

REFRACTIVE INDICES AND MOLAR REFRACTIONS IN SOLUTIONS (Çözeltilerde Kırılma indisleri ve molar kırılmalar)

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ÖZET

Bu çalışmada, benzene, methylbenzene ve 1,4-dimethylbenzene ile 1,2-dichloroethane çözeltilerinin kırılma indisleri 25°C de karışımların fonksiyonu olarak ölçüldü. Çözeltilerin molar kırılmaları 25°C de kırılma indislerinden hesaplandı.

Key Words: Refractive Index, Molar Refraction

ABSTRACT:

In this research, refractive indices of the solutions of 1,2-dichloroethane with benzene, methylbenzene and 1,4-dimethylbenzene were measured at 25°C as a function of mixtures. Molar refractions of the solutions were calculated from measurements of refractive indices at 25°C.

INTRODUCTION

The study of short lived complexes formed between polar solutes and aromatic solvents has gained considerable importance recently [1,2]. The nature of these interactions is dependent on the physical properties of the molecules involved. In order to look further, we measured refractive indices of these mixtures and calculated molar refractions. Deviations from additive law for molar refractions may be caused by association in mixtures. But they are not always good evidence of association particularly for weak complexes.

EXPERIMENTAL

Materials and Methods

The liquids used for the measurements in this study were provided by the Merck and Reidel Companies and their purity was 99 %. Refractive indices were measured using an Abbé refractometer. The refractometer was thermostated by circulating water through it from a thermostat. The temperature was maintained constant at (25.0 + 0.1)°C. The refractive indices of pure components are recorded in Table I at 25°C along with the corresponding literature values [3]. In addition, a temperature-controlled pycnometer was used in order to measure the densities of the pure components.

Molar refraction is related to refractive index by the relation:

$$R = \frac{(n^2 - 1)}{(n^2 + 2)} \cdot \frac{M}{d} \quad [1]$$

Molar refraction for a mixture is given by the relation:

$$R_{\text{mix}} = \frac{(n_{\text{mix}}^2 - 1)}{(n_{\text{mix}}^2 + 2)} \cdot \frac{(x_1 M_1 + x_2 M_2)}{d_{\text{mix}}} \quad [2]$$

where n_{mix} and d_{mix} are the refractive index and density of the mixture, x_1 and x_2 are mole fractions, M_1 and M_2 are molecular weights. The values of densities of the mixtures were obtained from volume measurements [4].

RESULTS AND DISCUSSION

The mixtures of varying mole fractions were prepared for the systems of 1,2-dichloroethane with benzene, methylbenzene, 1,4-dimethylbenzene and refractive indices of the mixtures were measured at 25°C. The molar refractions of the mixtures were calculated by the help of eq [2]. The refractive index and molar refraction values of the mixtures are recorded in Table II. In addition, molar refraction values have been plotted in Figure 1.

Table I. Refractive Indices of Pure Components

Substance	n_D	
	Exptl.	Lit. (3)
Benzene	1.4979	1.4979
Methylbenzene	1.4941	1.4941
1,4-Dimethylbenzene	1.4933	1.4932
1,2-Dichloroethane	1.4421	1.4422

TABLE II. REFRACTIVE INDICES AND MOLAR REFRACTIONS OF MIXTURES AT 25 °C

x_1	n_{mix}	$R_{mix}(cm^3)$	x_1	n_{mix}	$R_{mix}(cm^3)$
(x_1) 1,2-Dichloroethane + (x_2) Benzene					
0.0000	1.4979	26.203	0.6349	1.4625	22.860
0.1226	1.4911	25.544	0.7411	1.4565	22.315
0.2114	1.4961	25.068	0.8094	1.4527	21.972
0.3318	1.4794	24.433	0.9296	1.4460	21.370
0.4256	1.4741	23.939	1.0000	1.4421	21.022
0.5406	1.4677	23.342			
(x_1) 1,2-Dichloroethane + (x_2) Methylbenzene					
0.0000	1.4941	31.113	0.6472	1.4604	24.423
0.1138	1.4881	29.893	0.7512	1.4550	23.424
0.2282	1.4822	28.690	0.8365	1.4506	22.578
0.3092	1.4780	27.849	0.9198	1.4462	21.778
0.4305	1.4717	26.601	1.0000	1.4421	21.022
0.5216	1.4670	25.680			
(x_1) 1,2-Dichloroethane + (x_2) 1,2-Dimethylbenzene					
0.0000	1.4933	36.035	0.6272	1.4612	26.358
0.10.76	1.4877	34.308	0.7442	1.4551	24.891
0.2264	1.4817	32.439	0.8326	1.4507	23.380
0.3146	1.4771	31.069	0.9172	1.4463	22.177
0.4418	1.4706	29.122	1.0000	1.4421	21.022
0.5324	1.4660	27.762			

