Phase interference in antiferromagnetic quantum tunneling with
an arbitrarily directed magnetic field

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Abstract

The quantum interference effects induced by the topological phase are studied analytically in biaxial antiferromagnets with an external magnetic field at an arbitrarily angle. This study provides a nontrivial generalization of the Kramers degeneracy for equivalent double-well system to coherently spin tunneling at ground states as well as low-lying excited states for antiferromagnetic system with asymmetric twin barriers. The spin-phase interference effects are found to depend on the orientation of the magnetic field distinctly.

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One recent experiment on the molecular magnets Fe$_8$ showed a direct evidence of the topological part of the quantum spin phase (Berry phase) in the spin dynamics. The importance of the topological Berry phase in spin tunneling was elucidated by Loss et al., and von Delft and Henly. They showed that this term can lead to destructive (for half-integer total spins) and instructive (for integer spins) interference between opposite winding tunneling paths in single-domain ferromagnetic (FM) particles. While spin-parity effects are sometimes be related to Kramers degeneracy, they typically go beyond the Kramers theorem in a rather unexpected way. Similar spin-parity effects were found in antiferromagnetic (AFM) particles, where only the integer excess spins can tunnel but not the half-integer ones. The effects of magnetic field along the hard and medium axis were studied in AFM particles.

Theoretical results showed that in AFM particles the exchange energy is enhanced to the magnetic anisotropy, which leads to tunneling of the Néel vector a much stronger effect than tunneling of magnetization in FM particles. Therefore, the AFM particle is expected to be a better candidate for observing quantum tunneling than the FM particle with a similar size. Up to now theoretical studies on AFM tunneling have been focused on spin-phase interference between two opposite ground-state tunneling paths. The spin-phase interference between excited-level tunneling paths are unknown for AFM particles. Moreover, the previous works have been confined to the condition that the magnetic field be applied along the easy, medium, or hard axis, separately. The purpose of this letter is to study the resonant quantum tunneling and spin-phase interference at excited levels in AFM particles placed in a magnetic field at an arbitrary angle $\theta_H$. Therefore, our study provides a nontrivial generalization of the Kramers degeneracy for equivalent double-well system to coherently spin tunneling at ground states as well as low-lying excited states for AFM system with asymmetric twin barriers caused by the arbitrarily directed magnetic field.

The system of interest is a small ($\sim 5$nm in radius), single-domain, AFM particle at a temperature well below its anisotropy gap. According to the two-sublattice model, there is a strong exchange energy $\mathbf{m}_1 \cdot \mathbf{m}_2 / \chi_\perp$ between the two sublattices, where $\mathbf{m}_1$ and $\mathbf{m}_2$ are the magnetization vectors of the two sublattices with large, fixed and unequal magnitudes.
In the following, we assume that $m_1 > m_2$ and $m = m_1 - m_2 \ll m_1$. The system has the biaxial symmetry, with $\hat{x}$ being the easy axis, $\hat{y}$ being the medium axis, and $\hat{z}$ being the hard axis. The magnetic field is applied in the $ZY$ plane, at an arbitrary angle in the range of $0 \leq \theta_H < \pi/2$. In order to obtain the tunnel splitting for quantum coherence, we shall calculate the path integral:

$$\int D\{\theta\} D\{\phi\} \exp\left[-S_E(\theta, \phi)\right],$$

where $S_E$ is the effective Euclidean action for AFM tunneling,

$$S_E(\theta, \phi) = \frac{V}{\hbar} \int d\tau \left\{ \frac{1}{\gamma} \left( \frac{d\phi}{d\tau} \right)^2 - \frac{1}{\gamma} \left( \frac{d\phi}{d\tau} \right) \cos \theta + \frac{\chi_\perp}{2\gamma^2} \left( \frac{d\theta}{d\tau} \right)^2 \right\}.$$

