



# 应崇福论文选集

中国科学院声学研究所编

中国科学技术出版社



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## 前言

本选集选登了我 1998 年以前所发表的一些主要科学论文,共 44 篇。

在求学时代我一直攻读物理专业。大学毕业论文是关于当时出现不久的电视物理的一篇综述,硕士学位论文和博士学位论文分别是本选集中的第一和第二两篇文章,前者涉及统计力学,后者涉及电子物理。多年来我常想找个机会说明一下,硕士学位论文(1945 年)是在王竹溪先生指导下完成的,而且不只是一般指导,是耗费了王先生相当多的精力。王先生有好几件事令我留下深刻印象。精心指导论文,却只让在文章发表时写上 *gratitude for very kind help*,这是其中之一。另一件事是他的大褂。当时王先生常穿一件竹布大褂,时间一长,大褂早已在原浅蓝色上泛出红色。西南联大许多名教授,生活十分艰苦,却安贫乐道。正是王先生这样的旧大褂,延续、滋润着中国民族的精华。

1951 年我在美国布朗大学完成博士学位的要求后,因为回不了国,在应用数学系找了个临时工作。这样决定了我一生从事超声学研究的命运。1951 年以后我的论文便全涉及超声学,虽然在读书时,我没有读过声学。

1951 年到 1955 年,种种环境促使我的工作自然形成学科的前沿,并且几乎全是独立完成的。这里我要记下对 Rohn Truell 教授的怀念。他是我那时工作实验室的负责人,因此也是他在我回国受阻时给予了支持,并且在以后几年里比较尽力地为我提供研究上的便利。尤其是,在我准备回国时,他几次提到,将来要来中国看看,看是怎样一个国家,让我那样执著回归。可惜他在 1967 年早逝,我永远不能在北京接待他了。

1955 年底回国,1956 年到中国科学院工作。在本选集中却没有我 1956 年至 1978 年的论文。在这个阶段,前半部时间主要花在推广超声学和承担国家任务,所写文章没有送期刊发表。后半部时间则在文化大革命中度过。直到约 1978 年,才回到基础研究和应用研究,同一些同事和学生合作写了些常规的论文,这些构成本选集的大部内容。也许可以说,本选集不仅记录了我的学术努力,也多少记录了生涯的风风雨雨。

1980 年以后,论文涉及的面在超声学的总范围内还是相当广的。这原因是由于我的工作职务需要照顾的面比较宽。举个例说,指导医学超声研究的导师出国了,几批学生的论文只好由我来接替指导。近几年来则还有研究经费的因素。好在我多年来已习惯于“阵地转移”。

在本选集所刊登为数不多的论文中,我仅对极少数的外界反应有所了解。编号为 5,有关固体中球体的超声散射的 1956 年论文是其中最受重视的一篇。据中国科学院文献情报中心今年(1997 年)进行的检索,根据 SCI 统计,从 1980 年到 1996 年被国外刊物总共引用了 147 次,其中,1996 年(这是论文发表的 40 周年)是 7 次,1995 年是 9 次。这里缺少 1956~1979 年的数据。此外,有不少国内外期刊,SCI 并不统计;还有些作者则只是间接引用。

本选集的出版,得到了声学研究所两届领导和不少同志们的关怀、支持和劳动付出。也得到中国科学院学部的关怀。这里谨致深谢。

应素敏  
C. J. Ying 1997.8

## 内 容 简 介

本集选登了中国科学院院士应崇福自 1945 年至 1997 年发表的物理方面的主要论文,44 篇。应崇福院士自 1951 年起专业从事超声方面的研究,因此这些论文除 3 篇外都涉及超声学,包含固体中超声的散射、超声压电换能器的特性、压电体内瞬态声波的产生和传播、生物组织内声波的传播、激光超声在导体和非导体中的指向性图案、井孔声场、功率超声等方面。这些论文,除 1 篇是会议报告外,其余都发表在国内外学术期刊上,绝大多数属基础研究或应用基础研究的性质,其中 1 篇,发表已 40 年,迄今仍每年被引用 5 次以上。

# 应崇福院士论文选集

(1998 年以前)

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## 一、1945~1955 期间论文

# ON THE APPROXIMATE PARTITION FUNCTION IN THE STATISTICAL THEORY OF ADSORPTION \*

By C. F. Ying (应崇福)

