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Liquid Crystals

Publication details, including instructions for authors and subscription information: http://www.informaworld.com/smpp/title~content=t713926090

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To cite this Article Oh-e, M., Kondo, K. and Kando, Y.(1994) 'Theoretical consideration of the drop in threshold voltage at low frequencies in nematic liquid crystals', Liquid Crystals, 17: 1, 95 – 107 To link to this Article: DOI: 10.1080/02678299408036551 URL: http://dx.doi.org/10.1080/02678299408036551

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Theoretical consideration of the drop in threshold voltage at low frequencies in nematic liquid crystals

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(Received 25 August 1993; accepted 2 November 1993)

Research of numerical calculation and theoretical considerations have been applied in relation to the electrical double layer effect on the threshold voltage of liquid crystals in order to understand the drop in threshold voltage often observed at low frequencies. Decreased resistivity of an alignment film was found to contribute to this threshold voltage drop. Moreover, dielectric dispersion due to the electrical double layer at the interface between the liquid crystal and alignment film layers is thought to exist in the frequency range in which the drop in threshold voltage was obtained experimentally. Therefore Debye type dielectric dispersion of the electrical double layer in the system consisting of the nematic liquid crystal and the alignment film also influences the threshold behaviour at low frequencies.

1. Introduction

Significant interest in the transport of charge carriers in liquid crystals has led to many reports [1–12], because these properties in liquid crystals must be understood in order to have practical applications. At the interface between the liquid crystal and alignment film layers, interfacial polarization is induced by charge carrier transport; this was proposed from a study of MBBA capacitances at low frequencies [13, 14]. The existence of a capacitance other than the resistance–capacitance network of the liquid crystal has also been shown [15]. Moreover, formation of an electrical double layer has been suggested from results obtained with a reverse hysteresis loop in electrical capacitance–voltage measurements [16] and from the occurrence of a transient current peak [17].

Previously experimental results were reported for a drop in threshold voltage at low frequencies [18]. Investigation related this behaviour to the migration of electrical species in the liquid crystal layer. By considering charge carrier transport, the electrical double layer at the interface between the liquid crystal and alignment film layers, which causes interfacial polarization, was analysed and some understanding was gained about the behaviour at low frequencies.

In the present work, the effective voltage of the liquid crystal layer in the liquid crystal cell is calculated, assuming that pure liquid crystals behave as a parallel resistance–capacitance circuit. Then the effect of the electrical double layer, which is described by Gouy–Chapman's theory, on the threshold voltage is discussed [19].

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Finally, dielectric theory is applied to the system consisting of the liquid crystal and the alignment film, and the dielectric dispersion frequency of the electrical double layer is obtained.

2. Review of Gouy–Chapman's electrical double layer model

Gouy–Chapman's model [19], as shown in figure 1, involving the effect of diffusion, was introduced for the following static analysis, This model is characterized by the fact that the electrical species follow Boltzmann's distribution rule and the relation of electrical potential to electrical volume density can be expressed by Poisson's equation.

The time averaged concentration of a kind of charge carrier C_i is written as a function of Boltzmann's distribution

$$C_{\rm i} = C_{\rm i0} \exp\left(-\frac{W_{\rm i}}{RT}\right),\tag{1}$$

where *R* denotes the gas constant, *T* represents the absolute temperature and C_{i0} indicates the concentration of the charge carrier in the inner parts of the liquid crystal material. W_i is the work function, which is expressed as follows in a one-dimensional system

$$W_{i}(x) = Z_{i}F\phi = Z_{i}F(\phi(x) - \phi^{s}).$$
⁽²⁾

Here Z_i is the electrical charge number of the carrier, F symbolizes Faraday's constant, ϕ represents the electrical potential, ϕ^s corresponds to the internal electrical potential in the liquid crystal and x is the distance from the surface. With equation (1), the total electrical density ρ is written as follows:

$$\rho = \sum_{i} Z_{i} F C_{i0} \exp\left(-\frac{W_{i}}{RT}\right).$$
(3)



Figure 1. Gouy-Chapman's electrical double layer model.

