The Computation of Modified Landau-Lifshitz Equation under an AC Field

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Abstract: An accurate magnetization requires that both the reversible and irreversible components be modeled. The classical Landau-Lifshitz model deals with only the irreversible component of magnetization. We first subject the Landau-Lifshitz equation to an AC external field by performing a computation through the closed-form solution and the resulting hysteresis loop is displayed to show its deficiency. Then we modify the Landau-Lifshitz model into a new one by including a reversible part and an irreversible part accompanying with the switching criteria between these two states. With the new solutions we display the influence of parameters on the hysteresis loops of magnetic materials under AC fields.

Keyword: Landau-Lifshitz equation, Magnetization, Hysteresis loop

1 Introduction

In order to simulate the hysteretic phenomenon of ferromagnetic materials, there have been several physical models currently in use. However, the following model:

\[ \dot{M} = -\gamma M \times H_{\text{eff}} - \gamma \alpha \frac{M}{M_s} \times (M \times H_{\text{eff}}) \]  

proposed by Landau and Lifshitz (1935) is still the most popular one being used widely and plays a central role in the description of the micromagnetic dynamics of ferromagnetic media.

From the above equation it is apparent that \( M \cdot \dot{M} = 0 \); hence, the magnitude of magnetization vector \( M(t) \) is conserved, i.e., \( ||M(t)|| = M_s = \text{constant} \). Throughout this paper, a dot between two vectors stands for their scalar product, and \( ||\bullet|| \) denotes the magnitude of vector. The two material parameters of \( \gamma > 0 \) and \( \alpha \geq 0 \) are, respectively, the absolute value of gyromagnetic ratio and the damping constant [Bertotti, Mayergoyz and Serpico (2001)]. The effective field \( H_{\text{eff}} \) is the sum of applied field, demagnetizing field, anisotropy field and exchange field.

The Landau-Lifshitz equation is essential to the interpretation of the dynamics of domain wall [Zhai (1997)], ferromagnetic resonance [Fetisov, Patton and Sygonach (1999)], and the magnetization switching in thin film recording media [Schrefl, Fidler, Süss and Scholz (2000)]. Recently, some analytical results were obtained by Bertotti, Serpico and Mayergoyz (2001) and Bertotti, Mayergoyz and Serpico (2004) for the magnetic body exhibiting rotational symmetry about a certain axis and the external field being circularly polarized in the perpendicular plane. Besides that very few exact solutions are known for the nonlinear large magnetization motions. Usually, the majority of nonlinear studies are carried out by the numerical integration techniques [Serpico, Mayergoyz and Bertotti (2001); Krishnaprasad and Tan (2001); Frank (2004); Liu and Ku (2005); d’Aquino, Serpico and Miano (2005)].

In many technical applications of ferromagnetic materials the coercivity is considered to be one of the most important parameters in applied magnetism. The coercivity is responsible for the non-zero value of the external magnetic field required to reduce the total magnetic moment of the ferromagnetic sample to zero. The coercivity being in a close correlation with the hysteresis losses of the ferromagnetic material is usually determined from the width of the hysteresis loop as shown in Fig. 1, where \( M_s, M_r \) and \( H_c \) are respectively the saturation magnetization, the remanent magneti-
zation and the coercive magnetic field. In this paper we study the Landau-Lifshitz equation (1) under an AC field \( \mathbf{H}_{\text{eff}} = H_{\text{eff}}^0 \sin \Omega t \), where \( H_{\text{eff}}^0 \) is a constant amplitude vector in three positive directions and \( \Omega \) is the excitation frequency [Vértesy and Magni (2003)]. Rivkin and Ketterson (2006) have studied the magnetization reversal using various rf magnetic pulses, numerically showing that the switching is possible with simple sinusoidal pulses. Lee and Yuan (2007) have used an oscillating field to study the magnetization reversal, showing that the oscillating field reduces the coercivity significantly.

In this paper we first give an outline of the linear representation of the Landau-Lifshitz equation in Section 2. The linearization is important for getting the closed-form solution in Section 3. In Section 4 we display a magnetic hysteresis loop obtained from the Landau-Lifshitz equation to show that the solution does not give a physically relevant hysteresis loop and explain what the reason to cause this deficiency; hence, we propose a modification of the Landau-Lifshitz equation by including a reversible part and an irreversible part accompanying with the switching criteria between these two states. With the new solutions we show the influence of the parameters on the hysteresis loops, which are more closely correlated with the typical hysteresis loops for most magnetic materials. Finally, we draw some conclusions in Section 5.