*Department of Physics, Huachung University*

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### Abstract

Wang's theory for determining the approximate configurational partition function of the adsorbed layer is modified in two different ways. One is to assume that the configurational energy should be corrected; the other to advocate that the deficiency due to a wrong expression for the a priori probability of the central site is more significant. The configurational partition function is evaluated in both methods and the adsorption isotherm and the heat of adsorption computed for the case of quadratic lattice with dipole interaction. Values for the last two quantities when a uniform continuous distribution of the distant adsorbed particles is assumed are further given for comparison. The second method, which surpasses the first, is compared with Kirkwood's method in the case of hexagonal lattice with neighbour interaction. Numerical work is also carried out in this case.

### 1. Introduction

In Bethe's theory, as applied to the adsorption of gas particles<sup>\*\*1,2,3</sup>, the partition function for the whole assembly of adsorbed particles is not evaluated. What actually is evaluated is only the partition function of a small group of particles chosen out of the whole. Thus although the consequences deduced from this "local" partition function are assumed to hold for the entire adsorbed layer, the local partition function itself can evidently not be regarded as equivalent to the true partition function of the whole assembly. In certain problems, such as the critical condensation phenomenon, the latter, however, is indispensable. Wang<sup>4</sup> accordingly advanced a device for

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\* An abridged version of an M.Sc. dissertation, National Tsinghua University, 1945.

\*\* The term "particle" represents either a molecule or an atom as the case may be. In this paper the problem of dissociation is not considered.

1. R. Peierls, *Proc. Camb. Phil. Soc.* 32(1936), 471.

2. J. S. Wang, *Proc. Roy. Soc., A*, 161(1937), 127.

3. J. S. Wang, *Proc. Camb. Phil. Soc.*, 34(1938), 238.

4. J. S. Wang, "Free Energy in the Statistical Theory of Order-Disorder Transformation". *Science Reoort of National Tsinghua University*, series A. (1941). (printed but failed to appear).

its approximate evaluation without breaking from Bethe's scheme. Nevertheless the theory suffers from two defects. One is theoretical and the other practical. In the special case of neighbour interaction it leads to the conclusion that the condition of mathematical consistency is automatically satisfied so that a partition function always exists. This is not true in general. A second defect of the theory is that the numerical work involved is definitely laborious. It seems the theory should first be modified. Two different ways to effect this are suggested and formulated here. One way is to modify the energy expression (§2) and the other to modify the formulas of equilibrium distribution (§3).

## 2. The method of modifying the configurational energy expression

We shall still take the condition of mathematical consistency as our starting point, just as in Wang's paper<sup>4</sup>. The original expression for the configurational energy of the adsorbed layer was (Wang<sup>4</sup>, eq.(11))

$$E_c = \frac{1}{2} N \theta (U_0 + f_{1\tau} V_1) \quad (1)$$

where  $N$  is the total number of sites available for adsorption,  $\theta$  is the fraction of sites occupied by adsorbed particles,  $V_1$  is the interaction between two neighbouring adsorbed particles,  $U_0$  is the interaction of an adsorbed particle with all the adsorbed particles other than its nearest neighbours and is given by (Wang<sup>4</sup>, eq.(12), obtained upon the assumption of uniform distribution)

$$U_0 = \theta (U^* - z V_1) \quad (2)$$

$U^*$  being the interaction of an adsorbed particle with all other adsorbed particles when all sites are occupied and  $z$  being the number of sites neighbouring to any given site. The symbol  $f_{1\tau}$  is an abbreviation for the function  $f_1(e^\tau \epsilon)$ , where

$$\tau = -V_1/kT \quad (3)$$

and

$$f_1(x) = x \frac{d}{dx} \ln f(x) \quad (4)$$

$f(x)$  being the basic function in Wang's theory<sup>3</sup>(see eq.(23), §4 below). In Wang's paper<sup>4</sup> cited above the expression (2) was modified while (1) was assumed to be correct. Now a different procedure will be adopted. We shall modify the expression (1) for the configurational energy by adding a term  $NV_1w$  to it, but (2) will be retained:

