Eur. Phys. J. Appl. Phys. (2013) 63: 30301
DOI: 10.1051/epjap/2013130305

THE EUROPEAN
PHYSICAL JOURNAL
APPLIED PHYSICS

Regular Article

Epitaxy surface effect on the critical properties of a ferroelectric thin film

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Received: 27 June 2013 / Received in final form: 9 August 2013 / Accepted: 12 August 2013
Published online: 3 September 2013 –

Abstract. The characteristic properties, the critical surface transverse field $\Omega_{\rm sc}$ and the Curie temperature $T_{\rm c}$, for an N-layer ferroelectric thin film with epitaxy surface layers $N_{\rm s}$ described by the transverse field Ising model have been studied by the differential operator technique within the framework of effective-field theory with correlations. The analytical equation for the phase diagrams of ferroelectric thin films is derived. The surface transverse field $\Omega_{\rm s}$ dependence of the Curie temperature $T_{\rm c}$ in the ferroelectric thin film with different epitaxy surface layers is calculated. Meanwhile the epitaxy surface layer number $N_{\rm s}$ dependence of the critical surface transverse field $\Omega_{\rm sc}$ and the Curie temperature $T_{\rm c}$ is also examined. The results show that the critical properties depend heavily on modifications of interchange interactions and transverse fields in the epitaxy surface layer.

1 Introduction

In the previous works [1–4], the surface effects on the critical properties of ferroelectric thin films have been studied by assuming that interchange interactions and transverse fields in the epitaxy surface layers are different from those of the bulk layers. On the theoretical treatment, the transverse field Ising model (TIM) [5–9] has been believed to be a good candidate to investigate the critical properties of the ferroelectric thin film systems due to various surface modifications in the epitaxy surface layer. Within the framework of mean field approximation (MFA), Wang et al. [1] discussed the surface interchange interaction and surface layer number as well as thin film layer number dependence of the Curie temperature $T_{\rm c}$ in the ferroelectric thin film with a uniform transverse field in the epitaxy surface layers and bulk layers. Then, the possible modification to the transverse field in epitaxy surface layers was further extended by Sy [3]. By taking into account two types of surface modification, the effects of multi-surface modification on the Curie temperature $T_{\rm c}$ of ferroelectric thin films have been studied by the transfer matrix method [4]. In order to obtain more exact results than those of the MFA [10–13], the effective-field theory with correlations (EFT) [14–19] and the Green's function technique (GFT) [20,21] have been used to treat the TIM. Essaoudi et al. [16] discussed the dependence of the critical temperature on the thickness, interchange interactions and transverse fields by use of the effective field theory with a probability distribution technique. Recently, Arhchoui et al. [17] and our group [11] systematically studied the phase diagrams of ferroelectric thin films with two surface layers, respectively. Very recently, we calculated the crossover characteristic and three different phase transition regions in the phase diagram of the multiple-surface-layer ferroelectric thin film using the EFT [19]. As far as we know, however, the investigation of the critical properties (the critical surface transverse field $\Omega_{\rm sc}$ and the Curie temperature $T_{\rm c}$) in ferroelectric thin films with different epitaxy surface layers have not been carried out with the treatment of the EFT.

In the present article, the differential operator technique with correlations has been used to investigate the critical properties in an N-layer ferroelectric thin film with epitaxy surface layers $N_{\rm s}$ by taking into account the modifications of interchange interactions and transverse fields in the $N_{\rm s}$ epitaxy surface layers. Firstly, the analytical equation for the phase diagrams of ferroelectric thin films is derived. Then, the dependence of the Curie temperature $T_{\rm c}$ on the surface transverse field $\Omega_{\rm s}$ and the dependence of the critical surface transverse field $\Omega_{\rm sc}$ and the Curie temperature $T_{\rm c}$ on the epitaxy surface layer number $N_{\rm s}$ are obtained.