$E(\theta, \phi) = E_a(\theta, \phi) - mH_z \cos \theta - mH_y \sin \theta \sin \phi$, and $E_a(\theta, \phi)$ is the magnetocrystalline anisotropy energy. $V$ is the volume of the AFM particle, and $\gamma$ is the gyromagnetic ratio. $\theta$ and $\phi$ are the angular components of $\mathbf{m}_1$, which can also determine the direction of the Néel vector. $\tau = it$, and $m = h\gamma s/V$, where $s$ is the excess spin of the AFM particle due to the non-compensation of two sublattices. For this case, the magnetocrystalline anisotropy energy is $E_a(\theta, \phi) = K_\perp \cos^2 \theta + K_\parallel \sin^2 \theta \sin^2 \phi$, where $K_\parallel$ and $K_\perp$ are the longitudinal and the transverse anisotropy coefficients satisfying $K_\perp \gg K_\parallel > 0$. Therefore, the Néel vector is forced to lie in the $\theta = \theta_0$ plane, and the fluctuations of $\theta$ about $\theta_0$ are small. Introducing $\theta = \theta_0 + \alpha$, $|\alpha| \ll 1$, $E(\theta, \phi)$ reduces to

$$E(\alpha, \phi) = K_\perp \sin^2 \theta_0 \alpha^2 + K_\parallel \sin^2 \theta_0 (\sin \phi - \sin \phi_0)^2$$

$$+ 2K_2 \sin \theta_0 \cos \theta_0 (\sin \phi - \sin \phi_0)^2 \alpha,$$

where $\cos \theta_0 = mH_z/2K_\perp$, $\sin \phi_0 = mH_y/2K_\parallel \sin \theta_0 = h \sin \theta_H/\sqrt{1 - (\lambda h \cos \theta_H)^2}$, $\lambda = K_\parallel/K_\perp$, $h = H/H_0$, and $H_0 = 2K_\parallel/m$.

In the semiclassical limit, the dominant contribution to the transition amplitude comes from finite action solution (instanton) of the classical equation of motion. Since the configuration space of this problem is a circle, only two types of instantons must be taken into account. We use $A$ to denote the instanton passing through the barrier at $\theta = \theta_0$, $\phi = \pi/2$,
and $B$ as the instanton passing through the barrier at $\theta = \theta_0$, $\phi = 3\pi/2$. Correspondingly, there are two kinds of anti-instantons: $A^-$ and $B^-$. The small barrier at $\phi = \pi/2$ is $hU_S = hU (\phi = \pi/2) = K_{\parallel} V \sin^2 \theta_0 (1 - \sin \phi_0)^2$, and the large barrier at $\phi = 3\pi/2$ is $hU_L = hU (\phi = 3\pi/2) = K_{\parallel} V \sin^2 \theta_0 (1 + \sin \phi_0)^2$. Performing the Gaussian integration over $\alpha$, we can map the spin system onto a particle moving problem in one-dimensional potential well. Now the Euclidean transition amplitude of this system becomes

$$ K_E = \exp \left\{ -i \left[ S_{\text{tot}} - \left( 1 + \frac{\lambda}{2} + \frac{\lambda}{2} \sin^2 \phi_0 \right) s \cos \theta_0 - 2 S \left( \frac{S}{s} \right) \left( \frac{K_{\perp}}{J} \right) \cos \theta_0 \sin^2 \theta_0 \right] (\phi_f - \phi_i) \right\} $$

$$ \times \int d\phi \exp \left\{ - \int d\tau \left[ \frac{1}{2} \mathcal{M} \left( \frac{d\phi}{d\tau} \right)^2 + U (\phi) \right] \right\}, \quad (3) $$

where $S_{\text{tot}} = 2S - s$ is the total spins of two sublattices, and $S = m_1 V / h \gamma$ is the sublattice spins. In Eq. (3) we have taken $\chi_\perp = m_1^2 / J$, where $J$ is the exchange interaction between two sublattices. The effective mass and effective potential in Eq. (3) are

$$ \mathcal{M} = \frac{h S^2}{J V} \sin^2 \theta_0 \left[ 1 + \frac{1}{2 \sin^2 \theta_0} \left( \frac{J}{K_{\perp}} \right) \left( \frac{s}{S} \right)^2 \right], \quad (4a) $$

$$ U (\phi) = 2 \frac{K_{\parallel} V}{h} \sin^2 \theta_0 (\sin \phi - \sin \phi_0)^2. \quad (4b) $$

The potential $U (\phi) = U (\phi + 2n\pi)$ has an asymmetric twin barrier. The degenerate ground states are given by two different types of minima of $U (\phi)$ at $2n\pi + \phi_0$ and $(2n + 1)\pi - \phi_0$. $U (\phi)$ can be regarded as a superlattice consisting of two sublattices, and the energy spectrum can be obtained by applying the Bloch theorem and the tight-binding approximation. The translational symmetry is ensured by the possibility of successive $2\pi$ extensions.

