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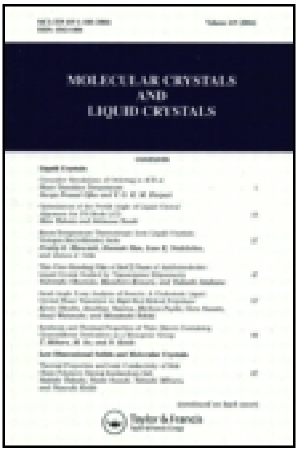

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Universal Nature of the Nematic Phase

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Universal Nature of the Nematic Phase

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In this paper, we synthesize and resume some previous results found on the universality of the nematic phase's anisotropic behavior. Throughout a linear transformation on the experimental data of some anisotropic macroscopic properties, we show that the corresponding points become distributed along universal lines that extend along the entire range of the nematic phase, from the nematic-isotropic phase-transition region until the nematic-crystalline phase-transition region. We also show that the universal curve of the order parameter is characterized by a singular power law behavior, with a single exponent that encompasses the entire range of the nematic phase.

Keywords Nematic phase; order parameter; universality

1. Introduction

Liquid crystals (LCs) were discovered in 1888, when Friedrich Reinitzer observed an apparent duplicity in the melting points of cholesteryl benzoate [1]. Since then, these materials are subject of intense research and, even today, the understanding of their properties appears to be quite challenging. Even the simplest of their phases, the nematic phase, is not fully understood. For example, the nature of the nematic-isotropic (NI) phase-transition has never been completely understood. Although it is surely a first-order phase-transition [2,3], it presents noticeable characteristics of a second-order phase-transition and a discussion about the existence of a 'critical nematic universal class' and the correspondent critical exponent have been subject of intense debate [4–11]. Recent discoveries [12–19] should increase the discussions about the criticality in the nematic phase. These discoveries are the theme of the present paper.

Here, we explicitly show that experimental data of different nematic liquid crystals (NLCs) present a universal behavior along the entire range of the nematic phase. Namely, we show that nematic anisotropic properties, like elasticity, viscosity, magnetic susceptibility and electric permittivity, have a common behavior for all compounds studied. We give special attention to those properties which are used to define the nematic order parameter (*S*), because *S* characterizes the anisotropy of the nematic material and, as consequence, all thermodynamical anisotropic properties of the NLCs. We show that the universal behavior of S is characterized by a singular power law behavior.

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Table 1. Abbreviation and scientific names of the NLC compounds studied