$$E_c/NV_1 = \frac{1}{2} \theta (\theta v^* + f_{1\tau}) + w \quad (5)$$

where

$$v^* = (U^*/V_1) - z \quad (6)$$

The quantity  $w$  will be determined by the condition of mathematical consistency, namely, that the partial differential equations<sup>4</sup>

$$\frac{1}{N} \frac{\partial}{\partial \tau} \ln Q = \frac{E_c}{NV_1} \quad (7)$$

$$\frac{1}{N} \frac{\partial}{\partial \theta} \ln Q = -\ln \xi \quad (8)$$

which determine the required configurational partition function  $Q(\theta, \tau)$ , should be integrable. This requires the relation

$$\frac{\partial}{\partial \theta} \frac{E_c}{NV_1} = -\frac{\partial}{\partial \tau} \ln \xi \quad (9)$$

to hold, in which  $E_c$  is given by (5) and  $\xi$  is determined by<sup>3</sup>

$$\frac{\theta}{1-\theta} = \xi e^{-U_0/kT} \frac{f_\tau}{f} \quad (10)$$

where  $f$  and  $f_\tau$  stand for  $f(\epsilon)$  and  $f(e^\tau \epsilon)$  respectively. The argument  $\epsilon$  in the function  $f$  is determined as a function of  $\theta$  and  $\tau$  by the equation

$$\frac{\theta}{1-\theta} = \frac{f_1}{z - f_{1\tau}} \quad (11)$$

Substituting (5) and (10) into (9), we obtain

$$\frac{\partial w}{\partial \theta} = \frac{\partial}{\partial \theta} \ln \frac{f_\tau}{f} - \frac{\partial}{\partial \theta} \left( \frac{1}{2} \theta f_{1\tau} \right) \quad (12)$$

To solve this equation we remember that (1) holds at  $\theta = 1$ . We can therefore take it as the boundary condition for  $w$  at  $\theta = 1$ . The solution is

$$w = \frac{1}{2}(z - \theta f_{1\tau}) - \int_\theta^1 \frac{\partial}{\partial \tau} \left( \ln \frac{f_\tau}{f} \right) d\theta \quad (13)$$

The configurational partition function  $Q$  is then, from (7) and (8),

$$\begin{aligned} \frac{1}{N} \ln Q = & \int_0^\tau \left\{ \frac{1}{2}(z + \theta^2 v^*) - \int_\theta^1 \frac{\partial}{\partial \tau} \left( \ln \frac{f_\tau}{f} \right) d\theta \right\}_{\theta=0} d\tau \\ & + \int_0^\theta \left\{ -\ln \frac{\theta}{1-\theta} + \ln \frac{f_\tau}{f} + \theta v^* \tau \right\} d\theta \end{aligned}$$

the path of integration being so chosen that  $\theta$  is first held constant at 0 while  $\tau$  varied from 0 to  $\tau$  and then  $\tau$  held constant at  $\tau$  while  $\theta$  varied from 0 to  $\theta$ , or

$$\frac{1}{N} \ln Q = -\theta \ln \theta - (1-\theta) \ln(1-\theta) + \frac{1}{2}(z + \theta^2 v^*)\tau - \int_\theta^1 \ln \frac{f_\tau}{f} d\theta \quad (14)$$

Equation(10) represents the adsorption isotherm in the present method. The heat of adsorption  $q$  can be calculated<sup>2</sup> either from the formula

$$q_0 - q = \frac{1}{N} \frac{\partial E_c}{\partial \theta} \quad (15)$$

or from

$$q_0 - 1 = -V_1 \frac{\partial}{\partial \tau} \ln \xi \quad (16)$$

for here the configurational partition function exists. The result is

$$\frac{q_0 - q}{V_1} = \frac{\partial}{\partial \tau} \ln \frac{f_\tau}{f} + \theta v^* \quad (17)$$

$q_0$  in these equations is the value of  $q$  at  $\theta = 0$ .

While yielding simpler formulas than the method of modifying  $U_0$  the present method can only be applied to the case of adsorption without super-lattice formation. In the case of superlattice, on the other hand, there are three equations expressing the condition of integrability<sup>4</sup>, of which one does not involve  $E_c$ . It is evident that the present method will not work if this particular equation is not satisfied.

