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MATHEMATICAL MODELING OF SPECTRAL CHARACTERISTICS OF OPTICAL COATINGS WITH SLIGHTLY INHOMOGENEOUS CHALCOGENIDE FILMS

A. Mitsa, V. Mitsa*, A. Ugrin

Uzhgorod National University, 54 Voloshyn Str., Uzhgorod 88000, Ukraine

The existence of high-refractive regions with inhomogeneous distribution of the refractive index over the depth at the interfaces of film – air (a near-surface region) and film – substrate (a transition region) for oxygen free non-crystalline films has been experimentally established. A structural model for these films has been proposed. The influence of transition and near – surface regions on spectral characteristics of a single quarter-wave layer and cut-off and narrow-band filters has been studied.

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1. Introduction

While synthesizing optical interference structures it is considered, as a rule, that the interfaces of film – substrate and film – film are sharp [1]. That is why the inhomogeneities at the interfaces of film – substrate are objectionable as they make the control of a geometric depth of the films being prepared difficult [1]. The necessity of optical coatings in new materials and wave-guide films has determined research of the refractive index and concentrational profile of components of oxygen – free films and oxygen materials and design of spectral characteristics of partially inhomogeneous films [2 – 8]. The aim of the present work is to study the distribution of elements over the depth of $(As_2S_3)_{1-x}(GeS_2)_x$ non-crystalline films, building the simplest model of the refractive index profile of the films based on a glassy (g) g-GeS_2 and modeling spectral characteristics of interference structures with a high- refractive layer based on $(GeS_2)_x(As_2S_3)_{1-x}$ glasses. The wavelengths of the most widely spread lasers or IR diodes – 480, 630, 750, 1000, 3000 nm are taken as operating wavelengths.

2. Experimental methods

SIMS profile was studied by using the device Cameca IMS 4 F. Taking into account etching rates of films by cesium ions, their depths, the time dependence of the intensity of As and Ge deposited ions was transformed into the depth dependence. Besides, the intensities of As and Ge distribution were normalized to sulphur intensity.

The data on the refractive index distribution profile of films were obtained by multi-angular ellipsometry using a laser ellipsometer in accordance with the methods described in [9]. The films were prepared by vacuum evaporation at the installation VUP – 5 к (Ukraine) at a temperature of the evaporator equal to 1225 K. A geometric depth of films was defined by the method Abélès.

3. Results

In Fig. 1 SIMS profile of elements distribution in $(GeS_2)_x$ $(As_2S_3)_{1-x}$ films is shown. An increased content of As and Ge with respect to the central part of the film is observed at the interface film – substrate. The diffusion of substrate material into a near-surface region is also observed. For increasing content of GeS_2 in films the size of the transition region increases from ~ 25 (x=0.3) to 30

* Corresponding author: mitsa@ univ.uzhgorod.ua

(x=0.7) nm. The dependences of Ge and As intensity at the interface may be extrapolated by using the relation I ~ ln(x).

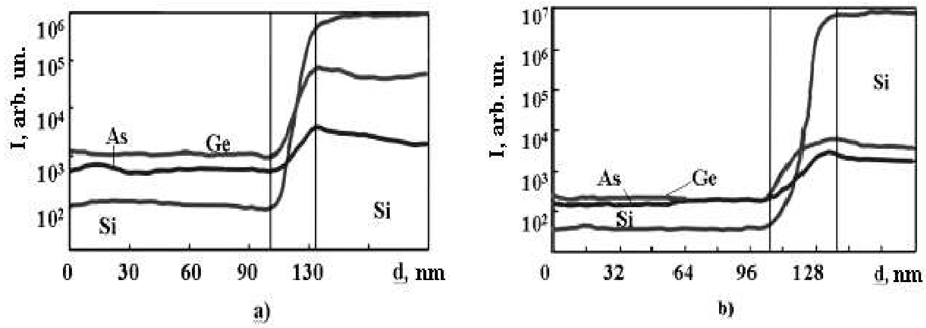

Fig. 1. SIMS profiles of $(GeS_2)_x(As_2S_3)_{1-x}$ films: a) x = 0,3; b) x = 0,7.

