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THE REFLECTIVE PROPERTIES OF BROADBAND OPTICAL MULTILAYERS*

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Binary multilayers with a certain kind of disorder may exhibit high reflection range over a very wide bandwidth. The dependencies of this bandwidth on the center wavelength, random layers sequence, refractive contrast of the coating materials, layer thickness errors and the angle of incidence have been studied numerically. Our simulations have shown that broadband reflectors can be designed for various applications. The reflective curves for staggered multilayers are also computed. The results show that the broadening effect of irregular multilayers is dominated by its degree of disorder. A broadband TiO_2-SiO_2 multilayer is fabricated. The measured width of the transmittance less than 1% for this multilayer covers 400 nm, from 700 to 1100 nm, which is just the tuning range of T_i: sapphire lasers.

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I. INTRODUCTION

Broad band optical reflectors are extensively demanded in tunable lasers and optical equipment. The high reflection range (HRR) of standard $\lambda/4$ stacks is limited to within the band gap and hence is rather narrow. For example a commercial femtosecond Ti:sapphire laser operating in the spectrum of 700–1100 nm requires more than three sets of ordinary dielectric reflectors of different center wavelengths to cover the tuning range. In addition, there are about ten mirrors in each set of reflectors. So it is very inconvenient to change the operating wavelength. Although metallic films possess a wider reflection range, their reflectance and the damage threshold are low. There have been various methods, suggested or demonstrated, to extend the reflection bandwidth, and the automatic design program has been also developed.[1,2] However, for most commercial tunable lasers the standard $\lambda/4$ stacks are still used. In the past few years the effect of light localization in disordered systems has induced considerable interest. Both theoretical[3−10] and experimental [11] studies have indicated that disorder can cause light localization in a one-dimensional system. The

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disorder of a multilayer can be achieved by randomly varying either the layer thickness or the layer refractive index. One of the major features of disordered multilayers is that its HRR is no longer restricted. Based on this effect, binary disordered multilayer reflectors with wide HRR have been proposed[12] and recently such a broadband reflector has been experimentally demonstrated.[13] To see if this kind of multilayer can be practically used as laser mirrors, we investigate numerically its reflective properties, mainly the dependences of the width and reflectance of HRR on the layer sequence, center wavelength, thickness errors, reflective contrast and incident angle, by the transfer matrix method. Then a high reflection mirror is designed and fabricated, that is especially suitable for tunable Ti:sapphire laser. The measurement of the mirror shows that its transmission may be lower than 1% in the wavelength range of 700–1000 nm, *i.e.*, the whole tuning range of a Ti:sapphire laser.

II. DESIGN OF BROADBAND DISORDERED MULTILAYER

The aim of our design is to obtain a multilayer with wide HRR and without any transmission dip in the HRR.

In the present case the disorder is caused by the change of the layer thickness. The layer thicknesses of a disordered multilayer are determined according to the degree of disorder D(defined as the mean square deviation of the random layer thicknesses from the average value), *i.e.*,

$$D = \frac{1}{H+L}\left[\frac{1}{m}\sum_{i=1}^{m}\left((H_i - H)^2 + (L_i - L)^2\right)\right]^{1/2},$$

where H_i and L_i are the real thicknesses of the i-th high and low refractive layers, respectively, and H and L are their average thicknesses, m the number of periods, λ_0 the center wavelength of reflection and θ the angle of incidence. To produce an optical reflector, the average layer thickness of each material is required to be $\lambda/4n$, where n is the corresponding refractive index.

