

# Modeling the Magnetization of an Ensemble of Chemically Inhomogeneous Superparamagnetic Titanomagnetite Particles

L. L. Afremov and A. V. Panov

*Far East State University, Vladivostok, Russia*

Received December 20, 1995; in final form, June 25, 1996

This paper continues the theoretical study of an ensemble of small, chemically inhomogeneous ferromagnetic particles [1–3] and is devoted to developing a more realistic model (compared to those presented, for example, in [4–8]) of a natural ferromagnet and to estimating various types of remanent magnetization, on the basis of the model.

We consider an ensemble of identical, noninteracting two-phase particles in an external field  $H$  at temperature  $T$ .

It was previously shown [1, 2] that, in the absence of an external magnetic field, the two-phase particle may exist in one of the following states: in the first ( $\uparrow\uparrow$ )-state, the magnetic moments of both phases are parallel and directed along the axis  $OZ$  (Fig. 1); in the second ( $\uparrow\downarrow$ )-state, the magnetizations of the phases are antiparallel and the magnetic moment  $m_1$  of the first phase is directed along the axis  $OZ$ ; in the third ( $\downarrow\downarrow$ )-state, both phase magnetizations are opposite to that in the first one; and in the fourth ( $\downarrow\uparrow$ )-state, the magnetic moments of the first and second phases are directed opposite to and along the axis  $OZ$ , respectively. The first and third states are metastable, since the free energy of the grain in these states is larger than that in the second and fourth states. All the states of a particle in an external magnetic field  $H$  directed along the axis  $OZ$ , except the first one, are metastable, but the most unstable one is the third state. The transitions from one state to another may occur at  $H$  larger than the corresponding critical fields given below: for the transition from the third to the second state,

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

for the transition from the third to the fourth state,

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

for the transition from the third to the first state,

$$H_{c3}^{(\uparrow\uparrow)} = \frac{a_1 I_{s1}^2 + a_2 I_{s2}^2 + 2(N_{12} - N_{21})I_{s1}I_{s2}}{(1 - \varepsilon)I_{s1} + \varepsilon I_{s2}} \quad (3)$$

for the transition from the fourth to the first state,

$$H_{c1}^{(\uparrow\downarrow)} = k_1 I_{s1} + \frac{2N_{11}I_{s1} + N_{21}I_{s2}}{1 - \varepsilon}, \quad (4)$$

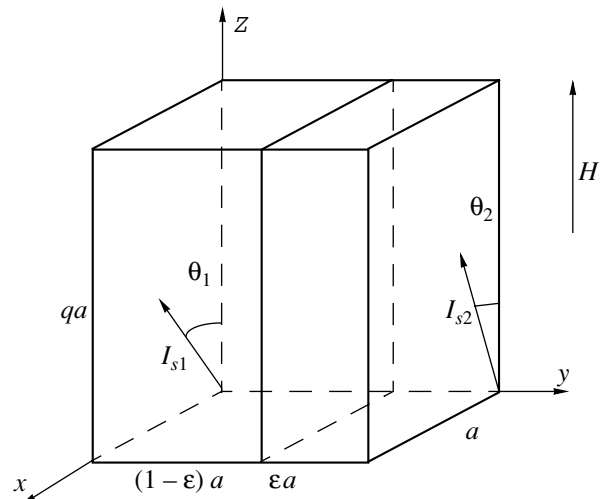
for the transition from the second to the first state,

$$H_{c2}^{(\uparrow\downarrow)} = k_2 I_{s2} + \frac{2N_{22}I_{s2} + N_{21}I_{s1}}{\varepsilon}, \quad (5)$$

and for the transition from the second to the fourth state,

$$H_{c4}^{(\downarrow\uparrow)} = \frac{a_1 I_{s1}^2 + a_2 I_{s2}^2 - 2(N_{12} - N_{21})I_{s1}I_{s2}}{|(1 - \varepsilon)I_{s1} - \varepsilon I_{s2}|}. \quad (6)$$

Here,  $I_{s1}$  and  $I_{s2}$  are the spontaneous magnetizations,  $k_1$  and  $k_2$  are the dimensionless constants of crystallographic anisotropy,  $1 - \varepsilon$  and  $\varepsilon$  are the relative volumes of the first and second phases, respectively,  $a_1 = 2N_{11} + k_1(1 - \varepsilon)$ ,  $a_2 = 2N_{22} + k_2\varepsilon$ , and  $N_{ik}$  are the demagnetization coefficients, determined by the form and relative sizes of the phases (see Application I).