Abbreviation	Scientific Name
mCB	4-n-(m alkyl)-4′-cyanobiphenyl
mOCB	4-n-(m alkoxy)-4′-cyanobiphenyl
PAAm	4–4′-di-(m alkoxy)azoxybenzene (PAA $m = 1$, PAP $m = 2$)
mAB	4–4′-di-n-(m alkyl)azoxybenzene
PCHm	4-cyano-4′-n-(m alkyl)-cyclohexanephenyl
PTTP-mm	4,4′-di-n-(m alkyl)diphenyldiacetylene
MBBA	4-methoxybenzylidene-4′-n-butylaniline
7CT	4-n-heptyl-4′-cyanotolane
CCH7	4′-n-heptyl-bicyclohexyl-4-carbonitrile
ME105	4-methoxy-benzoic acid 4′-n-pentyl-phenyl ester
ME605	4-n-hexyloxy-benzoic acid 4′-n-pentyl-phenyl ester
ZLI 1052	mixture from Merck
ROCM7037	4-(5-heptyl-pyrimidin-2-yl)-benzonitrile
LC-KN1	4-cyano-thiobenzoic acid S-(4′-n-pentyl-phenyl) ester
LC-KN2	4-n-pentyl-thiobenzoic acid S-(4′-cyano-phenyl) ester
LC-KN3	4-n-pentyl-benzoic acid 4′-n-hexyloxy-phenyl ester
LC-KN4	4-n-heptyl-thiobenzoic acid S-(4-cyano-phenyl) ester
LC-KN5	4-octanoic acid benzylidene-4′-aminobenzonitrile
N4	4-methoxy-4′-n-butylazoxybenzene (mix of two components)
OHMBBA	2-hydroxy-4-methoxybenzylidene-4′-butylaniline
7CE	4-n-heptyl-benzoic acid 4-cyano-phenyl
ZLI 1083	mixture from Merck
ROCM1850	4-pentyl-cyclohexanecarboxylic acid 4-cyano-phenyl ester
ROCM1870	4-heptyl-cyclohexanecarboxylic acid 4-cyano-phenyl ester
PEBAB	4-n-ethoxybenzylidene-4′-aminobenzonitrile
BBAB	4-n-butoxybenzylidene 4′-aminobenzonitrile
HBAB	4-n-hexyloxybenzylidene 4′-aminobenzonitrile
KN-Mix1	mixture defined in Ref. [21]
CCH 2,4	mixture defined in Ref. [22]
M597	mixture defined in Ref. [22]
PTTP-2244	mixture defined in Ref. [23]
PTTP-2345	mixture defined in Ref. [23]
9CN	4-n-nonylbenzoate 4′-cyano-phenyl
ROCM1951	4-pentyl-cyclohexanecarboxylic acid 4-methoxy-phenyl ester
ROCM1953	4-pentyl-cyclohexanecarboxylic acid 4-propoxy-phenyl ester
TPEB	trans-4-propyl cyclohexy-4 (trans-4-ethyl cyclohexyl) benzoate
TPPB	trans-4-propyl cyclohexy-4 (trans-4-propyl cyclohexyl) benzoate
TPPEB	trans-4-propyl cyclohexy-4 (trans-4-pentyl cyclohexyl) benzoate
7FBT	4′-heptyl-3-fluoro-4 isothiocyanatotolane
6OFBT	4′-hexyloxy-3-fluoro-4 isothiocyanatotolane
N5	mixture defined in Ref. [24]
EM	mixture defined in Ref. [25]

2. Universality and Non-Locality on the Nematic Phase

2.1 Order Parameter

We begin this section by exposing the universality of the order parameter profile in the classical nematic phase also discussing the approach used to obtain it. To do it, we have collected from the LCs' literature a great amount of experimental data. In this paper, we present 78 data sets of different measurements of the order parameter (S), each one covering entire range of the nematic phase. It is important to advise that in the approach used in this paper, not only the experimental data along the nematic phase is important, but also the precise determination of the NI phase-transition temperature (T_{NI}) and the nematic-crystalline (NK) phase-transition temperature (T_{NK}). The scientific names and abbreviations of the compounds studied and the references from which the data were collected are organized in Tables 1–7 in the appendix.

Once taken the experimental data, we introduce two linear transformations on them; a change in the temperature scale and a normalization of the data in a common temperature point in this scale. The aim of introducing this temperature scale is to afford a direct comparison of the thermal behavior of a given physical property of different compounds; in the usual temperature scale, the experimental data of different compounds are found in different temperature ranges. That is, we transform all temperature data of different

Table 2. Phase-transition temperatures and references for the data of mass magnetic susceptibility anisotropy shown in Figs. 1.a and 4

NLC	T_{NK} (°C)	T_{NI} (°C)	Reference
MBBA	19	45	[26]
PAA	118.2	135.3	[26,27]
PAP	136.8	168.4	[28,29]
4AB	22	31.9	[28,30]
5AB	24	67.5	[28,30]
5CB	24	35.3	[22,31]
7CB	28.5	42	[31,32]
7CE	44	56.5	[31,33]
7CT	58.5	67.5	[31,34]
PCH5	30	55	[31,35]
PCH7	30	59	[31,36]
CCH7	71	83	[31,37]
ME105	29.0	42.7	[38,39]
ME605	50.0	62.1	[38,40]
ZLI 1052	15.0	48.9	[38,41]
ROCM7037	45	51	[31,42]
LC-KN1	66.0	102.0	[38]
LC-KN2	75.5	99	[38]
LC-KN3	40.9	62.5	[38,43]
LC-KN4	82	92.4	[31,44]
N4	17.9	74.9	[28,31]
OHMBBA	44	64.6	[45]

