Wave scattering model for christiansen optical filters and control of glass homogeneity by the shelyubskii method.

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Christiansen optical filters with solid grains or liquid droplets immersed in a continuous phase of comparable refractive index were used as chromatic filters before replacement by interference filters. Christiansen filters are optically homogeneous for radiation in a narrow band strongly dependent upon temperature and now provide a method for characterizing glass homogeneity known as the Shelyubskii method [1] in the glass industry. Close to the Christiansen wavelength or temperature where liquid and particle refractive indices are equal, the spectral transmission becomes very sensitive to particle homogeneity as a result of scattering effects due to differences in particle refractive indices. Raman [2] developed a wave theory of the coherent transmission for an ideal detector of zero aperture angle by considering the phase changes among the waves passing the filter. Despite Imagawa's correction for the phase variance due to particle number fluctuations [3], Raman's theory does not predict the correct temperature dependence of the spectral transmission and invariably underestimates the variance of glass refractive index measured with Mach – Zehnder interferometry [4]. Varshneya *et al* [5] then advanced the controversial opinion that geometrical optics had to be used rather than wave optical theory to describe light scattering from large particles. In fact, a realistic wave theory of Christiansen filters has to account for the aperture angle of the detector and diffuse transmission through the scatttering medium. At the moment, a wave scattering model describing the spectral attenuation of Christiansen filters is still missing despite a very large number of experimental and theoretical contributions.

The present work examines critically the contradictions among previous theories and experiments on the basis of Monte Carlo simulations and statistical models of wave propagation in random scattering media. The anomalous diffraction theory for light scattering from large particles predicts a spectral coherent extinction in agreement with the Raman – Imagawa theory of weakly scattering media with an additional term describing scattering losses:

$$C = \alpha\ \mathrm{e}^{-\tau}\ \mathrm{e}^{-\frac{4\omega_\phi^2}{k\phi^2}\tau} \text{ with } \tau = \frac{k z \phi}{2}\left\langle\frac{Q_e}{d}\right\rangle \text{ and } \left\langle\frac{Q_e}{d}\right\rangle = \frac{2\pi^2 d}{\lambda^2}\left[(n_p - n_s)^2 + \omega^2\right] \text{ for } Q_e < 1/2$$

where α accounts for wall reflections and absorption losses, λ is the light wavelength, τ the optical thickness, z the slab thickness, Q_e the extinction efficiency from a single particle in fluid, ϕ the particle volume fraction, d the particle diameter, n_p the average particle refractive index, n_s the fluid refractive index, ω_ϕ the local variance in particle volume fraction, ω the variance in particle refractive indices and $k \approx 1$ a structure factor. Considering far field interferences of waves scattered in the forward direction, a photon diffusion approximation further gives the diffuse transmission D within the aperture angle Ω of the detector:

$$D = \alpha \ \mathrm{e}^{-\frac{4\omega_\phi^2}{k\phi^2}\tau} \sum_{s\geq 1} \frac{\tau^s}{s!}\left(1 - P(\Omega)^{1/s}\right)\mathrm{e}^{-\tau} \qquad \text{with } \tau = \frac{k\,z\phi}{2}\left\langle \frac{Q_e}{d} \right\rangle$$

where s is the scattering number of diffusion paths and $P(\Omega)$ the cumulative density probability for scattering angles from a single particle. In agreement with Monte Carlo simulations, the wave scattering model predicts non negligible diffuse transmission for relatively homogeneous glass spheres and small detector aperture angle (fig.1). Glass inhomogeneities significantly increase the diffuse contribution and the width of the spectral transmission curve defined in terms of temperature at half-height (fig.1)

