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Light scattering in silicon carbide nanocrystalline films

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The possibility of light scattering investigation in nanocrystalline SiC films has been studied. The scattering type and wavelength ranges where the absorption or scattering prevails have been determined. The particle sizes calculated from scattering spectra (10 and 19 nm) agree well with SiC crystallite sizes estimated from the structure analysis data.

Изучена возможность исследования эффектов рассеяния в плёнках нанокристаллического SiC. Определен тип рассеяния и диапазоны длин волн, в которых превалирует поглощение или рассеяние. Рассчитанные по спектрам рассеяния размеры частиц (10 и 19 нм) хорошо согласуются с оценками размеров кристаллитов карбида кремния по данным структурных измерений.

The present-day close attention to nanocrystalline objects is due to the fact that properties of nanostructures are unique and rather different from those of single crystals. A special interest is paid to nanocrystal silicon carbide films obtainable by direct ionic deposition [1]. The high stability of optical properties of silicon carbide films has made it possible to develop on basis of these films an optical temperature sensor which is very promising for application in the field of high-temperature optoelectronics [2]. However, experiments with deposition of silicon carbide films with thickness exceeding 2 μm have detected an irregular reflectivity of the films in ultraviolet and visible spectral regions. As supposed in [2], the effect of reflectivity deterioration may be caused by residual stresses in the nc-SiC structures that arise at deposition of films under study onto Al_2O_3 substrates. Such stresses may result in structural inhomogeneities and preconditions for light scattering and interference curve modulation. On the other hand, silicon carbide films consist of areas with SiC crystals of nanometer dimensions. These particles cause both light absorption and light scattering, whereby contribution of scattering to extinction coefficient has been underestimated previously. For these reasons, the purpose of this paper includes an experimental measurement of scattering effect in silicon carbide films, by spectrophotometry techniques, and reveal of spectral bands where dominating is one or another type of light scattering.

The study objects were samples of silicon carbide films obtained by direct ion deposition. The influence of sapphire substrate temperature and of deposited ion energy on structure and morphology of thin films have been studied in full detail in [3, 4]. For the measurements, films of I and II series (differing in structure) were taken. Structurally, the Series I films were composed of nanocrystal polytypes of 3C-SiC, 21R-SiC and carbon, all of average size $R \sim 10$ nm. The series II films contained nanocrystal polytypes of 3C-SiC and silicon, with amorphous disordered areas.

The spectral and photometric characteristics of the samples were measured in both transmitting and reflected light using a Perkin-Elmer Lambda-35 spectrophotometer provided with an integrating sphere, in order to evaluate the diffuse radiation component. The diffuse reflection spectra (measured with respect to primary standard sample) were subsequently used in calculations of extinction factors.

The light flux attenuation in an optically inhomogeneous medium is known to be a general result of light scattering and absorption effects [5]. If dimensions of inhomogeneities (such as nanocrystallites in our case) are considerably less than the relevant wavelength and spaces between particles are rather large the light flux passing through the layer of nc-SiC film must undergo the Rayleigh scattering. Such an assumption will be quite correct if (a) diameter of particles does not exceed 1/10 wavelength of incident light and (b) the particles are spatially distributed is a disordered manner [6]. In the Rayleigh scattering effect, the diffused light intensity is proportional to λ^{-4}. As to inhomogeneities of other dimensions, quite valid is dependence $I \sim \lambda^{-p}$, where $p < 4$ is reducing with increasing dimensions of inhomogeneities [5]. In real cases, there may sometimes arise specific areas with a great number of nanocrystals which can be regarded to as macro-inclusions of a size about λ. Here, the scattering effects can be described using either Mie theory or diffraction scattering theory [5]. In general case, the optical coefficient of light attenuation corrects for both effects of light absorption and scattering. In [7], it is shown, however, that if prevailing part in light transmission through optically inhomogeneous medium is played by absorption effect, the extinction spectrum should be proportional to λ^{-1}. In the case of predominant Rayleigh scattering, the extinction spectrum is proportional to λ^{-4}. Hence, on meeting the conditions of light scattering and absorption, equation for optical density factor can be written as