Table 3. Phase-transition temperatures and references for the data of electric permittivity anisotropy shown in Fig. 1.a

NLC	T_{NK} (°C)	T_{NI} (°C)	Reference
5CB	22.5	35	[46]
6CB	13.5	29	[46]
7CB	28.5	42	[46]
PCH3	36	46.2	[47,48]
PCH5	30	54.4	[35,47]
PCH7	30	58	[36,49]
ZLI 1083	−3	51.9	[47,50]
CCH7	71	84.1	[37,49]
ROCM1850	47	79	[49,51]
ROCM1870	55.1*	81	[49]
ROCM7037	45	51.3	[42,49]
PEBAB	105.5	128.9	[21]
BBAB	62.9	110.5	[21]
HBAB	54.5	100.9	[21]
6OCB	57	75.8	[52,53]
7OCB	53.5	73.5	[51,52]
LC-KN5	53.7	97.6	[21]
KN-Mix1	18	96.8	[21]

*Lowest temperature in the nematic phase at which the data have been measured.

compounds to a unique temperature scale defined by the limiting temperatures of the nematic phase of the respective compound. Namely, for the compounds bordered by an isotropic phase and by a crystalline phase, the nematic temperature scale (t_N) is defined as,

$$t_N = \frac{T - T_{NK}}{T_{NI} - T_{NK}},$$

in such a way that for all compounds it attains the values $t_N = 0$ at T_{NK}, and $t_N = 1$ at T_{NI}.

As we are uniquely interested in to compare the thermal profile of the physical properties of different NLCs compounds, we need to normalize their data. Here, all properties used to measure the profile of S will be normalized to 1 at $t_N = 0$. At this point, some important issues about t_N must be addressed. Firstly, t_N is an authentic temperature scale, constructed with two fixed points, it is not a reduced temperature, with a single fixed point as used, for example, in the study of continuous phase-transitions [2]. Furthermore, and in order to stresses that our approach is not the usual approach to study the NI critical-like behavior, the point used to normalize the data is not the temperature of the NI phase-transition [3]. Our data is normalized at the T_{NK}, the farthest temperature point from T_{NI}; it is important to have this information in mind when analyzing the data presented below. Finally, notice that we are not assuming a complete ordering with this normalization. We assume $S = 1$ at T_{NK} just to have a fixed value at this point. The results obtained with the S data are shown in Fig. 1.

Fig. 1 displays, in two figures, Fig. 1.a and Fig. 1.b, S experimental data of a total of 49 different NLCs', most of them measured by different authors at different epochs.

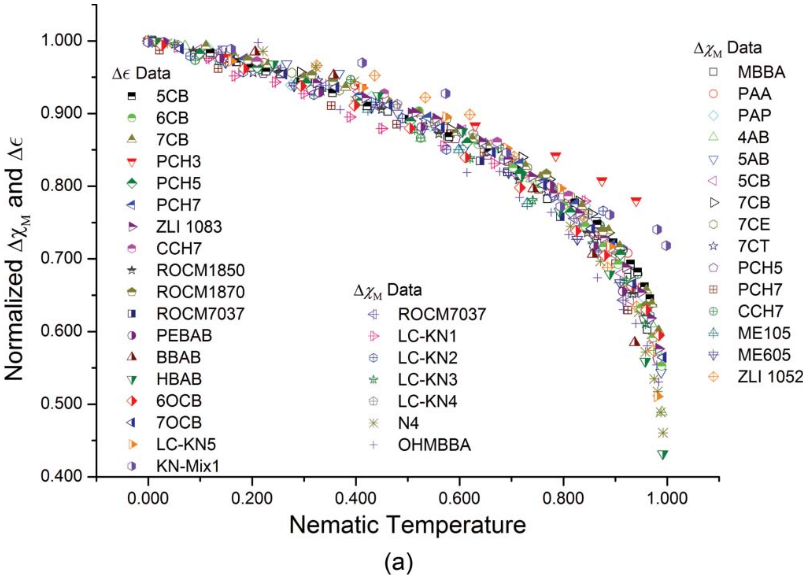

Figure 1. a. Global universal behavior of NLCs' scalar order parameter (S). Here we put together 22 data sets of mass magnetic susceptibility anisotropy ($\Delta\chi_M$) and 18 data sets of electric permittivity anisotropy ($\Delta\epsilon$), properties that usually are used to measure S. Among these 40 data sets, only 2 show a sensible deviation from the mainstream curve. Compounds names, phase-transition temperatures, and references are given at Tables 1–3. [From Ref. 12.]

