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再论铁电体的热-电-力耦合内场*

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摘要:基于铁电体的热-电-力耦合内场的性质研究,本文进一步探讨其内场的其他性质。由于力学因素使问题变得复杂,故忽略力学因素,以简化后的物性方程为出发点,以一维体系为例推导了内电势与温度之间的关系,计算了内电场和有关的热力学力,给出了确定内电势函数中的积分常数的方法。研究表明,内电势是温度的线性函数,其中的积分常数可由内电场、温度梯度及输运系数确定。这有助于更深入地了解铁电-顺电相变,特别是一级铁电-顺电相变的内在规律。

关键词:铁电体;内场;热-电-力耦合;非平衡态热力学
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Thermo-electro-mechanically coupled internal fields of ferroelectrics revisited*

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Abstract: Based on the properties studies of the thermo-electro-mechanically coupled internal fields of ferroelectrics, the other properties of these internal fields are further discussed in this paper. The mechanical factors are neglected because they make the problem more complicated. Taking the simplified physical property equation as the starting point, the relation between the internal electric potential and the temperature is derived by taking a one-dimensional system as an example. The internal electric field and the relevant thermodynamic forces are calculated, and the method for determining the integral constants in the internal electric potential function is given. It is shown that the internal electric potential is a linear function of the temperature, and the integral constants can be determined by the internal electric field, the temperature gradient and the transport coefficients. It is helpful to understand the inherent law of ferroelectric-paraelectric phase transition, especially the first order ferroelectric-paraelectric phase transition.

Keywords: ferroelectrics; internal field; thermo-electro-mechanical coupling; non-equilibrium thermodynamics
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0 引 言

大量微观粒子组成的凝聚物质体系在本质上是热-电(磁)-力耦合系统,因此,对凝聚物质体系的研究,是从单方面考虑过渡到对几乎所有方面同时考虑,这样才能更接近问题的本质。具体到铁电体:最初研究者关心其介电性质,这属于纯电学范畴;接着证实随着温度的改变,电极化状态会发生改变甚至发生铁电-顺电相变(以下简称"铁电相变"),这属于热-电耦合的范畴;后来研究表明在热或电性质改变的同时,还有体系的体积、形状等力学因素的改变,这就是比较全面的热-电-力耦合,甚至还可以和磁学因素发生耦合。

铁电体的热-电(磁)-力耦合已经被深入研究和认识。首先在理论上,Maugin[1]对铁电体的热-电-力耦合的本构关系给出了系统而明确的论述,这是处理相关问题的出发点;在固体力学方面,热-电(磁)-力耦合的研究在阐明诸如弹塑性变形、热传导、振动、能量转换、疲劳及断裂等问题上取得了一定的成果[2];在材料的制备及结构、性能表征方面,尤其是包括铁电性在内的"多铁性"研究方面,热-电(磁)-力耦合乃是重点研究的问题[3-7]。

铁电相变尤其是一级铁电相变能够体现明显的不可逆性,即"热滞"[8-9],但是长期以来都是用平衡态热力学的某种权宜性的修正(引入"亚稳态")来处理,即Landau-Ginzburg-Devonshire(LGD)理论[10-12]。Ai等[13]利用场论和分析力学的概念和方法证明了相变过程中亚稳态并不存在,因此,LGD理论不具有坚实的物理基础,同时利用非平衡变分原理证明了稳定态的存在,表明铁电相变过程是稳定态过程。相关的实验结果为稳定态的存在提供有力的依据[14-17]。因此,利用线性非平衡态热力学能较好地描述铁电相变过程[18-20]。铁电相变的线性非平衡态热力学理论仍是一个处于发展中的理论,但有其合理性的一面[21-24],目前,涉及相关领域的研究成果较少。

在实际应用中,当铁电相变发生时一般不外加电场或机械力,所涉及的物理场为"内场"。已有研究表明内电势与相变潜热及电极化强度有关,但这只是在相变温度附近才具有的关系[24];而从绝对零度到相变温度的范围,内电势与温度等因素的关系仍不明确。因此,本文对铁电体的热-电-力耦合内场的特性作进一步的深入探讨,以物性为突破口,尝试给出内电势与温度等因素的关系。