By solving the inverse ellipsometric problem based on multi-angle ellipsometry data at λ_0=632.8 nm one gets the following values of the size of regions and averaged refractive indices different from the central part of the $(GeS_2)_x(As_2S_3)_{1-x}$ films : 1) a near – surface region n_v=2.4–2.6, d_v=2.5–3 nm; 2) the central part of the film n_f=2.05–2.2, d_f= 90–100 nm; 3) a transition region of film – substrate n_p=2.3–2.45, d_p=30–40 nm. The latter is well agreed with the dimensions of a transition region obtained on the basis of studying SIMS-profile of films (Fig. 1) and structural study of these films given in [10]. The refractive indices of the central part of the films well agree with the refractive indices of respective bulk glasses and films [11,12]. E.g. refractive index (n) of film $Ge_{40}S_{60}$ is n= 2.60 at λ_0=632.8 nm. Proceeding from the results of ellipsometric studies the simplest profile of the refractive index of type a-GeS_2, the vacuum evaporated film may be presented as first step in the form of a three-layer layered-inhomogeneous film (Fig. 2). Taking into consideration that the concentration of arsenic and germanium with respect to sulphur in a transition region varies as ~ ln(x), one may admit that the increase in the refractive index in a transition region could follow such an approximation. To model the spectral characteristics of partially inhomogeneous films we proposed other variants of the variation in the refractive index in near-surface and transition regions.

Based on the method of multi-angular ellipsometric sudies at λ_0=632.8 nm a simplest layered-inhomogeneous model of the refractive index structure was proposed by us (Fig. 2). It includes near-surface, central and transition regions. We'll consider the given model both with a near-surface region and without it. The aim of this is firstly to study the influence of a transition region refractive index and that in a near-surface region on spectral characteristics of a single quarter-wave layer and interference filters at different operating wavelengths. Here, the possibility of absorption in Ge-rich sublayers was not taken into account.

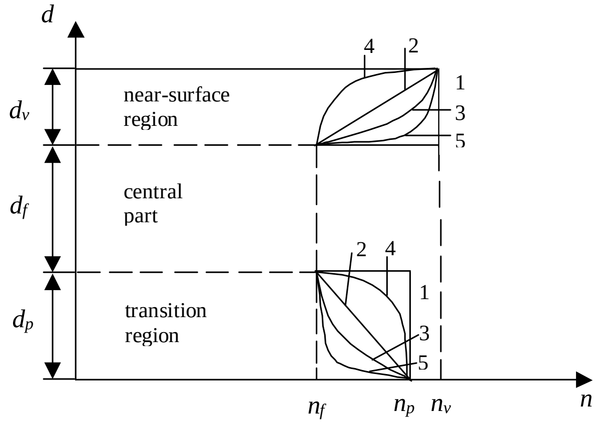

Fig. 2. The model of partially inhomogeneous film (the refractive index distributions: 1 - stepwise; 2 – linear; 3 – quadratic; 4 – logarithmic; 5 – exponential).

4. The construction of a mathematical model

After a thorough experiment it has been revealed that a geometric depth of a transition region may reach 30 nm and a near-surface one – 5 nm. The experimentally determined refractive indices of the central part of the film, a transition and near-surface regions at different operating wavelengths are given in Table 1.

Table 1. The values of refractive indices of the central part of the film, a transition and near-surface layers for different operating wavelengths.

№	λ_0, nm	n_f	n_p	n_v
1	480	2.1	2.6	2.6
2	630	2.05	2.5	2.5
3	750	2.05	2.45	2.45
4	1000	2.00	2.4	2.4
5	3000	2.00	2.4	2.4

Presetting the inhomogeneity of the refractive index with distribution of *n(z)* over the depth in a transition layer is done as follows. A transition layer is divided into m_t equal zones as to their depth and the value of the refractive index depending on the distribution type is equal to the value given in Table 2. The inhomogeneity in a near-surface layer is preset similarly.

Table 2. The refractive index value of *j*-zone of a near-surface and transition regions depending on the distribution.

