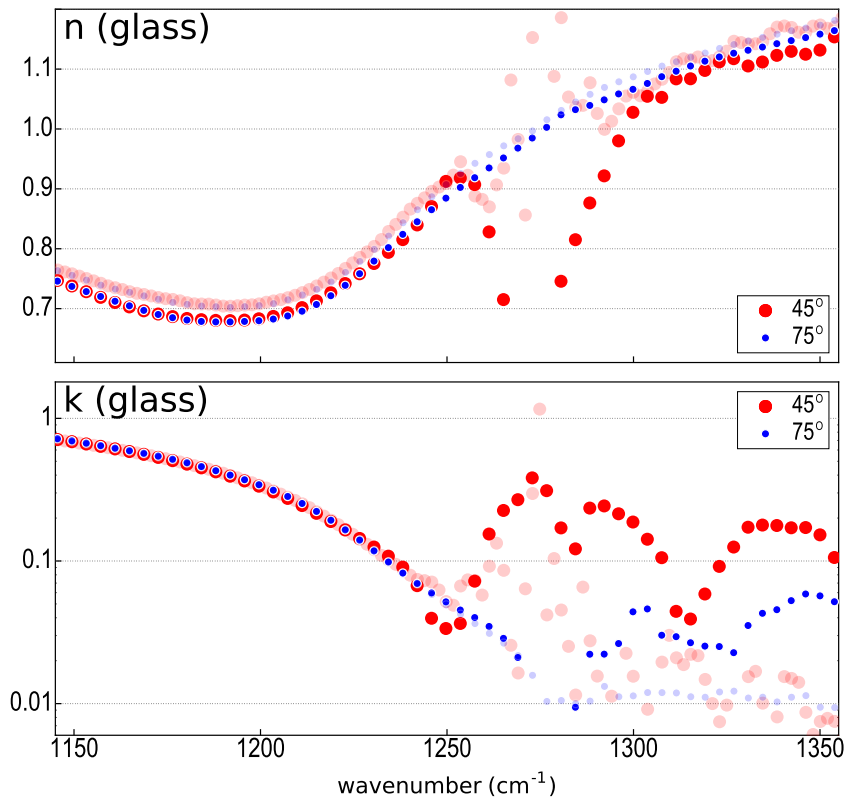


Figure S.2: Optical constants n and k obtained with the ellipsometric method (other glass sample, other ellipsometer).