### 2 Linear representation

Let us define a unit vector

\[
m := \frac{\mathbf{M}}{||\mathbf{M}||} = \frac{\mathbf{M}}{M_s},
\]

and use a new time scale \( t' = \gamma M_s t \) and a new field \( \mathbf{H} = H_{\text{eff}} / M_s \) for saving notations, and then Eq. (1) can be rearranged to

\[
\frac{dm}{dt'} = \hat{\mathbf{H}}m + \alpha \mathbf{H} - \alpha \mathbf{H} \cdot mm,
\]

where

\[
\hat{\mathbf{H}} := \begin{bmatrix}
0 & -H_3 & H_2 \\
H_3 & 0 & -H_1 \\
-H_2 & H_1 & 0
\end{bmatrix}
\]

is skew-symmetric, and \( H_i, i = 1, 2, 3 \), are three independent components of \( \mathbf{H} \).

Liu (2004) has proved that the Landau-Lifshitz equation (3) can be linearized to (see also Appendix A)

\[
\frac{dX}{dt'} = AX
\]

in the four-dimensional Minkowski space with \( X \in M^4 \) satisfying the cone condition of \( X^T g X = 0 \), where \( \tau \) denotes the transpose and \( g \) is a Minkowski metric given by

\[
g = \begin{bmatrix}
I_3 & 0_{3 \times 1} \\
0_{1 \times 3} & -1
\end{bmatrix}
\]

with \( I_3 \) the third order identity matrix. In above, we have defined

\[
X = \begin{bmatrix}
X^3 \\
X^0
\end{bmatrix} = \begin{bmatrix}
X^1 \\
X^2 \\
X^3 \\
X^0
\end{bmatrix} := X^0 \begin{bmatrix}
m \\
1
\end{bmatrix}
\]

as the augmented state vector, and

\[
A := \begin{bmatrix}
\hat{\mathbf{H}} & \alpha \mathbf{H} \\
\alpha \mathbf{H}^T & 0
\end{bmatrix}
= \begin{bmatrix}
0 & -H_3 & H_2 & \alpha H_1 \\
H_3 & 0 & -H_1 & \alpha H_2 \\
-H_2 & H_1 & 0 & \alpha H_3 \\
\alpha H_1 & \alpha H_2 & \alpha H_3 & 0
\end{bmatrix}
\]

as the system matrix, satisfying the Lie algebraic property of \( A^T g + gA = 0 \), which is known as the Lie algebra for the Lorentz group \( SO_o(3, 1) \) [Liu (2001)].
3 Closed-form solution

Now we search the closed-form solution of Eq. (1) under the AC field $\mathbf{H}_\text{eff} = \mathbf{H}_0^0 \sin \Omega t$. Let us define a new amplitude vector

$$\mathbf{H}^0 = \frac{\mathbf{H}_\text{eff}^0}{M_s},$$

and then to a constant linear system (15).

Equation (15) becomes

$$\frac{d\mathbf{X}}{d\tau} = \mathbf{B} \mathbf{X},$$

where

$$\mathbf{B} = \begin{bmatrix} \mathbf{B}_s^0 & \mathbf{B}_0^0 \\ (\mathbf{B}_0^0)^T & 0 \end{bmatrix} = \begin{bmatrix} \mathbf{H}^0 & \alpha \mathbf{H}^0 \\ \alpha (\mathbf{H}^0)^T & 0 \end{bmatrix}$$

is a constant matrix, satisfying the Lie algebraic property of $\mathbf{B}^T \mathbf{g} + \mathbf{g} \mathbf{B} = 0$.

There is a useful property for $\mathbf{B}$:

$$(\mathbf{B}_s^0)^k \mathbf{B}_0^0 = 0,$$

where $k$ is any positive integer.

Considering the transformation of time variables by

$$\frac{d\tau}{d\tilde{t}} = \sin \omega \tilde{t},$$

and then using Eq. (11) we obtain a constant linear system:

$$\frac{d\mathbf{X}}{d\tau} = \mathbf{B} \mathbf{X}.$$

Up to here we have transformed the nonlinear equation (1) to a time-varying linear system (5) and then to a constant linear system (15).