Distributions	The refractive index of j-zone of a transition region, n_{pj}	The refractive index of j-zone of a near-surface region, n_{vj}
Stepwise	n_p	n_v
Linear	$n_f+\frac{(n_p-n_f)}{m_p-1}\cdot(j-1)$	$n_v-\frac{(n_v-n_f)}{m_v-1}\cdot(j-1)$
Quadratic	$n_f+\frac{(n_p-n_f)}{(m_p-1)^2}\cdot(j-1)^2$	$n_v-\frac{(n_v-n_f)}{(m_v-1)^2}\cdot(j-1)^2$
Logarithmic	$n_f+\frac{(n_p-n_f)}{\ln(m_p)}\cdot\ln(j)$	$n_v-\frac{(n_v-n_f)}{\ln(m_v)}\cdot\ln(j)$
Exponential	$n_f+\frac{(n_p-n_f)}{e^{mp-1}-1}\cdot(e^{j-1}-1)$	$n_v-\frac{(n_v-n_f)}{e^{mv-1}-1}\cdot(e^{j-1}-1)$

The value of the refractive index in a transition region, *n(z),* will vary from n_f to n_p and in a near-surface – from n_v to n_f.

The geometric depth of the central part of the film (d_f) in this case is:

$$d_f(d_p,d_v)=\frac{1}{n_f}\left(\frac{\lambda_0}{k}-d_p\cdot n_{sp}-d_v\cdot n_{sv}\right),\text{ where } n_{sp}=\frac{1}{m_p}\sum_{j=1}^{mp}n_{pj}\ ,\ \ n_{sv}=\frac{1}{m_v}\sum_{j=1}^{mv}n_{vj}\ . \quad (1)$$

The variables n_{sp} **i** n_{sv} are the average values of the refractive index in a transition and a near-surface layer, respectively, m_p and m_v, are the number of partitions of a transition and near-surface layer, respectively; *k*=2, when a half-wave layer is considered and *k*=4, when a quarter-wave layer is considered; n_f is the refractive index of the central part of the film; n_{pj} **i** n_{vj} are the refractive indices of j-zone when partitioning a transition and near-surface region, respectively

(Table 1), λ_0 is an operating wavelength. For unambiguous understanding of formula (1) we'll consider k=2.

Let us model the influence of a transition and near-surface layer in a high-refractive component on spectral characteristics of the structures with the help of Abélès matrix method [1].

By choosing the refractive index n, a geometric depth of the layer d and the wavelength λ, as parameters, one may write a characteristic matrix of one layer like this [2-6]:

$$M_s(n,d,\lambda)=\begin{Vmatrix}\cos\delta(n,d,\lambda) & -\frac{i}{p}\sin\delta(n,d,\lambda)\\ -ip\sin\delta(n,d,\lambda) & \cos\delta(n,d,\lambda)\end{Vmatrix},$$

where $i=\sqrt{-1}$ and θ is the angle between a beam and perpendicular to the area of incidence.

The value $p=n\cos\theta$ for TE wave (s-polarization) and $p=\dfrac{n}{\cos\theta}$ for **TM** wave (***p***-polarization). We'll consider that the angle of incidence coincides with the perpendicular to the area of incidence ($\theta\approx 0$) and, respectively, $p=n$.

The characteristic matrices of a transition and near-surface layer will be equal to:

$$M_p(\lambda)=\prod_{j=0}^{mp-1}M_s(n_{pj},d_p,\lambda), \quad (2)$$

$$M_v(\lambda)=\prod_{j=0}^{mv-1}M_s(n_{vj},d_v,\lambda). \quad (3)$$

So, if one takes into account the availability of a near-surface and transition regions, then by using the formulas (1)-(3) one may put down a characteristic matrix of the given inhomogeneous structure [7]:

$$M(\lambda)=M_v(\lambda)\cdot M_s(n_f,d_f(d_p,d_v),\lambda)\cdot M_p(\lambda),$$

where n_f is the refractive index of the central part of the film, which is equal to the product of characteristic matrices of a near-surface layer, central part and a transition layer.

Let us consider multi-layered structures. The first to be considered is an elementary cut-off filter of the construction S-HLH..HLH.

We'll admit that a high-refractive layer comprises a transition and near-surface region. Then its characteristic matrix will be equal to:

$$M_H(\lambda)=M_v(\lambda)\cdot M_s(n_H,d_f(d_p,d_v),\lambda)\cdot M_p(\lambda), \quad (4)$$

where n_B is the refractive index of the central part of a high-refractive layer.