The design procedure of broadband disordered multilayer reflector is briefly described as follows. First, based on the requirements of the reflectance, high reflection width and center reflection wavelength, we select the degree of multilayer disorder D, the number of multilayer periods m, and the coating materials. Roughly speaking, a large D will result in a broad reflection range and, at the same time, low reflectance in this range, whilst more layer periods and large reflective contrast of coating materials will be favorable for a broader high reflection range. Secondly, after D, m and the coating materials are selected, the thicknesses of all the layers of the two materials may be obtained by a computer program, in which the average thicknesses of both materials should equal $\lambda/4n$ values, respectively, whilst the probability of each layer thickness used lies on a Gaussian curve with respect to the thickness. Of course, there are many possible layer thickness combination $\{H_i, L_i\}_1, \{H_i, L_i,\}_2, \cdots, i = 1$ to m , that simultaneously satisfy the selected D and m, but we have a third criterion that must be satisfied. The sequence of the layer thickness should be $H_1, H_2, \cdots, H_m$ with $H_1 < H_2 < \cdots < H_m$ or $H_1 > H_2 > \cdots > H_m$ and $L_1, L_2, \cdots, L_m$ with $L_1 < L_2 < \cdots < L_m$ or

$L_1 > L_2 > \cdots > L_m$, respectively. To get the widest bandwidth and high enough reflectance in this range we need to find out which is the best combination among $\{H_i, L_i\}_1, \{H_i, L_i, \}_2, \cdots$, by using the transfer matrix calculation, though the difference between the corresponding reflective curves is generally small. This is the third step. Following the above procedure, we finally obtain the thicknesses and sequence of all the layers.

One can see that in our design the degree of disorder D and the average layer thickness of $\lambda/4n$ are two key parameters. In principal, based on these two parameters the disordered multilayer may be worked out. This is quite different from the previous design methods stated in Refs.[1] and [2].

As an example, the designed thicknesses and sequence of a typical single disordered TiO_2-SiO_2 multilayer with D=0.15, m=30 and λ_0= 880 nm are given in Table 1, where only the thicknesses of TiO_2 layers are changed and those of all SiO_2 layers are kept identical.

Table 1. The designed thicknesses H_i (in units of nm) and sequence i of the TiO_2 layers for a binary TiO_2-SiO_2 disordered multilayer with D=0.15 and 30 periods. All the SiO_2 layers have the same thickness of $L = \lambda_0/4n_s$=151.7nm, where λ_0=880 nm. The first TiO_2 layer (i=1) is in direct contact with the optical grass substrate.

i	1	2	3	4	5	6	7	8	9	10
H_i	17.6	35.2	43.6	45	52.1	56.3	66.4	68.7	69.7	70.6
i	11	12	13	14	15	16	17	18	19	20
H_i	73	86	90.1	92.1	94	97.4	99.3	101	105	113
i	21	22	23	24	25	26	27	28	29	30
H_i	120	122	123	130	135	143	146	148	152	174

III. MAIN REFLECTIVE FEATURES OF A DISORDERED MULTILAYER

In this section we give some major features of single-disordered multilayers, where the TiO_2 layers are varied around an average value of $H = \lambda_0/4n_t$, and a constant SiO_2 layer thickness of $L = \lambda_0/4n_s$ is used.

The HRR for disordered multilayers may be easily shifted in the spectrum by merely changing the center wavelength λ_0. In Fig.1, curve *a* shows the reflection spectrum for a TiO_2-SiO_2(T-S) multilayer with D=0.15, m=30, $\theta = 0$ and λ_0=530 nm. Its HRR is as wide as the whole visible spectrum of 400–700 nm. Curve *b* is for a T-S multilayer with the same parameters as the previous one, except that $\lambda_0 = 880$ nm. The HRR is now located between 665–1175 nm and the bandwidth of R>99.5% is 470 nm (~680–1150 nm). It is wide enough to cover the tuning range of the commercial Ti:sapphire lasers, 700–1100 nm. Figure 2 gives the details of the above curve *b* for the range of R>99%. The corresponding spectrum resolution in the transfer matrix calculation is taken to be 0.1 nm.

