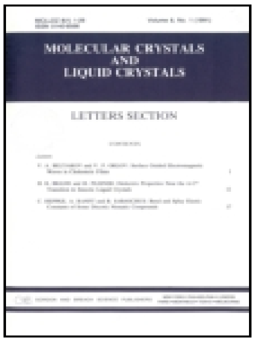

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Pressure effect on the cholesteric – smectic A transition in cholestryl myristate

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ABSTRACT

Cholesteryl myristate exhibits tricritical behavior under high pressures close to its cholesteric-smectic A (Ch/S_A) transition, as observed experimentally. We study here the thermodynamic properties of this liquid crystal by calculating the pressure dependence of the isothermal compressibility, thermal expansion and the specific heat close to the Ch/S_A transition. This calculation is performed at constant temperatures (80.8 and 105.0 °C) using the observed volume-pressure (V-P) data from the literature.

We find that the thermal expansion and the isothermal compressibility increase linearly, whereas the specific heat exhibits anomalous behaviour with increasing pressure close to the Ch/S_A transition in cholesteryl myristate. From the pressure dependence of the thermodynamic quantities studied, the Pippard relations are examined and their linear variations are obtained at various pressures at constant temperatures of 80.8 and 105.0°C for this liquid crystal close to the Ch/S_A transition. Our results also indicate that the tricritical point (TCP) is located at higher pressure (about 1 kbar) for the Ch/S_A transition in cholesteryl myristate, as suggested previously.

KEYWORDS

Cholesteric – smectic A transition; cholesteryl myristate; thermodynamic quantities; pippard relations

1. Introduction

Liquid crystals are the fourth state of the matter. They exhibit both the solid and liquid state properties. At the phase transition point, a discontinuity occurs in entropy and volume (first order transition). When entropy and volume exhibit continuous behavior (second order transition), the thermodynamic quantities such as specific heat, thermal expansivity and the isothermal compressibility show discontinuous behavior at the transition point. Concerning the presence of a tricritical point which is the intersection of a second-order and two first-order phase lines as examplified using the He^3 – He^4 mixtures by Griffiths [1], the nematic – smectic A (NA) transition in racemic A7 and 650 BC shows a crossover to a tricritical behavior [2]. Also, the smectic A – smectic C (AC) transition goes through a mean – field tricritical point [3–8]. Tricritical behavior of the smectic A to smectic C∗ transition has been studied for the liquid crystal mixtures previously [9, 10]. We have also studied first order and second order transitions [11–14] and, the tricritical transition [15] in liquid crystals as reported in our earlier works. In another work, we have calculated T – X phase diagram using the mean field theory for liquid crystals [16].

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Cholesteryl esters and their binary mixtures have been studied extensively near the phase transitions, as reported in the literature [17–21]. For the cholesteric types of liquid crystals the specific heat was measured near the smectic – cholesteric and cholesteric – isotropic liquid transitions, which are of a first – order type, in particular for the measurements of the cholesteryl myristate (CM) [22]. Experimentally, using the polarizing microscopy and thermal analyses, those transitions in CM have been studied [23]. Ultrasonic [24], thermodynamic and viscometric [25] measurements for the cholesteryl myristate and its binary mixtures have been conducted. Molecular diffusion coefficients [26] and components of the order parameter tensor [27] of cholesteryl myristate have been determined. In the isotropic phase of cholesteryl myristate, relaxation processes have been studied [28].

In regard to the binary mixtures of cholesteryl myristate, thermodynamics of the transition from the β-cholesteric phase (at low temperatures) to the α-cholesteric phase (high temperatures) has been studied for a mixture of cholesteryl chloride and the cholesteryl myristate [29]. For the binary mixtures of cholestanyl myristate – cholesteryl myristate and cholestanyl myristate - cholesteryl oleate (CO), thermal and structural properties have been investigated [30]. Structure of the binary mixtures of cholesteryl myristate and cholesteryl pelargonate [31] and also the structure of cholesteryl myristate and cholesteryl pentadecanoate solid solution [32] have been determined. Another binary mixture of cholesteryl myristate and dodecyloxybenzoic acid has been studied thermodynamically [33].

