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NEW INTERPRETATION OF LIGHT SCATTERING MEASUREMENTS IN NEMATIC LIQUID CRYSTALS

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NEW INTERPRETATION OF LIGHT SCATTERING MEASUREMENTS IN NEMATIC LIQUID CRYSTALS

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ABSTRACT

Light scattering measurements have caused doubts as to whether the Maier-Saupe theory of nematic liquid crystals is correct. It is shown that if an averaging process is supposed the measurements are in agreement with the theory. An idea is presented concerning the possibility of an error of the commonly accepted picture of the nematic phase.

АННОТАЦИЯ

Результаты экспериментов по рассеянию света поставили под сомнение справедливость теории жидких кристаллов Майера-Сопа. В настоящей статье показано, что при условии процесса усреднения эти измерения укладываются в рамки этой теории. Излагается наше мнение, согласно которому представления о нематической фазе мы считаем ошибочными.

KIVONAT

A fényszórási kisérletek kétségeket ébresztettek, hogy vajon helyes-e a folyadékkristályok Maier-Saupe elmélete. Jelen cikkben megmutatjuk, hogy egy átlagolódási folyamatot feltételezve ezek a mérések beilleszthetők az elmélet keretei közé. Közöljük elgondolásainkat, mely szerint a nematikus fázisról alkotott elképzelések hibát tartalmaznak.

INTRODUCTION

The Maier-Saupe (MS) model [1-3] is now a twenty-year-old theoretical interpretation of the nematic phase. It is a very successful and widely used model, and it shows good agreement with the temperature dependence of the order parameter measurements carried out by a great number of methods. The order parameter is a scalar parameter connected with the angular distribution function (ADF) of the molecular length-axes, its lowest nonzero (second) expansion coefficient on the Legendre polynominals. The expansion, in terms of Legendre polynomials, does not converge rapidly and although the higher order terms are badly needed to obtain more information on the ADF it is difficult to measure them.

Light scattering (LS) experiments were the first to succeed in measuring the next nonzero, i.e. the fourth, coefficient [4-7]. A very surprising finding was that its values are far below the theoretical predictions. These results caused some doubts concerning the validity of the MS model. The situation is controversial because the X-ray experiments [8,9] and coherent neutron scattering (CNS) measurements [10] give good agreement with the theory.

We should like to show a possible interpretation of the LS measurement. We will demonstrate that there are two different formalisms for the ADF in the spherical coordinata system in the cylindrical symmetric case. If an averaging process is supposed, that is at a different level for the X-ray and LS measurements compared with their wavelengths, it is possible to interpret both results. These results imply that the commonly accepted picture of the nematic phase needs correction.

DISCUSSION

In the general case an ADF can be written in the form $f(\Omega)$, where Ω covers the whole 4π angles. We also use the familiar spherical coordinate system where our function has the form $f(\vartheta,\varphi)$. The trouble starts if this ADF is cylindrically symmetric, independent of φ. In this case we can use the $f(\vartheta)_{\varphi}$ notation, using φ as a parameter, but we cannot ignore it. To eliminate φ from the formula we have to integrate it, using the integration rules of the spherical coordinate system:

$$d\Omega = d\vartheta \sin\vartheta d\varphi \tag{1}$$

and in this way we can get the $f(\vartheta)$ distribution function:

$$f(\vartheta) = \frac{1}{4\pi} \int_{0}^{2\pi} f(\vartheta)_{\varphi} \cdot \sin\vartheta d\varphi = \frac{1}{2} \sin\vartheta \, f(\vartheta)_{\varphi} \tag{2}$$

These two forms of the ADF have different meanings. The $f(\vartheta)_{\varphi}$ form means the probability of finding a molecule along a φ = const. line on the surface of the sphere at different ϑ orientation. The $f(\vartheta)$ form represents the probability of finding a molecule at ϑ orientation around the whole sphere. In the case of the ADF of a nematic liquid crystal the most dense by packed surface element is around $\vartheta = 0^{o}$ but the surface connected to the ϑ orientation is increases much more rapidly than $f(\vartheta)_{\varphi}$ decreases, and $f(\vartheta)$ will have the maximum far from the nematic director. The differences between the two ADF's are illustrated in Fig. 1. Both ADF's can be seen at a typical nematic temperature. We can see that the $f(\vartheta)_{\varphi}$ form has its maximum at $\vartheta = 0^{o}$; $f(\vartheta)$ at approximately 20^{o}. The existence of these two ADF's were realized by X-ray experts, and results have been published with both ADF's [8,9,11,12].