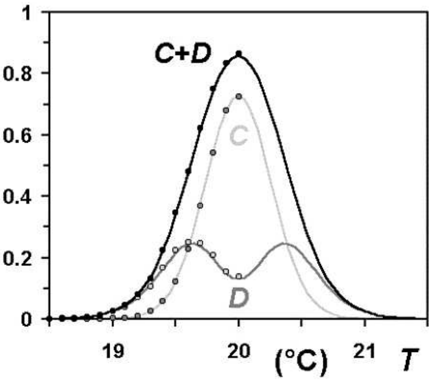

Figure 1 : Temperature dependence of spectral extinction C and diffuse transmission D of coherent light in a random packing of scattering glass spheres. Standart deviation of particle refractive indices $\omega = 10^{-4}$ and detector aperture angle Ω = 1mrad (d = 335µm, ϕ = 0.58, z = 5mm, ω_ϕ = 0.15, $\partial n_s / \partial T = 5\,10^{-4}$ and k =1). Numerical simulations (symbols) and wave scattering model.

The transmission curves of Christiansen filters were determined using an expanded He-Ne laser (beam diameter 5mm) and a detection system of small aperture angle $\Omega <$ 1.33mrad. The preparation process of the glass grains was standardised and the heating rate < 0.15°/mn of the scattering medium ensures negligible temperature gradients in the cell. For closely packed float glass grains in chlorobenzene, the wave scattering model describes experiments and predicts a standart deviation $\omega = 1.1\ 10^{-4}$ of glass refractive indices (fig.2*a*). The classical Raman – Shelyubskii theory ignores multiple scattering and then underestimates both the transmission of opaque media and the homogeneity of glass ($\omega = 4\ 10^{-5}$, fig.2*b*) as recently reported by Heidrich *et al* [4].

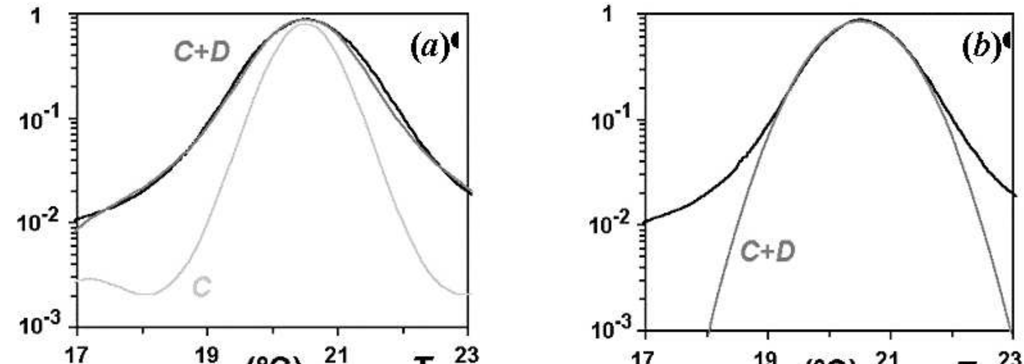

Figure 2 : Experimental transmission curves of float glass grains in Chlorobenzene Spectral extinction C and attenuation C+D derived either from the wave scattering model ($\omega = 1.1\ 10^{-4}$ (a)) or the Raman – Shelyubskii theory ($\omega = 4\ 10^{-5}$ (b)) (Ω =1.33mrad, d=375µm, ϕ=0.58, z =2mm, ω_ϕ = 0.085, $\partial n_s / \partial T = 5\,10^{-4}$ and k =1).

An extensive set of experiments was performed with float glass or Corning glass in chlorobenzene or brombenzol as a function of particle diameter, cell thickness and detector aperture angle (fig.3). In contrast with the predictions of Raman - Shelyubskii theory, the maximum transmittance C_o+D_o slightly increases with particle diameter (fig.3*a* and fig.3*b*).