Among the cholesteric esters, cholesteryl oleyl carbonate (COC) exhibits a change from a discontinuous to a continuous behavior at high pressures, as observed experimentally [34]. This transition which has both discontinuous (first order) and continuous (second order) features is the tricritical transition. As the liquid crystal of COC, cholesteryl myristate (CM) can also exhibit tricritical transition at high pressures, as pointed out previously [35, 36]. For cholesteryl myristate, cholesteric – smectic A (Ch/S_A) transition has been studied and its P-T phase diagram has been obtained experimentally [36]. In our earlier studies, we have calculated the T-X phase diagrams for binary mixtures of cholestaryl myristate - cholesteryl myristate and cholestaryl myristate – cholesteryl oleate [37] and we have analyzed the specific heat of cholesteryl myristate close to phase transions [38].

Various aspects of the cholesteric – smectic A (Ch/S_A) transition in cholesteryl myristate (CM) have been studied both experimentally and theoretically, as reported recently [39, 40]. In particular, high pressure studies of cholesteryl myristate in liquid crystals have focused on the thermodynamic properties of the Ch/S_A transition [35, 41].

In regard to the temperature and pressure dependence of the volume for the cholesteryl myristate and its binary mixtures, volumetric measurements have been performed by Pollmann and Schulte [36]. As explained in their work [36], they performed in particular, high pressure volume measurements up to 4 kbar by the glass piezometer containing the measuring substance by means of the mercury level which determines the volume change. As the pressure increases which compresses the measuring substance, the mercury level decreases in the glass piezometer. They were able to determine the change in the specific volume of the substance as accurate as $\pm$ 0.0001 cm^3/g [36].

The transition temperature was determined as $T_t = 98.6\,°C$ at the pressure of $P_t = 0.82$ kbar (transition pressure) for choleteryl myristate by means of the optical measuring method [36]. It has been stated that this transition temperature and pressure, the discontinuous change in the volume (ΔV_d) vanishes, whereas the P-V-T measurements dealing with the total change in volume (ΔV) in the transition region reveal that the remaining volume effect in the pretransition region above 0.82 kbar approaches zero [36]. So, the total volume (ΔV) decreases with increasing transition pressure (P_t) and the temperature (T_t) as the discontinuity in the volume (ΔV_d) disappears, as observed experimentally [36].

Thus, the transition occurs from the discontinuous to a continuous one in cholesteryl myristate according to the volume change, which suggests the presence of a tricritical point (TCP) in this liquid crystal. However, Semenchenko et al. [35] have indicated that extrapolating the volume change (ΔV) to zero at $P_t = 1.05$ kbar ($T_t = 107$ °C) locates in this pressure range the TCP in cholesteryl myristate according to their measurements at low pressures up to 0.8 kbar. Those coordinates of P_t and T_t refer to the transition between cholesteric (Ch) and smectic A (S_A) for the cholesteryl myristate.

On the other hand, Hermann et al. [41] have pointed out that the enthalpy changes at $P_t = 1.05$ kbar ($T_t = 107$ °C) while the change in volume disappears [36] for the Ch/S_A transition of cholesteryl myristate. Change in enthalpy can disappear at higher pressures (above 3 kbar) as pointed out previously [36]. The optical studies on long – chain cholesteryl n-alkanoates [42] have indicated that the Ch/S_A transition occurs as a continuous one at lower pressures [36]. This transition in cholesteryl myristate has been studied experimentally if the change in volume disappears at higher transition pressures [36].

In this study, our aim is to investigate the thermodynamic properties of the cholesteryl myristate close to the cholesteric – smectic A (Ch/S_A) transition. Due to the fact that the change in volume disappears in cholesteryl myristate at the transition temperature of $T_t = 98.6$ °C and the transition pressure of $P_t = 0.82$ kbar as observed experimentally [36], the pressure and/or temperature dependence of the thermodynamic quantities such as the specific heat, thermal expansion and the isothermal compressibility can give insight in some detail into the Ch/S_A transition in this liquid crystal. As in the volume change which disappear close to the Ch/S_A transition, changes in the specific heat, thermal expansion and the isothermal compressibility can characterize the kind of phase transition (continuous or discontinuous) in cholesteryl myristate under the effect of pressure and/or temperature. By assuming those thermodynamic quantities (specific heat, thermal expansion and the isothermal compressibility) exhibit similar critical behavior near the Ch/S_A transition in cholesteryl myristate, their linear variations under various temperature and pressure condititions (Pippard relations) can be examined. Furthermore, the existence of the tricritical point where the first order (discontinuous) lines coincide with a second order (continuous) line as stated above, can also be investigated according to the changes in the specific heat, thermal expansion and the isothermal compressibility at various temperatures and pressures close to the Ch/S_A transition in this liquid crystal.