The average orientation in three dimensional space is the nematic director. Most of the experimental methods are sensitive to the $f(\vartheta)$ distribution function, and veraging is carried out using this function. The average ϑ value is in the $20\text{-}35^{o}$ interval. Its location is strongly temperature dependent.

If measurements are performed with different correlation times or in other words with a different wavelength of the measuring process we can get a differently averaged ADF, depending on the activated molecular motions. The X-ray and coherent neutron scattering (CNS) methods have around 5000 times shorter wavelength and correlation time than the LS method. In this way we can expect that LS can see a more averaged ADF than that seen by the X-ray and CNS methods. Accepting that the last two methods are in good agreement with the theoretical predictions [8-10], we can say that these methods see almost the perfect ADF. The LS method sees some sort of averaged ADF, with a smaller half-width.

If that process is carried out ad infinitum with very long correlation time the distribution function will be very narrow, at the limit a Dirac delta at the average value. That is, the physical interpretation of the mathematical inequality found by Pershan [6] to be the lower limit of the $P_4(P_2)$ function. In this way with a different averaging level a continuous transition is possible between the theoretical $P_4(P_2)$ function and the Dirac delta limit.

We have simulated the averaging process by multiplying the $f(\vartheta)$ ADF by a Lorentzian line with different half-width. The Lorentzian lines were located on the average value of the ADF. The result can be seen in Fig. 2. The two limits represent the very wide Lorentzian line, the MS ADF, and the very narrow line, the Dirac delta limit. Both the Miyano A line and Pershan's data can be fitted well with an approximately 17° wide Lorentzian line multiplication.

CONCLUSION

It is concluded, that the LS measurements can be integrated into the MS theory if an averaging process is supposed. It seems to us there is some sort of error about the commonly accepted idea of the nematic phase, probably the $f(\vartheta)_{\varphi}$ ADF was used instead of the $f(\vartheta)$. This conclusion reflected by the molecular dynamics of the liquid crystals with the order fluctuation as the major dynamical process.

We believe that the real $f(\vartheta)$ allows other dynamical processes such as precession and libration. Moreover, we consider that these results decrease the difference between the nematic and smectic phases. The MS theory can be expanded to the smectic phases too.

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TEXT OF THE FIGURES

Fig. 1. The two different distribution functions in the spherical coordinate system in the cylindrical symmetric case.

Fig. 2. The $P_4(P_2)$ functions after the narrowing process. MS means the MS ADF, is the upper limit; δ represents the Dirac delta ADF, the lower limit. The numbers on the lines between these limits on the half-width of the Lorentzian lines are in degrees. The crosses represent Pershan's data, the line represent Miyano's data.