1 公式推导

铁电体的热-电-力耦合内场所满足的热力学方程和物性方程分别为[24]:

$$\nabla\cdot\boldsymbol{J}_q+\varphi_{\mathrm{i}}\nabla\cdot\boldsymbol{J}_P-\nabla\cdot(\boldsymbol{v}\cdot\boldsymbol{\sigma})+\rho\boldsymbol{f}\cdot\boldsymbol{v}=0, \tag{1}$$

$$\begin{aligned}&\boldsymbol{L}_{qq}\vdots\nabla\nabla\left(\frac{1}{T}\right)+\boldsymbol{L}_{qP}\vdots\nabla\nabla\left(\frac{\varphi_i}{T}\right)+\boldsymbol{L}_{q\sigma}\vdots\nabla\left[-\boldsymbol{v}\nabla\left(\frac{1}{T}\right)\right]+\\&\boldsymbol{L}_{qf}\vdots\nabla\left(-\frac{\boldsymbol{v}}{T}\right)+\varphi_{\mathrm{i}}\boldsymbol{L}_{Pq}\vdots\nabla\nabla\left(\frac{1}{T}\right)+\varphi_{\mathrm{i}}\boldsymbol{L}_{PP}\vdots\nabla\nabla\left(\frac{\varphi_i}{T}\right)+\\&\varphi_{\mathrm{i}}\boldsymbol{L}_{P\sigma}\vdots\nabla\left[-\boldsymbol{v}\nabla\left(\frac{1}{T}\right)\right]+\varphi_{\mathrm{i}}\boldsymbol{L}_{Pf}\vdots\nabla\left(-\frac{\boldsymbol{v}}{T}\right)-\\&\boldsymbol{L}_{\sigma q}\vdots\left\{(\boldsymbol{v}\nabla)\left[\nabla\left(\frac{1}{T}\right)\right]+\boldsymbol{v}\left[\nabla\nabla\left(\frac{1}{T}\right)\right]\right\}-\\&\boldsymbol{L}_{\sigma P}\vdots\left\{(\boldsymbol{v}\nabla)\left[\nabla\left(\left(\frac{\varphi_{\mathrm{i}}}{T}\right)\right)\right]+\boldsymbol{v}\left[\nabla\nabla\left(\frac{\varphi_{\mathrm{i}}}{T}\right)\right]\right\}+\\&\boldsymbol{L}_{\sigma\sigma}\cdot\vdots\left\langle(\boldsymbol{v}\nabla)\left[\boldsymbol{v}\nabla\left(\frac{1}{T}\right)\right]+\boldsymbol{v}\left\{\nabla\left[\boldsymbol{v}\nabla\left(\frac{1}{T}\right)\right]\right\}\right\rangle+\\&\boldsymbol{L}_{\sigma f}\vdots\left\{(\boldsymbol{v}\nabla)\frac{\boldsymbol{v}}{T}+\boldsymbol{v}\left[\nabla\left(\frac{\boldsymbol{v}}{T}\right)\right]\right\}+\boldsymbol{L}_{fq}\vdots\boldsymbol{v}\left[\nabla\left(\frac{1}{T}\right)\right]+\\&\boldsymbol{L}_{fP}\vdots\boldsymbol{v}\left[\nabla\left(\frac{\varphi_{\mathrm{i}}}{T}\right)\right]-\boldsymbol{L}_{f\sigma}\vdots\boldsymbol{v}\boldsymbol{v}\left[\nabla\left(\frac{1}{T}\right)\right]-\boldsymbol{L}_{ff}\vdots\frac{\boldsymbol{v}\boldsymbol{v}}{T}=0,\end{aligned} \tag{2}$$

式中:$\boldsymbol{J}_q$和$\boldsymbol{J}_P$分别为传导热流密度(q)和极化(P)电流密度;φ_{i}为内电势;$\boldsymbol{v}$、$\boldsymbol{\sigma}$和$\boldsymbol{f}$分别为速度、应力和体积力;ρ为密度;T为温度;$\boldsymbol{L}_{qq}$、$\boldsymbol{L}_{PP}$、$\boldsymbol{L}_{ff}$、$\boldsymbol{L}_{qP}$、$\boldsymbol{L}_{Pq}$、$\boldsymbol{L}_{qf}$、$\boldsymbol{L}_{fq}$、$\boldsymbol{L}_{Pf}$和$\boldsymbol{L}_{fP}$是9个2阶张量,$\boldsymbol{L}_{q\sigma}$、$\boldsymbol{L}_{\sigma q}$、$\boldsymbol{L}_{P\sigma}$、$\boldsymbol{L}_{\sigma P}$、$\boldsymbol{L}_{\sigma f}$和$\boldsymbol{L}_{f\sigma}$是6个3阶张量,$\boldsymbol{L}_{\sigma\sigma}$是一个4阶张量,这些张量都是输运系数。