### 3. The method of modifying the a priori probability expression

we shall in this section attempt to formulate a more suitable method. It is the method of modifying the quantity  $\xi$ . The physical background is as follows:  $\xi$  is the statistical weight or the a priori probability of the central site. Since the condition of integrability (9) is not satisfied we argue that the procedure of assigning a parameter  $\epsilon$  to each of the first shell particles is too rough and should be supplemented by assigning also a factor to the central site. We shall therefore add a factor  $e^\sigma$  to  $\xi$  and thus alter the equation (10) to

$$\frac{\theta}{1 - \theta} = \xi e^\sigma e^{\theta v^* \tau} \frac{f_\tau}{f} \quad (18)$$

The quantity  $\sigma$  will be determined by (9), in which the value of  $\xi$  determined by (18) should be used, while  $E_c$  is still given by (1). Thus we have the following differential equation for determining  $\sigma$ :

$$\frac{\partial \sigma}{\partial \tau} = \frac{\partial}{\partial \theta} \left( \frac{1}{2} \theta f_{1\tau} \right) - \frac{\partial}{\partial \tau} \ln \frac{f_\tau}{f} \quad (19)$$

In order to solve this equation we investigate the value of  $\sigma$  at  $\tau = 0$ . When  $\tau$  approaches zero,  $T$  approaches  $\infty$ , the Boltzmann factor in the configurational partition function approaches 1, and  $Q$  reduces to the expression for the case of no interaction between the adsorbed particles, when  $\xi$  is related to  $\theta$  by the equation  $\xi = \theta/(1 - \theta)$ , and consequently  $\sigma$  becomes 0 as  $\tau \rightarrow 0$ . With this boundary condition we obtain from (19)

$$\sigma = \ln \frac{f}{f_\tau} + \frac{1}{2} \int_0^\tau \frac{\partial}{\partial \theta} (\theta f_{1\tau}) d\tau \quad (20)$$

On first fixing  $\theta$  at zero, but varying  $\tau$  from 0 to  $\tau$  and then fixing  $\tau$  at  $\tau$  while varying  $\theta$  from 0 to  $\theta$ ,  $Q$  is found to be

$$\begin{aligned} \frac{1}{N} \ln Q = & \int_0^\tau \left\{ \frac{1}{2} \theta^2 v^* + \frac{1}{2} \theta f_{1\tau} \right\}_{\theta=0} d\tau \\ & + \int_0^\theta \left\{ -\ln \frac{\theta}{1 - \theta} + \sigma + \theta v^* \tau + \ln \frac{f_\tau}{f} \right\} d\theta \end{aligned}$$

or

$$\frac{1}{N} \ln Q = -\theta \ln \theta - (1 - \theta) \ln(1 - \theta) + \frac{1}{2} \theta^2 v^* \tau + \frac{1}{2} \int_0^\tau \theta f_{1\tau} d\tau \quad (21)$$

For the heat of adsorption we have

$$\frac{q_0 - q}{V_1} = \frac{1}{2} \frac{\partial}{\partial \theta} (\theta f_{1r}) + \theta v^* \quad (22)$$

It is to be noted that the present method has not the flaw the previous methods of modifying  $U_0$  and modifying  $E_c$  possess. The present method is numerically much simpler than Wang's original method and is also applicable to the case of superlattice. In the case of superlattice there are two  $\xi$ 's (see a recent paper of Wang<sup>5</sup>) and hence there will be two  $\sigma$ 's if the present method is applied. The three partial differential equations of the two  $\sigma$ 's expressing the condition of integrability can be verified to form a complete Jacobian system so that the mathematical consistency of the method is assured and the equations can be integrated.

It may be mentioned that the present method is mathematically equivalent to one recently proposed by Wang<sup>5</sup>, although they start from different physical principles.

#### 4. Numerical calculations for a quadratic lattice with dipole interaction

For a quadratic lattice with dipole interaction the function  $f(x)$  takes the form<sup>3</sup>

$$f(x) = 1 + 4x + (4e^{\frac{1}{2}\sqrt{2}r} + 2e^{\frac{1}{8}r})x^2 + 4e^{(\frac{1}{2}\sqrt{2} + \frac{1}{8})r}x^3 + e^{(\sqrt{2} + \frac{1}{4})r}x^4 \quad (23)$$

and  $z = 4$ ,  $v^* = 5.0336$  (this value of  $v^*$  has been calculated by Topping<sup>6</sup>). Theoretically the configurational partition function is fundamental, but experimentally only the adsorption isotherm and the heat of adsorption can be measured. We shall, therefore, calculate only the last two quantities.