2 The model and formalism

We consider a ferroelectric thin film system which has a three-dimensional simple cubic lattice structure consisting of N layers along the z-direction. Each layer is defined in the x-y plane and the pseudo-spin sites on the square lattice. Figure 1 is the schematic diagram for the ferroelectric thin film, the layer index number is denoted as

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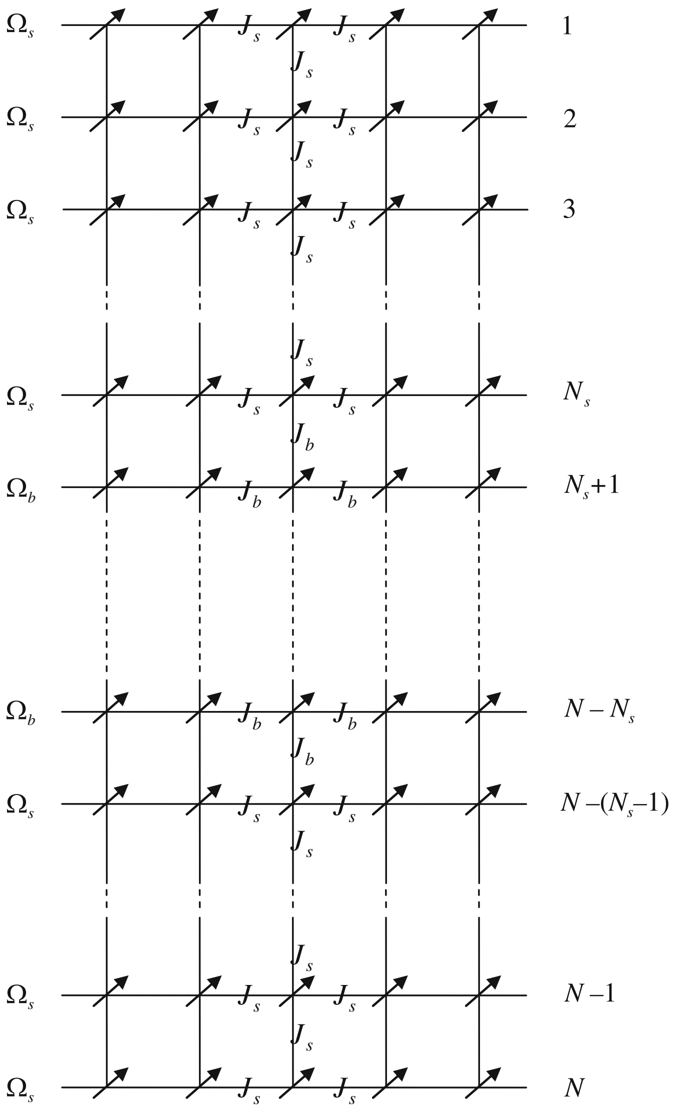

Fig. 1. Schematic diagram of an N-layer ferroelectric thin film with N_{s} epitaxy surface layers.

1, 2, 3, ..., N, where the top layers numbered 1, 2, 3, ..., N_{s}, and the bottom layers numbered $N-(N_{\mathrm{s}}-1)$, ..., $N-2$, $N-1$, N add up to $2N_{\mathrm{s}}$ layers which are defined as the epitaxy surface layers, whereas the other $N-2N_{\mathrm{s}}$ layers are defined as the bulk layers. The Hamiltonian of the system is [18]:

$$H=-\frac{1}{2}\sum_{ij}J_{ij}S_i^zS_j^z-\Omega_b\sum_{i\in \mathrm{b}}S_i^x-\Omega_{\mathrm{s}}\sum_{i\in \mathrm{s}}S_i^x, \quad (1)$$

where S_i^x (or S_i^z) is the component of the spin-$\frac{1}{2}$ operator at site i along the x (or z) direction, Ω_{s} (or Ω_{b}) is the transverse field in the epitaxy surface (or bulk) layer. J_{ij} is the nearest neighbour interchange interaction between the spin operators at sites i and j, and $J_{ij}=J_{\mathrm{s}}$ (or J_{b}) when the spin operators are in the epitaxy surface (or bulk) layer.

The pseudo-spin average value for the ith layer of the thin film can be obtained as follows [18]:

$$R_i=\langle S_i^z\rangle=\left\langle\prod_{\delta}\left[\cosh\left(DJ_{i,j+\delta}\right)+R_{j+\delta}\sinh(DJ_{i,j+\delta})\right]\right\rangle \times f_i\left(x\right)|_{x=0}\ , \quad (2)$$

where $\prod$ is the cumprod for the nearest neighbour of site i, $D=\partial/\partial x$ is the differential operator and $\exp\left(\alpha D\right) f\left(x\right)=f\left(x+\alpha\right)$. In the $2N_{\mathrm{s}}$ epitaxy surface layers, the function $f_i\left(x\right)$ is defined as

$$f_i(x)=f_{\mathrm{s}}(x)=\frac{x}{\sqrt{\left(2\Omega_{\mathrm{s}}\right)^2+x^2}}\tanh\left[\frac{\beta}{4}\sqrt{\left(2\Omega_{\mathrm{s}}\right)^2+x^2}\right], \quad (3)$$

whereas in the $N-2N_{\mathrm{s}}$ bulk layers, the function $f_i\left(x\right)$ is defined as

$$f_i(x)=f_{\mathrm{b}}(x)=\frac{x}{\sqrt{\left(2\Omega_{\mathrm{b}}\right)^2+x^2}}\tanh\left[\frac{\beta}{4}\sqrt{\left(2\Omega_{\mathrm{b}}\right)^2+x^2}\right], \quad (4)$$

where $\beta=1/(k_BT)$, T is the temperature.

