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Refractive index of Ge film at low temperature

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Abstract: Germanium (Ge) films with physical thickness of 1600nm was deposited on ZnSe substrates by an electron beam evaporation system. The transmittance of Ge film in the range of 2 to 15 μm was measured by a PerkinElmer FTIR cryogenic testing system from 80 K to 300 K with a step length of 20 K. Then, the relationship between the refractive index and wavelength in the 2 ~ 12 μm region at different temperatures was obtained by the full spectrum inversion method fitting. It can be seen that the relationship confirms to the Cauchy formula. The relationship between the refractive index of Ge film and the temperature / wavelength can be expressed as $n(\lambda, T) = 3.29669 + 0.00015T + 5.96834 \times 10^{-6}T^2 + \frac{0.41698}{\lambda^2} + \frac{0.17384}{\lambda^4}$, which was obtained by the fitting method based on the Cauchy formula. Finally, the accuracy of the formula was verified by comparing the theoretical value obtained by the formula with the measured result.

Key words: infrared film, Ge film, refractive index
PACS: 78.20.Ci

锗薄膜在低温下的折射率研究

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摘要:利用电子束蒸发方法在双面抛光的 ZnSe 基底上镀制单层 Ge 薄膜. 在 80 K ~ 300 K 温度范围内,采用 PerkinElmer Frontier 傅里叶变换红外光谱仪低温测试系统每 20 K 测量 Ge 单层在 2 ~ 15 μm 波长范围的透射率. 采用全光谱反演拟合方法得到 Ge 单层在不同温度下的折射率. 结果显示,Ge 单层折射率均随波长增大而减小,且变化趋势基本相同. 利用 Cauchy 色散公式对折射率波长色散关系进行拟合,得到 Ge 薄膜材料折射率温度/波长色散表达式为:$n(\lambda, T) = 3.29669 + 0.00015T + 5.96834 \times 10^{-6}T^2 + \frac{0.41698}{\lambda^2} + \frac{0.17384}{\lambda^4}$. 最后,验证了 Ge 单层膜折射率温度/波长色散公式的准确性.

关 键 词:Ge 膜;红外光学薄膜;折射率温度系数

中图分类号:O484.4　　**文献标识码**:A

Introduction

Infrared optical film is a key part of the optical system, especially the space camera and scanner, as it determines the image quality directly[1-4]. When the optical film is under cryogenic condition in space environment, its spectrum will drift significantly compared with the normal temperature, which is mainly caused by the temperature coefficient of the refractive index for the film materials[5-6]. Germanium (Ge) has proved to be the most useful material for applications as window or lens in the 2-15 μm region due to its excellent properties such as high refractive index and wide transparent region. The optical

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properties of Ge in the cryogenic temperature have been studied by the Infrared Multilayer Laboratory in the University of Reading, UK. Meanwhile, Ge film was widely used in infrared optical filters especially in the middle and far infrared region[7-9]. The refractive index of Ge film under cryogenic condition was commonly obtained by fitting and extrapolation[10-11]. However, it is relatively poor in terms of accuracy and efficiency, which would make sense of addressing these problems in manufacturing of high precision infrared filters.

This work is aimed at establishing a formula that can obtain the refractive index of Ge film in the transparent region quickly under different temperatures. By measuring the spectral transmittance of the Ge film, the refractive index and the parameters of the Cauchy dispersion formula at different temperatures can be derived, and then the relationship between the parameters and the temperatures was obtained by numerical fitting. Finally, the relationship between the refractive index of Ge film and the temperature/wavelength is received, and the accuracy of the formula is verified.