By applying Poisson's equation to this system, the next equation is derived with the permittivity of the system ε

$$\frac{d^2\phi}{dx^2} = -\frac{1}{\varepsilon} \sum_{i} Z_i F C_{i0} \exp\left(-\frac{W_i}{RT}\right).$$
(4)

On the other hand, the condition that the electrical charge is neutralized by summing up the total charge carriers, is expressed as follows:

$$\sigma = -\int_0^\infty \rho \, dx = \int_0^\infty \varepsilon \frac{d^2 \phi}{dx^2} \, dx = \varepsilon \left| \frac{d\phi}{dx} \right|_{x=0}^\infty = -\varepsilon \left| \frac{d\phi}{dx} \right|_{x=0},\tag{5}$$

where σ represents the surface electric density on the alignment film. With equation (5) and the solution of equation (4), the surface electrical density σ is written as follows

$$\sigma = \left[2\varepsilon RT \sum_{i} C_{i0} \left\{ \exp\left(-\frac{Z_{i}F\phi_{0}}{RT}\right) - 1 \right\} \right]^{1/2}.$$
 (6)

Here ϕ_0 corresponds to the electrical potential on the surface. Assuming that a pair of charge is dissolved in the liquid crystal layer, the capacitance caused by the electrical double layer C_{ED} is expressed as

$$C_{\rm ED} = \frac{\partial \sigma}{\partial \phi} = \sqrt{\left(\frac{2\varepsilon C_0 Z^2 F^2}{RT}\right) \cosh\left(\frac{ZF\phi_0}{2RT}\right)},\tag{7}$$

where C_0 and Z are rewritten terms of C_{i0} and Z_i , respectively. Moreover if the quadratic term is negligible, the surface electrical density is rewritten as follows:

$$\sigma = \sqrt{\left(\frac{2C_0 Z^2 F^2}{\epsilon RT}\right)} \epsilon \phi_0 = \kappa \epsilon \phi_0. \tag{8}$$

In this case, $1/\kappa$ has a length dimension, and it expresses the extent of the electrical double layer.

3. Results and discussion

3.1. Frequency dependence of effective voltage of the liquid crystal layer with and without the electrical double layer

Calculations of effective voltage, applied to the liquid crystal layer, were carried out for the system with and without the electrical double layer, in order to understand the layer function. Figure 2 shows an equivalent circuit for liquid crystal devices not involving the electrical double layer. The effective voltage over the liquid crystal layer V_{LC} is described as follows:

$$\frac{V_{\rm LC}}{V_0} = \frac{\frac{1}{R_{\rm AF}} + j\omega C_{\rm AF}}{\left(\frac{1}{R_{\rm LC}} + \frac{1}{R_{\rm AF}}\right) + j\omega (C_{\rm LC} + C_{\rm AF})} = \frac{R_{\rm LC}}{R_{\rm LC} + R_{\rm AF}} \cdot \frac{1 + j\omega C_{\rm AF} R_{\rm AF}}{1 + j\omega (C_{\rm LC} + C_{\rm AF}) \frac{R_{\rm LC} R_{\rm AF}}{(R_{\rm LC} + R_{\rm AF})}},$$
(9)

where V_0 is the external voltage and ω equals $2\pi f$, in which f denotes frequency. R_{LC} and C_{LC} are the resistance and capacitance of the liquid crystal layer, and R_{AF} and



Figure 2. An equivalent circuit for the liquid crystal cell. R_{LC} and C_{LC} stand for resistance and capacitance of the liquid crystal, respectively. R_{AF} and C_{AF} denote those values for the alignment film. V_{LC} , which is to be analysed, is the effective voltage applied to the liquid crystal layer.

Table 1. Summary of parameters used in the calculation by the system of figure 2.

| | Alignment film | Liquid crystal |
|---|---|---|
| Thickness (d/m) Relative permittivity ε_r Resistivity (ρ/Ω cm) | $5 \times 10^{-8} \\ 3 \\ 10^{9} - 10^{13}$ | $ \begin{array}{r} 6 \times 10^{-6} \\ 4 - 16 \\ 10^{7} - 10^{15} \end{array} $ |



Figure 3. Effects of the liquid crystal resistivity on the effective voltage over the liquid crystal layer. The thicknesses of the liquid crystal and alignment film layers were set at $6 \,\mu$ m and 500 Å, respectively. The alignment film resistivity was fixed at $10^{12} \,\Omega$ cm. The relative permittivities of the liquid crystal and alignment film were set at 8 and 3, respectively. Each $V_{\rm LC}$ was normalized by the result obtained with $10^{11} \,\Omega$ cm as the liquid crystal resistivity.