It can be verified from the above form of  $f(x)$  that the following relation holds:

$$f(x) = (e^{\mu r/2} x)^z f(e^{-\mu r} x_{-1}) \quad (24)$$

where  $z=4$  and

$$\mu = \frac{1}{2}\sqrt{2} + \frac{1}{8}$$

With the help of this relation it can be proved that

$$\frac{q_0 - q}{V_1} = \frac{1}{2}(z + v^*)$$

is an odd function of  $\theta - \frac{1}{2}$ , both in the method of modifying  $E_c$  and in the method of modifying  $\xi$ . Consequently it is only necessary to calculate  $q$  for  $\theta < \frac{1}{2}$ .

As has been pointed out by Wang himself<sup>4</sup>, the heat of adsorption as originally given for the case when the distant adsorbed particles are uniformly and continuously distributed<sup>3</sup> is in error.

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5. J. S. Wang, "Approximate Partition Function in Generalized Bethe's Theory of Superlattices", (to be published).

6. J. Topping, *Proc. Roy. Soc., A*, 114(1927), 67.

We shall calculate its correct numerical values. The correct formula is (Wang<sup>4</sup>, footnote 10)

$$\begin{aligned} \frac{q_0 - q}{V_1} = & \left(1 - \frac{2\pi}{u^2 G'}\right) \left\{ \phi_\tau - \phi + \frac{f_{1\tau}[(z - f_{1\tau})f_2 + f_1 f_{2\tau}]}{(z - f_{1\tau})f_2 + f_{1\tau} f_{2\tau}} \right. \\ & \left. + \frac{(f_1 - f_{1\tau})[(z - f_{1\tau})\phi_1 + f_{1\tau}\phi_{1\tau}]}{(z - f_{1\tau})f_2 + f_{1\tau} f_{2\tau}} \right\} \\ & + \frac{2\pi}{u^2 G'} \frac{f_{1\tau} f_{2\tau} + (z - f_{1\tau})\phi_1 + f_{1\tau}\phi_{1\tau}}{(z - f_{1\tau})f_2 + f_{1\tau} f_{2\tau}} + K a_1^2 \theta \frac{2\pi}{u} \left(1 + \frac{G}{u G'}\right) \end{aligned} \quad (25)$$

where

$$f_2 = f_2(\epsilon) = \epsilon \frac{d}{d\epsilon} \ln f_1$$

$$\phi = \left( \frac{\partial}{\partial \tau} \ln f \right)_\epsilon, \quad \phi_1 = \left( \frac{\partial}{\partial \tau} \ln f_1 \right)_\epsilon$$

the differentiation with respect to  $\tau$  in  $\phi$  and  $\phi_1$  being taken at constant  $\epsilon$ . Equation (25) was obtained by utilizing (16) and evidently would be the same as (17) were the two  $U_0$ 's not different.

On starting from (15) we can get for  $q$  in the same case of uniform continuous distribution another expression which will be different from (25) since (15) and (16) are now no more equivalent. We have  $U_0 = 2\pi\theta K a_1^2 V_1/u$  and consequently

$$\begin{aligned} \frac{q_0 - q}{V_1} = & \frac{f_{1\tau}}{2} \left\{ 1 - \frac{(z + f_1 - f_{1\tau})f_{2\tau}}{(z - f_{1\tau})f_2 + f_{1\tau} f_{2\tau}} \right\} \\ & + \frac{\pi}{u^2 G' \tau} \frac{1}{1 - \theta} \left\{ 1 - \frac{(z - f_{1\tau})(1 - f_1 + f_{1\tau})}{(z - f_{1\tau})f_2 + f_{1\tau} f_{2\tau}} \right\} \\ & + K a_1^2 \theta \frac{2\pi}{u} \left(1 + \frac{G}{2u G'}\right) \end{aligned} \quad (26)$$

Numerical values of this expression are also calculated.

In calculation we assign definite values to  $\tau$ . When  $\tau = -0.25$  the adsorption isotherms and variations of the heat of adsorption with  $\theta$  are shown in figures 1 and 2. It will be seen that for this value of  $\tau$  the two methods yield indistinguishable results. When  $\tau = -7.5$ , however, the corresponding values are different. These are shown, together with the values for uniform continuous distribution, in figures 3 and 4. For both values of  $\tau$  the two methods do not agree with the method assuming uniform continuous distribution.