由于考虑力学因素,方程(1)和(2)在形式上都比较复杂,以至于较难进行进一步的数学处理。但在很多情况下,只考虑热-电耦合,也不失为一种合理的近似。在这种近似下,即令$\boldsymbol{v}$,$\boldsymbol{\sigma}$和$\boldsymbol{f}$=0,方程(1)和(2)可以简化为[24]:

$$\nabla\cdot\boldsymbol{J}_q+\varphi_{\mathrm{i}}\nabla\cdot\boldsymbol{J}_P=0, \tag{3}$$

$$\left(\boldsymbol{L}_{qq}+\varphi_{\mathrm{i}}\boldsymbol{L}_{Pq}\right)\vdots\nabla\nabla\left(\frac{1}{T}\right)+\left(\boldsymbol{L}_{qP}+\varphi_{\mathrm{i}}\boldsymbol{L}_{PP}\right)\vdots\nabla\nabla\left(\frac{\varphi_{\mathrm{i}}}{T}\right)=0, \tag{4}$$

式中需要注意的是,$\nabla\nabla\neq\nabla^2=\nabla\cdot\nabla$。

在文献[24]中,已经以热力学方程(3)为基础作了一些定性的讨论,得到了若干与实验事实相符的结论,如对于一级铁电相变,相变潜热越大则电

极化强度越大;φ_i不仅是T的函数,而且与其他因素如应力也有关系,这体现了热-电-力耦合的本质,但若只考虑热-电耦合,不妨认为φ_i只是T的函数$\varphi_i(T)$。

目前尚未对物性方程(4)进行处理,主要原因在于其形式仍比较复杂,用数学物理方程的通常求解方法来求解仍很困难[25-26]。为了规避数学上的困难,不妨考虑一个简单情况,即一维体系。虽然一般来说,体系通常为三维的,但仍希望从一维体系出发讨论,能基本揭示问题的一般性质。

在一维情况下,$T=T(x),\nabla=\mathbf{i}\frac{\mathrm{d}}{\mathrm{d}x}$。首先来计算$\nabla\nabla\left(\frac{\varphi_i}{T}\right)$和$\nabla\nabla\left(\frac{1}{T}\right)$:

$$\nabla\nabla\left(\frac{\varphi_i}{T}\right)=\nabla\left[\varphi_i\nabla\left(\frac{1}{T}\right)+\frac{1}{T}\nabla\varphi_i\right]=$$

$$\nabla\varphi_i\nabla\left(\frac{1}{T}\right)+\varphi_i\nabla\nabla\left(\frac{1}{T}\right)+\nabla\left(\frac{1}{T}\right)\nabla\varphi_i+\frac{1}{T}\nabla\nabla\varphi_i=$$

$$\left[\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}\cdot\frac{\mathrm{d}T}{\mathrm{d}x}\cdot\left(-\frac{1}{T^2}\right)\cdot\frac{\mathrm{d}T}{\mathrm{d}x}+\varphi_i\cdot\frac{\mathrm{d}}{\mathrm{d}x}\left(-\frac{1}{T^2}\cdot\frac{\mathrm{d}T}{\mathrm{d}x}\right)+\left(-\frac{1}{T^2}\right)\cdot\frac{\mathrm{d}T}{\mathrm{d}x}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}\cdot\frac{\mathrm{d}T}{\mathrm{d}x}+\frac{1}{T}\cdot\frac{\mathrm{d}}{\mathrm{d}x}\left(\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}\cdot\frac{\mathrm{d}T}{\mathrm{d}x}\right)\right]\mathbf{ii}=$$

$$\left[-\frac{2}{T^2}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}\cdot\left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)^2+\varphi_i\cdot\frac{2}{T^3}\left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)^2+\varphi_i\left(-\frac{1}{T^2}\right)\cdot\frac{\mathrm{d}^2T}{\mathrm{d}x^2}+\frac{1}{T}\cdot\frac{\mathrm{d}^2\varphi_i}{\mathrm{d}T^2}\cdot\left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)^2+\frac{1}{T}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}\cdot\frac{\mathrm{d}^2T}{\mathrm{d}x^2}\right]\mathbf{ii}=$$

$$\left[\left(\frac{1}{T}\cdot\frac{\mathrm{d}^2\varphi_i}{\mathrm{d}T^2}-\frac{2}{T^2}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}+\frac{2}{T^3}\varphi_i\right)\cdot\left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)^2+\left(\frac{1}{T}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}-\frac{1}{T^2}\varphi_i\right)\cdot\frac{\mathrm{d}^2T}{\mathrm{d}x^2}\right]\mathbf{ii},\tag{5}$$

