

Theory of Magnetization of Two-Phase Superparamagnetic Particles: I. Magnetic States

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Abstract—A two-phase particle is analyzed for magnetic states. It is shown that the grain can reside in any of four states. Critical fields of magnetization reversal are calculated. A technique is developed to calculate the occupancy of magnetic states in an ensemble of fine two-phase particles.

INTRODUCTION

Lying at the basis of theoretical studies into the magnetic states of single-domain particles or of those close in size, the hypothesis that ferromagnetic grains are single-phase (chemically homogeneous) species is a simplification rather than a faithful reflection of reality. Depending on the way they are produced, fine particles can be surface-coated by the oxide of the principal or any other magnetic element [1–4]. It is likely that a polyphase magnetic system is formed in a different way—through the decomposition of solid solution, which produces adjacent phases, some enriched in and the others depleted of a magnetic material [5].

It is natural to expect that the chemical inhomogeneity of fine particles can significantly affect the stability of a state in which the magnetic moment is uniformly distributed and thus change the magnitude of the magnetic moment and the critical field of magnetization reversal.

The stability of the uniformly magnetized state of particles differing in shape is dealt with in [6–8]. There, a two-phase quasi-single-domain particle with uniaxial crystallographic anisotropy is used as a model, the surface layer is assumed to be sufficiently thin, and an analytical expression is derived for the critical field required to form a fanlike magnetic structure. It is further shown there that the formation of a surface layer with a low or high anisotropy brings on a change in the saturation magnetization of the quasi-single-domain grain. Unfortunately, the technique whereby the critical field of magnetization reversal is analyzed in the works cited is able to identify only the ground states corresponding to a given class of magnetic moment distribution functions. In fact, it leaves out the metastable states, although it is natural to expect that a two-phase particle may reside in states where the magnetic moments of the phases can take on parallel and antiparallel orientation. Depending on the relative position of the phases and by virtue of their interaction, one of the states, provided it is realizable, must be metastable even if the magnetic moments of the phases remain

homogeneous. The extension of the spectrum of equilibrium states must of necessity tell on the way an ensemble of fine particles is magnetized and on its magnetic properties.

This paper is an attempt to investigate the magnetization of an ensemble of chemically inhomogeneous fine particles by resorting to a simple and easy-to-grasp model.

1. THE MODEL OF A TWO-PHASE PARTICLE

Consider a particle, that consists of two crystallographically uniaxial, homogeneously magnetized ferromagnetic phases in the shape of a parallelepiped of base a^2 and height qa (Fig. 1). The parameters of the phases are as follows: I_{s1} and I_{s2} are the spontaneous magnetizations, k_1 and k_2 are the dimensionless constants of crystallographic anisotropy, and $1 - \epsilon$ and ϵ are the relative volumes of the first and second phases, respectively. To simplify matters, we assume that the transition region is infinitesimally thin, that the vectors I_{s1} and I_{s2} lie in the XOZ plane, and that the axes of crystallographic anisotropy of both ferromagnets are

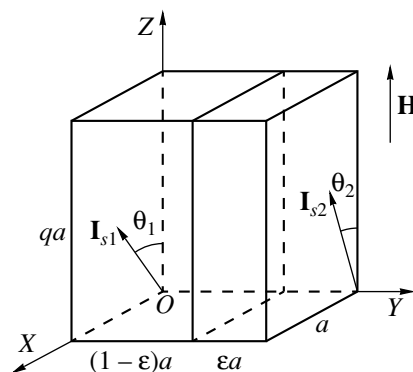


Fig. 1. Illustrating the model of a two-phase particle.

aligned parallel to the Z axis. The particle is placed in an external magnetic field \mathbf{H} directed along the Z axis.

We leave out of consideration the exchange and magnetoelastic interphase interactions. Their consideration is more or less justified only if the magnetic atoms in the boundary layer are distributed in a strongly random manner [9, 10].