**Fig. 1.** Model of a two-phase particle.

In view of the small volume of particles, the transitions from one state to another, in a field  $H$  smaller than the corresponding  $H_c$  from (1)–(6), are expected to be realized due to thermal fluctuations of the magnetic moments of the phases.

FREQUENCY OF TRANSITION FROM ONE EQUILIBRIUM STATE TO ANOTHER

The reorientation of the magnetic moment of one of the phases is determined by the potential barrier  $E_{ik}$  separating the  $i$ th and  $k$ th states. Following Neel [4], the frequency of transition from the  $i$ th to the  $k$ th state can be written in terms of  $E_{ik}$  as follows:

$$W_{ik} = f_0 \exp(-E_{ik}/k_b T), \tag{7}$$

where  $f_0 \sim 10^8\text{--}10^{10} \text{ s}^{-1}$  is the characteristic frequency of the attempts to overcome the potential barrier,  $k_b$  is Boltzmann's constant,  $E_{ik} = F_{k\max} - F_{i\min}$ ,  $F_{i\min}$  is the free energy of the equilibrium state in which the particle existed before the transition, and  $F_{k\max}$  is the maximum value of the free energy difference between the  $i$ th and  $k$ th states. For example, as  $H_{c2}^{(\uparrow\uparrow)} > 0$ ,

$$E_{12} = F(\theta_1 = 0, \theta_2 = \arccos[-(H\varepsilon + N_{21}I_{s1})/a_2I_s]) - F(\theta_1 = 0, \theta_2 = 0) = \frac{\varepsilon^2(H_{c2}^{(\uparrow\uparrow)} + H)^2}{2(k_2\varepsilon + 2N_{22})} qa^3, \tag{8}$$

where, according to Afremov *et al.* [1, 2],

$$F(\theta_1, \theta_2) = \frac{1}{2}(k_1I_{s1}^2(1 - \varepsilon)\sin^2\theta_1 + k_2I_{s2}^2\varepsilon\sin^2\theta_2)qa^3 - H(I_{s1}(1 - \varepsilon)\cos\theta_1 + I_{s2}\varepsilon\cos\theta_2)qa^3 + (N_{11}I_{s1}^2\sin^2\theta_1 + N_{22}I_{s2}^2\sin^2\theta_2)$$

$$+ N_{12}I_{s1}I_{s2}\sin\theta_1\sin\theta_2 + N_{21}I_{s1}I_{s2}\cos\theta_1\cos\theta_2)qa^3.$$

For  $H_{c2}^{(\uparrow\uparrow)} \leq 0$ , the  $(\uparrow\uparrow)$ -state is not realized, and the problem of finding  $W_{12}$  can be reduced to the problem on random wandering of the unit vector over a sphere. To the first approximation, its solution is  $W_{12} = f_0 \exp(-2H\varepsilon I_{s2} qa^3/k_b T)$ . The value of  $E_{ik}$  (and, consequently, the frequency  $W_{ik}$ ) for the remaining 11 transitions can be calculated in a similar way (see Application II).

MOTION EQUATION FOR THE STATE VECTOR AND ITS SOLUTION

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

$$\frac{dN_i(t)}{dt} = \sum_{k \neq i}^4 (-W_{ik}N_i + W_{ik}N_k) \tag{9}$$

with the initial conditions  $N_i(t=0) = n_i$ ,  $i, k = 1, \dots, 4$ .