而

$$\nabla\nabla\left(\frac{1}{T}\right)=\frac{\mathrm{d}}{\mathrm{d}x}\left(-\frac{1}{T^2}\cdot\frac{\mathrm{d}T}{\mathrm{d}x}\right)\mathbf{ii}=\left[\frac{2}{T^3}\cdot\left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)^2-\frac{1}{T^2}\cdot\frac{\mathrm{d}^2T}{\mathrm{d}x^2}\right]\mathbf{ii}。\tag{6}$$

由于参数的任意性,为使物性方程恒成立,由式(4)、(5)和(6)可知,$\left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)^2$项的系数和$\frac{\mathrm{d}^2T}{\mathrm{d}x^2}$项的系数必须都等于0,即:

$$\left(\boldsymbol{L}_{qq}+\varphi_i\boldsymbol{L}_{Pq}\right)\cdot\frac{2}{T^3}=-\left(\boldsymbol{L}_{qP}+\varphi_i\boldsymbol{L}_{PP}\right)\cdot\left(\frac{1}{T}\cdot\frac{\mathrm{d}^2\varphi_i}{\mathrm{d}T^2}-\frac{2}{T^2}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}+\frac{2}{T^3}\varphi_i\right),\tag{7}$$

$$\left(\boldsymbol{L}_{qq}+\varphi_i\boldsymbol{L}_{Pq}\right)\cdot\left(-\frac{1}{T^2}\right)=-\left(\boldsymbol{L}_{qP}+\varphi_i\boldsymbol{L}_{PP}\right)\cdot\left(\frac{1}{T}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}-\frac{1}{T^2}\varphi_i\right)。\tag{8}$$

观察式(7)和(8),可得出

$$\frac{T}{2}\cdot\left(\frac{1}{T}\cdot\frac{\mathrm{d}^2\varphi_i}{\mathrm{d}T^2}-\frac{2}{T^2}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}+\frac{2}{T^3}\varphi_i\right)=-\frac{1}{T}\cdot\frac{\mathrm{d}\varphi_i}{\mathrm{d}T}+\frac{1}{T^2}\varphi_i,\tag{9}$$

化简得

$$\frac{\mathrm{d}^2\varphi_i}{\mathrm{d}T^2}=0,\tag{10}$$

其解为

$$\varphi_i=AT+B\ (A,B\text{为不定常数})。\tag{11}$$

2　结果与比较

不妨简单地确定A和B。$T=0$时,$\varphi_i(T=0)=B$;$T=T_C$时(T_C为居里温度),$\varphi_i\left(T=T_C\right)=AT_C+\varphi_i(T=0)\rightarrow A=\left[\varphi_i\left(T=T_C\right)-\varphi_i(T=0)\right]\big/T_C$。因此

$$\varphi_i(T)=\frac{\varphi_i\left(T=T_C\right)-\varphi_i(T=0)}{T_C}T+\varphi_i(T=0)。\tag{12}$$

另外,考虑到$T=T(x)$,则内电场

$$E_i=-\frac{\mathrm{d}\varphi_i}{\mathrm{d}x}=-A\frac{\mathrm{d}T(x)}{\mathrm{d}x},\tag{13}$$

式中$\mathrm{d}T(x)/\mathrm{d}x$是温度梯度(数值),因此,由内电场和温度梯度可确定常数A。

可计算出热力学力$\boldsymbol{X}_P$和$\boldsymbol{X}_q$(分别与热力学流$\boldsymbol{J}_P$和$\boldsymbol{J}_q$相对应),考虑一维情况,可将矢量或张量看作一般变量,即

$$X_P=\nabla\left(\frac{\varphi_i}{T}\right)\Big/\mathbf{i}=\frac{\mathrm{d}}{\mathrm{d}x}\left[\frac{\varphi_i(x)}{T(x)}\right]=\frac{\mathrm{d}}{\mathrm{d}x}\left[\frac{AT(x)+B}{T(x)}\right]=-\frac{B}{\left[T(x)\right]^2}\cdot\frac{\mathrm{d}T(x)}{\mathrm{d}x},\tag{14}$$