In the adopted approximation, the free energy F of the grain placed in the external magnetic field H at zero absolute temperature ($T = 0$) may be written as the sum of the energy of crystallographic anisotropy

$$E_a = \frac{1}{2}(k_1 I_{s1}^2 (1 - \varepsilon) \sin^2 \theta_1 + k_2 I_{s2}^2 \varepsilon \sin^2 \theta_2) q a^3, \quad (1)$$

the energy of interaction between the magnetic moment and the internal magnetic field

$$E_m = (N_{11} I_{s1}^2 \sin^2 \theta_1 + N_{22} I_{s2}^2 \sin^2 \theta_2 + N_{12} I_{s1} I_{s2} \times \sin \theta_1 \sin \theta_2 + N_{21} I_{s1} I_{s2} \cos \theta_1 \cos \theta_2) q a^3, \quad (2)$$

and the energy of interaction between the magnetic moment and the external magnetic field

$$E_H = -H(I_{s1}(1 - \varepsilon) \cos \theta_1 + I_{s2} \varepsilon \cos \theta_2) q a^3. \quad (3)$$

In equation (2), N_{ik} are the demagnetizing factors determined by the shape and relative dimensions of the phases (see Appendix I).

2. EQUILIBRIUM STATES OF A TWO-PHASE PARTICLE

By applying the standard procedure to minimize the free energy $F = E_a + E_m + E_H$ in terms of θ_1 and θ_2 , we obtain a system of equations that define the equilibrium states of the magnetic moments of a grain

$$\begin{cases} a_1 \cos \theta_1 \sin \theta_1 + N_{12} j \cos \theta_1 \sin \theta_2 \\ -N_{21} j \cos \theta_2 \sin \theta_1 + \frac{H}{I_{s1}} (1 - \varepsilon) \sin \theta_1 = 0 \\ a_2 j \cos \theta_2 \sin \theta_2 + N_{12} \cos \theta_2 \sin \theta_1 \\ -N_{21} \cos \theta_1 \sin \theta_2 + \frac{H}{I_{s2}} \varepsilon j^2 \sin \theta_2 = 0, \end{cases} \quad (4)$$

where

$$a_1 = 2N_{11} + k_1(1 - \varepsilon), \quad a_2 = 2N_{22} + k_2 \varepsilon, \quad j = I_{s1}/I_{s2}.$$

We begin our analysis of solutions to (4) with the case $H = 0$.

2.1. Equilibrium States in the Absence of an External Field

It is an easy matter to see that for $H = 0$ the spectrum of solutions to (4) consists of three groups, namely,

(1) $\sin \theta_1 = \sin \theta_2 = 0$, (2) $\cos \theta_1 = \cos \theta_2 = 0$, and

(3) $\cos \theta_1 = \pm \sqrt{\frac{(a_1 b - N_{21})^2 - j^2 b^2 N_{12}^2}{(a_1 b - N_{21})^2 - N_{12}^2}}$, $\cos \theta_2 = \pm \frac{\cos \theta_1}{j b}$,

where

$$b = \frac{a_1 a_2 + N_{21}^2 - N_{12}^2 \pm \sqrt{(a_1 a_2 + N_{21}^2 - N_{12}^2)^2 - 4 a_1 a_2 N_{21}^2}}{2 a_1 N_{21}}.$$

The first group of solutions corresponds to a minimum of F , the second, to a maximum of F , and the third is not realized with the chosen relative orientation of the phases because it does not satisfy the extremum condition.

Thus, in the absence of an external field, a two-phase particle can reside in one of the states listed below:

–the first ($\uparrow\uparrow$) state, where the magnetic moments of both phases are aligned parallel and point along the OZ direction;

–the second ($\uparrow\downarrow$) state, where the phases are magnetized antiparallel and the magnetic moment of the first phase \mathbf{m}_1 points in the OZ direction;

–the third ($\downarrow\downarrow$) state, which differs from the first in that the magnetizations of the phases are aligned antiparallel relative to the OZ axis;

–the fourth ($\downarrow\uparrow$) state, where the magnetic moment of the second phase is aligned with the OZ axis and that of the first, against it.