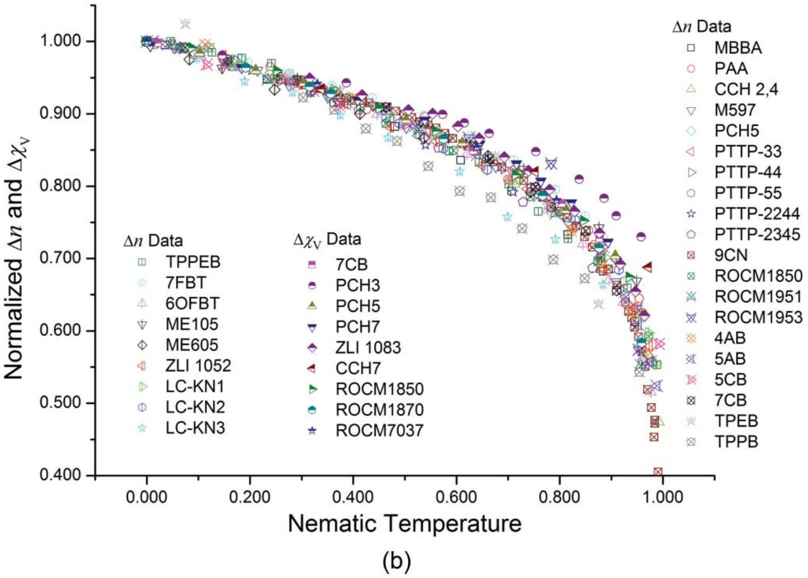

Figure 1. b. Global universal behavior of S. Here we are using 29 birefringence (Δn) data sets and nine volume susceptibility anisotropy ($\Delta\chi_V$). Now, among these 38 data sets, only 3 show a sensible deviation from the mainstream curve. Compounds names, phase-transition temperatures, and references are given at Tables 1, 4, and 5. [From Ref. 12.]

Table 4. Phase-transition temperatures and references for the data of birefringence shown in Fig. 1.b

NLC	T_{NK} (°C)	T_{NI} (°C)	Reference
MBBA	21	46.25	[54,55]
PAA	110	135	[25,56]
CCH 2,4	34	64	[22]
M597	65	114.4	[22]
PCH5	30	55.2	[22]
PTTP-33	107.5	131.9	[57]
PTTP-44	75.1	101.0	[23]
PTTP-55	86.0	111.3	[57]
PTTP-2244	55.0	98.5	[23]
PTTP-2345	40.0	105.0	[23]
9CN	27.8	59.3	[58]
ROCM1850	47.0	77.7	[59]
ROCM1951	40.7	71.1	[59]
ROCM1953	43.3	71.1	[59]
4AB	15	31.9	[34,56]
5AB	25.8	67.5	[34,56]
5CB	22.4	34.5	[32]
7CB	28.5	41.9	[32]
TPEB	94.0	134.0	[60]
TPPB	92.0	158.0	[60]
TPPEB	67.0	154.0	[60]
7FBT	26.6	43.6	[61]
6OFBT	32.5*	70.7	[61]
ME105	29.0	42.7	[39,62]
ME605	50.0	62.1	[40,62]
ZLI 1052	15.0	48.9	[41,62]
LC-KN1	66.0	102.0	[62]
LC-KN2	75.5	99	[62]
LC-KN3	40.9	62.5	[43,62]

*Lowest temperature in the nematic phase at which the data have been measured.