The polarization average value for the ith layer of an N-layer ferroelectric thin film with N_{s} epitaxy surface layers can be obtained on the base of equation (2). For the top epitaxy surface layers numbered 1, 2, 3, ..., N_{s} [18]:

$$R_1=\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_1\sinh\left(DJ_{\mathrm{s}}\right)\right]^4 \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_2\sinh\left(DJ_{\mathrm{s}}\right)\right]f_{\mathrm{s}}\left(x\right)|_{x=0}, \quad (5)$$

$$R_2=\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_1\sinh\left(DJ_{\mathrm{s}}\right)\right] \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_2\sinh\left(DJ_{\mathrm{s}}\right)\right]^4 \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_3\sinh\left(DJ_{\mathrm{s}}\right)\right]f_{\mathrm{s}}\left(x\right)|_{x=0}, \quad (6)$$

$$\vdots$$

$$R_{N_{\mathrm{s}}}=\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_{N_{\mathrm{s}}-1}\sinh\left(DJ_{\mathrm{s}}\right)\right] \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_{N_{\mathrm{s}}}\sinh\left(DJ_{\mathrm{s}}\right)\right]^4 \times\left[\cosh\left(DJ_b\right)+R_{N_{\mathrm{s}}+1}\sinh\left(DJ_b\right)\right]f_{\mathrm{s}}\left(x\right)|_{x=0}. \quad (7)$$

For the middle $N-2N_{\mathrm{s}}$ bulk layers:

$$R_i=\left[\cosh\left(DJ_b\right)+R_{i-1}\sinh\left(DJ_b\right)\right] \times\left[\cosh\left(DJ_b\right)+R_i\sinh\left(DJ_b\right)\right]^4 \times\left[\cosh\left(DJ_b\right)+R_{i+1}\sinh\left(DJ_b\right)\right]f_b\left(x\right)|_{x=0}. \quad (8)$$

For the bottom epitaxy surface layers numbered $N-(N_{\mathrm{s}}-1)$, ..., $N-2$, $N-1$, N:

$$R_{N-(N_{\mathrm{s}}-1)}=\left[\cosh\left(DJ_{\mathrm{b}}\right)+R_{N-N_{\mathrm{s}}}\sinh\left(DJ_{\mathrm{b}}\right)\right] \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_{N-(N_{\mathrm{s}}-1)}\sinh\left(DJ_{\mathrm{s}}\right)\right]^4 \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_{N-(N_{\mathrm{s}}-2)}\sinh\left(DJ_{\mathrm{s}}\right)\right] \times f_{\mathrm{s}}\left(x\right)|_{x=0}, \quad (9)$$

$$\vdots$$

$$R_{N-1}=\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_{N-2}\sinh\left(DJ_{\mathrm{s}}\right)\right] \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_{N-1}\sinh\left(DJ_{\mathrm{s}}\right)\right]^4 \times\left[\cosh\left(DJ_{\mathrm{s}}\right)+R_N\sinh\left(DJ_{\mathrm{s}}\right)\right]f_{\mathrm{s}}\left(x\right)|_{x=0}, \quad (10)$$

$$R_N = [\cosh(DJ_s) + R_{N-1}\sinh(DJ_s)] \times [\cosh(DJ_s) + R_N \sinh(DJ_s)]^4 f_s(x)|_{x=0}. \quad (11)$$

When the temperature closes to the Curie temperature, the polarization average values will tend to zero. Therefore, we can only consider the linear terms of R_i due to the higher-order terms of R_i tend to zero faster than the linear terms of R_i [18]. The analytical equation for an N-layer ferroelectric thin film with epitaxy surface layers N_s can be derived as follows:

(1) For even-layer-number thin films, $M = N/2$, and

$$(1 - 5k) D_{N_s,M-1} - k^2 D_{N_s,M-2} = 0. \quad (12)$$

(2) For odd-layer-number thin films, $M = (N+1)/2$, and

$$(1 - 4k) D_{N_s,M-1} - 2k^2 D_{N_s,M-2} = 0. \quad (13)$$

Obviously, the difference between the two analytical equations for even and odd number of layers is minor except for two coefficients.