Since we use the experimental V-P data [36] we are concerned with the presence of a tricritical point (TCP) at $T_t = 98.6$ °C and $P_t = 0.82$ kbar due to the discontinuous change toward the continuous change in volume of the cholesteryl myristate. This is obtained at various pressures at constant temperatures of 80.8 and 105.0 °C from the measurements of the volume changes [36]. Because of the fact that the thermodynamic quantities are derived on the basis of the volumetric measurements as a function of pressure at those constant temperatures, we restrict our investigation to the two constant temperatures for the Ch/S_A transition in cholesteryl myristate. When the experimental data are available in the literature for the pressure dependence of the specific volume V (cm^3/g) or the change in molar volume ΔV (cm^3/mol) at some other constant temperatures for the cholesteryl myristate close to the Ch/S_A, similar analysis given here can be performed for this liquid crystal.

In the present study, we analyze the experimental V-P data [36] at constant temperatures ($T = 80.8$ and 105.0°C). By analyzing the volume data of Pollmann and Schulte [36] we deduce the pressure dependence of the isothermal compressibility κ_T and the thermal expansion α_P for the cholesteric – smectic A transition in cholesteryl myristate. From the α_P vs. P and κ_T vs. P relations, the pressure dependence of the specific heat is obtained and then the Pippard relations are examined for the cholesteric – smectic A transition in this liquid crystalline system.

2. Calculations and results

The pressure dependence of the isothermal compressibility κ_T can be obtained from the V-P data using the definition

$$\kappa_T = -\left(\frac{1}{V}\right)\left(\frac{\partial V}{\partial P}\right)_T \tag{1}$$

By means of the thermodynamic relation

$$\frac{\alpha_P}{\kappa_T} = \frac{dP}{dT} \tag{2}$$

the pressure dependence of the thermal expansion α_P can also be obtained by knowing the slope value of dP/dT in molecular crystals.

In this study, we used the experimental data [36] for the specific volume V (cm^3/g) at various pressures at constant temperatures of T = 80.8°C and 105.0°C for the cholesteric-smectic A transition for the liquid crystal of cholesteryl myristate. Using the experimental $P_t - T_t$ diagram [36] close to the cholesteric-smectic A transition, we deduced the slope value of $dP_t/dT_t = 0.041$ bar/°C at 80.8°C and we calculated the pressure dependence of the thermal expansivity α_P by Eq. (2). We did the same analysis for T = 105.0°C using the experimental data [36] by deducing the slope value of $dP_t/dT_t = 0.049$ bar/°C above and below P_t in cholesteric myristate.

We also calculated the pressure dependence of the specific heat according to the thermodynamic relation

$$C_P - C_V = T\left({\alpha_P}^2/\kappa_T\right) \tag{3}$$

for the cholesteric – smectic A transition for cholesteryl myristate. For this calculation, we used the values of the isothermal compressibility κ_T and the thermal expansion α_P at various pressures for constant temperatures (80.8°C and 105.0°C) of this liquid crystal. We then examined the Pippard relations given by

$$C_P = TV\alpha_P\left(dP_t/dT_t\right) + T(dS/dT)_t \tag{4}$$

and

$$\alpha_P = \kappa_T\left(dP_t/dT_t\right) + (1/V)\left(dV/dT\right)_t \tag{5}$$

for the cholesteric-smectic A transition in cholesteric myristate. We plot $(C_P - C_V)/T$ as a function of $V\alpha_P$ in Figs. (1) and (2) at various pressures at constant temperatures of 80.8 and 105°C, respectively, as representative figures. Uncertainties in $(C_P - C_V)/T$ and in $V\alpha_P$ are also shown in those figures. From our plots, we extracted the values of the slope (dP_t/dT_t) according to Eq. (4), as given in Table 1.

For the second Pippard relation (Eq. 5), we plot α_P against κ_T for the cholesteric – smectic A transition in cholesteryl myristate. Figs. (3) and (4) give those plots of α_P vs. κ_T at various pressures, for temperatures of 80.8°C and 105°C, respectively, also as representative figures.