The first and third states are metastable because the free energy of the grain in these states, $F^{(\uparrow\uparrow)} = F^{(\downarrow\downarrow)} = N_{21} I_{s1} I_{s2}$, is greater than in the second and fourth states, where $F^{(\uparrow\downarrow)} = F^{(\downarrow\uparrow)} = -N_{21} I_{s1} I_{s2}$.

2.2. Equilibrium States of a Grain in an External Magnetic Field

In an external magnetic field \mathbf{H} directed along the OZ axis, all states except the first are metastable, but it is the third state that is the most unstable. Obviously, the grain can change from the third state to the second ($\uparrow\downarrow$), to the fourth ($\downarrow\uparrow$), or to the first ($\uparrow\uparrow$). The first two transitions imply the rotation of the magnetic moment of a phase, and the last, the rotation of the overall moment of the grain. Consider the transition to the second state. By putting $\theta_2 = \pi$ in the first equation of the system (4), we arrive at two types of solutions

$$\sin \theta_1 = 0 \text{ and } \cos \theta_1 = -\frac{H(1 - \varepsilon) + N_{21} I_{s2}}{a_1 I_{s1}}, \quad (5)$$

which for $H < (a_1 I_{s1} - N_{21} I_{s2})/(1 - \varepsilon)$ determine the ground state ($\theta_1 = 0$), a metastable state ($\theta_1 = \pi$), and the state in which the free energy F is maximum. For $H = k_1 I_{s1} + (2N_{11} I_{s1} - N_{21} I_{s2})/(1 - \varepsilon)$ the metastable state dis-

appears, because with $\theta_1 = \pi$ and $\theta_2 = \pi$ the free energy F is maximum. Therefore, we define the field

$$H_{c1}^{(\uparrow\uparrow)} = k_1 I_{s1} + \frac{2N_{11}I_{s1} - N_{21}I_{s2}}{1 - \epsilon} \quad (6)$$

as critical, that is, as one that causes a transition from the third to the second state to take place. Similarly, we can determine the critical fields for a transition from the third to the fourth state

$$H_{c2}^{(\uparrow\uparrow)} = k_2 I_{s2} + \frac{2N_{22}I_{s2} - N_{21}I_{s1}}{\epsilon}, \quad (7)$$

from the third to the first

$$H_{c3}^{(\uparrow\uparrow)} = \frac{a_1 I_{s1}^2 + a_2 I_{s2}^2 + 2(N_{21} - N_{12})I_{s1}I_{s2}}{(1 - \epsilon)I_{s1} + \epsilon I_{s2}}, \quad (8)$$

from the fourth to the first

$$H_{c1}^{(\uparrow\downarrow)} = k_1 I_{s1} + \frac{2N_{11}I_{s1} + N_{21}I_{s2}}{1 - \epsilon}, \quad (9)$$

from the second to the first

$$H_{c2}^{(\uparrow\downarrow)} = k_2 I_{s2} + \frac{2N_{22}I_{s2} + N_{21}I_{s1}}{\epsilon}, \quad (10)$$

and from the second to the fourth

$$H_{c4}^{(\downarrow\uparrow)} = \frac{a_1 I_{s1}^2 + a_2 I_{s2}^2 + 2(N_{21} + N_{12})I_{s1}I_{s2}}{|(1 - \epsilon)I_{s1} - \epsilon I_{s2}|}. \quad (11)$$

The critical fields for transitions from the first state to the third, $H_{c3}^{(\uparrow\uparrow)}$, and from the fourth to the second, $H_{c4}^{(\downarrow\uparrow)}$, are defined by equations (8) and (11), respectively.