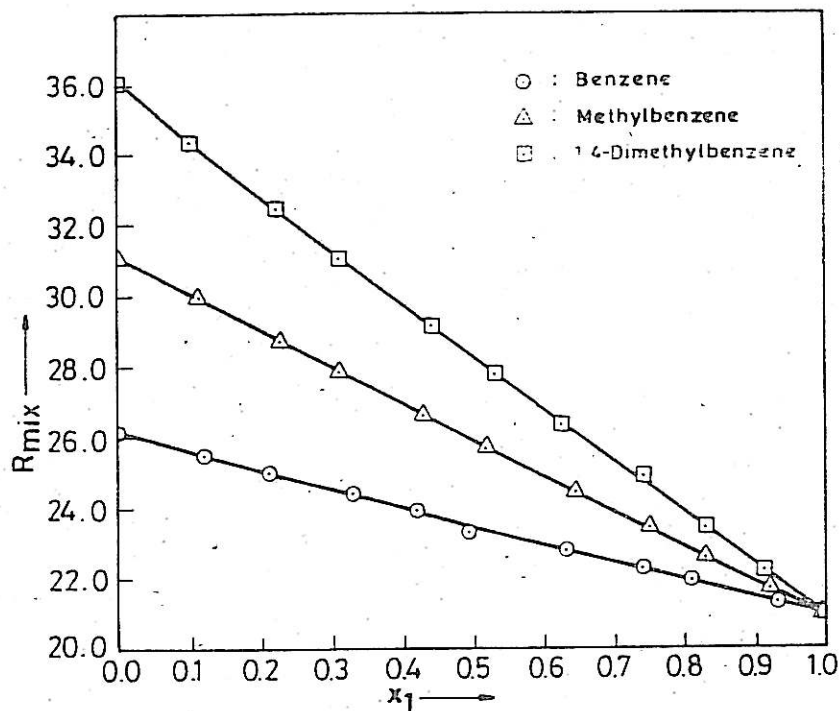


Figure 1. Plots of molar refractions (R_{mix}) of the mixture against mole fraction (x_1) of the 1,2-dichloroethane.

The lines in the figure are drawn to make the best fit of the calculated values. From Figure 1 it is evident that the plots of molar refraction against mole fraction are linear within experimental error. These systems do not possess a complexing nature. Only, there is a slight scatter of points from linear behavior in the high concentration region of the polar components. The deviations from linearity are due to dipole-dipole interactions. Because, although the aromatics are nonpolar, they possess Π -electrons which enable them to be easily polarized.

REFERENCES

1. Foster, R., "Organic Charge-Transfer Complexes", (Academic Press, New York), 1969.
2. Kuntz, I.D., Gasparro, F.P., Johnston, M.D. and Taylor, R.P., J. Am. Chem. Soc., 90 (1968).
3. Timmermans, J., "Physico-Chemical Constants of Pure Organic Compounds", Elsevier, New York, 1964.
4. Dhillon, M.S., Chugh, H.S., J. Chem. Thermodyn., 7, 359 (1975).
5. Nath, J., Sing, G., J. Chem. Eng. Data, 31, 115 (1986).

THE USE OF PHONONS TO STUDY MODULATION DOPED FIELD EFFECT TRANSISTOR (MODFET) STRUCTURES* Modfet Yapılarının İncelenmesinde Fononların Kullanılması

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ABSTRACT

Observation of phonons emitted by the electrons in GaAs-AlGaAs MODFET structures was used to study some features of the transport that are not normally accessible. In particular, two-dimensional (2D) behaviour of the devices could be distinguished from three dimensional (3D), by simply investigating the phonon emission of the channel electrons. We believe that, evidence was found for the creation of hot (non-equilibrium) phonons from the carrier concentration dependence of the phonon emission.