These data sets embrace measurements of mass magnetic susceptibility anisotropy ($\Delta\chi_M$), volume magnetic susceptibility anisotropy ($\Delta\chi_V$), electric permittivity anisotropy ($\Delta\epsilon$), and birefringence (Δn). The agreement shown by the 78 data sets is astonishing; only 5 present a considerable deviation from the mainstream curve. So, the first aspect that may be detached from these data is that, for the absolute majority of the cases, they coalescence along a unique curve, evidencing the existence of a single behavior of the order parameter, along the entire range of the nematic phase. This behavior is called as nematic universality. Most important is; the nematic universality is obtained throughout a linear mapping.

An essential aspect of the nematic universality is that it is not restricted to the neighborhood of a phase-transition; it connects two distinct phase-transitions, extending over the entire temperature domain of the nematic phase. Due to this fact, one can consider

Table 5. Phase-transition temperatures and references for the data of volume magnetic susceptibility anisotropy shown in Fig. 1.b

NLC	T_{NK} (°C)	T_{NI} (°C)	Reference
7CB	29.7	42.6	[34,49]
PCH3	36	46.2	[37,63]
PCH5	30	54.4	[35,47]
PCH7	30	58	[36,49]
ZLI 1083	−3	51.9	[47,50]
CCH7	71	83.3	[37,47]
ROCM1850	47	79	[49,51]
ROCM1870	55.1*	81	[49]
ROCM7037	45	51.3	[42,49]

*Lowest temperature in the nematic phase at which the data have been measured.

Table 6. Phase-transition temperatures and references for the data of elastics constants shown in Fig. 2

NLC	T_{NK} (°C)	T_{NI} (°C)	Reference
MBBA	19	45	[26]
PAA	110	135	[26]
5CB	24	35.3	[22,64]
7CB	28.5	41.9	[32,65]
ROCM7037	45	51	[42,49]
PCH7	30	59	[36,49]
CCH7	71	84.1	[37,49]

Table 7. Phase-transition temperatures and references of the data of viscosity coefficients shown in Fig. 3

NLC	T_{NK} (ºC)	T_{NI} (ºC)	Reference
5CB	22.4	34.5	[32,66]
N4	17.9	74.9	[31,66]
N5	−5.15	73.8	[24]
MBBA	20.9	45.9	[67,68]
PAA	118.2	135.3	[27,69]
EM	20*	48.8	[25,66]

*Lowest temperature in the nematic phase at which the data have been measured.

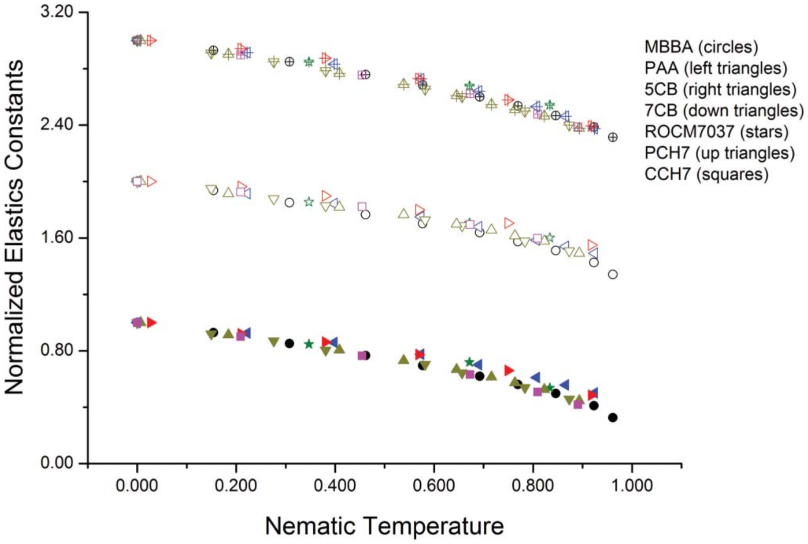

Figure 2. Global universality of the Frank's elastic constants. Solid, open and symbols with a cross inside correspond to K_{11}, K_{22} and K_{33} respectively. Due to the normalization, the absolute values of the elastic constants are irrelevant and they are plotted in three different positions for visual facilities. Compounds names, phase-transition temperatures, and references are given at Tables 1 and 6. [From Ref. 13.]

this universality as an intrinsic property of the nematic phase, and it should be a direct consequence of an unusual global critical behavior [5].