We solve Eq. (15) by decomposing it into two parts:

$$\frac{d\mathbf{X}_s}{d\tau} = \mathbf{B}_s \mathbf{X}_s + \mathbf{X}_0^0 \mathbf{B}_0^0,$$

$$\frac{d\mathbf{X}_0^0}{d\tau} = \mathbf{B}_0^0 \cdot \mathbf{X}_s.$$

Differentiating Eq. (17), inserting Eq. (16) for the differential of $\mathbf{X}_s$ and utilizing Eq. (13), we obtain

$$\frac{d^2\mathbf{X}_0^0}{d\tau^2} = ||\mathbf{B}_0^0||^2 \mathbf{X}_0^0.$$

The general solution of the above equation is

$$\mathbf{X}_0^0(\tau) = C_1 \cosh ||\mathbf{B}_0^0|| (\tau - \tau_i)$$

$$+ C_2 \sinh ||\mathbf{B}_0^0|| (\tau - \tau_i),$$

where $\tau_i$ is an initial time, and $C_1$ and $C_2$ are determined by

$$C_1 = \mathbf{X}_0^0(\tau_i), \ C_2 = \frac{1}{||\mathbf{B}_0^0||^2} \mathbf{B}_0^0 \cdot \mathbf{X}_s(\tau_i).$$

Taking advantage of $\mathbf{X}_0^0$ derived above, and using Eq. (16) the solution for $\mathbf{X}_s$ can be obtained as follows:

$$\mathbf{X}_s(\tau) = \exp[\mathbf{B}_s^0(\tau - \tau_i) \mathbf{X}_s(\tau_i)]$$

$$+ \int_{\tau_i}^\tau \exp[\mathbf{B}_s^0(\tau - \xi) \mathbf{X}_0^0(\xi)] d\xi \mathbf{B}_0^s,$$

where $\mathbf{X}_s(\tau_i)$ is an initial value of $\mathbf{X}_s$. By applying the Cayley-Hamilton theorem for $\mathbf{B}_s^0$ and through some calculations as given in Appendix B we get

$$\exp[\mathbf{B}_s^0(\tau - \tau_i)] = \mathbf{I}_3 + \sin ||\mathbf{H}^0|| (\tau - \tau_i) \frac{\mathbf{B}_s^0}{||\mathbf{H}^0||}$$

$$+ \left[1 - \cos ||\mathbf{H}^0|| (\tau - \tau_i)\right] \left(\frac{\mathbf{B}_s^0}{||\mathbf{H}^0||}\right)^2 ||\mathbf{H}^0||^2.$$

Inserting Eq. (19) for $\mathbf{X}_0^0$ into Eq. (21) and utilizing Eqs. (13) and (22), we obtain

$$\mathbf{X}_s(\tau)$$

$$= \left\{ \mathbf{I}_3 + \sin ||\mathbf{H}^0|| (\tau - \tau_i) \frac{\mathbf{B}_s^0}{||\mathbf{H}^0||}$$

$$+ \left[1 - \cos ||\mathbf{H}^0|| (\tau - \tau_i)\right] \left(\frac{\mathbf{B}_s^0}{||\mathbf{H}^0||}\right)^2 ||\mathbf{H}^0||^2 \right\} \mathbf{X}_s(\tau_i)$$

$$+ \left\{ \frac{C_1}{||\mathbf{B}_0^0||} \sinh ||\mathbf{B}_0^0|| (\tau - \tau_i)$$

$$+ \frac{C_2}{||\mathbf{B}_0^0||} \left[\cosh ||\mathbf{B}_0^0|| (\tau - \tau_i) - 1\right] \right\} \mathbf{B}_0^s.$$
We can insert
\[ \tau - \tau_i = -\frac{1}{\Omega} \left[ \cos \omega t' - \cos \omega t_i \right] \]
\[ = -\frac{\gamma M_s}{\Omega} \left[ \cos \Omega t - \cos \Omega t_i \right] \]
into Eqs. (19) and (23) and then divide the latter one by the former one to obtain \( \mathbf{m}(t) \) as follows:
\[
\mathbf{m}(t) = \left\{ \begin{array}{l}
I_3 + \sin \| \mathbf{H}^0 \| (\tau - \tau_i) \frac{B_s^t}{\| \mathbf{H}^0 \|} \\
+ \left[ 1 - \cos \| \mathbf{H}^0 \| (\tau - \tau_i) \right] \frac{(B_s^i)^2}{\| \mathbf{H}^0 \|^2} \right\} \mathbf{m}(t_i) \\
+ \left\{ \frac{1}{\| B_0^i \|} \sinh \| B_0^i \| (\tau - \tau_i) \\
+ \frac{B_0^i \cdot \mathbf{m}(t_i)}{\| B_0^i \|} \left[ \cosh \| B_0^i \| (\tau - \tau_i) - 1 \right] \right\} B_0^i \\
+ \left\{ \cosh \| B_0^i \| (\tau - \tau_i) \\
+ \frac{B_0^i \cdot \mathbf{m}(t_i)}{\| B_0^i \|} \sinh \| B_0^i \| (\tau - \tau_i) \right\}. \right. \]
(25)