Using the normalization condition

$$N_1 + N_2 + N_3 + N_4 = 1 \tag{10}$$

and eliminating  $N_4$  from (9), we rewrite set (9) in the matrix form

$$\frac{d\mathbf{N}}{dt} = \mathbf{W}\mathbf{N} + \mathbf{V}, \tag{11}$$

where the adopted notation is

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

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

It is convenient to write the solution to (11) with the help of the matrix exponent (see Application III):

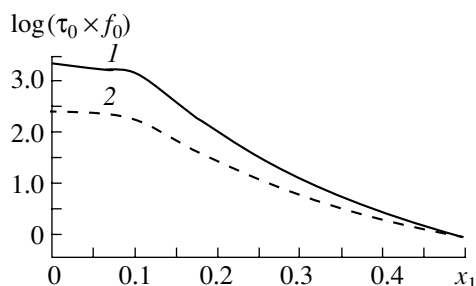
$$\mathbf{N}(t) = \exp(\mathbf{W}t)\mathbf{n}_0 + \int_0^t \exp(\mathbf{W}(t - \tau))d\tau \cdot \mathbf{V}. \tag{13}$$

Given the initial state vector  $\mathbf{n}_0$ , relations (12) and (13) completely determine the magnetic state population for an ensemble of two-phase particles.

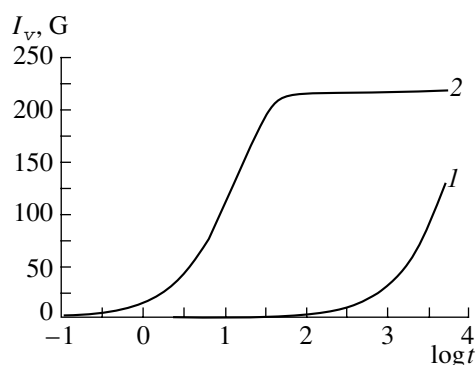
The ensemble magnetization can be calculated from the relation

$$I_v = c[(I_{s1}(1 - \varepsilon) + I_{s2}\varepsilon)(N_1 - N_3) + (I_{s1}(1 - \varepsilon) + I_{s2}\varepsilon)(N_2 - N_4)], \tag{14}$$

where  $c$  is the volume concentration of ferromagnetic particles.



**Fig. 2.** The relaxation time  $\tau_0$  versus the Ti concentration  $x_1$  for an ensemble of chemically homogeneous ( $\epsilon = 0$ , curve 1) and inhomogeneous ( $\epsilon = 0.3$ , curve 2) particles with  $x_2 = 0.6$ ,  $a = 10$  nm, and  $q = 1.5$ .



**Fig. 3.** Viscous magnetization of an ensemble of chemically homogeneous ( $\epsilon = 0$ , curve 1) and inhomogeneous ( $\epsilon = 0.3$ , curve 2) titanomagnetite grains with the parameters  $x_1 = 0.2$ ,  $x_2 = 0.6$ ,  $q = 1.5$ ,  $a = 18$  nm,  $H = 50$  Oe, and  $f_0 = 10^9$  s $^{-1}$  ( $t$  in seconds).

### MAGNETIZATION OF AN ENSEMBLE OF SMALL, CHEMICALLY INHOMOGENEOUS NONINTERACTING PARTICLES

Let us model the magnetization of an ensemble of small two-phase particles whose magnetic properties are similar to those of titanomagnetite. The incorrectness of the chosen magnet (because of the cubic symmetry of their crystal cell, the titanomagnetites are magnetically multiaxial) may be justified by the fact that, with an increase in the grain length, the equivalence of the crystallographic axes disappears, and, for  $q > 1.2$ , the "lightest" magnetization axis is controlled by the anisotropy of the shape [6, 9, 10]. Moreover, the use of the developed model may help to establish some classical relations for titanomagnetites, which are the principal carriers of rock magnetization.

In order to model the magnetization process, we use the linearly interpolated measurements of the spontaneous magnetization  $I_s = I_s(x)$  and the crystallographic anisotropy  $k = k(x)$  of titanomagnetites  $\text{Fe}_{3-x}\text{Ti}_x\text{O}_4$  ver-

sus the titanium concentration  $x$  [11]:

$$I_s(x) = (-598.5x + 485) \text{ G},$$

$$k = \begin{cases} 0.425(1.36 + 11.4x), & x < 0.1 \\ 0.425(2.89 - 3.9x), & x > 0.1. \end{cases} \quad (15)$$

In the calculations, the first and second (at the right of Fig. 1) phases were assumed to be depleted in and enriched with titanium, respectively ( $x_1 < x_2$ ).