We also compute for one value of  $\tau$  the quantity  $w$  of the method of modifying  $E_c$ . This measures the deviation from the ordinary expression of  $E_c$ . With the help of (24) it can be shown that  $w$  is an even function of  $\theta - \frac{1}{2}$ . The numerical values of  $w$  for  $\theta < \frac{1}{2}$  when  $\tau = -7.5$  are plotted in figure 5. Incidentally it may be remarked that the quantity  $\sigma$  of the method of modifying  $\xi$  is an odd function of  $\theta - \frac{1}{2}$ .

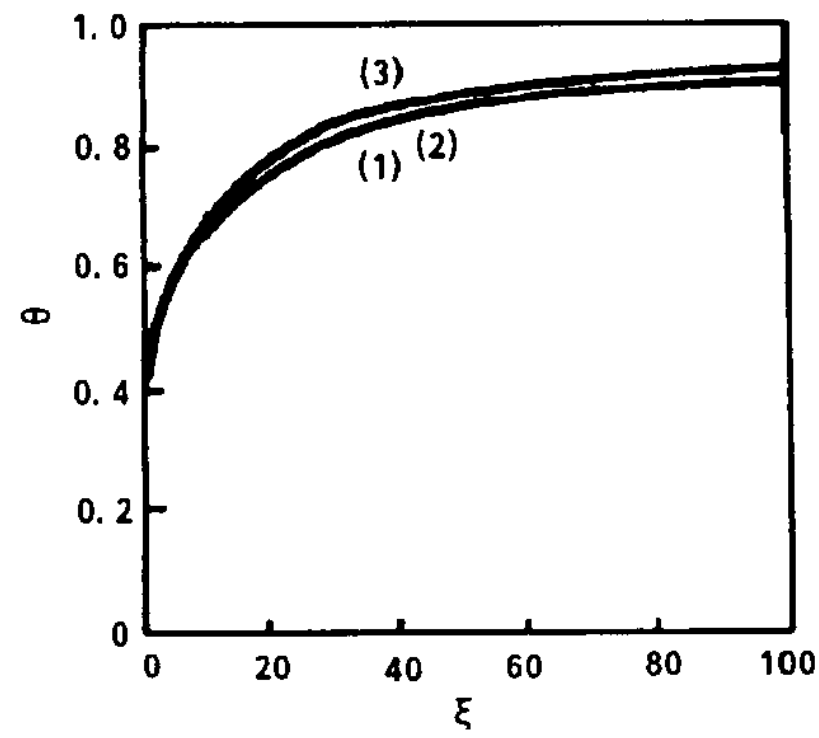


Fig.1—The adsorption isotherm for a quadratic lattice when  $\tau = -0.25$ .

Curve(1),the method of modifying  $E_c$ .

Curve(2),the method of modifying  $\xi$ .

Curve(3),the old theory of uniform continuous distribution.