3 Numerical results and discussions

In this section, we will investigate the critical surface transverse field Ω_{sc} and the Curie temperature T_c in a ferroelectric thin film with different epitaxy surface layers. According to the analytical equations (12) and (13), the phase diagram described by the relation between the Curie temperature and the surface transverse field can be calculated numerically. The results show which may be divided into two kinds of phase transition regions, the ferroelectric phase (F) which means that the polarization along the longitudinal direction ($S = \sum_{i=1}^{N} \langle S_i^z \rangle$) in the thin film is not equal to zero, and the paraelectric phase (P) which corresponds to $S = 0$ irrespective of any temperature. For simplicity and without loss of generality, the epitaxy surface layers of ferroelectric thin films varying from $N_s = 1$ to $N_s = 5$ will be taken into account in the following.

Figure 2 gives the phase diagram between the Curie temperature T_c and the surface transverse field Ω_s. It exhibits the dependence of the phase diagram on the epitaxy surface layer number, i.e., the curves of T_c vs. Ω_s for different numbers of epitaxy surface layer N_s. It is obvious that the Curie temperature T_c decreases monotonously with the increase of surface transverse field Ω_s for the ferroelectric thin film with a fixed epitaxy surface layer number N_s. Eventually, the curve of T_c vs. Ω_s and the horizontal coordinate Ω_s-axis will intersect at some point, and the value of Ω_s corresponding to this point is usually defined as the critical surface transverse field at which the T_c versus Ω_s curve reduces to zero. For the value of $\Omega_s > \Omega_{sc}$, the ferroelectric system is always paraelectric irrespective of any temperature. That is to say, any temperature can result in a transition from ferroelectric to paraelectric with increasing the surface transverse field. In contrast, when $\Omega_s < \Omega_{sc}$, the ferroelectric system is always ferroelectric phase unless the temperature is above a certain value. In other words, only larger temperature can result in a transition from ferroelectric to paraelectric with increasing the temperature. Meanwhile, Figure 2 also shows the dependence of the phase diagram on the epitaxy surface layer number. It is obvious that the ferroelectric phase region is the smallest for $N_s = 1$. When $N_s \geq 2$, the ferroelectric phase region is decreasing with the increasing of the epitaxy surface layer number. However, the paraelectric phase region is increasing with the decreasing of the epitaxy surface layer number.

In order to inspect the dependence of the critical surface transverse field Ω_{sc} and the Curie temperature T_c on the epitaxy surface layer number N_s in a ferroelectric thin film, Figures 3 and 4 are presented in the following.

Figure 3 gives the dependence of critical surface transverse field Ω_{sc} on the epitaxy surface layer number N_s for a ferroelectric thin film with some selected different surface interchange interaction J_s's. It can be found that there

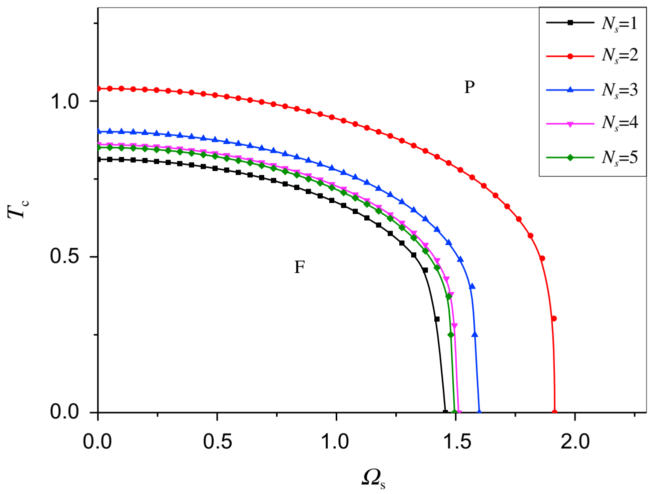

Fig. 2. The dependence of the phase diagram on epitaxy surface layers (T_c vs. Ω_s). $J_s = 1$, $J_b = 1$, $\Omega_b = 3.0$, $N = 30$.