 C_{AF} denote those values for the polymer alignment film layer. Parameters used in this calculation are summarized in table 1.

Figure 3 shows the calculated results for the relation between the resistivity of the liquid crystal and the effective voltage over the liquid crystal layer as a function of frequency. Liquid crystal resistivities larger than $1 \times 10^{10} \Omega$ cm showed no changes of the effective voltage with frequency, while liquid crystals with resistivities less than $1 \times 10^{10} \Omega$ cm produced a decrease in effective voltage with decreasing frequency. This behaviour corresponded to a higher threshold voltage needed to compensate for the voltage to reorient the liquid crystal molecules with decreasing frequency. This result, however, contradicted the drop in threshold voltage experimentally observed at low frequencies, even with liquid crystal resistivities higher than $1 \times 10^{11} \Omega$ cm.

On the other hand, the relation between the resistivity of the polymer alignment film and the effective voltage over the liquid crystal layer as a function of frequency is shown in figure 4. The decrease in alignment film resistivity produced an increase in effective voltage over the liquid crystal layer with decreasing frequency, in contrast to the case for the liquid crystal resistivity. This corresponded to a lower threshold voltage, which explained the drop in threshold voltage observed in the same region. However, with an alignment film resistivity of $1 \times 10^9 \Omega$ cm, the increase in effective voltage reached a maximum and then leveled off for decreasing frequencies of less than about 30 Hz. This meant that the external voltage was applied to the liquid crystal layer without loss of voltage due to the alignment film resistivity. Therefore, the much larger drop in threshold voltage, which was often observed experimentally, could not always be explained by the alignment film resistivity.

Figure 5(a) indicates the influence of the permittivity of liquid crystals on the effective voltage over the liquid crystal layer. The effective voltage over the liquid crystal layer decreased with increasing permittivity of the liquid crystals. However, the



Figure 4. Effects of the alignment film resistivity on the effective voltage over the liquid crystal layer. The thicknesses of the liquid crystal and alignment film layers were set at $6 \,\mu m$ and 500 Å, respectively. The liquid crystal resistivity was fixed at $10^{11} \,\Omega \,cm$. The relative permittivities of the liquid crystal and alignment film were set at 8 and 3, respectively. Each $V_{\rm LC}$ was normalized by the result obtained with $10^{12} \,\Omega \,cm$ as the alignment film resistivity.



Figure 5. Effects of the liquid crystal permittivity on the effective voltage over the liquid crystal layer (a), and its dependence on frequency with typical relative permittivities of the liquid crystal (b). The thicknesses of the liquid crystal and alignment film layers were set at $6 \,\mu m$ and 500 Å, respectively. The resistivities were fixed at 10^{11} and $10^{12} \,\Omega$ cm. The alignment film permittivity was set at 3. Each $V_{\rm LC}$ was normalized by the result obtained with 8 as the liquid crystal permittivity.

effective voltage was frequency independent from 10 Hz to 1 kHz for a particular permittivity of the liquid crystal material, as shown in figure 5(b). There is generally no relaxation for longitudinal or transverse permittivity of liquid crystals at low frequencies. These facts clarified why the drop in threshold voltage at low frequencies could not be explained by the permittivity of the liquid crystals. Considering the symmetrical circuit of the system and no relaxation for the permittivity of the alignment film at low frequencies, the permittivity of the alignment film could not also be a cause of the drop in threshold voltage.

Numerical and theoretical analyses with equation (9) suggested that the variation of the effective voltage over the liquid crystal layer, dependent on frequency, was governed by the time constant which is equal to the products of resistance and



Figure 6. An equivalent circuit for the liquid crystalline system involving the electrical double layer. R_{LC} and C_{LC} stand for resistance and capacitance of the liquid crystal, respectively. R_{AF} and C_{AF} denote those values for the alignment film. C_{ED} symbolizes capacitance of the electrical double layer. V_{LC} , which is to be analysed, is the effective voltage over the liquid crystal layer.

