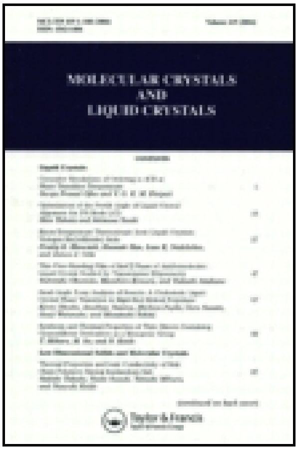

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Light Reflection in Cholesteric Mixtures

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Light Reflection in Cholesteric Mixtures†‡

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The pitch of cholesteryl propionate–acetate mixture has been measured as a function of temperature and concentration. The twisting power of pure cholesteryl propionate and cholesteryl acetate (in mixture) is calculated by using the phenomenological equation, derived by Bak and Labes. The temperature dependence of twisting power is found to be linear in a temperature range far away from transitions, which can be explained by Keating's formula.

INTRODUCTION

The relationship between the cholesteric pitch and concentration has been studied theoretically by de Gennes,[1] Goosens[2] and experimentally by Adams, *et al.*[3,4] Bak and Labes,[5,6] by extending the concept of the long-range distortions induced by chiral molecules in a nematic matrix to multi-component liquid crystal mixtures, have derived an equation governing the pitch-concentration relation. It is a phenomenological equation and seems that it can explain the experimental data quite well. For a binary cholesteric–cholesteric mixture, the equation can be written as:

$$\frac{M_A + (M_B - M_A)w_A}{2pd} = N_0\left\{\left(\frac{M_B}{M_A}\beta_A - \frac{M_A}{M_B}\beta_B\right)w_A + \frac{M_A}{M_B}\beta_B + \Delta\beta_{AB}(1 - w_A)w_A\right\}. \tag{1}$$

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where w_A = weight fraction of the cholesteric component A; and N_0 is Avogadro's number

$$d = \frac{d_B}{1 + (d_B/d_A - 1)w_A}$$

is the density (g/cm^3) of the mixture and $d_{A(B)}$ is the density of the pure cholesteric component $A(B)$. The parameter $\Delta\beta_{AB}$ is defined as:

$$\Delta\beta_{AB} \equiv 2\beta_{AB} - \left[\left(\frac{M_B}{M_A}\right)\beta_A + \left(\frac{M_A}{M_B}\right)\beta_B\right] \tag{2}$$

where $\beta_{A(B)}$ is the effective (or mean) molecular twisting power between $A(B)$ molecules only. When β is positive (negative), the helical structure is right (left) handed.

EXPERIMENTAL

A modified Schimadzu double beam spectrophotometer was employed to measure the reflected peak of the film at a 20° reflected (incident) angle. The datum was taken at certain equilibrium temperature which was obtained by cooling the sample.

The sample was sandwiched between slide glass and cover glass and heated by a block heater. The temperature was controlled within 0.1°C and the reflected wavelength was measured with an accuracy of ±5 Å. The cholesteric compounds were purchased from Merck and used without further purification. Their transition temperatures were measured by Reichert Thermovar. Cholesteryl propionate (CP) shows a melting point at $T = 96$°C and a clear point at $T = 114$°C. Cholesteryl acetate (CA) shows no mesophase phase and becomes normal liquid at $T = 116.5$°C when heating from the solid. During the cooling from normal liquid, the transition temperature to cholesteric mesophase is 94.5°C and due to its very narrow temperature range, the solidified temperature cannot be measured. The mixture was obtained by mixing the cholesteric compounds in chloroform solution. In order to obtain a uniform color display, the glasses were cleaned by cleaning solution and rubbed.

The pitch is determined according to

$$\lambda = \bar{n}P \cos\frac{1}{2}\left[\sin^{-1}\left(\frac{\sin I}{n}\right) + \sin^{-1}\left(\frac{\sin R}{n}\right)\right] \tag{3}$$

where λ = wavelength of reflected light; $I(R)$ = angle of illumination (viewing); $\bar{n}$ = average index of refraction of cholesteric mixture and P

is the resultant pitch of the mixture. This equation was derived by Fergason[7] on the basis of a model of regions of local order imbedded in a material with refractive index $\bar{n}$. The molecular twisting power (β) is computed by Eq. (1).

RESULTS AND DISCUSSION

Figure 1 represents a typical reflected peak observed by the spectrophotometer when one scans the wavelength at a constant temperature. The lower curve is the background when the sample is removed. The wavelength of maximum intensity can be determined within ± 5 Å by manual adjustment. The angle of illumination and viewing is 20 degrees with the normal to the sample. The dependence of reflected wavelength on temperature for a cholesteryl acetate and propionate mixture at different weight ratios is shown in Figure 2. By Eq. (3), the resultant pitch was calculated by letting $\bar{n} = 1.5$ and its temperature dependence is shown in Figure 3. Eq. (1) can be written as

$$N_0 \beta_A = \frac{M_A}{2pd_A} \tag{4}$$

when $w_B = 0$.