2.3. Ground and Metastable States of a Two-Phase Particle

Equations (6) and (7) suggest that the critical fields H_c may take on both positive and negative values. A negative critical field should be construed as implying the unfeasibility for the particle to reside in the $(\uparrow\uparrow)$ or $(\downarrow\downarrow)$ state. Should a particle find itself in one of them, then at $H_{c1}^{(\uparrow\uparrow)} \leq 0$ it will spontaneously pass from the $(\downarrow\downarrow)$ state to the second or fourth state ($H_{c2}^{(\uparrow\uparrow)} \leq 0$). The foregoing may be given a simple geometrical interpretation. Let the ensemble consist of particles differing in phase size ϵ and phase length-to-width ratio q . Then to each particle we may assign a point on the $\{\epsilon, q\}$ phase plane (see Fig. 2). The curve such that $H_{c1}^{(\uparrow\uparrow)}(\epsilon, q) = 0$ and $H_{c2}^{(\uparrow\uparrow)}(\epsilon, q) = 0$ will separate the phase plane into two parts. The particles whose representative points fall under the curve can reside only in one of the states where the magnetic moments of the phases are aligned

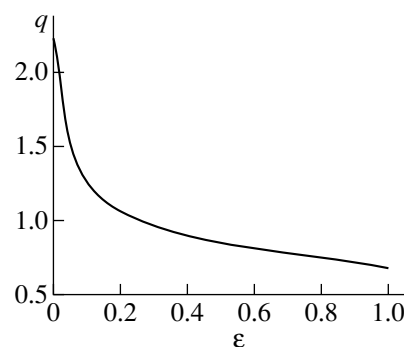


Fig. 2. Magnetic state (ϵ, q) diagram of an ensemble of $\text{Co}_x\text{Zn}_{2-x}\text{Ba}_3\text{Fe}_{24}\text{O}_4$ particles. Cobalt concentration in the first phase, $x_1 = 0$; in the second, $x_2 = 0.5$.

antiparallel, i.e., there is no metastability. The particles represented by points above the curve can reside in both the ground and metastable states.

Quite clearly, the metastability depends on the relative strength of the interphase magnetostatic interaction and the interaction of spin magnetic moments with the crystal field. As the grain length-to-width ratio q increases, the strength of the interphase magnetostatic interaction decreases, and this enhances the role of the crystallographic anisotropy that governs both the parallel and antiparallel alignment of phase magnetizations to the same extent.

The foregoing investigation of the equilibrium states in which a two-phase particle can exist provides a good stepping stone toward a study of an ensemble of such particles.

3. THE STATE DISTRIBUTION OF TWO-PHASE PARTICLES

Consider an ensemble of identical noninteracting two-phase particles placed in an external magnetic field \mathbf{H} at some temperature T . Since the particles are small in volume, the thermal fluctuations experienced by the magnetic moments of the phases may be expected to initiate transitions between states in a field H that is weaker than any critical field of the spectrum of H_c discussed above [see (11) through (16)].

3.1. Frequency of Transitions between Equilibrium States

A chance for the magnetic moment of a phase to change its alignment is determined by the height of the potential barrier E_{ik} that separates the i th and k th states. Proceeding as in [11], we take the frequency of transitions from the i th to the k th state to be expressed in terms of E_{ik} as

$$W_{ik} = f_0 \exp(-E_{ik}/k_B T), \quad (12)$$

where f_0 ranging from 10^7 to 10^{10} s^{-1} is the characteristic frequency of attempts to overcome the potential barrier, k_B is Boltzmann's constant, $E_{ik} = F_{k \max} - F_{i \min}$, where $F_{i \min}$ is the free energy of the equilibrium state in which the particle resided prior to the transition, and $F_{k \max}$ is the maximum free energy that separates the i th and k th states. For example, with $H_{c2}^{(\uparrow\uparrow)} > 0$, we have

$$E_{12} = F(\theta_1 = 0, \theta_2 = \arccos[-(H\varepsilon + N_{21}I_{s1})/a_2I_{s1}]) - F(\theta_1 = 0, \theta_2 = 0) = \frac{\varepsilon^2(H_{c2}^{(\uparrow\uparrow)} + H)^2}{2(k_2\varepsilon + 2N_{22})}qa^3. \quad (13)$$

If $H_{c2}^{(\uparrow\uparrow)} \leq 0$, then, as noted earlier, the $(\uparrow\uparrow)$ state is unfeasible, and the problem of finding W_{12} reduces to the problem of the random walk of a unit vector over a sphere, whose solution to a first approximation is $W_{12} = f_0 \exp(-2H\varepsilon I_{s2}qa^3/k_B T)$. In a similar way, we can calculate E_{ik} for the remaining 11 transitions (see Appendix II) and, thus, for the frequency W_{ik} .