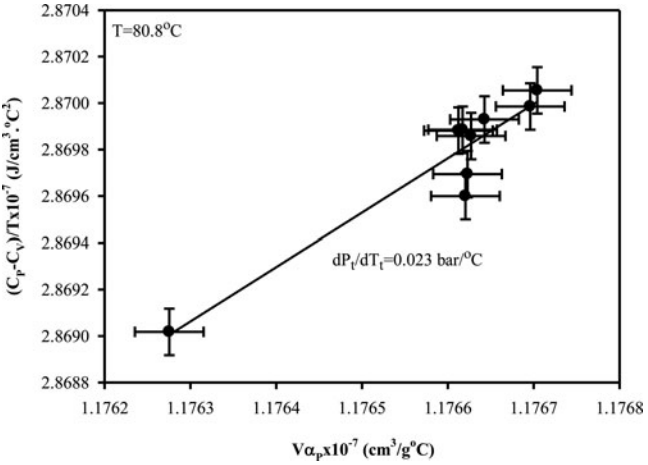

Figure 1. $(C_P - C_V)/T$ as a function of $V\alpha_P$ at various pressures (T = 80.8°C) according to the first Pippard relation (Eq. 4) close to the cholesteric – smectic A transition in cholestric myristate.

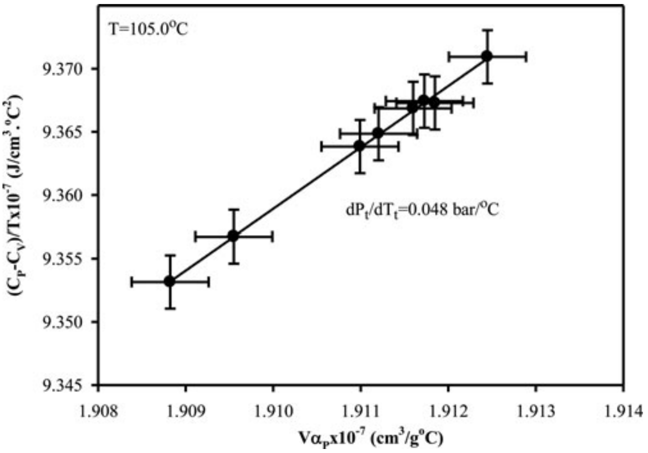

Figure 2. $(C_P - C_V)/T$ as a function of $V\alpha_P$ at various pressures (T = 105.0°C) according to the first Pippard relation (Eq. 4) close to the cholesteric – smectic A transition in cholestric myristate.

Table 1. Values of the slope (dP_t/dT_t) (Eqs. 4 and 5) and the value of the intercept, $(1/V)(dV/dT)_t$ (Eq. 5) for the cholesteric myristate close to the cholesteric – smectic A transition (Figs. 1 – 4). The observed values of the slope dP_t/dT_t which we deduced from the P_t vs. T_t phase diagram [36] with the transition temperature (T_t) and pressures (P_t) [36] at constant temperatures (T = 80.8 and 105.0 °C) are also given here.

T (°C)	T_t (°C)	P_t (kbar)	dP_t/dT_t (bar/°C) (Eq. 4)	dP_t/dT_t (bar/°C) (Eq. 5)	dP_t/dT_t (bar/°C) Observed [35]	$-(1/V)(dV/dT)_t \times 10^{-4}$ (°C^{-1})(Eq. 5)
80.8	98.6	0.82	0.023	0.041	0.041	9.0
105.0	105.0	1.13	0.048	0.049	0.049	4.0

Uncertainties in the α_P and κ_T are given in those figures. From our plots (Figs. 3 and 4), we also extracted the values of the slope (dP_t/dT_t) and the intercept $(1/V)(dV/dT)_t$ according to Eq. (5), as given in Table 1.

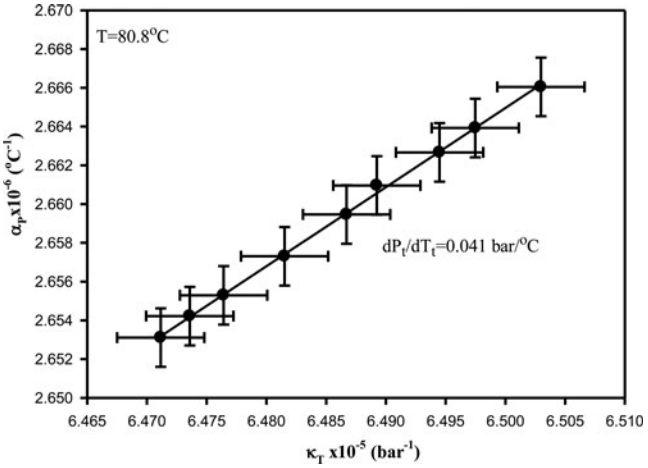

Figure 3. Thermal expansion (α_P) as a function of the isothermal compressibility (κ_T) at various pressures (T = 80.8°C) according to the second Pippard relation (Eq. 5), close to the cholestric – smectic A transition in cholesteric myristate.