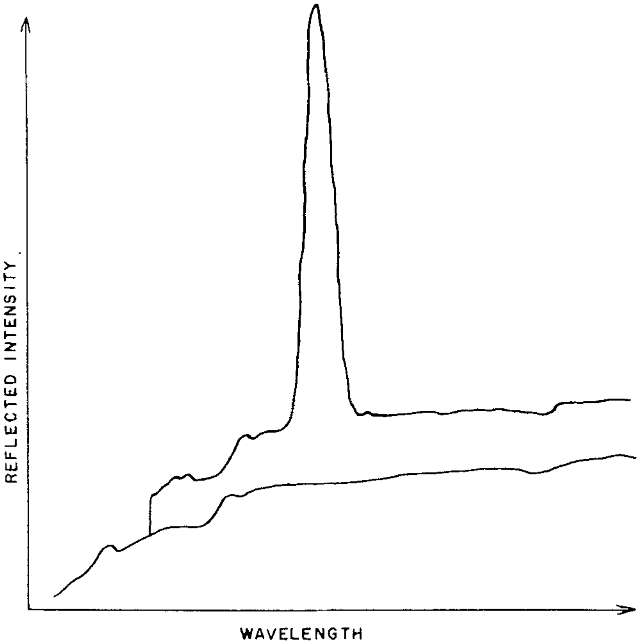

FIGURE 1 Typical reflected peak obtained by the spectrophotometer. The lower curve is the background.

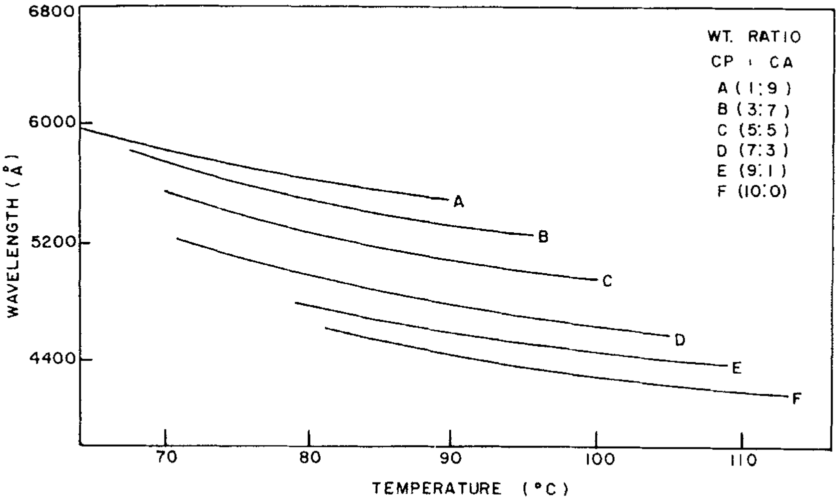

FIGURE 2 The dependence of the reflected wavelength of cholesteryl propionate–acetate mixture on temperature at different weight ratios.

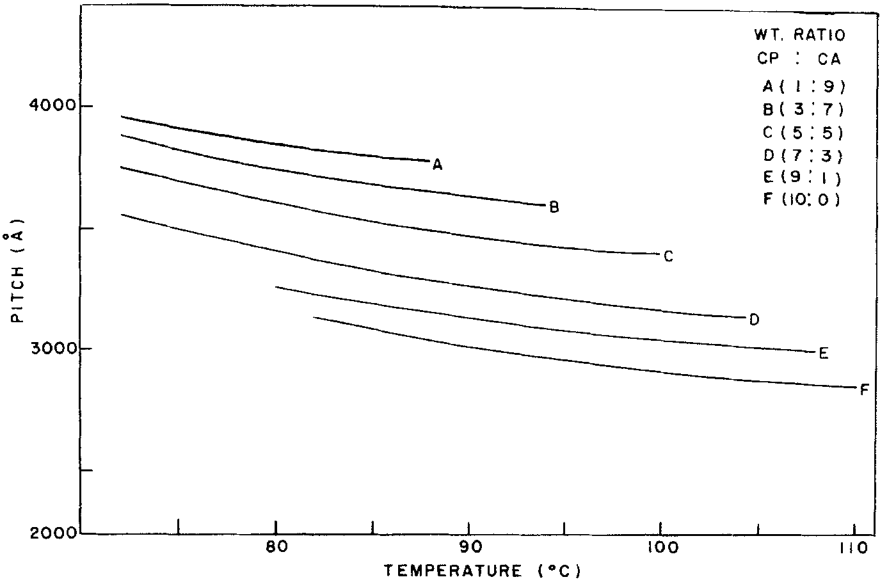

FIGURE 3 The dependence of the pitch of cholesteryl propionate–acetate mixture on temperature at different weight ratios.