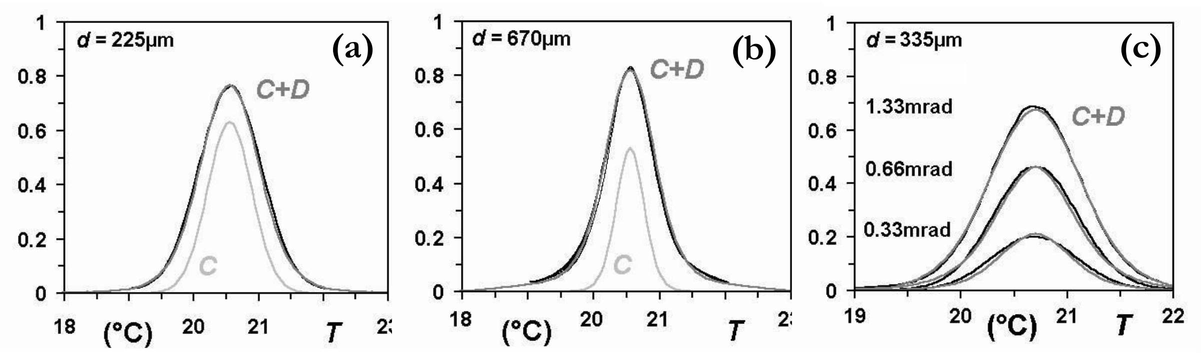

Figure 3 : Experimental transmission curves, spectral extinction C and attenuation C+D (a) float glass in chlorobenzene with d = 375µm and ω = 1.5 10^{-4}, (b) float glass in chlorobenzene with d = 670µm and ω = 1.1 10^{-4}, (c) Corning glass in brombenzol with d = 335µm and ω = 2.2 10^{-4}.

Multiple scattering effects and non linear variations of extinction efficiency with the dielectric contrast indeed result in a complex dependence of the maximum transmittance C_o+D_o of Christiansen filters with mean particle diameter and light wavelength in good agreement with phenomenological formula derived by Afghan *et al* [6] and Hoffmann *et al* [7]:

$$C_o + D_o \approx \mathrm{e}^{-\beta \frac{z\,d}{\lambda^2\,\Omega^{1/2}}\omega^2} \quad \text{for} \quad d < \frac{\lambda}{\pi\,\omega} \qquad \text{and} \qquad C_o + D_o \approx \mathrm{e}^{-\frac{z\,\omega^{1.7}}{d\,\Omega^{1/2}}} \quad \text{for} \quad d > \frac{\lambda}{\pi\omega}$$

For closely packed grains, the variance ω_ϕ in local particle volume fraction is about 0.07 (fig.4*a*). The weak decrease of the variance in glass refractive indices further reflects some inhomogeneity within the volume of the grains (fig.4*b*).

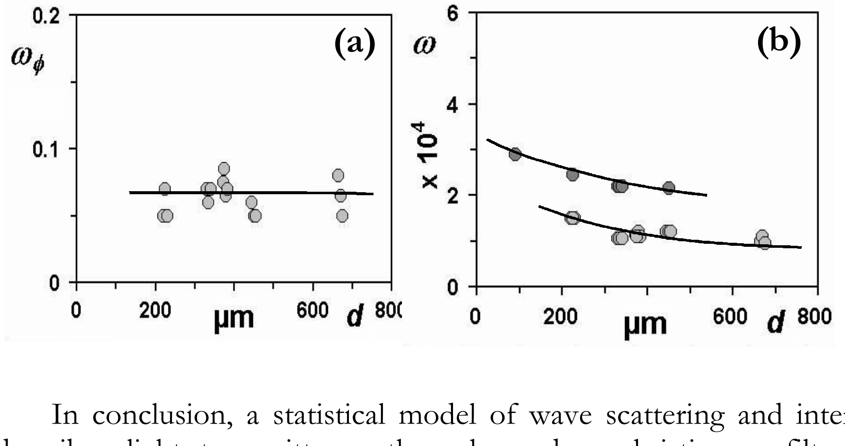

Figure 4 : Variance of local particle volume fraction ω_ϕ (*a*) and particle refractive indices ω (*b*) derived from transmission curves for float glass (○) and Corning glass (●).

In conclusion, a statistical model of wave scattering and interference phenomena describes light transmittance through random christiansen filters and accounts for reported inadequacies of the Raman – Shelyubskii theory. The scattering model provides a convenient way to determine the variance of glass refractive index for relatively small aperture angle in the forward direction. The present statistical treatment of far field interferences indeed becomes inaccurate for large detector aperture angle and strongly scattering media.

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