The sequence of the random layers is a crucial factor for getting a broadband reflector without any transmitted peak in the HRR. Three reflection curves shown in Fig.3 come from three T-S multilayers. They have the same D=0.15, m=30, $\theta = 0$ and λ_0=880 nm, but different sequences of the random TiO_2 layers, while all SiO_2 layers have the same

thickness of $\lambda_0/4n_s$. For curve a (solid line), the sequence of TiO_2 layers is $H_1, H_2, \cdots, H_{30}$ with $H_1 < H_2 < \cdots < H_{30}$, where H_i is the thickness of the i-th TiO_2 layer. In this case

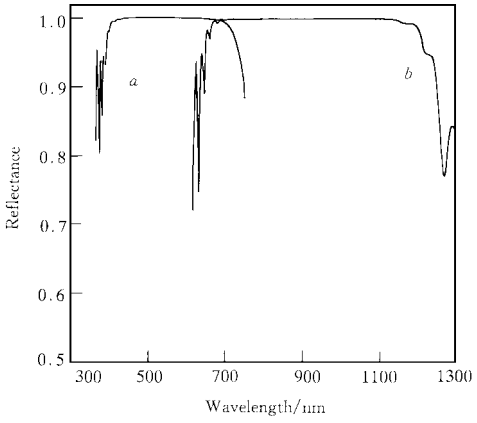

Fig.1. The reflection spectra of TiO_2-SiO_2 disordered multilayers with $\lambda_0 = 530$ nm (curve a) and 880 nm (curve b).

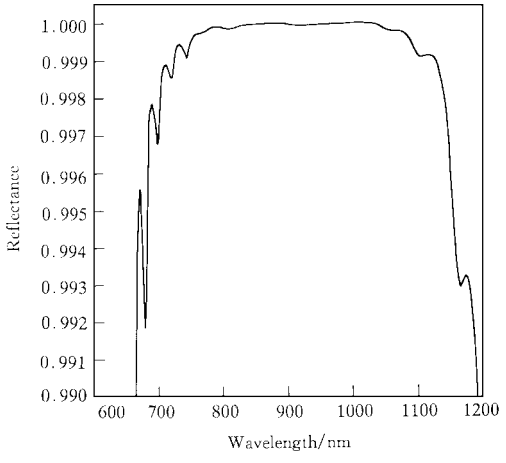

Fig.2. The high reflection range of $R > 99\%$ for a TiO_2-SiO_2 disordered multilayer with $\lambda_0 = 880$ nm.

there are no transmitted peaks in the whole HRR. However, if this sequence is broken,the transmitted peaks in the whole HRR. However, if this sequence is broken,the transmitted peaks occur, as indicated by curve b (dotted line), where the sequence is $H_1, H_{30}, H_2, H_{29}, \cdots, H_{15}, H_{16}$, and curve c (dashed line), where the sequence is $H_1, \cdots, H_5, H_{26}, \cdots, H_{30}, \cdots, H_{11}, \cdots, H_{15}, H_{16}, \cdots, H_{20}$. We have also calculated the reflection spectra for the disordered multilayers with different values of D, m and λ_0 and found that a broadband reflector

can be obtained, when a moderate D is taken and the random layers are arranged in progressive order of their thicknesses, even though the maximum thickness difference between layers may be as big as a factor of ten.

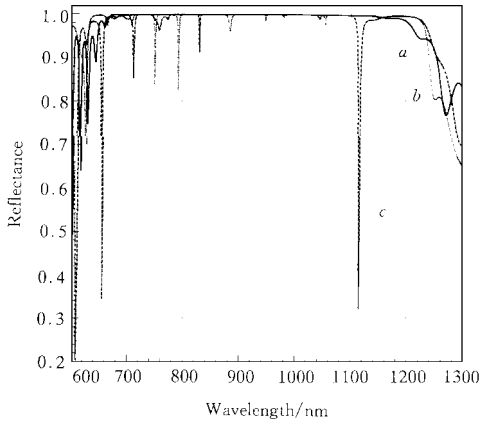

Fig.3. The reflection spectra of TiO_2-SiO_2 disordered multilayers with D=0.15, m=30, λ_0 = 880 nm as a function of different thickness sequences of the TiO_2 layers.

