The optical constants of the films MgBaF$_4$ in the range of 1.3–16 micron

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Abstract. A BaF$_2$ films, characterized by low refraction coefficient in the infrared region of the spectrum, are widely used in the production technology of optical structures. Their serious drawback is the low mechanical strength and moisture resistance. This paper presents the results of an experimental study of the optical properties of a new type of MgBaF$_4$ films made from a vacuum-sintered mixture of MgF$_2$ and BaF$_2$ to a homogeneous composition.

1. Introduction
Fluoride films obtained by vacuum deposition have high transparency and low refraction coefficient in the middle infrared (IR) region of the spectrum [1]. Therefore, they are widely used as film-forming materials and are used in the composition of multilayer interference coatings. Among the films of fluorides, BaF$_2$ films have the minimum absorption rate. Their serious drawback is the low mechanical strength and moisture resistance. In papers [2, 3] there were studies of the optical films BaF$_2$ doped with small amounts (up to 1–2 %) of MgF$_2$. It has been shown that this reduces the optical scattering losses.

This paper presents the results of experimental studies of the optical properties of MgBaF$_4$ films. The films were produced in vacuum at the VU-2M installation by electron-beam evaporation (ELI-22) of a material from a graphite crucible [4]. Control of the thicknesses of the deposited films and the deposition rate of the condensate was carried out by the photometric method [5]. The thickness was determined by the extremes in the transmission spectrum in those areas where the absorption in the films is absent (or minor), and the dispersion of the refraction coefficient is small. As a film-forming material, a mixture of MgF$_2$ and BaF$_2$ sintered in vacuum to a homogeneous composition was used.

A plate made of zinc selenide of CVD brand was used as a substrate for spraying [6]. Such a substrate practically does not absorb at wavelengths up to 14–15 microns, but above 20 microns has an absorption close to 100% [2]. Study of the optical properties of the film MgBaF$_4$ was done in the range of 1.3–16 microns.

Spectra of substrates and films were measured on Fourier spectrophotometers FSM 1201 and Bruker VERTEX-70 in the company LLC "Tidex". These spectra were used to find refractive indices and extinction coefficients, which are the real and imaginary part of the complex refraction coefficient $N=n+ik$ [6] for the substrate ($n_s$, $k_s$) and film ($n_f$, $k_f$). For this purpose, original methods based on finding the reflection spectra $R$ and transmission spectra $T$, with the correction of these spectra for absorption, in the substrate [7] and the film [8, 9] were used.
2. Results of studies of optical films with the correction for absorption. Frequency spectra or refractive and extinction coefficients for MgBaF₄ films

In Figure 1 the measured transmission spectra of \( T \) (curve 1), reflection \( R \) (curve 2) and absorption \( A \) (curve 3) of the film on a zinc selenide substrate are given. The same figure shows the transmission spectra of \( T_{\text{corr}} \) (curve 4) and reflection spectra of \( R_{\text{corr}} \) (curve 5) found for the MgBaF₄ film, adjusted for absorption by the method presented in [8]. In accordance with this method, the search for optical constants of the film was carried out in several stages. In the first stage the measurements of the reflection spectra of the \( R_s \) and transmittance \( T_s \) of the substrate. The geometric thickness of the \( h_s \) substrate was also determined. From the reflection spectra of the \( R_s \), transmittance \( T_s \) and absorption \( A_s = 1 - R_s - T_s \) of the substrate \( n_s \) and \( k_s \) were the optical constants (OC) of the substrate. The method of finding optical constants is described in [7]. Found \( n_s \) and \( k_s \) of the substrate were subsequently used for the calculation of all spectra using the program Film Analysis [8]. The program provides an introduction to the database of refractive indices and extinction coefficients of the substrate \( (n_s, k_s) \) and film \( (n_f, k_f) \), as well as film thickness \( h_f \) and substrate thickness \( h_s \).

Let us consider the method of correction of absorption spectra. The models of absorbing film and substrate are used for correction. Let us mark with \( R_0 \) and \( T_0 \) the reflection and transmission spectra of a nonabsorbing film on a nonabsorbing substrate. From the law of conservation of energy, \( R_0 \) and \( T_0 \) can be expressed through the measured values of \( R_{\text{exp}}, T_{\text{exp}}, \) and \( A_{\text{exp}} \) as follows:

\[
R_0 = R_{\text{exp}} + A'_{fs} \left( k_s, k_f, n_s, n_f, h_s, h_f \right) = R_{\text{exp}} + f_r A_{\text{exp}}; \\
T_0 = T_{\text{exp}} + A'_{fs} \left( k_s, k_f, n_s, n_f, h_s, h_f \right) = T_{\text{exp}} + f_t A_{\text{exp}},
\]

(1)

\( A'_{fs} \) and \( A'_{fs} \) is the assessment of the contribution of absorption in the spectra of reflection and transmission, which is expressed in the total absorption \( A_{\text{exp}} \) and correction functions \( f_r \) and \( f_t \).