1 Theoretical basis and methods

The interference effect will happen when lights spread into transparent film. The spectral transmittance through a monolayer film can be expressed as $t = 4n_0 n_s / |n_0 B + C|^2$ [10,12], where $\begin{pmatrix} B \\ C \end{pmatrix} = \begin{pmatrix} \cos \cos\alpha & \frac{i \sin \sin\alpha}{N} \\ i N \sin \sin\alpha & \cos \cos\alpha \end{pmatrix} \begin{pmatrix} 1 \\ n_s \end{pmatrix}$, $\alpha = 2\pi N d/\lambda$, $N = n - ik$, λ is the incident wavelength, n_0 and n_s are the refractive indices of the air and substrate, respectively. It is obvious that the transmittance t is a multivariate function of the parameters n, k and d of the thin film. According to the theory of optical film and the extremum $T_{\max}$ and $T_{\min}$ of the spectrum, the refractive index of the film can be expressed as

$$n_f = (A + (A^2 - n_0^2 n_s^2)^{1/2})^{1/2} \quad , \quad (1)$$

where $A = n_0^2 + n_s^2/2 + 2n_0 n_s (1/t_{\min} - 1/t_{\max})$, n_0 and n_s are the refractive indices of the air and substrate, respectively.

As the initial value, the result of Eq. 1 was used to calculate the value of the refractive index of the film, and the designed thickness was taken as the initial thickness of the film. Then, the wavelength range $\lambda_{\min} - \lambda_{\max}$ was divided into S intervals. The evaluation function was constructed by the difference of the actual measured value $\tilde{t}(\lambda)$ and the theoretical value of $t(\lambda)$, using the minimum of Merit as the objective function to get the optical parameters of the interval i, where the range of the optical parameters of the film was set as the constraint condition to construct the model of the optimization:

$$\min Merit(i) = \int_{\lambda_{lbi}}^{\lambda_{ubi}} \omega(\lambda)[t(\lambda) - \tilde{t}(\lambda)]\mathrm{d}\lambda \quad , \quad (2)$$

$$\text{s. t.} \begin{cases} n_{lbi} \leqslant n_i \leqslant n_{ubi} \\ d_{lbi} \leqslant d_i \leqslant d_{ubi} \end{cases} \quad , \quad (3)$$

where Eq. 2 is the objective function, Eq. 3 is the constraint condition, the lower and upper bounds of interval i are λ_{lbi} and λ_{ubi}, respectively, and $\omega(\lambda)$ is the weight factor of λ. An integrated optimization algorithm based on the nonlinear least squares method and the improved genetic algorithm is used to solve the equation. Then, the optical parameters of interval i were obtained. Finally, the optical parameters of the remaining S-1 intervals can be received in turn, and the optical parameters of the whole wavelength range of the film were obtained.

Double-sided polished single crystal ZnSe wafers with the size of 15×15 mm^2 were selected as substrates. Then Ge single layer film with thickness of 1600 nm was coated on ZnSe substrates by electron beam evaporation using a Denton automatic optical coating machine. During the coating process, the pressure was 3.0×10^{-3} Pa, substrate temperature was 453 K, and the deposition rate was 2 Å/s. At last, the transmittance of Ge single film in 2 ~ 15 μm region was measured by a Perkin Elmer FTIR cryogenic testing system within temperature range 80 ~ 300 K, with a step length of 20 K.

2 Results and discussions

The transmitted spectra of the Ge film at 80, 140, 200, 260 and 300 K are shown in Fig. 1. It can be seen that the peaks and valleys of the spectrum drift towards short wavelength entirely when the temperature decreased. The drift distance of the valley with longer wavelength is much larger than the peak. This can be ascribed to positive temperature coefficient of refractive index for Ge film, which leads to a spectra blue shift at a lower temperature, which is different from the PbTe film[13]. Meanwhile, the change rate of refractive index of Ge film with temperature is bigger in long wavelength region than in short wavelength region. In addition, there is a serious absorption in 10 ~ 15 μm region estimated by the lower peak of the transmittance.

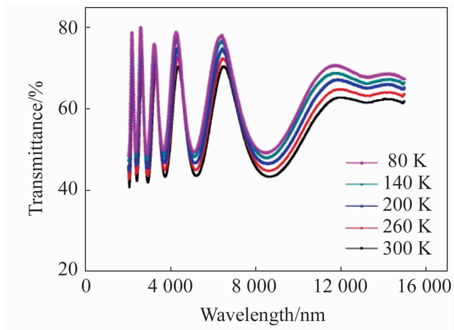

Fig. 1 The transmitted spectra of Ge films at 80, 140, 200, 260 and 300 K
图1 Ge膜在80 K、140 K、200 K、260 K和300 K温度下的光谱图

Figure 2 shows the relationship between the refractive index and wavelength obtained by full spectrum inversion fitting method at 80, 140, 200, 260 and 300 K in the 2 ~ 12 μm region. It can be seen that the refractive index of Ge film decreases with longer wavelength and the trend is almost the same for different temperatures. Moreover, the relationship between the refractive index and wavelength confirms Cauchy formula which can be used by the fitting method.