Figure 4 is obtained by inserting the pitch value of pure cholesteryl propionate, namely the data of curve F on Figure 3, to equation (4). An approximate density $d_A = 1$ g/cm^3 and $M_A = 443$ g were used. Away from the transition temperatures, the data can be fitted in a straight line. It implies that there is a linear dependence of twisting power of cholesteryl propionate on temperature. Near the transition temperatures, the twisting power will go to zero as expected. Because the mesophase temperature range of cholesteryl acetate is so narrow, one couldn't measure the pitch directly. We estimated the pitch for pure cholesteryl acetate by using the data of Figure 3 to plot a graph of $1/p$ versus concentration of cholesteryl acetate. By extrapolating, the pitch values of pure cholesteryl acetate at different temperatures were obtained and its twisting power is shown on Figure 5. It apparently obeys the linear relation with temperature.

Masubuchi, *et al.*[8] reported that there is a linear dependence of index of refraction or density on temperature for cholesteric mixture. The value $\bar{n}/d = 1.5/1 = 1.5$, which was used to calculate the twisting power, seems to be a reasonable one. It also has been shown by Masabuchi *et al.* that the influence on the pitch due to the temperature dependence of refractive index and density was quite small whem compared to the twisting angle change caused by temperature itself. The accuracy for the twisting power of cholesteryl propionate should be within 10%.

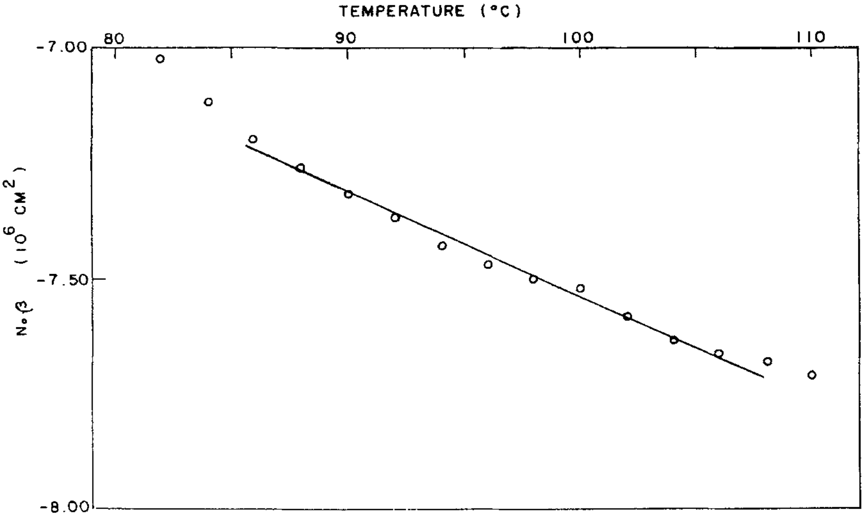

FIGURE 4 The variation of calculated molar twisting power of cholesteryl propionate with temperature.

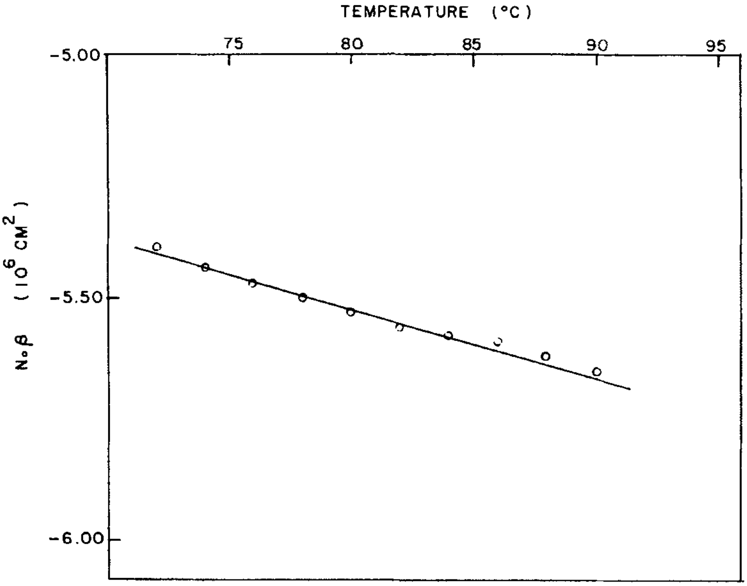

FIGURE 5 The variation of estimated molar twisting power of cholesteryl acetate (in the mixture) with temperature.

We have used Eq. (1) to compute the values of $N_0\Delta\beta_{AB}$ and it is found that their values are in the order of 10^5 cm^2 which is quite small when it is compared with $N_0\beta_{A(B)}$. It means that the deviation of twisting power (β_{AB}) between CP and CA from the average value of β_A and β_B is small.

In conclusion, for the temperature range far away from the transition temperatures, the data for the twisting power of CP and CA can be fitted in a linear relation with temperature. This relation can be explained by Keating's phenomenological theory[9] which regards the twisting of cholesteric molecules as the rotational analog of thermal expansion.

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