Figure 1. The measured transmission spectra of (1), reflection (3), and absorption (5), transmission spectra (2) and reflection (4), calculated considering the correction of the spectra on the absorption.

Correction functions are defined as [2]:

\[
f_r = \frac{R_0 - R_{\text{exp}}}{A_{\text{exp}}}, \\
f_t = \frac{T_0 - T_{\text{exp}}}{A_{\text{exp}}}. 
\]

Finding \( f_r \) and \( f_t \) is an inverse problem, comparable in complexity to finding the optical constants of the film. In its direct form, it is an insoluble problem due to the ambiguity of the connection between the reflection and transmission coefficients of the film on the substrate and the optical constants of the film.

To find \( f_r \) and \( f_t \) for specific spectra, we use numerical simulation. It lies in the fact that the spectra of the film models on the substrate are calculated first. The models are constructed so that the spectra
of the film model on the substrate approximately correspond to the experimental spectra. An exact match, as shown in [8], is not required. The model includes the value of $h_f$, film thickness found from the experimental spectrum, its average refraction coefficient $<n>$, and absorption in the regions of 2–4 microns and above 10 microns. This model corresponds to the real spectra of the fluorides.

The spectra of the model used on a zinc selenide substrate are shown in figure 2. Comparison with experimental absorption spectra shows their difference, which is not fundamental.

The $R_{mod}$ reflection, $T_{mod}$ transmission and $A_{mod}$ absorption spectra of the film model on the substrate are calculated under the assumption of absorption in the substrate and film (shown in figure 2, curves 1, 2, 3).

**Figure 2.** Spectra of transmission (1), reflection (2), absorption $A$ (3), and transmission (4) and reflection (5) spectra obtained with the absorption correction for the film model on the substrate

Also, the reflection spectra of $R_{0mod}$ and transmission of $T_{0mod}$ film model on the substrate are calculated under the assumption of zero absorption.

The correction functions for the model are defined as:

$$f_{r,mod} = \frac{(R_{0mod} - R_{mod})}{A_{mod}},$$
$$f_{t,mod} = \frac{(T_{0mod} - T_{mod})}{A_{mod}}.$$

Using the obtained correction functions for the models, we carry out the correction of the experimental spectra by equations (1) using equations (2). Considering the different absorption models of films, it is possible to estimate the correction error by comparing the numerical values of the correction functions.

Finally, to find the reflection $R_{0corr}$ and transmission $T_{0corr}$ spectra corrected for absorption, we use the equations

$$R_{0corr} = R_{exp} + f_{r,mod} + A_{exp};$$
$$T_{0corr} = T_{exp} + f_{t,mod} + A_{exp}.$$

The absorption-adjusted reflection and transmission spectra of the film without absorption are shown in figure 2 (curves 4, 5). These spectra were used to determine the dependence of the refraction coefficient on the wavelength shown in figure 3.

To calculate the optical constants corrected for absorption, the methods of finding the optical constants of non-absorbing films described in [7], using the software FilmAnalisis [8], were used.

To do this, we used the step-by-step method as follows. Initially, the refraction coefficient and film thickness using the program FilmAnalisis [8] are located on the region of the spectrum initially free
from absorption. This refraction coefficient is used as the first approximation in the transition to the neighboring part of the spectrum. Thus, the whole spectrum is traversed and the dispersion of $n_f$ is found.

![Graph showing the refraction coefficient $n$ vs. wavelength $\lambda$ in microns.](image)

**Figure 3.** The dependence of the refraction coefficient on a wavelength.

The extinction coefficient was found as follows. In figure 4 the absorption spectra of the substrate with $A_{ss}$ film (curve 1), the absorption spectrum of the substrate without $A_s$ film (curve 2) and the difference of these spectra $A_{ss}-A_s$ (curve 3) are presented.

![Graph showing absorption spectra of substrate with film, substrate without film and the difference in absorption.](image)

**Figure 4.** The absorption spectra of substrate with film, of substrate without film and the difference between the values of these spectra.

Graphical analysis shows that the absorption is in the film define as

$$A_f = A_{ss} - A_s$$

In work [5] the equation connecting absorption in the $A_f$ film with the extinction coefficient $k_f$ is given. The results of the calculation of $k_f$ is shown in figure 5.

![Graph showing the extinction coefficient $k$ vs. wavelength $\lambda$ in microns.](image)

**Figure 5.** The dependence of the extinction coefficient on a wavelength.

3. Conclusions

Thus, the obtained data made it possible to calculate the dependences of the optical constants – the refractive index $n$ and the extinction coefficient $k$ on the wavelength in the range from 1 to 16 microns. MgBaF$_4$ films have absorption in the IR region of the spectrum, comparable with barium fluoride films. The maximum absorption of 3 microns due to water and water vapor in the pores did not exceed the absorption in pure foams of barium fluoride (5–6 %). However, MgBaF$_4$ films have advantages such as moisture resistance and high mechanical strength, so they can be used as film-forming materials with low refractive index in the spectrum range up to 16 microns.
References

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