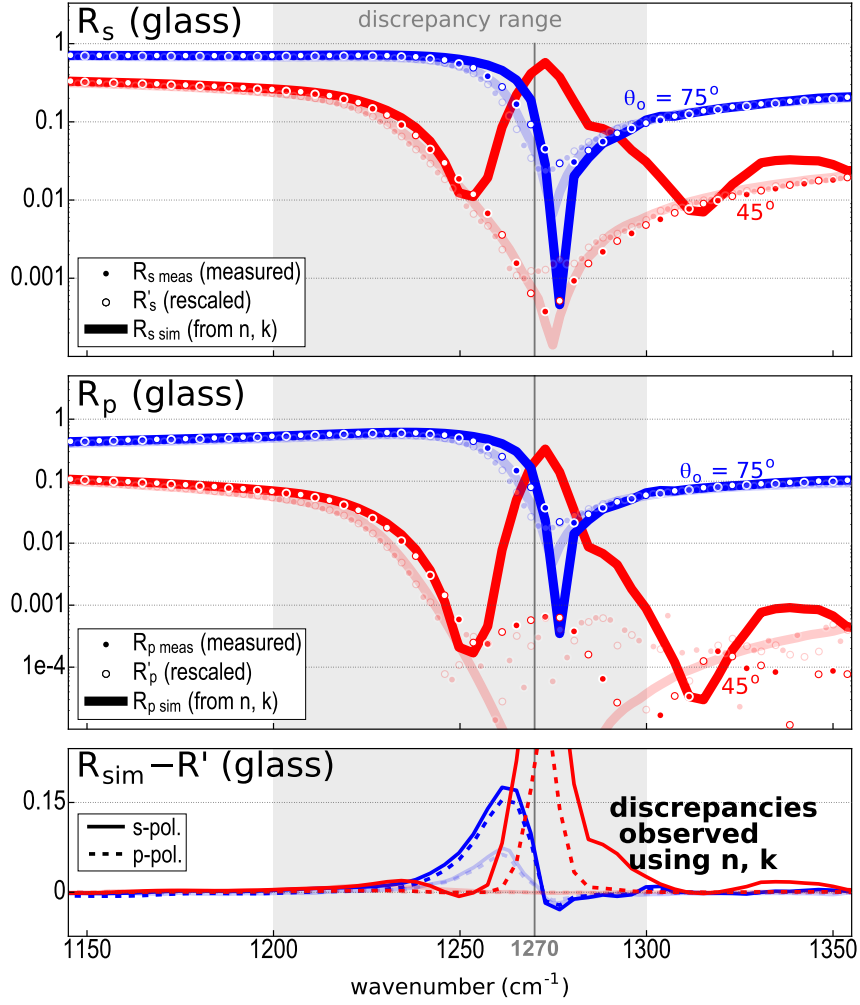


Figure S.3: Discrepancies observed between the polarized reflectance spectra measured (rescaled) and calculated with n and k from the ellipsometric method (other glass sample, other ellipsometer).

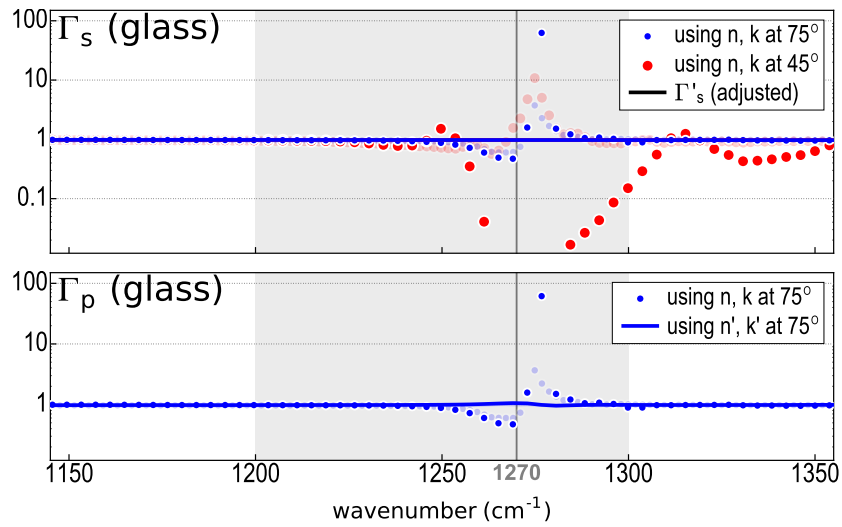


Figure S.4: Specularity factor Γ , analyzed independently in polarization planes s and p (other glass sample, other ellipsometer). The noisiest p curves are omitted for clarity.

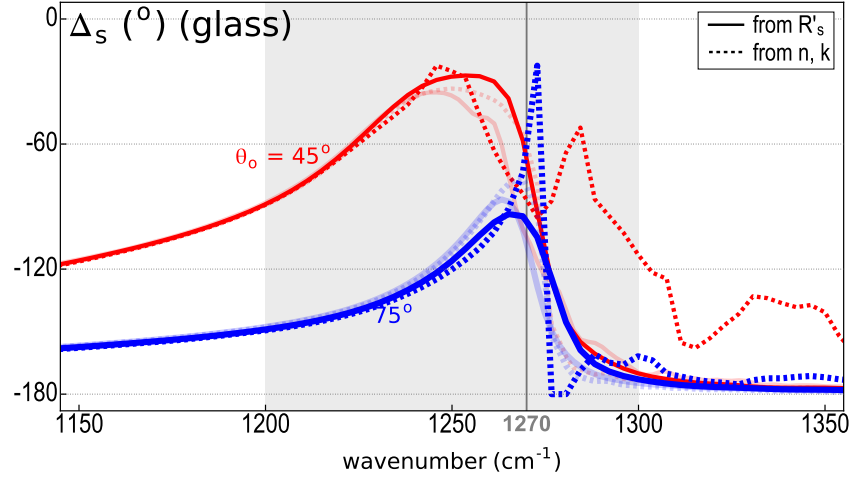


Figure S.5: Phase Δ_s of the s -polarized reflection coefficient r_s (other glass sample, other ellipsometer).