The HRR and the reflectance of a disordered multilayer depend strongly on the refractive contrast of the two coating materials. The reflection spectra for multilayers with the same D=0.15, m=30, θ=0 and λ_0= 1000 nm, but made of different coating materials: TiO_2-SiO_2; ZnS-MgF_2(Z-M) and antimonite-cryolite(A-C), have been computed. Their

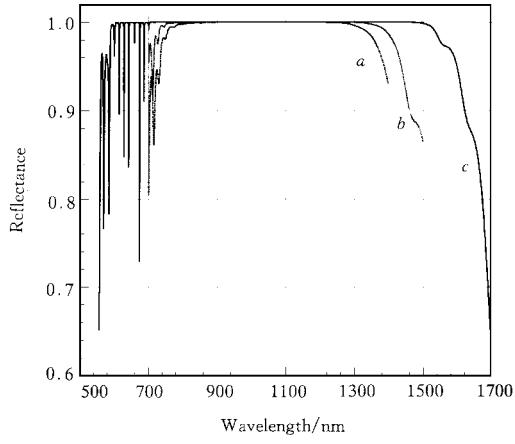

Fig.4. The dependence of high reflection bandwidth on the refractive contrast of the coating materials. Carve a :TiO_2-SiO_2; b : ZnS-MgF_2; c : antimonite-cryolite.

refractive contrasts are 1.57; 1.70 and 2.1, respectively. Both the HRR and reflectance

increase obviously with the refractive contrast, as in Fig.4. The widths of R >90% for T-S, Z-M and A-C are, respectively, 580, 661 and 835 nm. The corresponding widths of R >99.5% are 530, 625 and 822 nm.

In practices, errors in the layer thickness are unavoidable. It is therefore necessary to know how these changes alter the reflectance and the HRR of a disordered multilayer. We assume that the thickness errors are random and statistically independent with a maximum value δ, *i.e.*, the errors are chaotically taken between $+\delta$ and $-\delta$. The errors are then randomly added to the designed layer thicknesses for both high and low refractive materials and the corresponding reflection spectrum is calculated. In Fig.5 curves a, b, c and d are the reflection spectra for the T-S multilayers with the same D=0.15, m=30, $\theta = 0, \lambda_0 = 880$ nm, but different δ=0, 5, 10 and 15 nm, respectively. Curves a, b and c almost overlap in the HRR of 700–1150 nm. This result means that the tolerance in layer thickness for an actually disordered multilayer is around 10 nm, which is not a problem in conventional coating procedures.

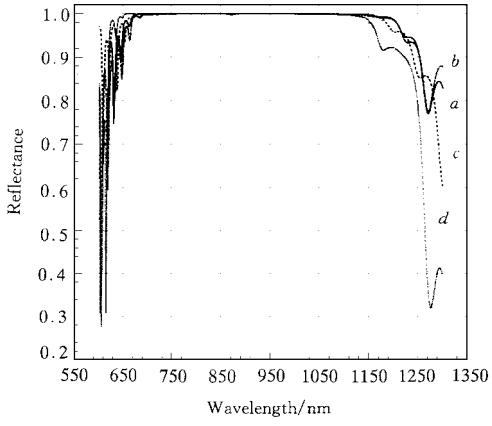

Fig.5. Variation of reflection spectrum as a function of errors in layer thickness. Curves a, b, c and d correspond to maximum error δ=0, 5, 10 and 15 nm, respectively.

The disordered multilayers may be used under oblique incidence too. The HRR for s-waves will shift slightly towards the short wavelength direction and at the same time become wider, as the angle of incidence increases. Figure 6 gives the reflection spectra of a TiO_2-SiO_2 binary multilayer with D=0.15, m=30 and $\lambda_0 = 880$ nm for five angles of incidence. The five curves from right to left correspond to the angles of incidence θ =0, 10, ,20, 30 and 40 degrees. Their center reflection wavelengths are around 880, 877, 865, 848 and 820 nm, and their bandwidths for $R > 99.5\%$ are 470, 471, 507, 515 and 530 nm, respectively.