The periodic instanton configuration $\phi_p$ satisfies the equation of motion $\frac{1}{2} \mathcal{M} \left( \frac{d\phi_p}{d\tau} \right)^2 - U (\phi_p) = -E$, where $E > 0$ can be viewed as the classical energy of the pseudoparticle configuration. Then the periodic instanton $A$ solution for $0 \leq E \leq U_S$ is $\sin \phi_A = \frac{1 - \xi_1 \sin^2 (\omega_1 \tau, k_1)}{1 + \xi_1 \sin^2 (\omega_1 \tau, k_1)}$, $\sin (\omega_1 \tau, k_1)$ is the Jacobian elliptic sine function of modulus $k_1 = \sqrt{(1 - \alpha)(1 + \beta) / (1 + \alpha)(1 - \beta)}$, where

$$ \alpha = \sin \phi_0 + \frac{hE}{K_{\parallel} V \sin^2 \theta_0}, \quad \beta = \sin \phi_0 - \frac{hE}{K_{\parallel} V \sin^2 \theta_0}, \quad \xi_1 = (1 - \alpha) / (1 + \alpha), \quad \omega_1 = \omega_0 / g_1, \quad \omega_0 = \sqrt{2 K_{\parallel} V / h \mathcal{M} \sin \theta_0}, \quad g_1 = 2 / \sqrt{(1 + \alpha)(1 - \beta)}. $$

The associated Euclidean action is

$$ S_A = \int_{-\beta}^{\beta} d\tau \left[ \frac{1}{2} \mathcal{M} \left( \frac{d\phi_A}{d\tau} \right)^2 + U (\phi_A) \right] = W_A + 2E\beta, \quad (5a) $$

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where
\[
W_A = 4M\omega_1 \left[ E(k_1) + \frac{(k_1^2 - \xi_1)}{\xi_1}K(k_1) + \frac{(\xi_1^2 - k_1^2)}{\xi_1}\Pi(k_1, \xi_1) \right].
\] (5b)

Here \(K(k_1), E(k_1), \) and \(\Pi(k_1, \xi_1)\) are the complete elliptic integral of the first, second, and third kind, respectively. The similar method can be applied to the periodic instanton \(B\), and the result is \(S_B = W_B + 2E\beta\) for \(0 \leq E \leq U_S\), where
\[
W_A = 4M\omega_1 \left[ E(k_1) + \frac{(k_1^2 - \xi_1)}{\xi_1}K(k_1) + \frac{(\xi_1^2 - k_1^2)}{\xi_1}\Pi(k_1, \xi_1) \right],
\] (6)

and \(\xi_2 = (1 + \beta) / (1 - \beta)\). While for \(U_S \leq E \leq U_L\), the result is \(\tilde{S}_B = \tilde{W}_B + 2E\beta\), where
\[
\tilde{W}_B = 2M\xi_3 (1 + \alpha) \omega_2 \left[ \frac{1}{k_2^2 - \xi_3}E(k_2) - \frac{1}{\xi_3}K(k_2) + \frac{k_2^2 + \xi_3^2 + 2k_3^2\xi_3}{\xi_3}\Pi(k_2, \xi_3) \right],
\] (7)

with \(\omega_2 = \frac{\omega(E_n)}{\alpha}, \xi_3 = (1 + \beta) / (1 - \beta), k_2 = \sqrt{2/(\alpha - \beta)}\), and \(k_2^2 = \frac{(\alpha - 1)(1 + \beta)}{2(\alpha - \beta)}\).