3.2. The Equation of "Motion" for the State Vector and Its Solution

We introduce the occupancy vector normalized to unity, $N(t) = \{N_1(t), N_2(t), N_3(t), N_4(t)\}$. If the initial state $\mathbf{n}_0 = \{n_1, n_2, n_3, n_4\}$ of an ensemble of two-phase particles is unstable, then a transition to equilibrium may be regarded as a discrete-state Markov process, which is described by a system of four equations

$$\frac{dN_i(t)}{dt} = \sum_{k \neq i}^4 (-W_{ik}N_i + W_{ki}N_k) \quad (14)$$

subject to the initial conditions $N_i(t=0) = n_i, i, k = 1, 2, 3, 4$.

By using the normalization condition

$$N_1 + N_2 + N_3 + N_4 = 1 \quad (15)$$

and eliminating N_4 from (14), we may rewrite it (14) in matrix notation as

$$\frac{dN}{dt} = \mathbf{W}\mathbf{N} + \mathbf{V}, \quad (16)$$

where

$$\mathbf{W} = \begin{pmatrix} -(W_{12} + W_{13} + W_{14} + W_{41}) & W_{21} - W_{41} & W_{31} - W_{41} \\ W_{12} - W_{42} & -(W_{21} + W_{23} + W_{24} + W_{42}) & W_{32} - W_{42} \\ W_{13} - W_{42} & W_{23} - W_{43} & -(W_{31} + W_{32} + W_{34} + W_{43}) \end{pmatrix},$$

$$\mathbf{N} = \begin{pmatrix} N_1 \\ N_2 \\ N_3 \end{pmatrix}, \quad \mathbf{V} = \begin{pmatrix} W_{41} \\ W_{42} \\ W_{43} \end{pmatrix}. \quad (17)$$

It is convenient to write the solution of (16) using the matrix exponential function (see Appendix III)

$$\mathbf{N}(t) = \exp(\mathbf{W}t)\mathbf{n}_0 + \int_0^t \exp(\mathbf{W}(t-\tau))d\tau\mathbf{V}. \quad (18)$$

Given the initial state vector \mathbf{n}_0 , equations (17) and (18) will completely define the occupancy of magnetic states in an ensemble of two-phase particles.

APPENDIX I

Magnetostatic Energy of a Two-Phase Grain

The magnetostatic energy of a grain may be regarded as the energy of interaction between magnetic charges with a surface density

$$\varepsilon_m = \frac{1}{2} \iint_{S,S'} \frac{(\mathbf{I}_s(\mathbf{r})d\mathbf{S})(\mathbf{I}_s(\mathbf{r}')d\mathbf{S}')}{|\mathbf{r} - \mathbf{r}'|}. \quad (I.1)$$

Because the problem is symmetric, we may rewrite it (I.1) as

$$\varepsilon_m = \frac{1}{2} \sum_{i,k=1}^8 \iint_{S_i,S_k} \frac{(\mathbf{I}_s(\mathbf{r}_i)d\mathbf{S}_i)(\mathbf{I}_s(\mathbf{r}_k)d\mathbf{S}_k)}{|\mathbf{r}_i - \mathbf{r}_k|}, \quad (I.2)$$

where \mathbf{r}_i are the coordinates of points on the surface S_i (see Fig. 1).