From the above equation \( \mathbf{M}(t) = M_s \mathbf{m}(t) \) can be calculated. In the next section we will use the above equation to simulate the magnetic hysteresis.

4 A modification

In the following calculations we are fixed the gyromagnetic ratio to be \( \gamma = 221021 \) m/As, the saturated magnetization to be \( M_s = 1000000 \sqrt{3} \) A/m and the three components of the initial values of \( \mathbf{M} \) to be \( M_1 = M_2 = 1000000 \) A/m and \( M_3 = -1000000 \) A/m. In addition that we use \( \alpha = 0.004, \Omega = 150000 \) rad/s, and \( H_{\text{eff}}^0(1) = H_{\text{eff}}^0(2) = 0 \) A/m and \( H_{\text{eff}}^0(3) = 1500 \) A/m in Fig. 2.

As shown in Fig. 2 the hysteresis loop computed by the Landau-Lifshitz equation is not practical, since the dissipative term always works when \( \alpha > 0 \), and which can be seen that the hysteresis loop attains its saturation state very soon. Although the third component \( M_3 \) can turn its direction from negative to positive under the vertical AC field, however, the third component \( M_3 \) is standing on a constant value too long even the vertical AC field changes its direction from positive to negative. Recalling that \( M_s H_{\text{eff}}^0 / \| H_{\text{eff}}^0 \| \) is a limiting state of the Landau-Lifshitz equation when \( H_{\text{eff}}^0 \) acts in one direction. Therefore, we can say that the hysteresis loop calculated by the Landau-Lifshitz equation has a drawback that the magnetization orbit is confined near to a limiting state too long even the external AC field changes its direction to cause the limiting state vector changing its direction. This situation makes the hysteresis curve in Fig. 2 obtained from the Landau-Lifshitz equation is quite not similar to the usual one as shown in Fig. 1.

The above conditions give us an incentive to modify the Landau-Lifshitz equation. As discussed by Della Torre (1999) the dynamical magnetization process can be divided into a reversible part and an irreversible part. Thus, we modify the Landau-Lifshitz equation into two parts:
\[
\dot{\mathbf{M}} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\gamma \alpha}{M_s} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}}), \quad \text{if } \| \mathbf{M} \| = M_s \quad \text{and} \quad \mathbf{H}_{\text{eff}} \cdot \mathbf{M} > 0, \quad (26)
\]
\[
\dot{\mathbf{M}} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \gamma \alpha M_s \mathbf{H}_{\text{eff}}, \quad \text{if } \| \mathbf{M} \| < M_s \quad \text{or} \quad \mathbf{H}_{\text{eff}} \cdot \mathbf{M} \leq 0. \quad (27)
\]
The first equation corresponds to the irreversible part, while the second equation corresponds to the reversible part. Eq. (26) is the same as Eq. (1), but equipped with a saturation condition $\|\mathbf{M}\| = M_s$ and a switching condition $\mathbf{H}_{\text{eff}} \cdot \mathbf{M} > 0$.

From Eq. (26) we have

$$\dot{\mathbf{M}} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\gamma \alpha}{M_s} [\mathbf{M} \cdot \mathbf{H}_{\text{eff}} \mathbf{M} - \|\mathbf{M}\|^2 \mathbf{H}_{\text{eff}}].$$

(28)

By using $\|\mathbf{M}\|^2 = M_s^2$ in the irreversible state, the above equation further reduces to

$$\dot{\mathbf{M}} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \gamma \alpha M_s \mathbf{H}_{\text{eff}} - \frac{\gamma \alpha}{M_s} \mathbf{M} \cdot \mathbf{H}_{\text{eff}} \mathbf{M}.$$

(29)

It can be seen that the last term in the above disappears in Eq. (27). The last term is a dissipative one, and correspondingly the term $\gamma \alpha M_s \mathbf{H}_{\text{eff}}$ is a conservative one. Liu (2000) has established a general setting of the dynamical system, which is controlled by the difference of the conservative force and the dissipative force.