2.2 Elastic Constants

The above results evidence that the nematic phase's anisotropy presents a universal behavior. An important issue is to verify if similar universal behavior is found in other physical properties that have the nematic anisotropy as fundamentals. Fig. 2 shows the results for the elastic constants. We are using de Gennes definition of K_{11}, K_{22} and K_{33} [3] and the same nematic temperature scale defined above. We also normalize the data of each elastic constant at T_{NK}. In order to put all elastic constant in the same graph, the elastic constant K_{11} was normalized to the value 1, K_{22} has to the value 2 and K_{33} to 3. Again, as detached above, the results reveal an astonishing similar behavior for each elastic constant; each one follows a unique pattern that agrees with the idea that there is a universal profile for the physical properties having the anisotropy of the nematic phase as fundament.

2.3 Miesowicz's Coefficients

Now we investigate if the nematic universality can also be seem in the viscosity coefficients. In the form exposed above, our results are inconclusive; when we fix the value of the Miesowicz's coefficients, for example, at the NK phase-transition temperature, we do not obtain a well defined universal behavior. We understand that there are two reasons for this behavior. First, the nematic viscosity may not be solely function of the order parameter, other physical parameters may be important to its determination. Furthermore, as we said above, our approach can only give decisive conclusions if we have a precise value of the viscosity

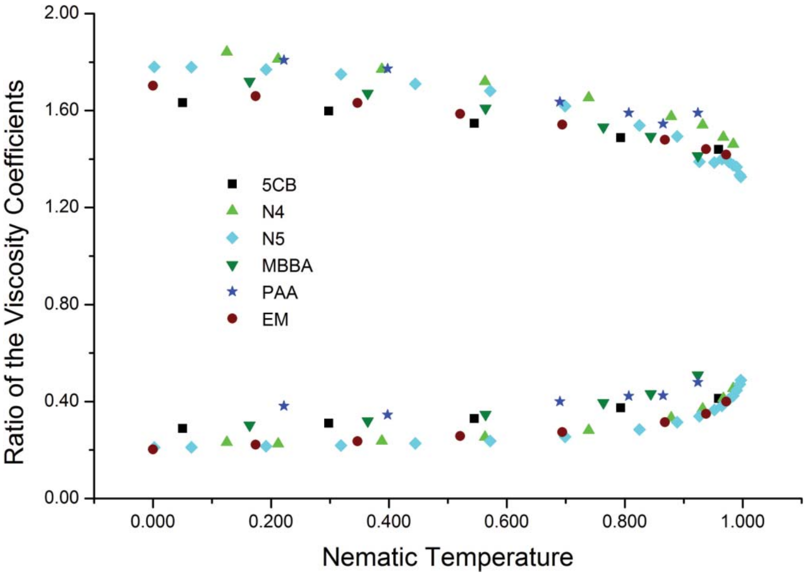

Figure 3. In this figure we show the universality of the ratios between the Miesowicz's coefficients, m_3/m_1 and m_3/m_2. In the upper half of the figure we see the universal curve corresponding to the ratio m_3/m_2 and at the lower half we have the same result for m_3/m_1. Compounds names, phase-transition temperatures, and references are given at Tables 1 and 7.

at the NK phase-transition temperature. The values of viscosity at this temperature are scarce and difficult to be obtained. To overcome these difficulties, we take ratios between the Miesowicz's coefficients. The results obtained are shown in Fig. 3. The coefficients are referred as m_1, m_2 and m_3, and we are using the notation of Helfrich [20]. Again, as happened with the order parameter and elasticity data, the ratios of the Miesowicz's coefficients are displayed along a two curves that suggest the existence of a universal profile.

3. Characterization of the Nematic Phase Universal Behavior

The above studies we show that the physical properties defining the order parameter, the elastic constants and the viscosity coefficients have, for each of them, a unique profile along the entire range of the nematic phase. If we assume that the elastic constants and viscosity coefficients are function of the order parameter, the uniqueness in each of their profiles becomes intelligible; as they are function of the same universal function, the order parameter, they are also universal. Consequently, to understand to universality observed on these physical quantities, we must first understand the universality of the order parameter. That is, we must find, in the physical fundamentals of the nematic phase, the reason why the order parameter is universal.