Table 2. Summary of parameters used in the calculation by the system of figure 6.

| | Alignment film | Liquid crystal |
|---|--------------------|------------------------------------|
| Thickness (d/m) | 5×10^{-8} | 6×10^{-6} |
| Relative permittivity ε_r | 3 | 8 |
| Resistivity (ρ/Ω cm) | 1011 | 1012 |
| Charge carrier concentration $(C_0/\text{mol dm}^{-3})$ | | 10 ⁻¹² -10 ¹ |

capacitance components of the liquid crystal and the alignment film, respectively. If the relation $C_{AF}R_{AF} = C_{LC}R_{LC}$ is valid, there is no frequency dependence for the effective voltage applied to the liquid crystal layer. On the other hand, the relation $C_{AF}R_{AF} > C_{LC}R_{LC}$ produces a smaller effective voltage for the liquid crystal layer at low frequencies, which corresponds to the observed larger threshold voltage. Conversely, a larger effective voltage over the liquid crystal layer is obtained, and corresponds to the observed smaller threshold voltage, if $C_{AF}R_{AF} < C_{LC}R_{LC}$ is valid. In the present case, the drop in threshold voltage could be explained to some extent by the decrease in resistivity of the alignment film. However, there was a limit to this, as described in figure 4. A much smaller threshold voltage at low frequencies was often observed experimentally.

Figure 6 shows an equivalent circuit for liquid crystal devices including the electrical double layer. A capacitance corresponding to the electrical double layer was inserted directly between the liquid crystal and alignment film layers. Parameters used in the calculation, are summarized in table 2. The effective voltage over the liquid crystal layer is described as follows:

$$\frac{V_{\rm LC}}{V_0} = \frac{\frac{1}{R_{\rm AF}} + j\omega(C_{\rm AF} + C_{\rm ED})}{\left(\frac{1}{R_{\rm LC}} + \frac{1}{R_{\rm AF}}\right) + j\omega(C_{\rm LC} + C_{\rm AF} + C_{\rm ED})}$$



Figure 7. Relation between the charge carrier concentration and the thickness of the electrical double layer. Parameters summarized in table 2 were used in this calculation.

$$= \frac{R_{\rm LC}}{R_{\rm LC} + R_{\rm AF}} \cdot \frac{1 + j\omega(C_{\rm AF} + C_{\rm ED})R_{\rm AF}}{1 + j\omega(C_{\rm LC} + C_{\rm AF} + C_{\rm ED})\frac{R_{\rm LC}R_{\rm AF}}{(R_{\rm LC} + R_{\rm AF})}}.$$
(10)

Here $C_{\rm ED}$ symbolizes the capacitance of the electrical double layer and corresponds to equation (7). The analysis of this circuit revealed the same tendencies for the variation of the effective voltage applied to the liquid crystal layer with the resistance and capacitance components of the liquid crystals and alignment film, as those in the system without the electrical double layer. If $(C_{\rm AF} + C_{\rm ED})R_{\rm AF} = C_{\rm LC}R_{\rm LC}$ is valid, there is no dependence of the effective voltage on frequency.

The thickness of the electrical double layer is changeable. Figure 7 shows the extension of the electrical double layer as a function of charge carrier concentration. With increase in the charge carrier concentration which forms the electrical double layer, this layer becomes thinner in order to increase the capacitance component. However, the actual effect of charge carrier concentration in the electrical double layer is described in equation (7). Figure 8 (*a*) indicates the effective voltage over the liquid crystal layer as a function of the charge carrier concentration. The effective voltage over the liquid crystal layer decreased with decreasing charge carrier concentration below 10^{-5} mol dm⁻³. But no clear frequency dependence was found at each concentration as shown in figure 8 (*b*).

3.2. Dielectric dispersion of the electrical double layer

Dielectric theory was applied to the system composed of the liquid crystal and alignment film layers. Figure 9 shows a model of two dielectric layers in which the thickness of the liquid crystal layer is one hundred times larger than that of the alignment film layer, which is typical for liquid crystal cells. In this system, the applied voltage is divided by the capacitance of each dielectric layer and the conductive current is not the same between them. As a result, charge carriers are accumulated at the interface between the liquid crystal and alignment film layers.