### RELAXATION TIME AND THE VISCOUS MAGNETIZATION OF AN ENSEMBLE OF NONINTERACTING PARTICLES

The three eigenvalues of matrix  $\mathbf{W}$ , determined from the equation  $\det|\mathbf{W} - \lambda\mathbf{E}| = 0$ , where  $\mathbf{E}$  is the unit matrix, may be interpreted as the inverse relaxation times  $\tau_i$ . Furthermore, the smallest  $\tau$  and greatest  $\tau_0$  characterize the life spans of the most unstable and most stable states, respectively.

Figure 2 presents the calculated dependences of the relaxation time  $\tau_0$  on the titanium content  $x_1$  in the first phase of an ensemble of two-phase particles.

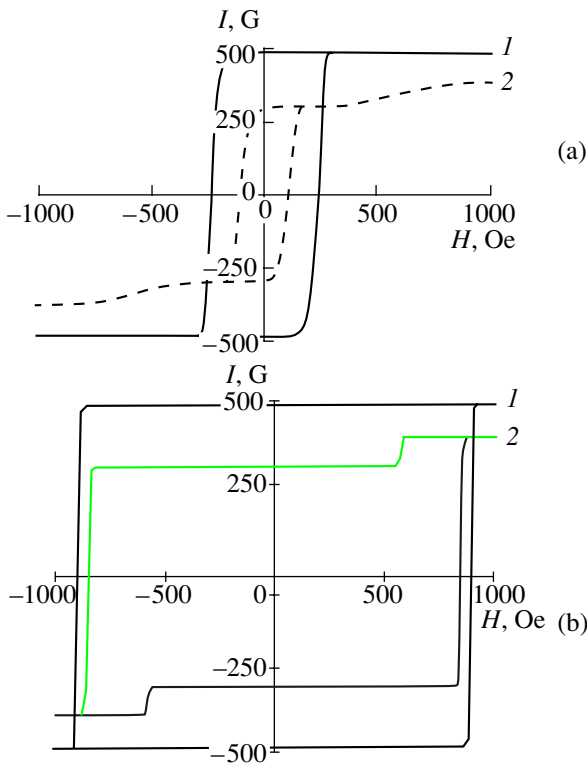
Decreasing the chemical inhomogeneity of the grain (increasing  $x_1$ ) results in a decrease in  $\tau_0$  (Fig. 2); the relaxation time being practically invariable in a range of  $x_1 < 0.1$  and sharply falling for  $x_1 > 0.1$ . Such a behavior of the relaxation time is due to a decrease in  $I_s(x)$  and to a nonmonotone variation in the crystallographic anisotropy constant  $k(x)$ .

The viscous magnetization  $I_v(t)$  versus  $\log(t)$  was calculated based on our model of interacting particles (Fig. 3). As would be expected, a decrease in the relaxation time, while augmenting the chemical inhomogeneity, results in greater viscous magnetization.

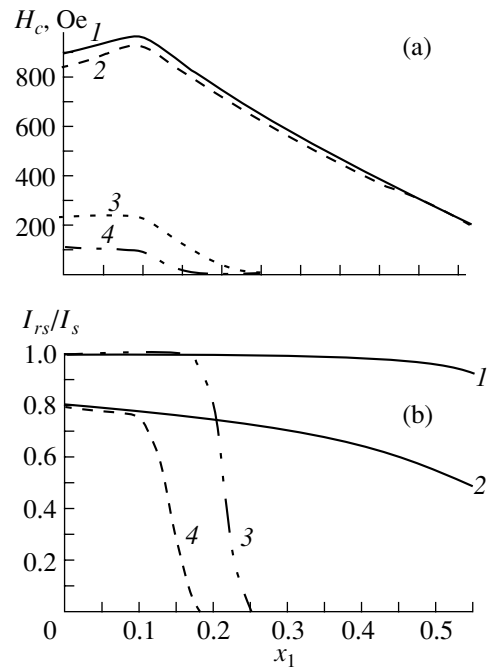
### THE HYSTERETIC PROPERTIES AND REMANENT MAGNETIZATION OF AN ENSEMBLE OF NONINTERACTING TWO-PHASE GRAINS

In the superparamagnetic state, small particles may exhibit hysteretic properties only on the condition that either time elapses after the external magnetic field is turned off or the time for which the field drops to zero,  $H = 0$ , is smaller than the relaxation time. On account of this, we used the following modeling procedure: the magnetic field varied discretely by a few steps up to the maximum value, and the magnetization which formed at each step for a time shorter than the relaxation time was determined from relations (13) and (14), where the initial state vector was taken to be equal to the finite state vector of the preceding step. It is natural that, with an increase in the grain size, a similar situation can be realized in a reasonable time interval (comparable to the time interval of measurements).