Because the magnetization vector components of the first and second phases are expressed in terms of the direction cosines, $\mathbf{I}_{s1} \{I_{s1} \sin \theta_1, 0, I_{s1} \cos \theta_1\}$, $\mathbf{I}_{s2} \{I_{s2} \sin \theta_2, 0, I_{s2} \cos \theta_2\}$, it follows that

$$\varepsilon_m = N_{11}I_{s1}^2 \sin^2 \theta_1 + N_{22}I_{s2}^2 \sin^2 \theta_2 + N_{12}I_{s1}I_{s2} \sin \theta_1 \sin \theta_2 + N_{21}I_{s1}I_{s2} \cos \theta_1 \cos \theta_2, \quad (I.3)$$

where

$$N_{11} = \frac{4}{q} \left\{ \int_0^{1-\varepsilon} (1-\varepsilon-y)[f_1(y) - f_2(y)]dy + \frac{1}{2}(1-q)(1-\varepsilon)^2 \left[\ln(1-\varepsilon) - \frac{3}{2} \right] \right\},$$

$$N_{22} = \frac{4}{q} \left\{ \int_0^\varepsilon (\varepsilon - y)[f_1(y) - f_2(y)] dy + \frac{1}{2}(1 - q)\varepsilon^2 \left[\ln \varepsilon - \frac{3}{2} \right] \right\},$$

$$N_{12} = \frac{4}{q} \left[\int_0^\varepsilon f_1(y) y dy + \varepsilon \int_\varepsilon^{1-\varepsilon} f_1(y) y dy + \int_{1-\varepsilon}^1 (1 - y) f_1(y) y dy \right]$$

$$+ 2[\varepsilon^2 \ln \varepsilon + (1 - \varepsilon)^2 \ln(1 - \varepsilon) + 3\varepsilon(1 - \varepsilon)],$$

$$N_{21} = \frac{4}{q} \left[\int_0^\varepsilon f_2(y) y dy + \varepsilon \int_\varepsilon^{1-\varepsilon} f_2(y) y dy + \int_{1-\varepsilon}^1 (1 - y) f_2(y) y dy \right]$$

$$+ \frac{2}{q} [\varepsilon^2 \ln \varepsilon + (1 - \varepsilon)^2 \ln(1 - \varepsilon) + 3\varepsilon(1 - \varepsilon)],$$

$$f_1(y) = q \ln \left\{ \frac{(\sqrt{q^2 + y^2} + q)\sqrt{1 + y^2}}{(\sqrt{1 + q^2 + y^2} + q)|y|} \right\} - \sqrt{q^2 + y^2}$$

$$+ \sqrt{1 + q^2 + y^2} - \sqrt{1 + y^2} - |y| + q \ln y,$$

$$f_2(y) = q \ln \left\{ \frac{(\sqrt{1 + y^2} + q)\sqrt{1 + y^2}}{\sqrt{1 + q^2 + y^2}|y|} \right\} - \sqrt{q^2 + y^2}$$

$$+ \sqrt{1 + q^2 + y^2} - \sqrt{1 + y^2} - |y| + \ln y.$$

APPENDIX II

Expressions for Energy Barriers

$$E_{12} = \begin{cases} \frac{\varepsilon^2 (H_{c2}^{(\uparrow\uparrow)} + H)^2}{2(k_2\varepsilon + 2N_{22})}, & H_{c2}^{(\uparrow\uparrow)} > 0 \\ 2H\varepsilon I_{s2}, & H_{c2}^{(\uparrow\uparrow)} < 0, \end{cases} \quad (II.1)$$

$$E_{14} = \begin{cases} \frac{(1 - \varepsilon)^2 (H_{c1}^{(\uparrow\uparrow)} + H)^2}{2(k_1(1 - \varepsilon) + 2N_{11})}, & H_{c1}^{(\uparrow\uparrow)} > 0 \\ 2H(1 - \varepsilon)I_{s2}, & H_{c1}^{(\uparrow\uparrow)} < 0, \end{cases} \quad (II.2)$$