The total current of the system is expressed by conductive and displacement



Figure 8. Effects of the charge carrier concentration in the liquid crystal on the effective voltage over the liquid crystal layer (a), and its dependence on frequency (b).

currents. Displacement current dD/dt has a time variation according to the dielectric flux density D

$$i = \sigma_{\rm LC} E_{\rm LC} + \frac{dD_{\rm LC}}{dt} = \sigma_{\rm AF} E_{\rm AF} + \frac{dD_{\rm AF}}{dt},\tag{11}$$

where σ and *E* are the conductivity and electric field. Subscripts LC and AF denote the liquid crystal and alignment film layers. Then with $D_{LC} = \varepsilon_{LC}E_{LC}$, $D_{AF} = \varepsilon_{AF}E_{AF}$, and the external voltage V_0 expressed by $(E_{LC} + E_{AF}/100) \cdot d_{LC}$, the following equation concerning the electric field in the liquid crystal layer is obtained:

$$\frac{dE_{\rm LC}}{dt} + \frac{\sigma_{\rm LC} + 100\sigma_{\rm AF}}{\varepsilon_{\rm LC} + 100\varepsilon_{\rm AF}} E_{\rm LC} = \frac{100\sigma_{\rm AF}}{\varepsilon_{\rm LC} + 100\varepsilon_{\rm AF}} \cdot \frac{V_0}{d_{\rm LC}}.$$
(12)

By applying the first condition, $E_{\rm LC}(0) = \{100\varepsilon_{\rm AF}/(\varepsilon_{\rm LC} + 100\varepsilon_{\rm AF})\}(V_0/d_{\rm LC})$, which was



Figure 9. A model of two dielectric layers in the liquid crystalline system.

introduced by considering that the applied voltage over the system at the first stage was divided by the capacitance component of each dielectric layer, the solution of equation (12) can be obtained

$$E_{\rm LC}(t) = \frac{100\sigma_{\rm AF}}{\sigma_{\rm LC} + 100\sigma_{\rm AF}} \cdot \frac{V_0}{d_{\rm LC}} + \left(\frac{100\varepsilon_{\rm AF}}{\varepsilon_{\rm LC} + 100\varepsilon_{\rm AF}} - \frac{100\sigma_{\rm AF}}{\sigma_{\rm LC} + 100\sigma_{\rm AF}}\right) \frac{V_0}{d_{\rm LC}} \exp\left(-\frac{t}{\tau}\right), \tag{13}$$

where

$$\tau = \frac{\varepsilon_{\rm LC} + 100\varepsilon_{\rm AF}}{\sigma_{\rm LC} + 100\sigma_{\rm AF}}.$$
(14)

After the same treatment is carried out to obtain the time-dependent electric field in the alignment film layer $E_{AF}(t)$ as above, the electrical density q(t) at the interface between the liquid crystal and alignment film layers is described as follows. Here the electrical density is expressed by the difference in conductive currents between the liquid crystal and the alignment film layers

$$q(t) = \int_{0}^{t} (i_{\rm LC} - i_{\rm AF}) dt = 100 \int_{0}^{t} \left[\frac{\sigma_{\rm LC} \varepsilon_{\rm AF} - \sigma_{\rm AF} \varepsilon_{\rm LC}}{\varepsilon_{\rm LC} + 100 \varepsilon_{\rm AF}} \cdot \frac{V_0}{d_{\rm LC}} \cdot \exp\left(-\frac{t}{\tau}\right) \right] dt$$
$$= 100 \left(\frac{\sigma_{\rm LC} \varepsilon_{\rm AF} - \sigma_{\rm AF} \varepsilon_{\rm LC}}{\varepsilon_{\rm LC} + 100 \varepsilon_{\rm AF}} \right) \cdot \frac{V_0}{d_{\rm LC}} \cdot \tau \left\{ 1 - \exp\left(-\frac{t}{\tau}\right) \right\}, \tag{15}$$

where i_{LC} and i_{AF} are the conductive current for the liquid crystal and alignment film layers.

Generally the dielectric flux density is described as follows, using a relaxation function $\alpha(t)$ of the electric field

$$D(t) = \varepsilon_{\infty} E(t) + \int_0^t E(s)\alpha(t-s) \, ds, \qquad (16)$$



Figure 10. Dielectric dispersion and absorption expressed by equation (17). ω_d is the dispersion frequency. ε_{∞} and ε_s are permittivities under infinite frequency and non-vibrating fields, respectively.