A low-refractive layer will have a characteristic matrix equal to:

$$M_L(\lambda)=M_s(n_L,d_f(d_p,d_v),\lambda), \quad (5)$$

where n_H is the refractive index of a low-refractive layer.

Taking this into account we can put down a characteristic matrix of (2k+1) - layered structure of type S-HLH..HLH:

$$M(\lambda)=\prod_{i=1}^{k}(M_H(\lambda)\cdot M_L(\lambda))\cdot M_H(\lambda). \quad (6)$$

Let us consider a narrow-band filter with the construction S-HL..2H..LH. If one considers that a high-refractive layer comprises a transition and a near-surface region, then its characteristic matrix is defined by the formula (4), and that of a high-refractive layer – by formula (5). The characteristic matrix of (4k+1) - layered structure of type S-HLH..2H..HLH may be set by the formula:

$$M(\lambda)=\prod_{i=1}^{k}(M_H(\lambda)\cdot M_L(\lambda))\cdot M_s\left(n_H,\frac{1}{n_H}\left(\frac{\lambda_0}{2}-d_p\cdot n_{sp}-d_v\cdot n_{sv}\right),\lambda\right)\cdot\prod_{i=1}^{k}(M_L(\lambda)\cdot M_H(\lambda)). \quad (7)$$

Knowing the characteristic matrix of the whole structure, we can easily find the transmission coefficient:

$$T(\lambda)=4/\left(2+\frac{n_0}{n_s}M_{11}^2(\lambda)+\frac{n_s}{n_0}M_{22}^2(\lambda)+n_0n_sM_{12}^2(\lambda)+\frac{1}{n_0n_s}M_{21}^2(\lambda)\right), \quad (8)$$

where n_0, n_s are the refractive indices of environment and substrate, respectively.

Taking into account the above theoretical data let us study how a transition, and near-surface will influence the spectral characteristics of a single-layered quarter-wave structure at different operating wavelengths. For such calculations the corresponding software was developed.

5. The results of the mathematical modeling

A model experiment has shown that in a single quarter-wave layer the transmission coefficient in the presence of a transition and near-surface regions increases (Figs. 3-4). A stepwise distribution of the refractive index influences most of the spectral characteristics. One gets logarithmic, linear, quadratic and exponential distributions. This affirmation is valid for all the considered wavelengths both in the presence of a transition region and in that of transition and near-surface regions.

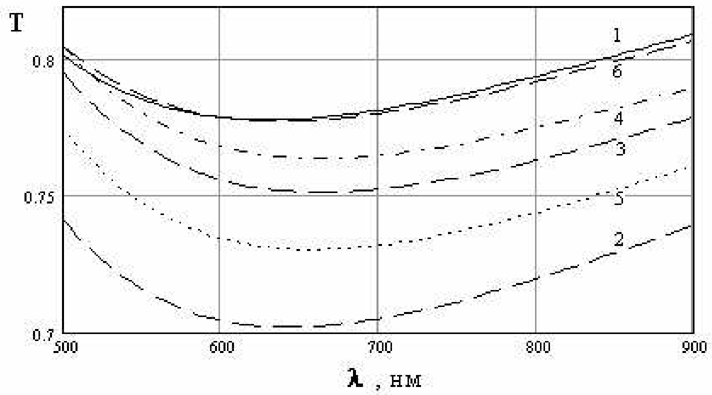

Fig. 3. Spectral characteristics of a single quarter-wave layer (λ_0=630 nm) in an ideal case and in the presence of a transition region with different distributions of the refractive index: 1 – an ideal case; 2 – with a stepwise distribution of the refractive index; 3 – with a linear distribution of the refractive index; 4 – with a quadratic distribution of the refractive index; 5 – with a logarithmic distribution of the refractive index; 6 – with an exponential distribution of the refractive index.

By calculation methods one gets the numerical characteristics related to the influence of near-surface and transition regions on spectral characteristics of the structure.

In the presence of transition and near-surface regions with a stepwise distribution of the refractive index the transmission coefficient at an operating wavelength λ_0=480 nm decreases for ΔT = 0.125, at λ_0 = 630 nm for ΔT = 0.076, at λ_0 = 750 nm for ΔT = 0.051, at λ_0=1000 nm for $\Delta T = 0.031$ and at λ_0=3000 nm for ΔT = 0.005. So, with the increase of the operating wavelength the value of the transmission coefficient in the minimum point differs less from the ideal case.