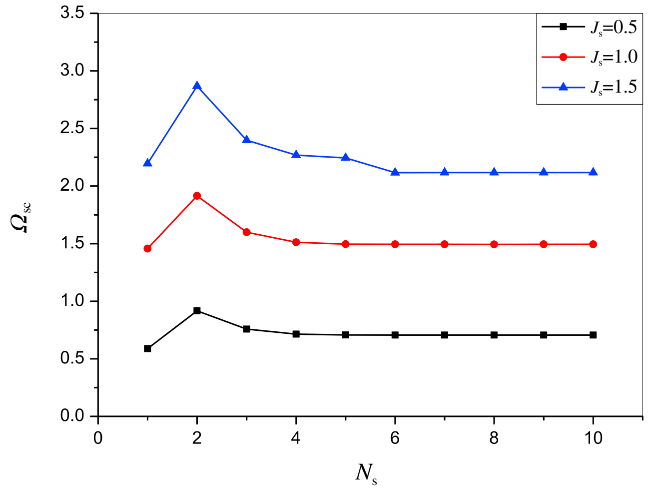

Fig. 3. The dependence of critical surface transverse field Ω_{sc} on the epitaxy surface layer number N_s. $J_b = 1.0$, $\Omega_b = 3.0$, $N = 30$.

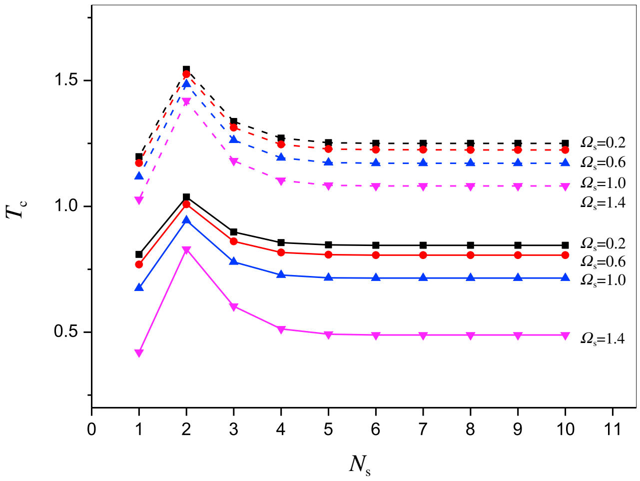

Fig. 4. The dependence of Curie temperature T_c on the epitaxy surface layer number N_s. $J_b = 1.0$, $\Omega_b = 3.0$, $N = 30$. $J_s = 1.0$ (solid line), $J_s = 1.5$ (dash line).

exists a direct relation between the epitaxy surface layer number N_s and the critical surface transverse field Ω_{sc} for the thin film with a fixed surface interchange interaction J_s from Figure 3. When the epitaxy surface layer number $N_s \geq 2$, the value of critical surface transverse field Ω_{sc} is monotonously decreasing with increasing of the epitaxy surface layer number N_s. Moreover, the value of critical surface transverse field Ω_{sc} will infinitely close to the corresponding value of the thin film with $N_s = 1$ as possible as. On the other hand, Figure 3 also gives the dependence of critical surface transverse field Ω_{sc} on the surface interchange interaction J_s. It is obvious that the larger the surface interchange interaction J_s, the larger the critical surface transverse field Ω_{sc} for the thin films with a fixed epitaxy surface layer number N_s.

Figure 4 gives the dependence of Curie temperature T_c on the epitaxy surface layer number N_s for a ferroelectric thin film with different surface transverse field Ω_s and surface interchange interaction J_s. It is obvious that the corresponding properties for the relation between the Curie temperature T_c and the epitaxy surface layer number N_s, and the dependence of Curie temperature T_c on the surface transverse field Ω_s and the surface interchange interaction J_s are similar to those of the critical surface transverse field Ω_{sc}.

4 Conclusions

In this work, the critical properties of an N-layer ferroelectric thin film with epitaxy surface layers N_s have been investigated by the use of the differential operator technique with correlations. Based on the analytical equation for the phase diagrams of ferroelectric thin films with different epitaxy surface layers, the effects of the surface transverse field Ω_s on the Curie temperature T_c are calculated. Furthermore, the effects of the epitaxy surface layer number N_s on the critical surface transverse field Ω_{sc} and the Curie temperature T_c are discussed in detail. We hope that these results may be useful for theoretical or experimental investigations of ferroelectric thin films with multiple epitaxy surface layers.

Project supported by the Special Funds of the National Natural Science Foundation of China (Grant No.: 11247208).