The coercivity  $H_c$  and the remanent saturation magnetization  $I_{rs}$ , determined from the hysteresis loop



**Fig. 4.** Hysteresis loops for a system of two-phase particles with the parameters  $x_1 = 0$ ,  $x_2 = 0.6$ ,  $q = 1.5$ ,  $f_0 = 10^9 \text{ s}^{-1}$ ,  $\epsilon = 0$  (curve 1), and  $\epsilon = 0.3$  (curve 2). (a)  $a = 15 \text{ nm}$ ,  $T = 10 \text{ s}$ ; (b)  $a = 50 \text{ nm}$ ,  $T = 10^3 \text{ s}$ ;  $T$  is the period of the field  $H$  variation.



**Fig. 5.** (a) Coercivity  $H_c$  and (b) the ratio of remanent saturation magnetization to spontaneous magnetization,  $I_{rs}/I_s$ , versus the titanium concentration  $x_1$  ( $x_2 = 0.6$ ,  $q = 1.5$ ). (1)  $\epsilon = 0$ ,  $a = 50 \text{ nm}$ , (2)  $\epsilon = 0.3$ ,  $a = 50 \text{ nm}$ , (3)  $\epsilon = 0$ ,  $a = 15 \text{ nm}$ , (4)  $\epsilon = 0.3$ ,  $a = 15 \text{ nm}$ .

(Fig. 4), are presented in Fig. 5. A slight change in the hysteresis parameters for  $x_1 < 0.1$ , and their abrupt drop for  $x > 0.1$ , is due to the nonmonotonic behavior of the crystallographic anisotropy parameter (increase in  $k(x)$  for  $x < 0.1$  and its decrease for  $x > 0.1$ , according to (15)). Such a variation in the coercivity and in the remanent saturation magnetization is ultimately related to a decrease in  $\tau_0(x_1)$ , which is caused by reducing the inhomogeneity degree of the titanium distribution over the grain volume (Fig. 2).

CONCLUSION

Increasing the chemical inhomogeneity of titanomagnetite grains, while decreasing the titanium content in one of the phases, leads to an increase in the relaxation time of a system of such particles and, as a result, to an increase in the hysteresis parameters, the coercivity, and the remanent saturation magnetization.

APPLICATION I.  
MAGNETOSTATIC ENERGY  
OF A TWO-PHASE GRAIN

The magnetostatic energy of a grain can be thought of as the energy of interaction between the magnetic

charges of the surface density

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

Using the symmetry of the problem, we can rewrite (I.1) as follows:

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

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

The magnetization components of the first and second phases are expressed in terms of the directing cosines:  $\mathbf{I}_{s1} = \{I_{s1} \sin \theta_1, 0, I_{s1} \cos \theta_1\}$ ,  $\mathbf{I}_{s2} = \{I_{s2} \sin \theta_2, 0, I_{s2} \cos \theta_2\}$ , and, therefore,

$$\begin{aligned} \epsilon_m = & N_{11} I_{s1}^2 \sin^2 \theta_1 + N_{22} I_{s2}^2 \sin^2 \theta_2 \\ & + N_{12} I_{s1} I_{s2} \sin \theta_1 \sin \theta_2 + N_{21} I_{s1} I_{s2} \cos \theta_1 \cos \theta_2, \end{aligned} \quad (\text{I.3})$$

where

$$\begin{aligned} N_{11} = & \frac{4}{q} \left\{ \int_0^{1-\epsilon} (1-\epsilon-y)[f_1(y) - f_2(y)] dy \right. \\ & \left. + \frac{1}{2} (1-q)(1-\epsilon)^2 \left[ \ln(1-\epsilon) - \frac{3}{2} \right] \right\}; \end{aligned}$$