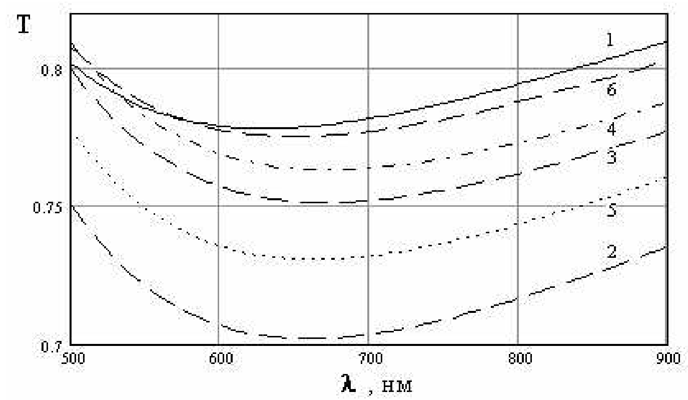

Fig. 4. Spectral characteristics of a single quarter-wave layer (λ_0=630 nm) in an ideal case and in the presence of a near-surface and transition regions with different distributions of the refractive index: 1 – an ideal case; 2 – with a stepwise distribution of the refractive index; 3 – with a linear distribution of the refractive index; 4 – with a qudratic distribution of the refractive index; 5 – with a logarithmic distribution of the refractive index; 6 – with an exponential distribution of the refractive index.

Let us consider the largest possible shifts of the minimum point for a quarter-wave single layer at different operating wavelengths. At an operating wavelength λ_0=480 nm the largest shift performs a quadratic distribution for $\Delta\lambda = 35$ nm, at λ_0=630 nm, quadratic for $\Delta\lambda = 43$ nm, at λ_0=750 nm, linear for $\Delta\lambda$=46 nm, at λ_0= 1000 nm, stepwise for $\Delta\lambda = 79$ nm and at λ_0=3000 nm, stepwise for $\Delta\lambda = 163$ nm. So, with increasing an operating wavelength in the presence of a transition and near-surface regions the minimum point shifts to the region of long waves.

The fact that a near-surface layer does not practically influence the transmission coefficient in the minimum point and realizes only shifting to the region of long waves has appeared to be very interesting.

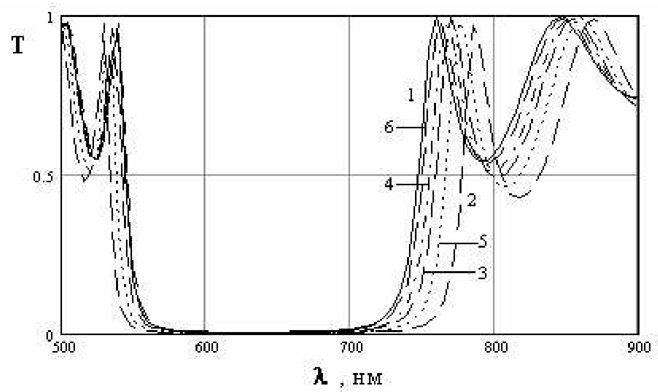

Fig. 5 Spectral characteristics of 1 17-layered structure S-HLH..HLH (λ_0=630 nm) in an ideal case and in the presence of a near-surface and transition regions with different distributions of the refractive index: 1 – an ideal case; 2 – with a stepwise distribution of the refractive index; 3 – with a linear distribution of the refractive index; 4 – with a qudratic distribution of the refractive index; 5 – with a logarithmic distribution of the refractive index; 6 – with an exponential distribution of the refractive index.

The influence of a near-surface and transition regions on spectral characteristics in different constructions may be studied by considering a 17-layered cut-off filter S-HLH..HLH (Fig.5) and a 17-layered narrow-band filter S-HLH..2H..HLH (Fig. 6).