ÖZET

GaAs-AlGaAs MODFET yapılarının normal metodlarla ulaşılamayan ve transportla ilgili bazı özellikleri, kanalı oluşturan elektronların fonon emisyonu kullanılarak çalışıldı. Özellikle, bu aygıtların kanallarını oluşturan elektronların fonon emisyonu incelenerek elektron gazının 2-boyutlu davranışları 3-boyutlu davranışlarından ayırtı edilebildi. Fonon emisyonunun taşıyıcı konsantrasyonuna bağlılığından, sıcak fononların oluşturulmasına ait delil bulduğumuza inanıyoruz.

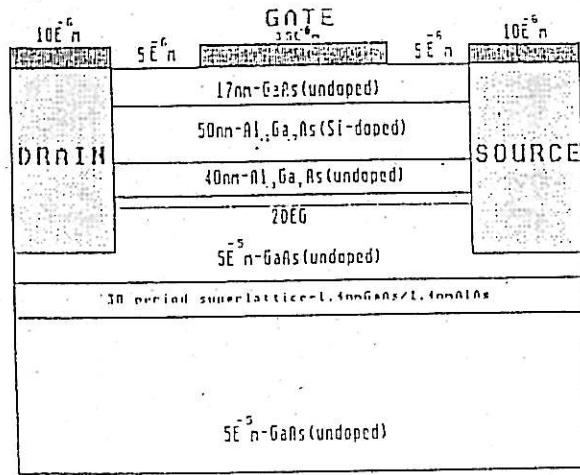
1. INTRODUCTION

The interactions between electrons and phonons play an important role in modern semiconductor devices, especially through the energy loss process which determines carrier mobility [1]. In modulation-doped heterostructures, where elastic scattering by ionised impurities is weak, at high electric field it is the emission of polar optic phonons which limits the mobility, whilst low-field mobility at low temperatures is determined ultimately by the interaction with acoustic phonons [2] Direct observation of the phonons which are involved in the scattering processes is capable of yielding valuable information regarding detailed mechanisms which complements and enhances that obtainable from mobility measurements. Instead of the averages taken over all phonon polarisations, directions and coupling parameters that are involved in the mobility data, direct phonon data possess spatial and temporal resolution through which the detailed physics of the interaction with phonons of different types can be probed [3]. In the present paper, we show how a study of the phonons can be used to assist the modelling of heterostructure-based devices, in particular the modulation-doped field effect transistor (MODFET) [4].

* The experiments were carried out in the University of Lancaster, U.K.

2. THE SAMPLES AND EXPERIMENTAL DETAILS

The normally-on (depletion-type) MODFETs used in these experiments were fabricated from heterostructures grown by Molecular Beam Epitaxy (MBE) on 480 μm thick semi-insulating GaAs substrates, polished on both faces. Figure 1 gives a schematic cross-sectional view of the layer structure.



0.48nm-SI GaAs Substrate (100)

Figure 1. Schematic cross-sectional view of a typical MODFET.

The devices themselves were produced by photolithography and had a gate length of 35 μm and width of 65 μm with total source-drain separation of 45 μm . To detect the phonons, several $65 \times 65 \mu\text{m}$ superconducting aluminium bolometer patterns were defined on the opposite side of the wafers at various angles relative to the normal of the MODFET using an infra-red mask aligner. The carrier concentration and mobility of the basic heterostructure were determined using the van der Pauw-Hall technique and found to be 2.3×10^{15} , $3.3 \times 10^{15} \text{ m}^{-2}$ and 0.81 , $19.6 \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ at temperatures of 300 K and 77K, respectively.

In figure 2, the room temperature characteristics of the particular MODFET, to which the phonon data relate, is shown.