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

$$N_{12} = \frac{4}{q} \left[ \int_0^\varepsilon f_1(y) y dy + \varepsilon \int_\varepsilon^{1-\varepsilon} f_1(y) y dy + \int_{1-\varepsilon}^1 (1 - y) f_1(y) y dy \right] + 2[\varepsilon^2 \ln \varepsilon + (1 - \varepsilon)^2 \ln(1 - \varepsilon) + 3\varepsilon(1 - \varepsilon)];$$

$$N_{21} = \frac{4}{q} \left[ \int_0^\varepsilon f_2(y) y dy + \varepsilon \int_\varepsilon^{1-\varepsilon} f_2(y) y dy + \int_{1-\varepsilon}^1 (1 - y) f_2(y) y dy \right] + \frac{2}{q}[\varepsilon^2 \ln \varepsilon + (1 - \varepsilon)^2 \ln(1 - \varepsilon) + 3\varepsilon(1 - \varepsilon)];$$

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

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

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

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

APPLICATION II.  
EXPRESSIONS FOR THE POTENTIAL BARRIERS

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

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

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

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

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

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

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

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

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

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

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

$$E_{42} \left\{ \begin{array}{l} \frac{(H_{c4}^{(\downarrow\uparrow)} - H)^2 |I_{s1}(1 - \epsilon) - I_{s2}\epsilon|}{2H_{c4}^{(\downarrow\uparrow)}} \\ H_{c4}^{(\downarrow\uparrow)} > H, \quad \epsilon < \frac{I_{s1}}{I_{s1} + I_{s2}} \\ -2H|I_{s1}(1 - \epsilon) - I_{s2}\epsilon|, \quad H_{c4}^{(\downarrow\uparrow)} > H, \quad \epsilon < \frac{I_{s1}}{I_{s1} + I_{s2}} \\ \frac{(H_{c4}^{(\downarrow\uparrow)} + H)^2 |I_{s1}(1 - \epsilon) - I_{s2}\epsilon|}{2H_{c4}^{(\downarrow\uparrow)}}, \quad \epsilon > \frac{I_{s1}}{I_{s1} + I_{s2}} \end{array} \right. \quad (\text{II.12})$$

APPLICATION III.  
THE MATRIX COMPONENT EXPRESSED  
IN TERMS OF THE TRANSITION MATRIX

In order to solve the problem stated in the title of this application, we use the Sylvester–Lagrange interpolating polynomial, which, in the absence of the multiple roots of the matrix  $\mathbf{W}$  ( $\lambda_0 \neq \lambda_1 \neq \lambda_2$ ), takes the form

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

If  $\mathbf{W}$  has two multiple roots, for example,  $\lambda_0 = \lambda_1 \neq \lambda_2$ , we have

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

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

$$\begin{aligned} \alpha &= \left[ \frac{r(\lambda)}{\psi(\lambda)} (\lambda - \lambda_1) \right]_{\lambda = \lambda_1} = \frac{\exp(\lambda_1 t)}{(\lambda_1 - \lambda_2)^2}, \\ \beta &= \left[ \frac{r(\lambda)}{\psi(\lambda)} (\lambda - \lambda_1)^2 \right]_{\lambda = \lambda_2} = \frac{\exp(\lambda_2 t)}{\lambda_1 - \lambda_2}, \\ \gamma &= \frac{\partial}{\partial \lambda} \left[ \frac{r(\lambda)}{\psi(\lambda)} (\lambda - \lambda_1)^2 \right]_{\lambda = \lambda_2} \\ &= \frac{[t(\lambda_2 - \lambda_1) - 1] \exp(\lambda_2 t)}{(\lambda_1 - \lambda_2)^2}. \end{aligned}$$

In the case of three multiple roots ( $\lambda_0 = \lambda_1 = \lambda_2$ ),  $\psi(\lambda) = (\lambda - \lambda_1)^3$ ,

$$\begin{aligned} r(\lambda) &= \left[ \frac{\alpha}{(\lambda - \lambda_1)^3} + \frac{\beta}{(\lambda - \lambda_1)^2} + \frac{\gamma}{(\lambda - \lambda_1)} \right] \psi(\lambda), \quad (\text{III.3}) \\ \alpha &= [r(\lambda)]_{\lambda = \lambda_1} = \exp(\lambda_1 t), \\ \beta &= \frac{\partial}{\partial \lambda} [r(\lambda)]_{\lambda = \lambda_1} = t \exp(\lambda_1 t), \end{aligned}$$

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

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