For double-disorder multilayers, where both the TiO_2 and SiO_2 layers are changed around their average values $\lambda_0/4n$, respectively, the reflective properties are similar to that of a single-disordered multilayer, when an appropriate D is taken.

The above simulations give a clear evidence that these disordered multilayers may

possess a rather wide HRR in a certain wavelength range and may be used under different conditions.

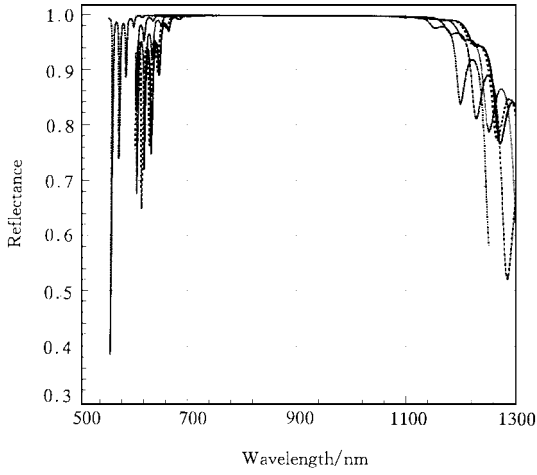

Fig.6. Variation of reflection spectrum as a function of angle of incidence. The curves from right to left correspond to θ=0, 10, 20, 30 and 40 degrees, respectively.

IV. COMPARISON BETWEEN DISORDERED AND STAGGERED MULTILAYERS

The staggered multilayer was suggested as one of the broadband reflection mirrors, where the layer thickness was staggered in such a way as to form an arithmetic or a geometric progression. This is equivalent to gradually varying the center reflection wavelength of a multilayer.[14,15] At that time, because of less layers and limited thickness difference used, a series of transmitted dips might exist in the reflection curve. Now we increase both the number of layers and the thickness difference between the thickest and thinnest layer of staggered multilayers. To compare with our disordered multilayers the staggered TiO_2-SiO_2 multilayers with thirty periods are designed as follows. All the SiO_2 layers have the same thickness of $L = \lambda_0/4n_s$, while the thicknesses of the TiO_2 layers vary according to $H_i = (\lambda_0 + q_i\Delta)/4n_t$, where $\lambda_0 = 880$ nm and $q_i (i = 1$ to $30)$ are taken to be $-15, -14, \cdots, -1, 0, 1, \cdots, 14$, totally thirty values. Then the layers are arranged in ascending order from the innermost layer. And Δ is a parameter which determines the thickness difference of the TiO_2 layers, *i.e.*, Δ is related to the corresponding degree of disorder D. From the definition of D stated in the section II, it can be known that Δ=0, 13, 26, 40 and 55 nm correspond to D=0, 0.05, 0.1 0.15 and 0.21, respectively.

Figure 7 gives the calculated reflection curves of multilayers with different Δ. The high reflection range of a multilayer becomes wider as Δ increases. The width of $R > 99\%$ are about 240, 320, 400 and 500 nm for $\Delta = 0$, 13, 26 and 40 nm, respectively. However, when

the value of Δ is too big, for instance 55 nm, the reflectance in high reflection range may drop to lower than 99 %.