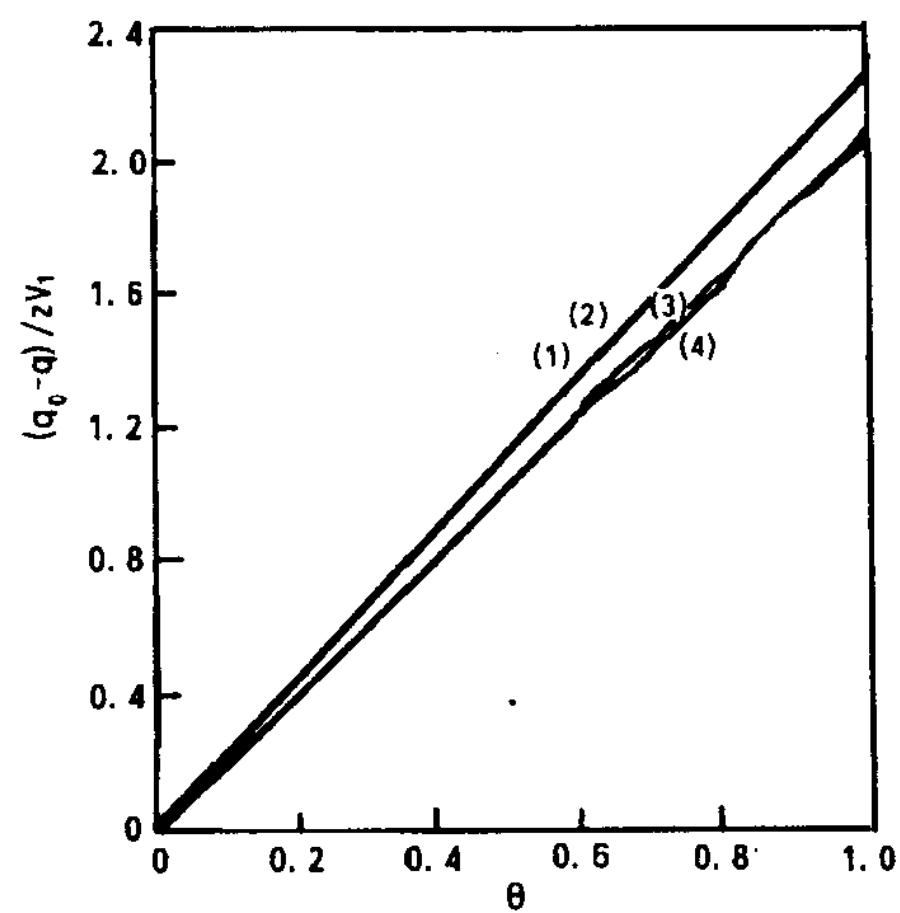


Fig.2—Variation of the heat of adsorption with  $\theta$  for a quadratic lattice when  $\tau = -0.25$ .

Curve(1),the method of modifying  $E_c$ .

Curve(2),the method of modifying  $\xi$ .

Curve(3),the old theory of uniform continuous distribution,from eq.(16).

Curve(4),the old theory of uniform continuous distribution,from eq.(15).



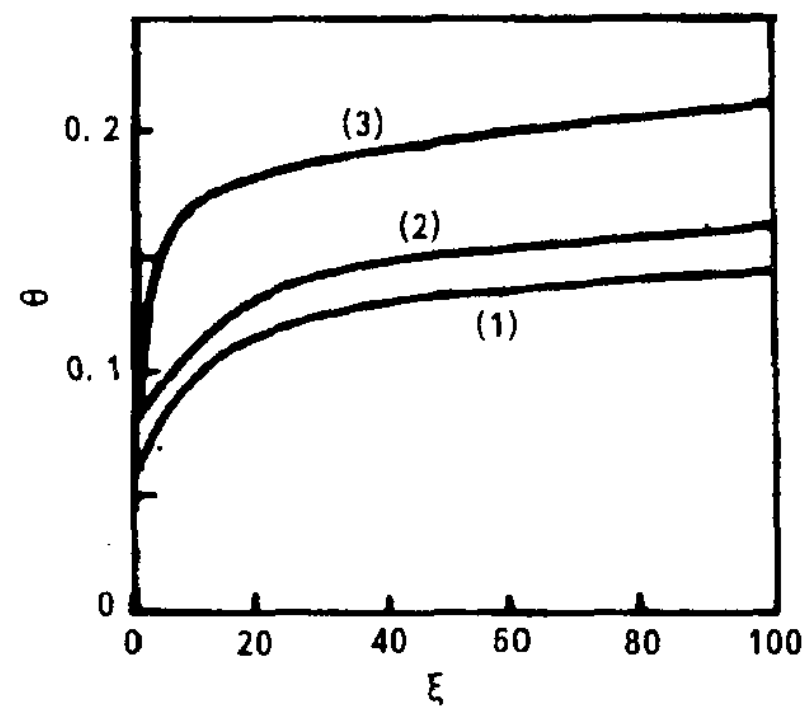


Fig.3—The adsorption isotherm for a quadratic lattice when  $\tau = -7.5$ ,  
Curves numbered as in fig.1.