Reversible and irreversible changes in the magnetization may occur together during the magne-
tization process. The irreversible processes are usually associated with the dissipation of energy through domain wall motion or moment switching in single domain particles, while the reversible processes are usually linked with moment rotation or domain wall displacement in a single potential well. Consideration of the energy changes involved in these different processes leads to a natural separation between the magnetization gained for the system involving generation of heat (irreversible magnetization) and that where no heat is generated (reversible magnetization).

Let us compare Eq. (27) with the modified Bloch equation, which takes the relaxation effect into account:

\[ \mathbf{M} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\chi_0}{\tau_y} \mathbf{H}_{\text{eff}} - \frac{1}{\tau_y} \mathbf{M}, \]

where \( \chi_0 \) is the susceptibility of the magnetic material and \( \tau_y \) is the relaxation time during the precession. If we let \( \chi_0 / \tau_y = \gamma \alpha M_s \) and ignore the last term in Eq. (30), which reflects the irreversibly relaxed phenomenon of magnetization, then we get Eq. (27). In passing, we note the differences between the modified Bloch equation and the Landau-Lifshitz equation: Eq. (30) is linear but Eq. (26) is nonlinear, and \( ||\mathbf{M}|| = M_s \) is an invariance of Eq. (26) but not an invariance of Eq. (30). These two equations are identical only when \( \mathbf{H}_{\text{eff}} \) is proportional to \( \mathbf{M} \).

For Eq. (27) we have the following closed-form solution:

\[
\mathbf{M}(t) = \left\{ \mathbf{I} + \sin||\mathbf{H}^0||(\tau - \tau_i) \frac{\mathbf{B}_1^2}{||\mathbf{H}^0||} \right. \\
\left. + \left[1 - \cos||\mathbf{H}^0||(\tau - \tau_i)\right] \frac{(\mathbf{B}_1^0)^2}{||\mathbf{H}^0||^2} \right\} \mathbf{M}(t_i) \\
+ \alpha(\tau - \tau_i) \mathbf{H}_{\text{eff}}. \tag{31}
\]

where \( \mathbf{B}_1^0 \) was defined by Eq. (12) and \( \tau - \tau_i \) was defined by Eq. (24).

Now, we are in a good position to simulate the magnetization hysteresis with the above formulæ: Eq. (25) applicable in the irreversible state and Eq. (31) applicable in the reversible state. We use the same parameters values as those used in Fig. 2 to show a typical magnetization response in Fig. 3. As shown in Fig. 3(a) the magnetization magnitude \( ||\mathbf{M}|| \) is varied between the irreversible state with \( ||\mathbf{M}|| = M_s \) and the reversible state with \( ||\mathbf{M}|| < M_s \). As shown in Figs. 3(b) and 3(c) the first and the second components of \( \mathbf{M} \) have high frequency oscillation, while the hysteresis loop shown in Fig. 3(d) for the vertical component reveals a certain stable behavior. Furthermore, the first and the second components almost hold a constant oscillating amplitude during the reversible state and upon entering the irreversible state they fast tend to zero after the first half cycle of the AC input field and then remain in the zero values until the end of the input; hence, in the steady state the direction of magnetization is along only in the vertical direction, and the switching of magnetization direction is rather fast when the AC field changes its direction.

When keeping all the parameters values unchanged, in the following calculations we merely change \( \alpha = 0.01 \) by considering a larger damping constant in Fig. 4, \( \Omega = 300000 \) rad/s by considering a higher exciting frequency in Fig. 5, and \( \mathbf{H}^0_{\text{eff}}(3) = 2000 \) A/m by considering a larger amplitude of AC field in Fig. 6. As usual a larger damping constant renders a smaller hysteresis loop, a higher exciting frequency renders a larger hysteresis loop, and of course a larger amplitude of excitation leads to a larger hysteresis loop.

![Figure 4: The hysteresis loop is plotted under \( \alpha = 0.01, \Omega = 150000 \) rad/s, and \( \mathbf{H}^0_{\text{eff}}(3) = 1500 \) A/m.](image)
5 Conclusions

According to the linearization of the Landau-Lifshitz equation derived by Liu (2004), we have derived here a closed-form solution of the magnetization when subjected to an AC field along the vertical direction, which can simulate the switching of magnetization direction of a magnetic thin film. However, we found that the Landau-Lifshitz equation can not simulate the hysteresis loop of usual type very well. Therefore, we proposed a modification by considering the decomposition of the magnetization into an irreversible part and a reversible part and derived the switching criteria of these two states. According to the closed-form solutions we have simulated the magnetic hysteresis loops, which are closer to the real ones under different parameters values.