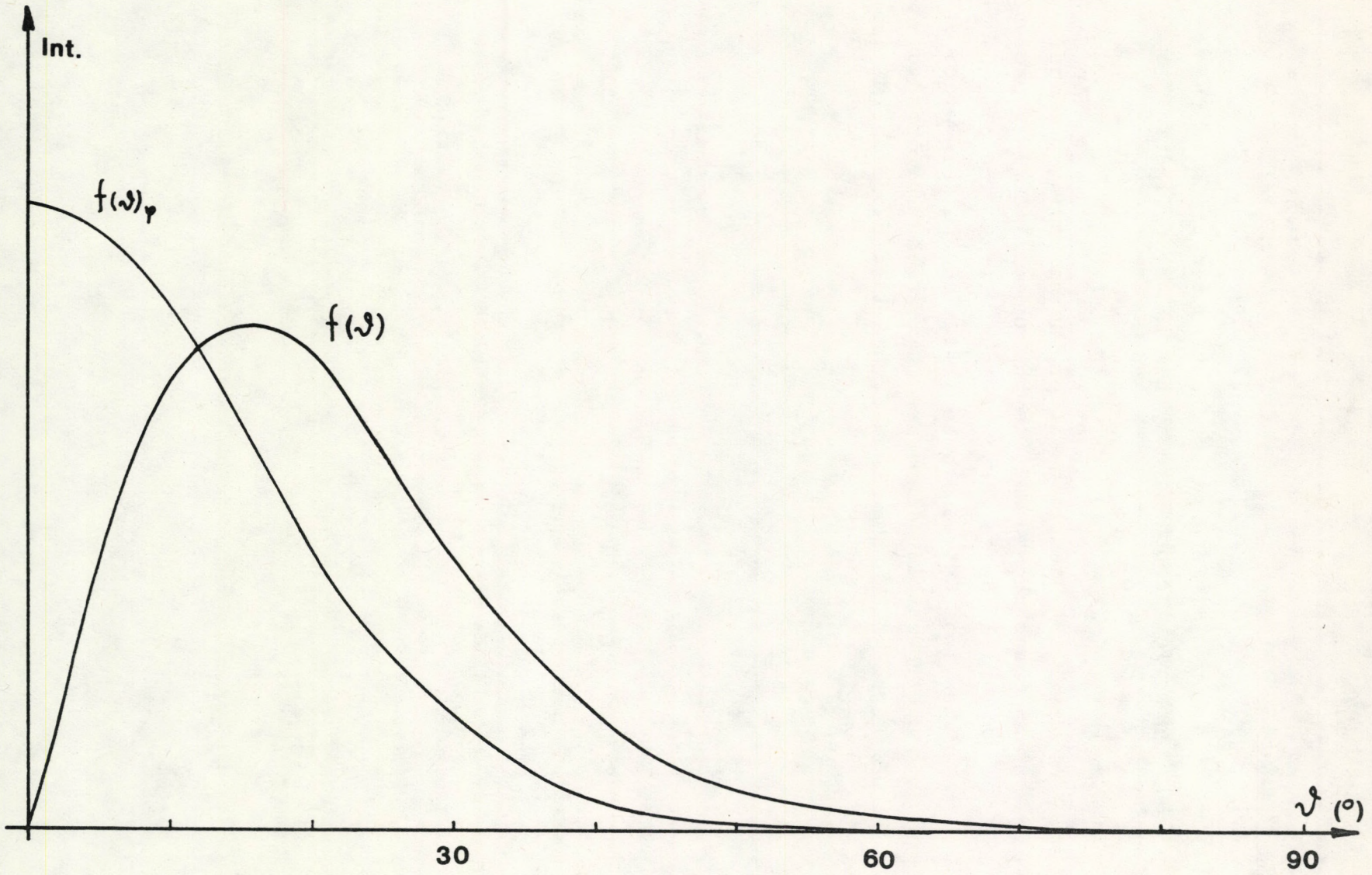
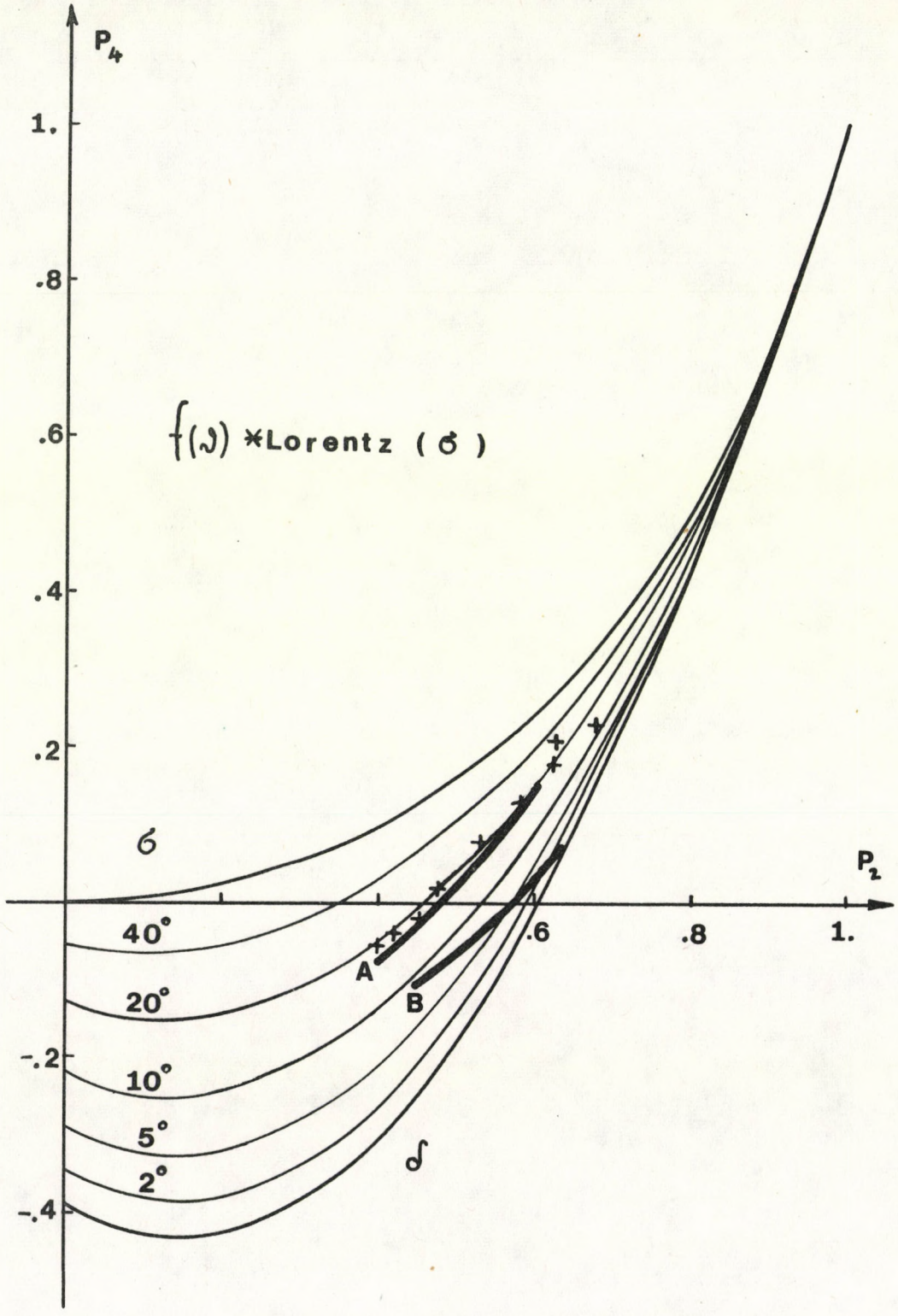