To further improve the characterization of the universal order parameter profile, we take a di-log display of the $\Delta\chi_M$ data shown in Fig. 1. In order to linearize that curve, at the abscissas we put the data corresponding to $\ln(1–t_N)$ and at the ordinates we put $\ln(\text{Normalized } \Delta\chi_M)$. The resulting curve reveals an important property of S: Its profile is characterized globally by a single exponent. The connection between the angular coefficient

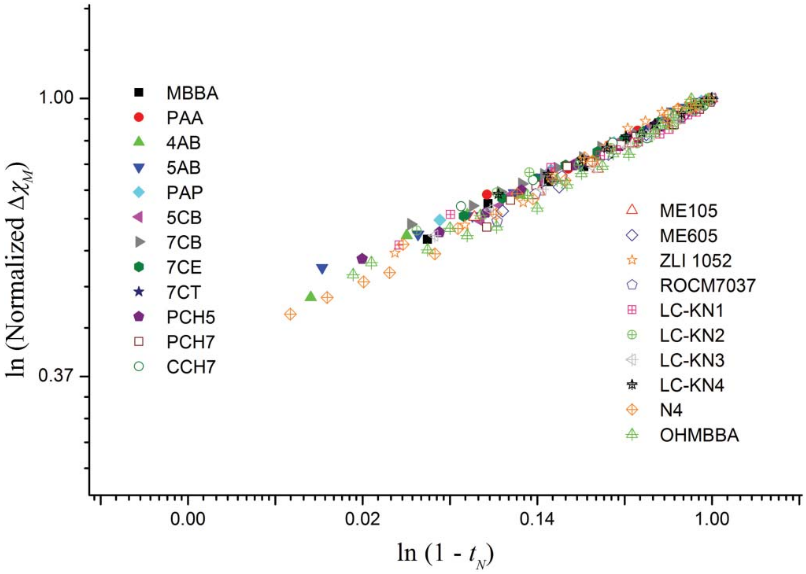

Figure 4. Linearized graph of the $\Delta\chi_M$ data shown in Fig. 1.a. This result shows that the nematic phase order parameter should have a constant exponent along the entire range of the nematic phase. The connection between the angular coefficient of this curve and the critical-like order parameters exponents (β) is not direct. Compounds names, phase-transition temperatures, and references are given at Tables 1 and 2.

of the curve shown in Fig. 4 and critical-like exponents expected to the NI phase transition is not direct. Let us explain why.

The dependence of S which is compatible with the weak first-order character of the NI phase-transition [9] is given by

$$S(T) = S^{**} + c_0 \left(T^{**} - T\right)^{\beta}, \tag{1}$$

where S^{**} is the value of S at the effective second order critical temperature T^{**}, β is the critical-like exponent and c_0 is a constant. The linearization process used to obtain Fig. 4 takes S^{**} as being null. Moreover, when we take $(1–t_N)$ to the ordinates, in fact we are choosing T^{**} as being T_{NI}. Due to these definitions, the resulting angular coefficient is not equal to the β of Eq. (1). In the next paper we deal with the calculation of β. Anyway, the results shown in Figs. 1 and 4 suggest an unusual critical-like universal behavior to the nematic phase: S has a global universal behavior and the exponent of the corresponding universal curve appears to be constant along the entire range of the phase.

4. Final Remarks and Conclusions

In this work we have collected a large number of experimental data sets that suggest that the anisotropic behavior of the nematic phase presents a global universal behavior. Especially important is the behavior of the order parameter, which seems to follow a power law behavior characterized by a constant exponent. As the properties of the nematic phase are dependent on the order parameter, the universality observed to its profile should have present in other properties. Figs. 2 and 3 confirm this expectation.

Finally, we observe that it is not enough the measure the angular coefficient of the straight line observed in Fig. 4 to determine β. Due to the linearization procedure, the results obtained are not compatible with the weak first-order character of the NI phase-transition and, consequently, the angular coefficient is not the critical-like exponent β. The determination of β is the main theme of the following paper.

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