In Fig.8 three pairs of reflection curves are shown and each of them comes from a disordered and a staggered multilayers possessing the same D. The widths of high reflection range are almost the same if the multilayers have the identical degree of disorder. The

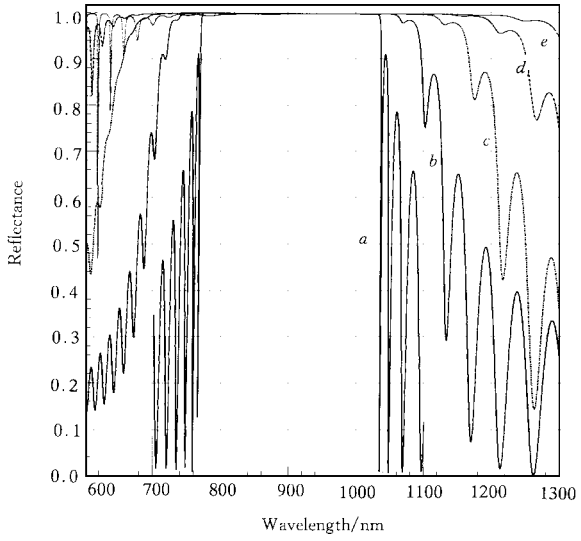

Fig.7. The reflection spectra of TiO_2-SiO_2 staggered multilayers with different values of Δ. Curves a, b, c, d and e correspond to Δ=0, 13, 26, 40 and 55 nm, respectively.

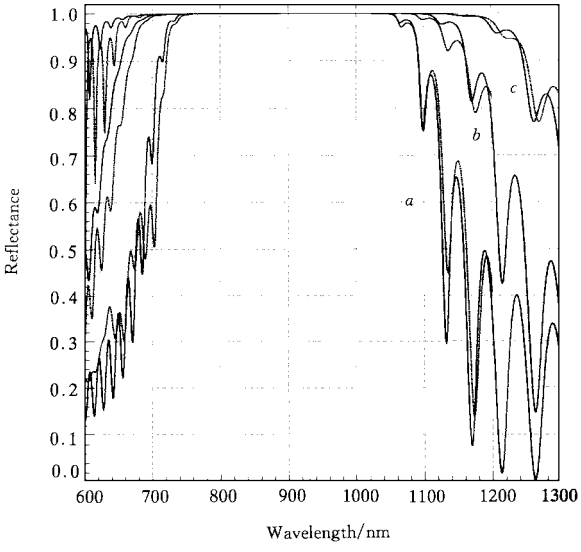

Fig.8. The comparison of reflection spectra between disordered (dotted line) and staggered (solid line) multilayers. curves a : D=0.05, curves b : D=0.1, curves c : D=0.15.

broadening of reflective bandwidth seems to be independent of the methods by which the irregularity of multilayers is made. This implies that the degree of disorder D might be a measure of broadening effect for these irregular multilayers.

V. TRANSIMISSION MEASUREMENT OF A REAL BROADBAND MULTILAYER

To give an experimental evidence that such multilayer can be used as a high reflective mirror for tunable Ti:sapphire lasers, we prepared a TiO_2-SiO_2 multilayer with λ_0=880 nm, D=0.15 and M=24. The thinknesses of all SiO_2 layers are the same of $\lambda_0/4n_s$, while those of the TiO_2 layers are varied and the average value is $\lambda_0/4n_t$. The designed thickness, in units of nm, and the sequence of twenty four TiO_2 layers are listed as follows: 17.7, 35.3, 43.7, 52.5, 66.4, 68.9, 69.9, 70.8, 73.2, 86.3, 92.3, 94.2, 97.6, 99.6, 106, 113, 120, 121, 122, 130, 143, 148, 153 and 174. The thinnest layer directly contacts the surface of the optical glass substrate. This multilayer was fabricated by a conventional electron-beam coating method, where layer thickness was monitored by a quartz oscillator and controlled by a microcomputer.