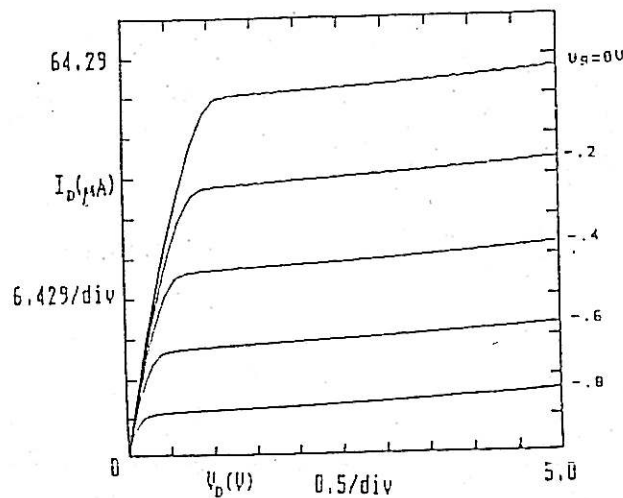


Figure 2: The characteristics of the device obtained by using a conventional AC technique,

to the usual capacitance relation [5]

$$n(x) = \frac{C}{q} [V_g - V_T - V(x)] \quad (1)$$

where C is the capacitance per unit area between the channel and gate, calculated to be $1.14 \times 10^{-3} \text{ Fm}^{-2}$ in the particular device studied, the electronic charge, $n(x)$ is the sheet carrier concentration, and $V(x)$ is the potential at a distance x along the 2 DEG channel. From the room temperature characteristics we measured a threshold voltage V_T of -1.0 V , whilst at about 1.5 K a value of -0.3 V was determined from the phonon emission data, as will be discussed below. The shift in the threshold voltage is attributed to the changing occupation of deep donor traps, DX centres, which are present in the AlGaAs layer [6] and this interpretation was supported by the observation of the effect of light. When the device was illuminated an immediate increase was seen in both phonon signal and drain current with a recovery time of a few seconds. Generally, however, the sample was held in the dark since the light caused sample heating which prevented accurate temperature stabilisation. The simple charge control model is valid only up to a critical gate voltage, above which neutralisation of donors [7] in the AlGaAs causes $n(x)$ and hence drain current to saturate. There is uncertainty as to the effect of further increasing V_g . It has been suggested that, depending on the exact structure, this may result either in population of the doped AlGaAs layer (MESFET conduction) [8], or in a rapid increase of carrier concentration in the 2DEG channel [9]. We shall see that phonon experiments are able to yield significant information on this question.

In order to study the emitted phonons, the carriers were excited by short (10ns) voltage pulses applied between source (earthed) and drain, whilst the gate-source potential was kept fixed. A 50 ohm resistor was connected in parallel across the source drain for improved (but still imperfect) matching to the transmission line. The pulses had to be short enough to allow time resolution of different phonon polarisations and spatial of different phonon directions. However, 10 ns is still slow compared to the dynamics of the carriers, so that they could still be regarded as being in equilibrium. To detect the phonons the temperature was stabilized on the superconducting transition of the Al bolometers ($\sim 1.7 \text{ K}$) with a bolometer resistance of 50 ohm for matching. Detected signals were pre-amplified by a B and H 3GHz-wide bandwidth amplifier and captured, averaged and stored by an EG and G digital boxcar integrator.

3. RESULTS AND DISCUSSIONS

A typical signal from the bolometer directly opposite the MODFET following a 1 V excitation pulse is shown in figure 3.

The main feature is a broad peak whose arrival time corresponded to the transverse acoustic (TA) phonons. A bolometer at an inclination of 30° , on the other hand, showed an additional smaller peak, which corresponds to the velocity of longitudinal acoustic (LA) phonons. The presence of the LA signal only on the 30° bolometer was related to the anisotropy of the piezoelectric coupling. Increasing the power input to the electrons increased the magnitude of the signals with no significant changes in signal shape or arrival time. Phonons could be detected for all source-drain pulse amplitudes greater than 50 mV, corresponding to power dissipation in the channel of less than $10 \mu\text{W}$.

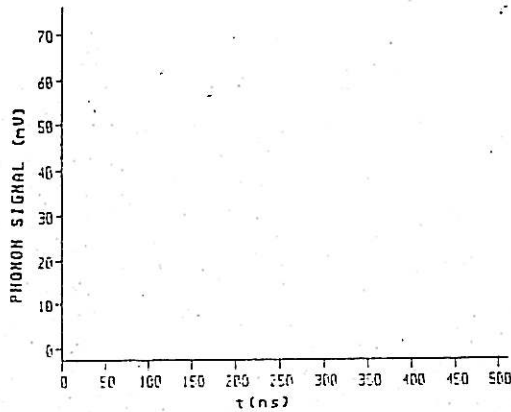


Figure 3: Typical phonon emission signal for $\vartheta = 0^0$ bolometer.