3. Discussion

We studied here the pressure dependences of the isothermal compressibility κ_T, thermal expansion α_P and the specific heat for the cholesteric-smectic A transition in cholesteryl myristate. Using the experimental data for the molar volume at various pressures for constant temperatures of 80.8°C and 105.0°C, we calculated the pressure dependences of κ_T, α_P and $C_P - C_V$ at these two constant temperatures. From our calculations, we obtained that the κ_T and α_P increase with increasing pressure toward the smectic A phase for constant temperatures studied, whereas variation of the specific heat $(C_P - C_V)$ with the pressure is anomalous. This anomalous behaviour can then characterize the cholesteric-smectic A transition in cholesteric myristate. At the transition pressure ($P_t = 0.82$ kbar, $T_t = 98.6$ °C), the cholesteric

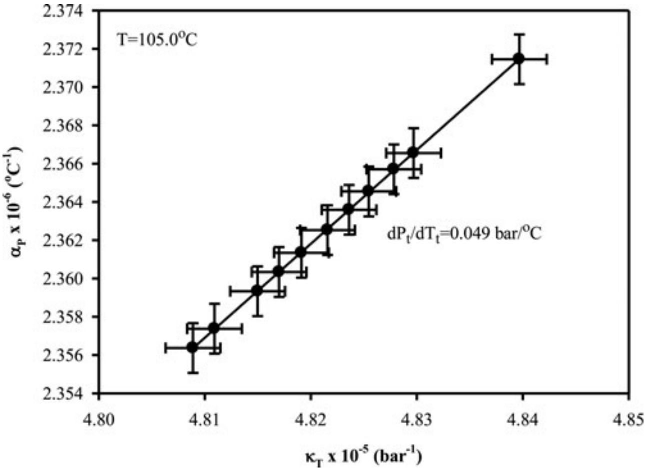

Figure 4. Thermal expansion (α_P) as a function of the isothermal compressibility (κ_T) at various pressures (T = 105.0°C) according to the second Pippard relation (Eq. 5), close to the cholestric – smectic A transition in cholesteric myristate.

phase of the cholesteric myristate should coexist with the smectic A phase [36]. As the transition pressure P_t is raised to $P_t = 1.13$ kbar at T = 105°C, a rapid decrease in the volume change ΔV takes place [36] which indicates a continuous transition toward the cholesteric phase in cholesteric myristate. This can increase the anomaly in the specific heat associated with a continuous (second order) transition from the smectic A (low temperature phase) to the cholesteric phase (high temperature phase) in this liquid crystal.

Using the pressure dependence of the κ_T, α_P and $C_P - C_V$, we examined the Pippard relations (Eqs. 4 and 5) for the cholesteric-smectic A transition in cholesteric myristate. As shown in Figs. (1 – 4), we obtained a linear variation of $(C_P - C_V)/T$ against $V\alpha_P$ (Eq. 4) for this liquid crystal at various pressures for constant temperatures of 80.8°C and 105°C (Figs. 1 and 2), as expected. We also obtained a linear variation of α_P against κ_T as shown in Figs (3) and (4), which is also expected from Eq. (5) for the cholesteric-smectic A transition in cholesteryl myristate.