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References


Appendix A

In this appendix we derive Eq. (5). Upon defining the integrating factor of

Eq. (3) can be arranged to

\[
\frac{d}{dt'}(X^0 m) = X^0 \dot{H}m + \alpha X^0 H. \tag{A2}
\]

On the other hand, from Eq. (A1) it follows that

\[
\frac{d}{dt'}X^0 = \alpha X^0 H \cdot m. \tag{A3}
\]

Let us introduce

\[
X = \begin{bmatrix} X^0 \\ m \end{bmatrix} := \begin{bmatrix} X^0 \\ X^1 \\ X^2 \\ X^3 \\ X^0 \end{bmatrix} = \begin{bmatrix} X^0 m_1 \\ X^0 m_2 \\ X^0 m_3 \\ X^0 \end{bmatrix} \tag{A4}
\]

such that Eqs. (A2) and (A3) are combined together into a single equation (5).

Appendix B

In this appendix we derive Eq. (22). From Eq. (12) we obtain a characteristic equation for \( B_s^i \):

\[
\det(\lambda I_3 - B_s^i) = \lambda^3 + ||H^0||^2 \lambda = 0, \tag{B1}
\]

where \( \det \) denotes the determinant and \( \lambda \) is the eigenvalue of \( B_s^i \). By the Cayley-Hamilton theorem \( B_s^i \) also satisfies the above equation, that is,

\[
(B_s^i)^3 + ||H^0||^2 B_s^i = 0. \tag{B2}
\]

Splitting the exponential series of \( \exp[B_s^i(\tau - \tau_i)] \) into odd and even powers, leads to

\[
\exp[B_s^i(\tau - \tau_i)] = I_3 + (\tau - \tau_i)B_s^i + \frac{(\tau - \tau_i)^3}{3!} - \frac{(\tau - \tau_i)^5}{5!} (B_s^i)^5 + \ldots + \frac{(\tau - \tau_i)^{2n+1}}{(2n+1)!} (B_s^i)^{2n+1} + \ldots + \frac{(\tau - \tau_i)^2}{2!} (B_s^i)^2 + \frac{(\tau - \tau_i)^4}{4!} (B_s^i)^4 + \ldots + \frac{(\tau - \tau_i)^{2n}}{(2n)!} (B_s^i)^{2n} + \ldots \tag{B3}
\]
However, by applying $B_s$ to Eq. (B2) repeatedly, we obtain the following recurrence relations:

$$(B_s^i)^3 = -\|H^0\|^2 B_s^i, \quad (B_s^i)^4 = -\|H^0\|^2 (B_s^i)^2,$$

$$(B_s^i)^5 = \|H^0\|^4 B_s^i, \quad (B_s^i)^6 = \|H^0\|^4 (B_s^i)^2, \ldots \quad (B4)$$

Replacing the higher order terms in Eq. (B3) by the above formulae we obtain

$$\exp[B_s^i(\tau - \tau_i)] = I_3 + \left[ \frac{(\tau - \tau_i)^3 \|H^0\|^3}{3!} + \frac{(\tau - \tau_i)^5 \|H^0\|^5}{5!} + \cdots + \frac{(-1)^n(\tau - \tau_i)^{2n+1} \|H^0\|^{2n+1}}{(2n+1)!} \frac{B_s^i}{\|H^0\|} \right]

\left[ \frac{(\tau - \tau_i)^2 \|H^0\|^2}{2!} - \frac{(\tau - \tau_i)^4 \|H^0\|^4}{4!} + \cdots + \frac{(-1)^{n+1}(\tau - \tau_i)^{2n} \|H^0\|^{2n}}{(2n)!} \right] \frac{(B_s^i)^2}{\|H^0\|^2}. \quad (B5)$$

Recalling that

$$\sin \theta = \sum_{k=0}^{\infty} (-1)^k \frac{\theta^{2k+1}}{(2k+1)!},$$

$$1 - \cos \theta = \sum_{k=1}^{\infty} (-1)^{k+1} \frac{\theta^{2k}}{(2k)!},$$

and from Eq. (B5) we can derive Eq. (22).