Now we turn to the calculation of level splittings of excited states. For a particle moving in a double-well-like potential \(U(x)\), the WKB formula gives the tunnel splittings of the \(n\)th degenerate excited levels or the imaginary parts of the \(n\)th metastable levels as \(\Delta E_n\) (or \(\text{Im} E_n\)) = \(\frac{\omega(E_n)}{\pi} \exp(-W)\) where \(\omega(E_n) = 2\pi/t(E_n)\). \(t(E_n) = \sqrt{2m \int_{x_1(E_n)}^{x_2(E_n)} \frac{dx}{\sqrt{E_n - U(x)}}}\) is the period of the real-time oscillation in the potential well, where \(x_{1,2}(E_n)\) are the turning points for the particle oscillating inside the potential \(U(x)\). For the present case, we find that the level splittings for instantons \(A\) and \(B\) in the domain \(0 \leq E \leq U_S\) are \(\Delta E_{A(B)} = \frac{2}{t_{A(B)}(E)} \exp(-W_{A(B)})\), where \(t_A(E) = t_B(E) = \frac{2}{\omega_1(E)}K(k_1'), \) and \(k_1' = \sqrt{1 - k_1^2}\). For \(U_S \leq E \leq U_L\), the imaginary parts of the metastable energy levels are \(\text{Im} E = \frac{2}{t_B(E)} \exp(-2\tilde{W}_B)\), where \(\tilde{t}_B(E) = \frac{2}{\omega_2(E)}K(k_2'), \) and \(k_2' = \sqrt{1 - k_2^2}\).

Then we discuss the low energy limit of the level splitting. By using the small oscillator approximation for energy near the bottom of the potential well, \(\mathcal{E}_n = (n + 1/2)\Omega, \Omega = \sqrt{d^2U/d\phi^2}_{\phi=\phi_0}/M = \sqrt{2K\langle V \rangle/\hbar M} \sin \theta_0 \cos \phi_0\), Eqs. (5b) and (6) are expanded as
\[
W_{A(B),n} = W_{A(B),0} - \left(n + \frac{1}{2}\right) + \left(n + \frac{1}{2}\right)
\times \ln \left(\frac{n + 1/2}{2^{7/2} \sqrt{\frac{2\pi}{\int S}} \sqrt{1 + \frac{1}{2\sin^2 \theta_0} \left(\frac{I}{K_1}\right)}} \left(\frac{g}{\alpha}\right)^2 \sin^2 \theta_0 \cos^3 \phi_0\right),
\] (8a)
The topological term, we derive the secular equation as
\[ \mathcal{W}_{A(B),0} = 2^{3/2} \sqrt{\frac{K_B}{J}} S \sin^2 \theta_0 \sqrt{1 + \frac{1}{2 \sin^2 \theta_0} \left( \frac{J}{K_\perp} \right) \left( \frac{s}{S} \right)^2} \times \left( \cos \phi_0 \mp 2 \sin \phi_0 \arcsin \sqrt{1 - \sin \phi_0} \right), \quad (8b) \]

where “−” for the instanton A, and “+” for the instanton B. Therefore, the tunnel splittings of nth excited levels are found to be
\[ \hbar \Delta \mathcal{E}_{A(B),n} = \frac{2 \cos \phi_0}{\sqrt{n!}} \frac{\sqrt{K_\perp} J V}{S} \frac{1}{\sqrt{1 + \frac{1}{2 \sin^2 \theta_0} \left( \frac{J}{K_\perp} \right) \left( \frac{s}{S} \right)^2}}^{n+1/2} \exp \left( -\mathcal{W}_{A(B),0} \right). \quad (9) \]

Eq. (8b) shows that at finite magnetic field the WKB exponent for instanton A, \( \mathcal{W}_{A,0} \), is smaller than that for instanton B, \( \mathcal{W}_{B,0} \), because the barrier through which instanton B must tunnel is higher than that for instanton A.