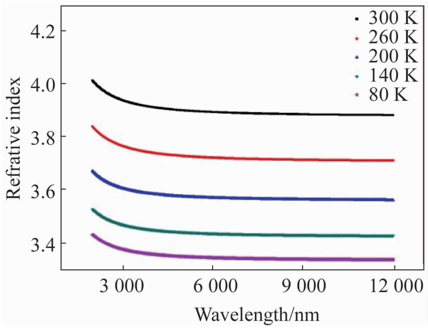

Fig. 2　The relationship between the refractive index and wavelength at 80, 140, 200, 260 and 300 K in the 2 ~ 12 μm region
图 2　在 2 ~ 12μm 波段范围内 80 K、140 K、200 K、260 K 和 300 K 温度下的折射率与波长关系

The Cauchy formula can be expressed as $n(\lambda) = A_n + \frac{B_n}{\lambda^2} + \frac{C_n}{\lambda^4}$, in which there are three unknown parameters A_n, B_n and C_n[10]. Then the relationships between the unknown parameters and the temperature T are researched below.

The relationship between A_n and T obtained by the binomial fitting method is shown in Fig. 3. It can be expressed as $A_n = 3.29669 + 0.00015T + 5.96834 \times 10^{-6} T^2$, where the square of correlation coefficient R is 0.99636, and the standard error is 0.00245. Consequently, the difference between A_{nmax} and A_{nmin} is 0.5447 within the temperature range of 80 ~ 300 K, and the temperature coefficient of A_n is 0.00248 K^{-1}.

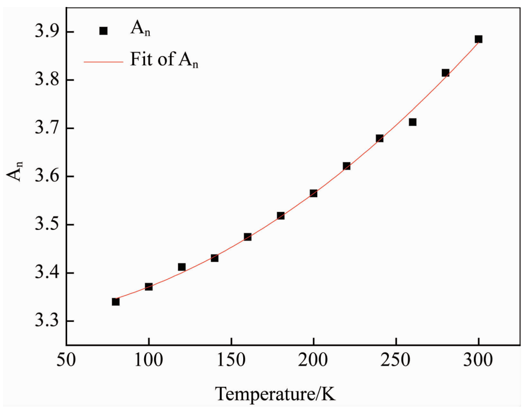

Fig. 3　The relationship between A_n and T
图 3　参量 A_n 与温度 T 的关系拟合图

The relationship between B_n/C_n and T are shown in Fig. 4. It can be seen that the temperature has lesser effect on B_n and C_n, which can be taken as an average of their value, respectively. The maximum effects of the B_n and C_n on the temperature coefficients of the refractive index for Ge film are 1.42×10^{-5} and 4.67×10^{-7} K^{-1}, respectively.

Based on the above analysis, the relationship between the Ge film's refractive index and temperature / wavelength was obtained by fitting method based on the