$$\lg(1/T) = D_\lambda = B\lambda^{-p}, \qquad (1)$$

where $p \le 4$, and B is a parameter which depends on nanoparticle dimensions and on ε factor. Thus, for Rayleigh scattering, $B = 32\pi^4 r^3[(\varepsilon - 1)/(\varepsilon + 2)]^2 d$ [8], where r is the nanoparticle radius; ε, dielectric constant of SiC film; d – the SiC film thickness. Using equation (1), one can obtain the expression to evaluate the coefficient p:

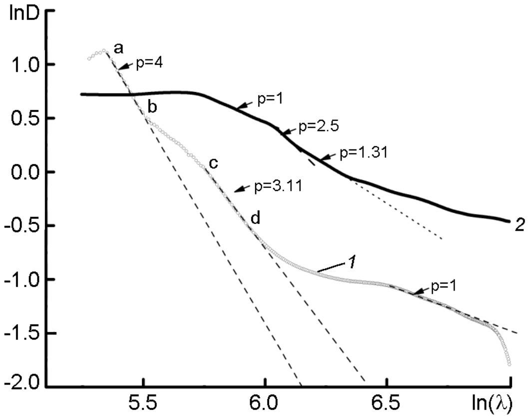

Fig. 1. Dependences of logarithm of the optical density of the SiC films on the logarithm of the wavelength of light.

$$\ln(D_\lambda) = \ln B - p\ln(\lambda). \qquad (2)$$

Linearity and value of coefficient p in $\ln D_\lambda$ and $\ln\lambda$ coordinates are the main criteria for the presence of scattering centers (nanocrystals). This fact can also be used to reveal a relationship between light absorption and scattering factors. Equation (2) makes it possible to determine not only the type of scattering itself, but a relevant wavelength ranges where the prevailing factor is either light absorption or scattering. Fig. 1 represents the $\ln(D_\lambda) = f(\ln(\lambda))$ dependence for samples of SiC films, both I and II series. For the I series sample with about 10 nm crystallites, the curve *1* is calculated using Eqs. (1, 2) under assumption of the Rayleigh scattering. The calculated plot is seen to coincide with the straightened section ($a - b$) of the experimental curve. The calculated power index $p \approx 4$. Hence, there must exist an area of extinction coefficient dependence where the Rayleigh scattering effect takes place. The second straightened section of experimental curve corresponds to particle size (about 19 nm) and to somewhat modified value of (p), such as ≈ 3.11. The calculated particle sizes (10 nm and 19 nm) are in good agreement with SiC crystallite size estimated from the structure measurements. As to the long-wave range ($p \approx 1$), it is the absorption factor can be assume to prevail.

The second curve indicates presence of absorption ($p \approx 1$) and absence of scattering.

In visible and ultra-violet spectral regions, an unusual character of reflection spectra was also observed. Thus, within spectral region below 400 nm, a dramatic drop of reflection coefficient is observed for silicon carbide films. Such a drop is explained in [6] as a result of light scattering, so as the shorter are the wavelengths, the greater is scattering effect that results eventually in decreased reflection.

Thus, application of modern spectroscopy techniques using an integrating sphere to assess the diffuse component of radiation provides a further study of light scattering effects. Using mutually complementing methods of diffuse reflection and transmission, one can determine the spectral regions where one or another type of light scattering predominates. This methodology can be applied to identify the scattering types and to reveal interrelation between absorption and scattering in complex nanocrystalline silicon carbide films.

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Розсіювання світла нанокристалічними плівками карбіду кремнію

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Вивчено можливість спектрофотометричного дослідження ефектів розсіювання світла у плівках нанокристалічного SiC. Визначено тип розсіювання та діапазони довжин хвиль, де переважає поглинання або розсіяння. Розраховані розміри часток (10 та 19 нм) добре узгоджуються з оцінками розмірів кристалітів SiC за даними структурних вимірювань.