Fig. 1



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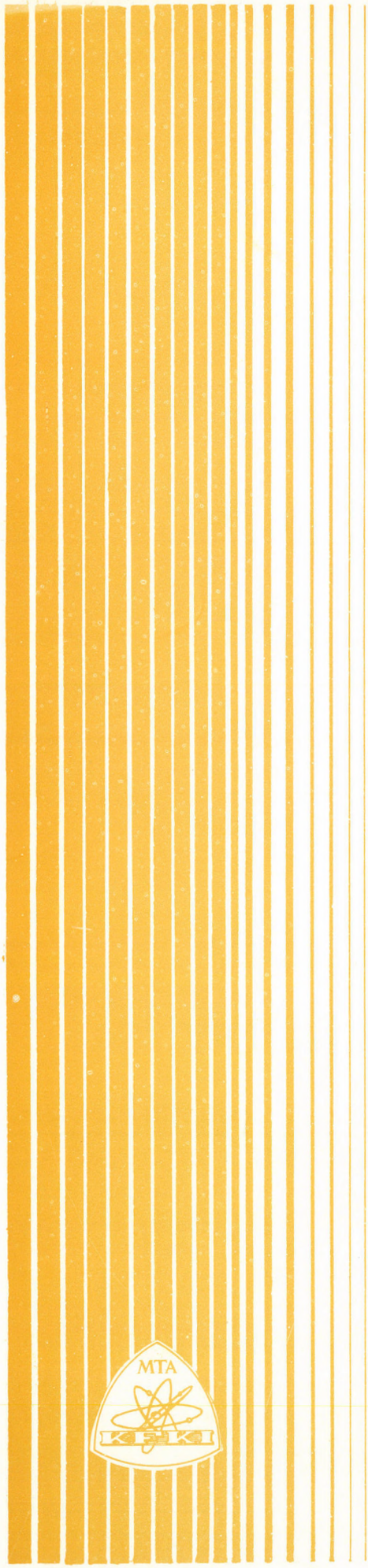

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