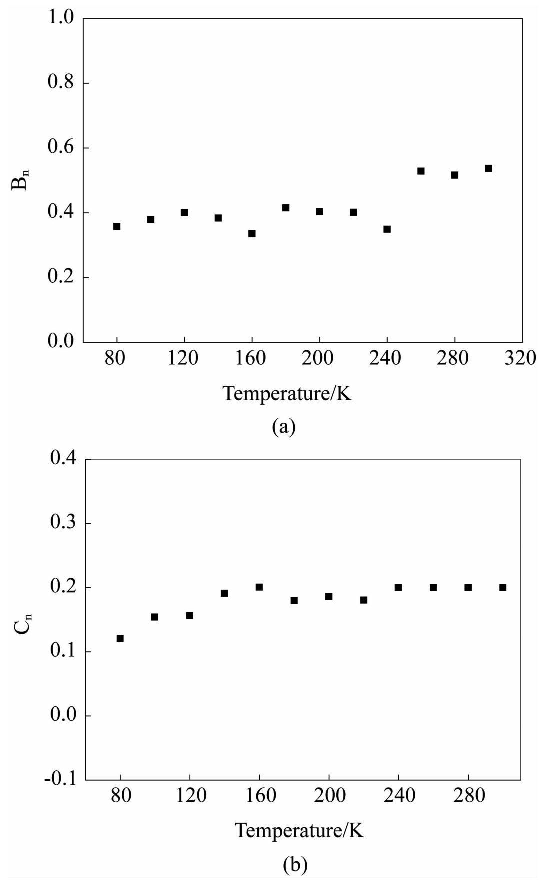

Fig. 4　The relationships between (a) B_n and (b) C_n and T
图 4　(a) B_n 和 (b) C_n 分别随温度变化关系图

Cauchy formula that can be expressed as:

$$n(\lambda, T) = 3.29669 + 0.00015T + 5.96834 \times 10^{-6} T^2 + 0.41698/\lambda^2 + 0.17384/\lambda^4 \quad . \quad (4)$$

The temperature coefficient of the refractive index for Ge film is approximately 0.00248 K^{-1}. Compared with the results of Ge substrate from the Infrared Multilayer Laboratory, the data of film is much larger, which is mainly due to poor density of the film.

Figure 5 shows the comparison of the designed spectra by the formula and the measured spectra at 80 K and 300 K. As we can see, the measured spectra coincide exactly with the designed value at 80 K and 300 K in the 2 ~ 10 μm region, which proves the accuracy of the temperature coefficient of the refractive index for Ge film. Because of the absorption of the Ge film, there is a deviation between the calculated spectra and the measured spectra in the 10 ~ 12 μm region.

3　Conclusions

In summary, the relationship between the refractive index of Ge film and the temperature / wavelength was obtained. The formula was established by a fitting method based on the Cauchy formula. The temperature coefficient of the refractive index for Ge film is approximately 0.00248 K^{-1}. These results can be used to calculate the refractive index of Ge film at different temperatures within the wavelength range of 2 ~ 10 μm, which is benefi-

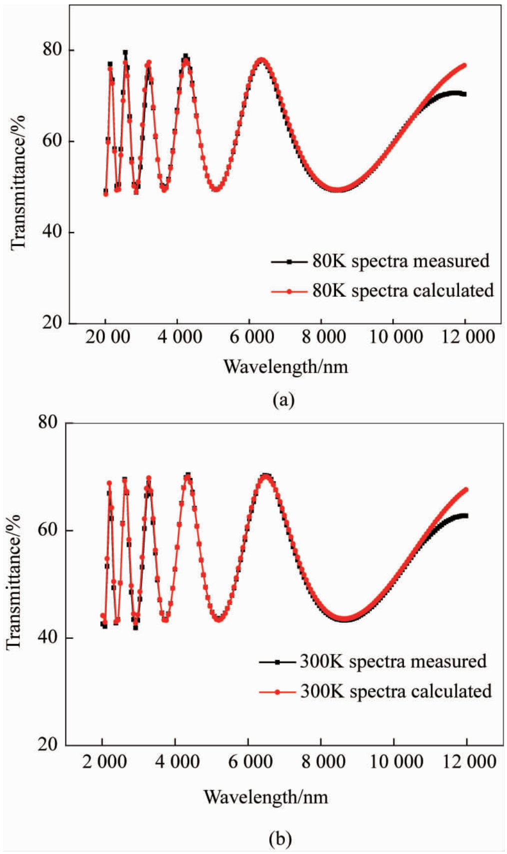

Fig. 5 The contrast of the designed spectra by the formula and the measured spectra at (a) 80 K (b) 300 K
图5 (a)80 K 和(b)300 K 温度下实测光谱与公式得到的理论值的对比图

cial for the manufacturing of optical devices with high temperature stability.

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