From our plots, the dP_t/dT_t values which we extracted can be compared with the observed values (Table 1). Since we used initially the observed values which we deduced from the $P_t - T_t$ phase diagram [36] to calculate the pressure dependence of the thermal expansion (Eq. 2), the slope values of the dP_t/dT_t according to Eq. (5) are the same as the observed ones (Table 1). This requires the validity of the first Pippard relation (Eq. 4) which can be tested (Figs. 1 and 2) by comparing the slope values of dP_t/dT_t at constant temperatures (80.8 and 105.0°C) with the observed values (Table 1). Our slope value of $dP_t/dT_t = 0.048$ bar/°C is very close to the observed one at T = 105.0°C, whereas our value of $dP_t/dT_t = 0.023$ bar/°C is far from the observed value of 0.041 bar/°C at T = 80.8°C (Table 1). This is due to the fact that at the transition pressure of $P_t = 0.8$ kbar at T = 80.8 °C the specific volume V has some small discontinuity, whereas at the transition pressure of $P_t = 1.13$ kbar at T = 105.0 °C it almost disappears with a continuous change from the cholesteric phase to the smectic A phase as the pressure increases, as observed experimentally for the cholesteric myristate [36]. As a result of this observed behaviour, our value of $dP_t/dT_t = 0.048$ bar/°C at T = 105.0 °C (Eq. 4) agrees with the observed value for a continuous Ch/S_A transition, whereas our value of $dP_t/dT_t = 0.023$ bar/°C at T = 80.8 °C indicates a discontinuous Ch/S_A transition with a small discontinuity in the volume [36] giving an observed value of $dP_t/dT_t = 0.041$ bar/°C. So, we can conclude that on the basis of the volume changes as observed at various pressures [36], the cholesteric - smectic A transition is of a first order (weak discontinuity) at 80.8 °C, which changes its character toward a second order (continuity) at 105.0 °C for cholesteryl myristate. Thus, it is expected that a tricritical point (TCP) occurs between the transition pressures of $P_t = 0.8$ and 1.13 kbar at constant temperatures of 80.8 and 105.0 °C according to our dP_t/dT_t values (Eq. 4) (Table 1) on the basis of the observed volume changes [36]. Since our dP_t/dT_t value at T = 105.0 °C predicts the observed value (Table 1), most likely the tricritical point occurs close to the transition pressure of $P_t = 1.13$ kbar due to the discontinuous change in volume (T = 80.8 °C) toward a continuous change (T = 105.0 °C) for cholesteryl myristate. This prediction in fact agrees with the expectation of Semenchenko et al. [35] as $P_t = 1.05$ kbar ($T_t = 107$ °C) at which the transition volume approaches zero for cholesteryl myristate.

In regard to a crossover from a discontinuous to the continuous behavior which occurs in cholesteryl myristate as the pressure increases (from the cholesteric to the smectic A phase), it has been pointed out that the existence of the change in the total volume (ΔV) is a pretransitional effect [36] as stated above. This should correspond to the change in enthalpy (ΔH) as measured by Hermann et al. [41], which does not disappear at higher pressures (up to 3 kbar). Thus, the Clasius – Clapeyron equation (ratio of the ΔV to the ΔH) which gives rise to the slope dP_t/dT_t for the Ch/S_A transition in cholesteryl myristate would be valid at low pressures (below $P_t = 0.82$ kbar) as pointed out previously [36].

Since the Clasius – Clapeyron equation describes a first order transition (in the presence of the latent heat), this discontinuous change in the volume (ΔV) and in enthalpy (ΔH) decreases and eventually diminishes toward a second order (continuous) transition in this liquid crystal at higher pressures up to 3 kbar. Most likely, during this transition from the cholesteric to the smectic A phase as the pressure is increased, cholesteryl myristate is expected to exhibit a tricritical point (TCP) at about $P_t = 1$ kbar as the first order (discontinuous) transition changes toward a second order (continuous) one.

4. Conclusions

The thermodynamic properties of the cholesteryl myristate were studied close to the cholesteric – smectic A (Ch/S_A) transition by calculating the specific heat, thermal expansion and the isothermal compressibility as a function of pressure at the two constant temperatures (80.8 and 105.0 °C). For this calculation, the experimental V – P data were used from the literature. On the basis of the experimental data we obtained that the thermal expansion and the isothermal compressibility increase, whereas the specific heat exhibits anomalous behaviour as the pressure increases from the cholesteric to the smectic A phase in this crystalline system. As pointed out from the volume measurements, our results also indicate that a tricritical point (TCP) exists at about 1 kbar because of a first order (discontinuous) change toward a second order (continuous) change in cholesteryl myristate.

We also obtained a linear variation of the specific heat with the thermal expansion and a linear variation of the thermal expansion with the isothermal compressibility (Pippard relations) close to Ch/S_A transition in this liquid crystal. This linear variation of the thermodynamic quantities and the slope (dP_t/dT_t) values which were deduced from the Pippard relations, describe the observed behaviour and also support the idea that a possible tricritical point (TCP) exists in cholesteryl myristate. Similar approach can be applied to those liquid crystals which exhibit tricritical behavior as the cholesteric myristate and its binary mixtures.

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