 Table 3.
 Summary of parameters used in the calculation of dielectric dispersion frequency of the electrical double layer.

| | Alignment film | Liquid crystal |
|---|------------------------|------------------------|
| $\begin{tabular}{lllllllllllllllllllllllllllllllllll$ | 3 10 ⁻¹¹ | 8 10 ⁻¹¹ |

where ε_{∞} is the permittivity at infinite frequency. The first term $\varepsilon_{\infty} E$ expresses the immediate response by the electric field and the second term corresponds to overlapping effects from the continuously applied electric field. In this case, if the relaxation function $\alpha(t)$ and the time-dependent electric field E(t) are expressed as $\alpha(t) = A \exp(-t/\tau)$ and $E(t) = E_0 \exp(j\omega t)$ respectively (τ : a function of time; A, E_0 : constants), then Debye's equation, which expresses the frequency dependence of the complex permittivity, is valid [20]

$$\varepsilon(\omega) - \varepsilon_{\infty} = \frac{\varepsilon_{\rm s} - \varepsilon_{\infty}}{1 + j\omega\tau},\tag{17}$$

where ε_s denotes the permittivity under the non-vibrating field. The function for Debye's equation is depicted in figure 10. Dielectric dispersion took place when the frequency of the external field was close to a reciprocal of the relaxation time constant τ . The form of equation (15) suggested that interfacial polarization due to charge carriers in the liquid crystal and alignment film layers followed an exponential type relaxation expressed in the same way as $\alpha(t) = A \exp(-t/\tau)$, with the time constant described in equation (14). Therefore the relaxation of the interfacial polarization in the liquid crystalline system could follow Debye's equation.

The most important point seen in this study was that the relaxation time of interfacial polarization in the liquid crystalline system could be estimated with the time constant τ expressed by equation (14). If dielectric dispersion due to the electrical double layer takes place, the capacitance component can be considered to increase with decreasing frequency. Consequently, it produces a smaller impedance of the electrical layer and

a larger effective voltage over the liquid crystal layer. With the time constant τ expressed by equation (14), the dispersion frequency ω_d is approximated as follows, using the parameters in table 3:

$$\omega_{\rm d} = \frac{1}{\tau} = \frac{\sigma_{\rm LC} + 100\sigma_{\rm AF}}{\varepsilon_{\rm LC} + 100\varepsilon_{\rm AF}} = \frac{10^{-11} \,\Omega^{-1} \,{\rm cm}^{-1} + 100 \times 10^{-11} \,\Omega^{-1} \,{\rm cm}^{-1}}{(8 + 100 \times 3) \times 8 \cdot 854 \times 10^{-12} \,{\rm Fm}^{-1}} = 37 \,{\rm Hz}.$$
 (18)

This result suggested that dielectric dispersion could occur in the frequency range in which the drop in threshold voltage occurred. Thus the decrease in impedance of the electric double layer caused by the dielectric dispersion could affect the threshold behaviour at low frequencies and so produce the observed drop in threshold voltage.

4. Conclusions

The drop in threshold voltage experimentally observed at low frequencies was analysed by static and dynamic considerations of interfacial polarization in the liquid crystalline system. Two possible causes were found. One was the decrease in alignment film resistivity. The other was dielectric dispersion of the interfacial polarization due to charge carriers in the system consisting of the liquid crystal and alignment film layers. The dielectric dispersion caused by the electrical double layer in the liquid crystalline system followed Debye's equation of complex permittivity. Its frequency was generally in the frequency range around that for which the drop in threshold voltage was often observed.

Finally more effort should be given to obtaining experimental support for the theoretical considerations, and to developing these using the anisotropy of the liquid crystals. As the drop in threshold voltage has been found to depend on the materials constituting the liquid crystal and the alignment film, further analyses must be carried out to understand the material effects on threshold behaviour at low frequencies.

The authors are especially grateful to Mr N. Kikuchi and Mr S. Hasegawa of Electron Tube and Devices Division and Dr B. O. Myrvold of Hitachi Research Laboratory for helpful discussions. They also wish to thank Drs S. Oh-hara and S. Numata for their valuable suggestions.

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