At this level the phonons are expected to be acoustic in character, with a thermal distribution of energies, whilst at higher voltages the emission of the zone centre longitudinal optic mode of energy 36 meV predominates. However, such phonons decay rapidly (~ 7 ps) into LA phonons, which themselves downconvert into lower frequency TA phonons [10]. In addition, for frequencies above 1THz, isotopic (Rayleigh) scattering prevents ballistic propagation [11]. Thus for all but the very lowest excitation voltages and certainly in the normal regime of operation of the MODFET as a device, the phonon (TA) flux is thoroughly randomised and contains little spectral information about the original emission process. Because of the finite parasitic resistance of the source and drain contact pads, the voltage dropped across the channel would be significantly different to that applied to the drain. The actual potential difference could be inferred from variation in phonon signal with drain voltage, by making the reasonable assumption that the total phonon power, integrated over all polarisations, frequencies and directions within the TA feature of Figure 3, was directly proportional to the electrical power dissipated within the MODFET channel. Clearly, a significant amount of energy must also be dissipated by the resistance of the contact pads. However, the resulting phonons arriving at the bolometer were undoubtedly spread out over a much longer time interval than the main TA peak. This would be expected, firstly, because the pad areas are much larger than the area of the 2 DEG channel so that pulses will suffer considerable geometric broadening. In addition, the phonons will be generated purely by Joule heating in very low region in which phonons and electrons are in thermal equilibrium, as opposed to the direct emission by the 2DEG. Finally, because of the very high defect density, the emitted phonons will suffer strong diffusive scattering and hence delay in arrival time.

As a transistor, the MODFET is normally operated for maximum transconductance in the charge control regime defined by [1]. However, there has been considerable interest in attempting to understand the physics that prevents the unrestricted increase of $n(x)$ through further increasing V_g .

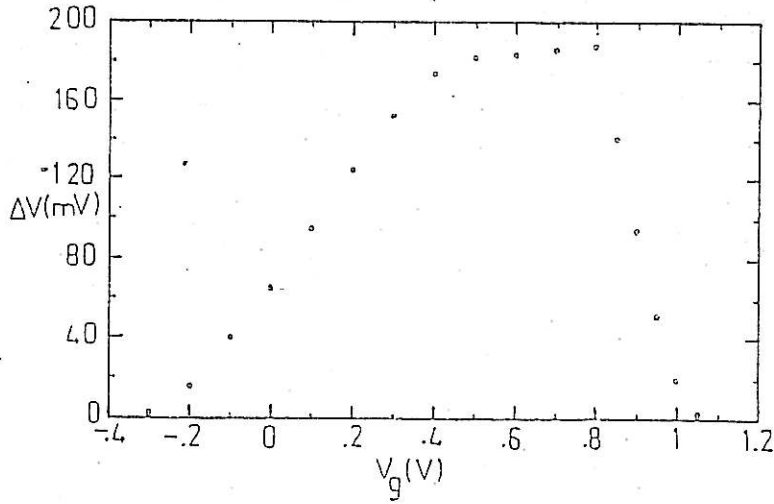


Figure 4: Gate voltage dependence of the emitted phonon signal, ΔV .

Figure 4 shows for our MODFET the data for the phonon signal, ΔV , versus V_g taken in this range. with pulse drain voltage, V_D , constant at 3.0 V, that is, in the saturated region of the characteristic. The flattening of the phonon curve is well known to be due to the neutralisation of Si donors in the AlGaAs [7], and it is clear that, apart from deviations due to the parasitic resistances, the phonon signal also mirrors this effect. The voltage defining the end of this plateau is given by [8].

$$V_p = \frac{n_o q}{C} \quad (2)$$

Where n_o is the equilibrium 2DEG concentration, obtained when the AlGaAs contains a significant region in which the conduction band is flat. For our device, n_o is calculated to be $5.4 \times 10^{15} \text{m}^{-2}$ and hence $V_p = 0.71 \text{ V}$.