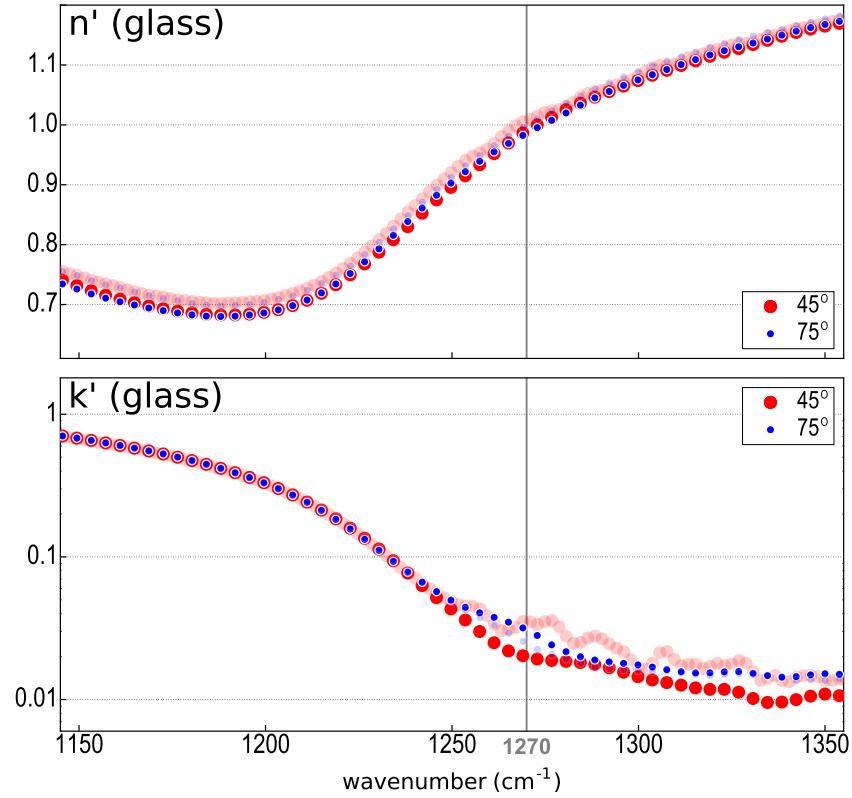


Figure S.6: Optical constants n' and k' obtained from R'_s with the alternative method (other glass sample, other ellipsometer).