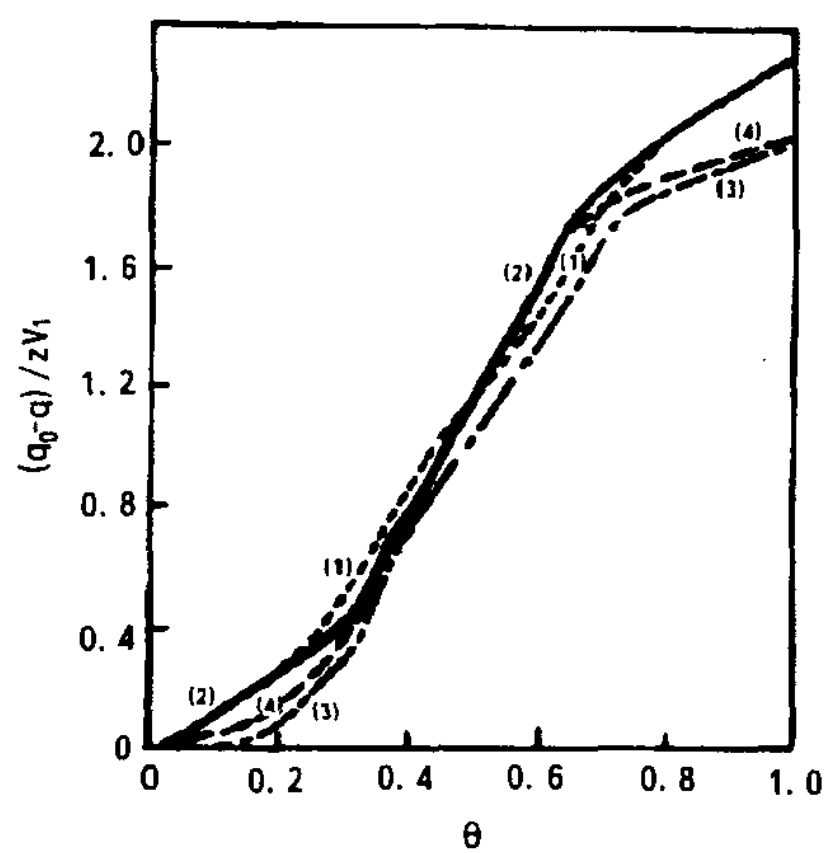


Fig.4—Variation the heat of adsorption with  $\theta$  for a quadratic lattice  
when  $\tau = -7.5$ , Curves numbered as in fig.2.

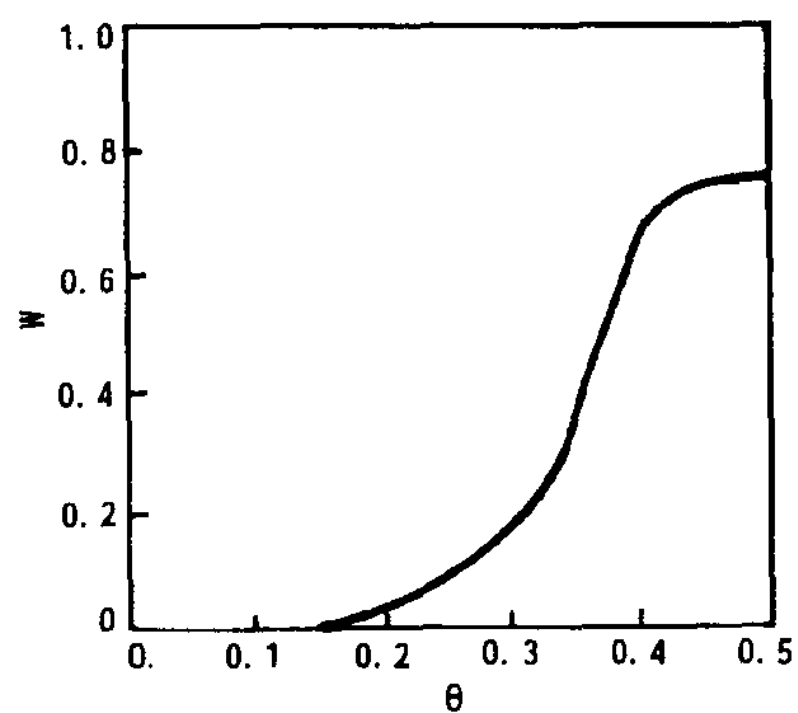


Fig. 5—Variation of the correction term of  $E_c$  with  $\theta$   
for a quadratic lattice when  $\tau = -7.5$ .