It is less clear what happens as V_g is increased beyond V_p ; two different effects are predicted, depending on the exact structure of the device. Firstly, when V_g approaches V_B , the gate-Schottky barrier height, the appearance of gate current is expected⁹. In our experiments, this was observed both directly, and as a discontinuity in drain current, which otherwise remains constant. Secondly, it has been predicted that, as V_g increases beyond V_p , the conduction band itself in the AlGaAs falls below the Fermi energy, resulting in increased carrier concentration in the AlGaAs, and hence parallel conduction. At the same time the 2DEG concentration is expected to remain constant at the equilibrium value [7-9].

Our data are not at first sight consistent with this model. Although the total drain current observed does increase slightly with increasing V_g , the 2DEG channel current as indicated by the phonon signal—the small corrections for the changes that occur in voltage across the channel as its resistance varies do not effect this conclusion—falls rapidly. Clearly, this decrease in current might indicate a decrease in either carrier density or mobility.

An alternative possibility, proposed by Ponse et al [9] on the basis of a model in which quasi-Fermi level bending is taken fully into account, is that the 2DEG carrier density actually increases for $V_g > V_p$. The argument is that the carrier density in the regime is controlled by the gate diffusion (drift) current, not by the gate voltage.

Then the explanation for the decrease in phonon signal in this regime is sought in terms of a change in carrier mobility rather than in carrier density. The observations would then be explained in terms of the shift between elastic and inelastic scattering contributions [12], μ_E and μ_I respectively, to the total mobility, μ , of the carriers. By Matthiessen's rule.

$$\mu^{-1} = \mu_E^{-1} + \mu_I^{-1} \quad (3)$$

If the number of 2DEG carriers is constant, then the drain current is determined by μ , whereas the emitted phonon power gives a handle directly on μ_I , since phonon emission is the mechanism of energy loss by the electrons. The primary effect of varying V_g would be to increase μ_I , thus decreasing phonon emission, whilst decreasing μ_E by a similar amount and thus keeping μ , and hence current approximately constant. A possible explanation of the change in mobility is that the rapid increase in 2DEG carrier concentration proposed by Ponse et al⁹, results in a hot phonon bottleneck [13-14], thus inhibiting optic phonon emission by the electrons and resulting in an increased μ_I , such a conclusion would be extremely interesting since "hot phonons", have not been directly observed before.

In summary, the emission of the phonons was studied in a MODFET structure. The gate voltage dependence of the phonon emission demonstrated a very clear transition from a 2DEG channel to a 3DEG one. By studying the emission data, we were able to extract information about the transport parameters in a MODFET device.

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REFERENCES

1. Shah J, Pinczuk A, Stormer HL, Gossard A, and Weigmann W, Appl. Phys. Lett. 44,322(1984).
2. Daniels ME, Ridley BK and Emeny M, Solid-State Electronics, 32, 1207 (1989).
3. Wigmore JK, Erol M, Sahraoui-Tahar M, Wilkinson CDW, Davies JH and Stanley C, Semicond. Sci. Technol. 6, 837 (1991).
4. Drummond TJ, Masselink WT and Morkoç H, Proceedings of the IEEE 74, 6, 773 (1986).
5. Delagebeaudeuf D and Linh NT, IEEE Trans Electron. Devices, ED-29,955 (1982).

6. YOUNG RJ, ROBINSON GT, LEE K and SHUR MS, J. VAC. SCI. TECHNOL. B1, 190 (1984).
7. Masselink WT, Drummond TJ, Klem J, Kopp W, Chang YC, Ponce F and Morkoç H, IEEE Trans. Electron Devices. ED-32, 713 (1985).
8. Lee K, Shur MS, Drummond TJ and Morkoç H, IEEE. Trans. Electron Devies, ED-31, 29(1984).
9. Ponce F, Masselink WT and Morkoç H, IEEE Trans. on Electron Devices, ED-32, 1017 (1985).
10. Tamura S and Maris H.J, Phys. Rev. B-31, 2595 (1985).
11. Tamura S, Pyhs. Rev. B-31, 2574 (1985).
12. Marchetti MC, Lai W and Lax M, Solid State Electron, 31, 677(1988).
13. Ridley BK, Semicond. Sci. Technol, 4, 1142 (1989).
14. Price PJ, Physica 134 B, 164(1985).