It is noted that \( \hbar \Delta \mathcal{E}_{A,n} \) or \( \hbar \Delta \mathcal{E}_{B,n} \) is only the level shift induced by tunneling between degenerate excited states through a single barrier. The periodic potential \( U(\phi) \) can be regarded as a one-dimensional superlattice with the sublattices A and B. The general translation symmetry results in the energy “band” structure which is formally the same as that of a one-dimensional tight-binding model in solid state physics, and the energy spectrum could be determined by the Bloch theorem. The Bloch states for sublattices A and B can be written as \( \Phi_A(\xi, \phi) = \frac{1}{\sqrt{L}} \sum_n e^{i \xi \phi_n} \varphi_A(\phi - \phi_n) \), and \( \Phi_B(\xi, \phi) = \frac{1}{\sqrt{L}} \sum_n e^{i (\xi \phi_n + a)} \varphi_B(\phi - \phi_n - a) \), where \( \phi_n = 2n\pi + \phi_0 \), \( L = N(a+b) \), \( a = \pi - 2\phi_0 \), and \( b = \pi + 2\phi_0 \). Then the total wavefunction \( \Psi_\xi(\phi) \) is a linear combination of the two Bloch states, \( \Psi_\xi(\phi) = a_A(\xi) \Phi_A(\xi, \phi) + a_B(\xi) \Phi_B(\xi, \phi) \). Including the phase contributions of the topological term, we derive the secular equation as
\[ \begin{bmatrix} \mathcal{E}_n - E(\xi) & e^{i(\xi - \mu)a} \Delta \mathcal{E}_{A,n} + e^{-i(\xi - \mu)b} \Delta \mathcal{E}_{B,n} \\ e^{-i(\xi - \mu)a} \Delta \mathcal{E}_{A,n} + e^{i(\xi - \mu)b} \Delta \mathcal{E}_{B,n} & \mathcal{E}_n - E(\xi) \end{bmatrix} \begin{bmatrix} a_A(\xi) \\ a_B(\xi) \end{bmatrix} = 0, \quad (10a) \]

where
\[ \mu = S_{\text{tot}} - \left( 1 + \frac{\lambda}{2} + \frac{\lambda}{2} \sin^2 \phi_0 \right) s \cos \theta_0 - 2S \left( \frac{S}{s} \right) \left( \frac{K_\perp}{J} \right) \cos \theta_0 \sin^2 \theta_0, \quad (10b) \]
and $E_n = (n + 1/2) \Omega$. The Bloch wave vector $\xi = 0$ in the first Brillouin zone. Therefore, the eigenvalues of Eq. (10a) are

$$E_\pm = E_n \pm \sqrt{(\Delta E_{A,n})^2 + (\Delta E_{B,n})^2 + 2(\Delta E_{A,n})(\Delta E_{B,n}) \cos \Theta},$$

(11a)

where

$$\Theta = 2\pi \left\{ s \left[ 1 + \left( \frac{\lambda}{2} + \frac{\lambda}{2} \sin^2 \phi_0 \right) \cos \theta_0 \right] + 2S \left( \frac{S}{s} \right) \left( \frac{K_1}{J} \right) \cos \theta_0 \sin^2 \theta \right\}. \quad (11b)$$

The tunnel splitting of $n$th excited level is

$$\Delta E_n = 2\sqrt{(\Delta E_{A,n})^2 + (\Delta E_{B,n})^2 + 2(\Delta E_{A,n})(\Delta E_{B,n}) \cos \Theta}. \quad (11c)$$

We can rederive this result by calculating the transition amplitude, or by the effective Hamiltonian method.

At zero magnetic field, $\sin \phi_0 = 0$, $\cos \theta_0 = 0$, $\Delta E_{A,n} = \Delta E_{B,n}$, the tunnel splitting is suppressed to zero for the half-integer excess spins, which is in good agreement with the Kramers theorem. The presence of a magnetic field perpendicular to the plane of rotation of magnetization yields an additional contribution to the topological phase, resulting constructive and destructive interferences alternatively for both integer and half-integer excess spins. Tunneling is thus periodically suppressed. At finite magnetic field, the tunneling spectrum of the degenerate $n$th excited levels depends on the parity of excess spins $s$, 

$$\Delta E_n = 2\left\{ (\Delta E_{A,n})^2 + (\Delta E_{B,n})^2 \pm 2(\Delta E_{A,n})(\Delta E_{B,n}) \right\}^{1/2} \cos \left[ 2\pi \left( \left( 1 + \frac{\lambda}{2} + \frac{\lambda}{2} \sin^2 \phi_0 \right) s \cos \theta_0 + 2S \left( \frac{S}{s} \right) \left( \frac{K_1}{J} \right) \cos \theta_0 \sin^2 \theta \right] \right\} \quad (12)$$