$$E_{41} = \begin{cases} \frac{(1 - \varepsilon)^2 (H_{c1}^{(\uparrow\downarrow)} - H)^2}{2(k_1(1 - \varepsilon) + 2N_{11})}, & H_{c1}^{(\uparrow\downarrow)} > H \\ -2H(1 - \varepsilon)I_{s2}, & H_{c1}^{(\uparrow\downarrow)} < H, \end{cases} \quad (II.3)$$

$$E_{34} = \begin{cases} \frac{\varepsilon^2 (H_{c2}^{(\uparrow\uparrow)} - H)^2}{2(k_2\varepsilon + 2N_{22})}, & H_{c2}^{(\uparrow\uparrow)} > H \\ -2H\varepsilon I_{s2}, & H_{c2}^{(\uparrow\uparrow)} < H, \end{cases} \quad (II.4)$$

$$E_{21} = \begin{cases} \frac{\varepsilon^2 (H_{c2}^{(\uparrow\downarrow)} - H)^2}{2(k_2\varepsilon + 2N_{22})}, & H_{c2}^{(\uparrow\downarrow)} > H \\ -2H\varepsilon I_{s2}, & H_{c2}^{(\uparrow\downarrow)} < H, \end{cases} \quad (II.5)$$

$$E_{32} = \begin{cases} \frac{(1 - \varepsilon)^2 (H_{c1}^{(\uparrow\uparrow)} - H)^2}{2(k_1(1 - \varepsilon) + 2N_{11})}, & H_{c1}^{(\uparrow\uparrow)} > H \\ -2H(1 - \varepsilon)I_{s2}, & H_{c1}^{(\uparrow\uparrow)} < H, \end{cases} \quad (II.6)$$

$$E_{23} = \frac{(1 - \varepsilon)^2 (H_{c1}^{(\uparrow\downarrow)} + H)^2}{2(k_1(1 - \varepsilon) + 2N_{11})}, \quad (II.7)$$

$$E_{43} = \frac{\varepsilon^2 (H_{c2}^{(\uparrow\downarrow)} + H)^2}{2(k_2\varepsilon + 2N_{22})}, \quad (II.8)$$

$$E_{13} = \frac{(H_{c3}^{(\uparrow\uparrow)} + H)^2 (I_{s1}(1 - \varepsilon) + I_{s2}\varepsilon)}{2H_{c3}^{(\uparrow\uparrow)}}, \quad (II.9)$$

$$E_{31} = \begin{cases} \frac{(H_{c3}^{(\uparrow\uparrow)} - H)^2 (I_{s1}(1 - \varepsilon) + I_{s2}\varepsilon)}{2H_{c3}^{(\uparrow\uparrow)}}, & H_{c3}^{(\uparrow\uparrow)} > H \\ -2H(I_{s1}(1 - \varepsilon) + I_{s2}\varepsilon), & H_{c3}^{(\uparrow\uparrow)} < H, \end{cases} \quad (II.10)$$

$$E_{24} = \begin{cases} \frac{(H_{c4}^{(\downarrow\uparrow)} - H)^2 |I_{s1}(1 - \varepsilon) - I_{s2}\varepsilon|}{2H_{c4}^{(\downarrow\uparrow)}}, & H_{c4}^{(\downarrow\uparrow)} > H, \varepsilon > \frac{I_{s1}}{I_{s1} + I_{s2}} \\ -2H |I_{s1}(1 - \varepsilon) - I_{s2}\varepsilon|, & H_{c4}^{(\downarrow\uparrow)} > H, \varepsilon > \frac{I_{s1}}{I_{s1} + I_{s2}} \\ \frac{(H_{c4}^{(\downarrow\uparrow)} + H)^2 |I_{s1}(1 - \varepsilon) - I_{s2}\varepsilon|}{2H_{c4}^{(\downarrow\uparrow)}}, & H_{c4}^{(\downarrow\uparrow)} > H, \varepsilon < \frac{I_{s1}}{I_{s1} + I_{s2}} \\ \varepsilon < \frac{I_{s1}}{I_{s1} + I_{s2}}, & \end{cases} \quad (II.11)$$