Because of calibration difficulty in the infrared range the accurate reflection spectrum could not be achieved. And our previous experiment has verified that the absorption of TiO_2-SiO_2 multilayer with 24 periods is negligible. Therefore, we measured the transmission spectrum for such a multilayer by a FR infrared spectrometer. Figure 9 gives the calculated and measured curves. The measured width with a transmittance less than 1% may cover

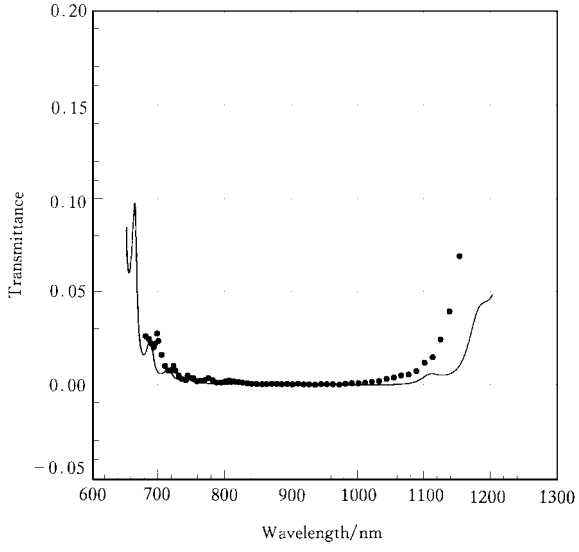

Fig.9. The transmission spectra for a prepared disordered TiO_2-SiO_2 multilayer with λ_0=880 nm, D=0.15 and M=24. Its thickness and sequence are given in the text. solid line: calculated, spots: measured.

about 400 nm (from 700 to 1100 nm), which is just the tuning range of Ti: sapphire lasers. The agreement between calculation and measurement is reasonable. The deviation at the longer wavelength side might be attributed to the thickness error of the real multilayer.

VI. CONCLUSION

Our simulation have shown that different broadband disordered multilayer reflectors can be designed for different applications and the broadening effect of an irregular multilayer is dominated by its degree of disorder. Furthermore, the measurement for a real TiO_2-SiO_2 multilayer has demonstrated that such a kind of broadband multilayer may be used in tunable Ti: sapphire lasers.

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REFERENCES

[1] M. A .Macleod, Thin-Film Optical Filters (American Elsevier, New York, 1969).
[2] Tang Jinfa and Zheng Quan, Applied Thin-Film Optics (Shanghai Science and Technology Publishing House,1984),(in Chinese).
[3] P. Sheng, B. White, Z. Q Zhang and G. Papannicolaou, in "Scattering and Localization of Classical Waves in Random Media", edited by P. Sheng (World Scientific, Singapore, 1990), p.563.
[4] K. M. Yoo and R. R. Alfano, *Phys.Rev.*, **B39**(1989),5806; *Opt. Lett.*, **16**(1991), 1823.
[5] A. Kondilis and P. Tzanetakis, *Phys.Rev.*, **B46**(1992), 15426; *J. Opt. Soc. Am.*, **A11**(1994), 1661.
[6] A. R. McGurn, K. T. Christensen, F. M. Mueller and A. A. Maradudin, *Phys. Rev.*, **B47**(1993),13120.
[7] J. M. Frigerio, J. Rivory and P. Sheng, *Opt. Commun.*, **98** (1993),231.
[8] M. M. Sigalas and C. M. Soukoulis, *Phys. Rev.*, **B51**(1995), 2780.
[9] A. Kondilis and E. N.Economou, *Solid State Commun.*, **95** (1995),885.
[10] Zhao-Qing Zhang, *Phys.Rev.*,**B52**(1995),7960.
[11] Zhang D. Z., Hu W., Zhang Y. L. Li Z. L., Cheng B. Y. and Yang G. Z., *Phys. Rev.*, **B50**(1994),9810.
[12] K. M. Yoo and R. R. Alfano, *Appl. Opt.*, **28**(1989),2456.
[13] Daozhong Zhang, Zhaolin Li, Wei Hu and Bingying Cheng, *Appl. Phys. Lett.*, **67**(1995),2431.
[14] O. S. Heavens and H. M. Liddell, *Appl. Opt.*, **5**(1966),373.
[15] S. Penselin and A. Stuedel, *Z. Phys.*, **142**(1995),21.