where “+” for integer $s$, and “−” for half-integer $s$. The spin-parity effect and the oscillation of the tunnel splitting with the field is shown in Fig. 1. Another important observation is that only the $\hat{z}$ component of the magnetic field (i.e., along the hard axis) can lead to the oscillation of the tunnel splitting for the highly anisotropic case. As shown in Fig. 2, even a small misalignment of the field with the $\hat{z}$ axis can completely destroy the oscillation effect, and the oscillation is absent when the field is along the medium axis. For small $\theta_H$ the tunnel splitting oscillates with the field, whereas no oscillation is shown up for large $\theta_H$. In the latter case, a much stronger increase of tunnel splitting with the field is shown.
This strong dependence on the orientation of the field can be observed for ground-state resonance as well as excited-state resonance. As a result, we conclude that the spin-phase interference effects depend on the orientation of the external magnetic field distinctly. This distinct angular dependence, together with the oscillation of the tunnel splittings with the field, may provide an independent experimental test for the spin-phase interference effects in AFM particles. In Fig. 3, we plot the tunnel splittings of the ground-state level and the first excited level as a function of the magnetic field for integer excess spins. It is clearly shown that the splitting is enhanced by quantum tunneling at the excited levels. Detailed calculations of the thermodynamic quantities of the system show that the specific heats oscillate with the magnetic field and are strongly parity dependent of excess spins at sufficiently low temperatures. Due to the topological nature of the Berry phase, these spin-parity effects are independent of details such as the magnitude of excess spins, the shape of the soliton and the tunneling potential.

More recently, Wernsdorfer and Sessoli have measured the tunnel splittings in the molecular Fe$_8$ clusters with the help of an array of micro-SQUIDs, and have found a clear oscillation in the tunnel splittings. Similar spin-phase interference effects observed in ferromagnetic Fe$_8$ cluster are found theoretically in this letter for AFM particles, which may bring a new insight to test the manifestation of quantum effects at a macroscopic level and the influence of quantum phases on the tunneling behaviors of spin systems.

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Figure Captions:

Fig. 1 The relative tunnel splitting of the first excited level \((n = 1) \Delta \varepsilon_1/\Delta \varepsilon_{A,0} (H = 0)\) as a function of \(H/H_0\) for integer \((s = 10)\) and half-integer \((s = 10.5)\) excess spins at \(\theta_H = 0^\circ\). Here \(S = 1000, K_1/J = 0.002\), and \(\lambda = K_2/K_1 = 0.02\).

Fig. 2 The relative tunnel splitting of the first excited level \((n = 1) \Delta \varepsilon_1/\Delta \varepsilon_{A,0} (H = 0)\) as a function of \(H/H_0\) for \(\theta_H = 0^\circ, 1^\circ, 3^\circ, 5^\circ, 10^\circ, 20^\circ\) and \(90^\circ\), respectively. Here \(s = 10, S = 1000, K_1/J = 0.002\), and \(\lambda = K_2/K_1 = 0.02\).
Fig. 3 The relative tunnel splitting $\Delta \varepsilon_n/\Delta \varepsilon_{A,0} (H = 0)$ of the ground-state level ($n = 0$) and the first excited level ($n = 1$) as a function of $H/H_0$ for integer ($s = 10$) excess spins at $\theta_H = 0^\circ$. Here $S = 1000$, $K_1/J = 0.002$, and $\lambda = K_2/K_1 = 0.02$. 
The graph illustrates the function $\frac{\Delta \epsilon_1}{\Delta \epsilon_{A,0}}(H=0)$ as a function of $H/H_0$. Two curves are shown, one for $s=10$ (solid line) and another for $s=10.5$ (dashed line). The y-axis is on a logarithmic scale, ranging from $10^{-1}$ to $10^3$, and the x-axis ranges from 0 to 1 on a logarithmic scale. The graph shows oscillations as $H$ increases.
\[\frac{\Delta \varepsilon_1}{\Delta \varepsilon_{A,0}} (H=0)\]

Graph showing the variation of \(\frac{\Delta \varepsilon_1}{\Delta \varepsilon_{A,0}}\) as a function of \(H/H_0\) for different \(\theta_H\) values.

- \(\theta_H = 0^0\) (solid line)
- \(\theta_H = 1^0\) (dashed line)
- \(\theta_H = 3^0\) (dash-dotted line)
- \(\theta_H = 5^0\) (dotted line)
- \(\theta_H = 10^0\) (dash-dot-dotted line)
- \(\theta_H = 20^0\) (dashed line)
- \(\theta_H = 90^0\) (dotted line)
$\frac{\Delta \varepsilon_n}{\Delta \varepsilon_{A,0}(H=0)}$ vs $\frac{H}{H_0}$

- $n=0$
- $n=1$