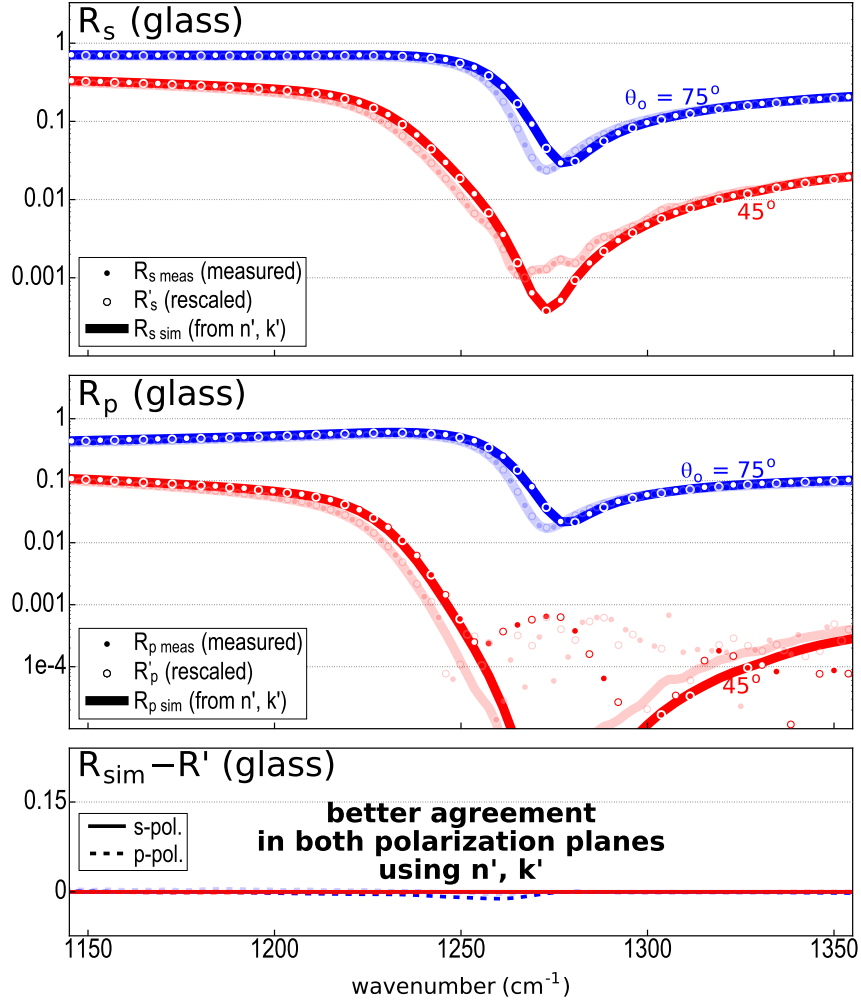


Figure S.7: Comparison of the polarized reflectance spectra measured (rescaled) and calculated with n' and k' from the alternative method (other glass sample, other ellipsometer).

Depolarization

The depolarization is another spectral parameter measured and provided by the acquisition software controlling the ellipsometer described in the main paper. It represents the loss of polarization brought to the fully polarized incident beam due to the reflection by the sample. A significant depolarization value generally implies less valid ellipsometric measurements.

The top part of Fig. S.8 presents the spectral curves of the depolarization in all measurement cases discussed in the main paper (two samples, four angles, same color code). For comparison purposes, the curves of the s -polarized specularity factor Γ_s are reported in the bottom part of Fig. S.8. Observations are as follows.

For the glass sample (left), in the grayed limited discrepancy region, spectral modulations are observable both in the depolarization curves and in Γ_s . The depolarization varies with the angle of observation, as the specularity factor does. At 75° (blue curves), the spectral modulation is more obvious in Γ_s than it is in the depolarization curve. For the ammonium sulfate sample (AS, right), in the grayed discrepancy region and at all angles investigated, spectral modulations are obvious in Γ_s while they are absent or drowned in the noise in the depolarization curves. As discussed in the main paper, Γ_s is expected to decrease monotonically with wavenumber, and the unexpected spectral modulations shown here are due to problematic values, on limited spectral ranges, of the optical constants calculated with the ellipsometric method. Therefore, as shown in Fig. S.8, the depolarization parameter is less sensitive than the specularity factor when searching for spectral regions where the optical constants calculated with the ellipsometric method can be improved.

Note that for both samples, a depolarization of about 1% (plus noise) is generally observed outside the discrepancy regions, which is relatively low and tends to show that the ellipsometric measurements acquired with the specific instrument used and the specific samples analyzed are generally valid, except on limited spectral ranges such as the discrepancy regions discussed in the main paper.

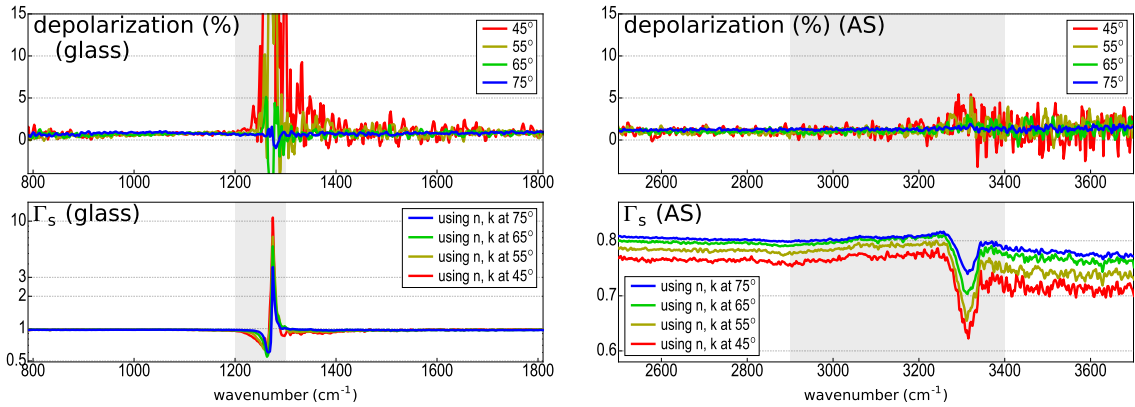


Figure S.8: Depolarization (top) and s -polarized specularity factor Γ_s (bottom) for glass (left) and ammonium sulfate (AS, right), at four angles.