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Appendix I

$$D_{1,M} = k^{M-1}\left[\left(1-4k_1\right)B_{M-1} - k_2 B_{M-2}\right], \tag{A1}$$

$$D_{2,M} = k^{M-2}\left\{\left[\left(1-4k_3\right)\left(1-4k_4\right) - k_3k_4\right] \times B_{M-2} - k_5\left(1-4k_3\right)B_{M-3}\right\}, \tag{A2}$$

$$D_{3,M} = k^{M-3}\left\{\left[\left(1-4k_3\right)\left(1-4k_4\right)\left(1-4k_6\right) - k_3k_6\left(1-4k_4\right) - k_4k_6\left(1-4k_3\right)\right]B_{M-3} + \left[k_3k_5k_6 - k_5\left(1-4k_3\right)\left(1-4k_6\right)\right]B_{M-4}\right\}, \tag{A3}$$

$$D_{4,M} = k^{M-4}\left\{\left[k_3k_4k_6^2 - k_6^2\left(1-4k_3\right)\left(1-4k_4\right) + \left(1-4k_3\right)\left(1-4k_4\right)\left(1-4k_6\right)^2 - k_4k_6\left(1-4k_3\right)\left(1-4k_6\right) - k_3k_6\left(1-4k_4\right)\left(1-4k_6\right)\right]B_{M-4} + \left[k_5k_6^2\left(1-4k_3\right) + k_3k_5k_6\left(1-4k_6\right) - k_5\left(1-4k_3\right)\left(1-4k_6\right)^2\right]B_{M-5}\right\}, \tag{A4}$$

$$D_{5,M} = k^{M-5}\left\{\left[k_4k_6^3\left(1-4k_3\right) + \left(1-4k_3\right)\left(1-4k_4\right)\left(1-4k_6\right)^3 + k_3k_6^3\left(1-4k_4\right) - k_4k_6\left(1-4k_3\right)\left(1-4k_6\right)^2 - 2k_6^2\left(1-4k_3\right)\left(1-4k_4\right)\left(1-4k_6\right) + k_3k_4k_6^2\left(1-4k_6\right) - k_3k_6\left(1-4k_4\right)\left(1-4k_6\right)^2\right]B_{M-5} + \left[2k_5k_6^2\left(1-4k_3\right)\left(1-4k_6\right) - k_3k_5k_6^3 + k_3k_5k_6\left(1-4k_6\right)^2 - k_5\left(1-4k_3\right)\left(1-4k_6\right)^3\right]B_{M-6}\right\}, \tag{A5}$$

$$B_M = \sinh\left[\left(M+1\right)\phi\right]/\sinh\left(\phi\right), \tag{A6}$$

$$\cosh\left(\phi\right) = \left(1/k-4\right)/2, \tag{A7}$$

$$k_1 = \cosh\left(DJ_{\mathrm{b}}\right)\cosh^3\left(DJ_{\mathrm{s}}\right)\sinh\left(DJ_{\mathrm{s}}\right)f_{\mathrm{s}}\left(x\right)|_{x=0}, \tag{A8}$$

$$k_2 = \cosh^4\left(DJ_{\mathrm{s}}\right)\sinh\left(DJ_{\mathrm{b}}\right)f_{\mathrm{s}}\left(x\right)|_{x=0}, \tag{A9}$$

$$k_3 = \cosh^4\left(DJ_{\mathrm{s}}\right)\sinh\left(DJ_{\mathrm{s}}\right)f_{\mathrm{s}}\left(x\right)|_{x=0}, \tag{A10}$$

$$k_4 = \cosh\left(DJ_{\mathrm{b}}\right)\cosh^4\left(DJ_{\mathrm{s}}\right)\sinh\left(DJ_{\mathrm{s}}\right)f_{\mathrm{s}}\left(x\right)|_{x=0}, \tag{A11}$$

$$k_5 = \cosh^5\left(DJ_{\mathrm{s}}\right)\sinh\left(DJ_{\mathrm{b}}\right)f_{\mathrm{s}}\left(x\right)|_{x=0}, \tag{A12}$$

$$k_6 = \cosh^5\left(DJ_{\mathrm{s}}\right)\sinh\left(DJ_{\mathrm{s}}\right)f_{\mathrm{s}}\left(x\right)|_{x=0}, \tag{A13}$$

$$k = \cosh^5\left(DJ_{\mathrm{b}}\right)\sinh\left(DJ_{\mathrm{b}}\right)f_{\mathrm{b}}\left(x\right)|_{x=0}. \tag{A14}$$