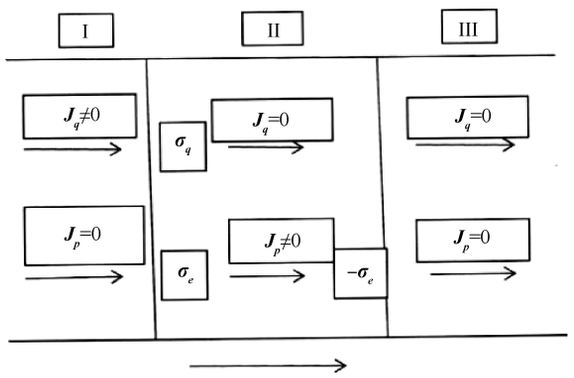
$$X_q=\nabla\left(\frac{1}{T}\right)\Big/\mathbf{i}=\frac{\mathrm{d}}{\mathrm{d}x}\left[\frac{1}{T(x)}\right]=-\frac{1}{\left[T(x)\right]^2}\cdot\frac{\mathrm{d}T(x)}{\mathrm{d}x}。\tag{15}$$

比较式(14)和(15),可得

$$X_P=BX_q。\tag{16}$$

研究显示,通过考虑相变体系内特定区域、特定过程中热力学流和力之间的关系,可将常数B用

物性参数,即输运系数表示出来。一级铁电相变发生时,3个不同区域内的热(或电)的流$\boldsymbol{J}_q$($\boldsymbol{J}_P$)和局域产生$\boldsymbol{\sigma}_q$($\boldsymbol{\sigma}_e$)如图1所示。



注:I为相变已发生区域;II为相变正在发生区域;
III为相变将要发生区域。

图1　3个不同区域内的热(或电)流$\boldsymbol{J}_q$($\boldsymbol{J}_P$)和局域产生$\boldsymbol{\sigma}_q$($\boldsymbol{\sigma}_e$)[24]

在升、降温过程中,对于相变正在发生区域,即区域II,都有[18-19]

$$\boldsymbol{J}_q^{\mathrm{II}} = \boldsymbol{L}_{qq}^{\mathrm{II}}\cdot X_q^{\mathrm{II}} + \boldsymbol{L}_{qP}^{\mathrm{II}}\cdot X_P^{\mathrm{II}} = 0, \tag{17}$$

可推出

$$B = -\frac{\boldsymbol{L}_{qq}^{\mathrm{II}}}{\boldsymbol{L}_{qP}^{\mathrm{II}}}。 \tag{18}$$

在降温过程中,对于相变已发生区域,即区域I(为铁电相),有[18-19]

$$\boldsymbol{J}_P^{\text{I-ferr}} = \boldsymbol{L}_{PP}^{\text{I-ferr}}\cdot \boldsymbol{X}_P^{\text{I-ferr}} + \boldsymbol{L}_{Pq}^{\text{I-ferr}}\cdot \boldsymbol{X}_q^{\text{I-ferr}} = 0, \tag{19}$$

可推出

$$B = -\frac{\boldsymbol{L}_{Pq}^{\text{I-ferr}}}{\boldsymbol{L}_{PP}^{\text{I-ferr}}}。 \tag{20}$$

由此可知,φ_i可以在理论上确定。而在文献[24]中,得出$\varphi_i = -\boldsymbol{\sigma}_q/\boldsymbol{\sigma}_e \propto l/P$,其中$\boldsymbol{\sigma}_q$和$\boldsymbol{\sigma}_e$分别为热量和电荷的局域产生,$l$和$P$分别为相变潜热和电极化强度,而$l$和$P$都是实验可观测量。因此,本研究确定了有关的物性参量,如输运系数。

3　结　论

本文进一步揭示了φ_i的更多特征,即φ_i是温度T的线性函数;φ_i中的不定常数A和B可由内电场、温度梯度及输运系数确定。对于一级铁电相变,温度梯度大时,内电场$\boldsymbol{E}_i$,热力学力$\boldsymbol{X}_P$和$\boldsymbol{X}_q$也大,这比较符合相界即相变正在发生区域II的情况,此处相关物理量的变化率出现极大值。而对于二级铁电相变的情况,由于没有“热滞”,其比较简单,可完全由平衡态热力学处理。

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