$$E_{42} = \begin{cases} \frac{(H_{c4}^{(\downarrow\uparrow)} - H)^2 |I_{s1}(1 - \varepsilon) - I_{s2}\varepsilon|}{2H_{c4}^{(\downarrow\uparrow)}}, \\ H_{c4}^{(\downarrow\uparrow)} > H, \quad \varepsilon < \frac{I_{s1}}{I_{s1} + I_{s2}} \\ -2H |I_{s1}(1 - \varepsilon) - I_{s2}\varepsilon|, \\ H_{c4}^{(\downarrow\uparrow)} > H, \quad \varepsilon < \frac{I_{s1}}{I_{s1} + I_{s2}} \\ \frac{(H_{c4}^{(\downarrow\uparrow)} + H)^2 |I_{s1}(1 - \varepsilon) - I_{s2}\varepsilon|}{2H_{c4}^{(\downarrow\uparrow)}}, \\ \varepsilon > \frac{I_{s1}}{I_{s1} + I_{s2}}. \end{cases} \quad (II.12)$$

APPENDIX III

Expression of a Matrix Exponential Function in Terms of the Transition Matrix

To achieve the task stated in the title of this Appendix, we take advantage of the Sylvester–Lagrange interpolation polynomial. If the matrix **W** does not have multiple roots ($\lambda_0 \neq \lambda_1 \neq \lambda_2$), this polynomial may be written as

$$r(\lambda) = \sum \exp(\lambda_k t) \frac{\prod_{i \neq k} (\lambda - \lambda_i)}{\prod_{i \neq k} (\lambda_i - \lambda_k)}. \quad (III.1)$$

If **W** has two multiple roots, say, $\lambda_0 \neq \lambda_1 \neq \lambda_2$, then

$$r(\lambda) = \left[\frac{\alpha}{(\lambda - \lambda_1)} + \frac{\beta}{(\lambda - \lambda_2)^2} + \frac{\gamma}{(\lambda - \lambda_2)} \right] \Psi(\lambda), \quad (III.2)$$

where $\Psi(\lambda) = (\lambda - \lambda_1)(\lambda - \lambda_2)^2$ is the minimal polynomial, and

$$\alpha = \left[\frac{r(\lambda)}{\Psi(\lambda)} (\lambda - \lambda_1) \right]_{\lambda = \lambda_1} = \frac{\exp(\lambda_1 t)}{(\lambda_1 - \lambda_2)^2},$$

$$\beta = \left[\frac{r(\lambda)}{\Psi(\lambda)} (\lambda - \lambda_1)^2 \right]_{\lambda = \lambda_2} = \frac{\exp(\lambda_2 t)}{\lambda_1 - \lambda_2},$$

$$\gamma = \frac{\partial}{\partial \lambda} \left[\frac{r(\lambda)}{\Psi(\lambda)} (\lambda - \lambda_1)^2 \right]_{\lambda = \lambda_2} = \frac{[t(\lambda_2 - \lambda_1) - 1] \exp(\lambda_2 t)}{(\lambda_1 - \lambda_2)^2}.$$

If there are three multiple roots ($\lambda_0 = \lambda_1 = \lambda_2$), then

$\Psi(\lambda) = (\lambda - \lambda_1)^3$, and

$$r(\lambda) = \left[\frac{\alpha}{(\lambda - \lambda_1)^3} + \frac{\beta}{(\lambda - \lambda_1)^2} + \frac{\gamma}{\lambda - \lambda_1} \right] \Psi(\lambda), \quad (III.3)$$

where

$$\alpha = [r(\lambda)]_{\lambda = \lambda_1} = \exp(\lambda_1 t),$$

$$\beta = \frac{\partial}{\partial \lambda} [r(\lambda)]_{\lambda = \lambda_1} = t \exp(\lambda_1 t),$$

$$\gamma = \frac{\partial^2}{\partial \lambda^2} [\exp(\lambda_1 t) (\lambda - \lambda_1)^2]_{\lambda = \lambda_1} = \frac{1}{2} t^2 \exp(\lambda_1 t).$$

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