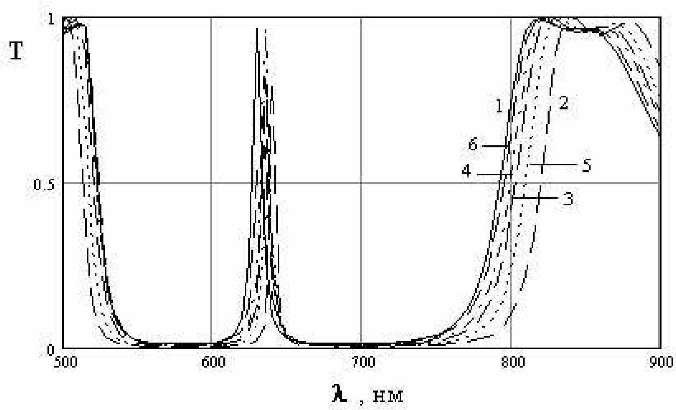

Fig. 6. Spectral characteristics of a 17-layered structure S-HLH..2H..HLH (λ_0=630 nm) in an ideal case and in the presence of a near-surface and transition region with different distributions of the refractive index: 1 – an ideal case; 2 – with a stepwise distribution of the refractive index; 3 – with a linear distribution of the refractive inde; 4 – with a quadratic distribution of the refractive index; 5 – with a logarithmic distribution of the refractive index; 6 – with an exponential distribution of the refractive index.

The studies have shown that the availability of a near-surface and transition regions with different types of the refractive index distributions results in the increase of the reflection range (Fig. 5). Again, because a stepwise distribution of the refractive index influences the widening of a spectral range, then – a logarithmic, linear, quadratic and exponential dependences are produced. It should be noted that at a stepwise distribution of the refractive index in a near-surface and transition regions and at an operating wavelength λ_0=630 nm the reflection range (T<0.1) increases for 46.39 nm (Fig. 5, curve 2). When the operational wavelength increases the difference of spectral range width decreases at different distributions of the refractive index, and its shift into the region of long waves increases.

A similar fact is observed while studying a 17-layered narrow-band filter S-HLH..2H..HLH (Fig. 6). The types of distributions influence the spectral characteristics in the same sequence as in the previous case. Again, the reflection range increases and the shift into the region of long waves takes place. The halfwidth of a spectral transmission range decreases and the maximum increases and shifts towards higher wavelengths (Table 5).

If a near-surface and transition regions with a stepwise distribution of the refractive index exist, then at an operational wavelength λ_0=630 nm the transmission in the maximum point increases from 0.959 to 0.99, the value of the wavelength in the maximum shifts from that selected in an ideal case at 630 nm to 640.695 nm, and the halfwidth of transmission decreases from 6.164 to 2.931 nm (Table 3).

Table 3 The influence of a partially high-refractive layer on spectral characteristics of a 17-layer narrow-band filter S-HLH..2H..HLH.

Distributions	Operating wavelength shift $\Delta\lambda$, nm	Transmission coefficient, T_{max}	Halfwidth of a spectral transmission range (nm)
Ideal	0	0.959	6.164
Stepwise	10.695	0.99	2.931
Linear	6.048	0.972	4.792
Quadratic	5.030	0.966	5.409
Logarithmic	7.300	0.982	3.874
Exponential	2.676	0.957	6.033

The increase in the operating wavelength when studying the same narrow-band filter S-HLH..2H..HLH results in the situation similar to that with a cut-off filter S-HLH..HLH.

Let us consider the case when the number of layers in filters S-HLH..HLH and S-HLH..2H..HLH increases at the operational wavelength λ_0=630 nm. Thus, the availability of a transition and near-surface regions with a stepwise distribution of the refractive index in a high-refractive component for a 25-layered cut-off filter S-HLH..HLH results in the increase of the reflection range for 43.214 nm with regard to an ideal case, and for a 25-layered narrow-band S-HLH..2H..HLH the halfwidth of the transmission decreases from 1.121 to 0.365 nm. As it is seen these deviations are smaller than in the case with 17-layered structures.

Further increase in the layers also decreases the difference between an ideal case of sharp interfaces between films and in the presence of a transition and near-surface regions in a high-refractive component. This phenomenon is observed at all possible operational wavelengths.

6. Conclusions

The types of refractive index distributions of a near-surface and transition regions influence the spectral characteristics in the order: stepwise, logarithmic, linear, quadratic and exponential ones. They cause the decrease in the transmission of a quarter-wave layer and shift the minimum point to the region of long waves. For cut-off filters the reflection range increases if the above regions exist. For narrow-band filters the reflection range increases and the halfwidth of a spectral transmission range of the filter decreases.

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