Fast and Robust Algorithm for the Minimisation of the Energy of Spin Systems

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An optimization algorithm based on orthogonal matrix transformations is presented for the minimisation of the energy of a magnetic system with respect to spin orientations. When combined with the limited-memory Broyden-Fletcher-Goldfarb-Shanno (LBFGS) algorithm and a line search procedure, it significantly outperforms the frequently used damped spin dynamics approach. Comparison between LBFGS, conjugate-gradient and velocity projection algorithms employing the orthogonal spin optimisation is also presented. The performance and robustness of the new algorithm is demonstrated in calculations where magnetic Skyrmion states are identified by energy minimisation starting from random orientations of the spins.

I. INTRODUCTION

Simulations of magnetic materials typically start with the identification of the stable and/or metastable states of the system. This means finding spin configurations corresponding to minima on the energy surface characterising the magnetic system. In many cases such calculations are based on damped spin dynamics obtained by integrating the deterministic Landau-Lifshitz (LL) equation\(^{(1)}\)

\[
\frac{d\hat{s}_i}{dt} = -\frac{\gamma}{\mu_i} \hat{s}_i \times \mathbf{b}^\text{eff}_i - \frac{\alpha \gamma}{\mu_i} \hat{s}_i \times \hat{\mathbf{b}}^\text{eff}_i,
\]  

(1)

where \(\hat{s}_i\) is a unit vector defining the direction of the magnetic moment of spin \(i\)th, \(\gamma\) is gyromagnetic ratio, \(\mu_i\) is the length of a magnetic moment, \(\mathbf{b}^\text{eff}_i\) is the effective field.

\[
\frac{d\hat{s}_i}{dt} = -\frac{\alpha \gamma}{\mu_i} \hat{s}_i \times \hat{s}_i \times \mathbf{b}^\text{eff}_i.
\]  

(2)

A trajectory corresponding to the integration of this equation is also depicted in Fig. 1 (labeled LL-dis). The path obtained corresponds essentially to steepest descent on the energy surface. Convergence to the energy minimum requires much fewer energy and gradient evaluations and therefore smaller computational effort than the damped dynamics approach. Near the minimum, however, the gradient is small and the method requires many evaluations in the last part of the trajectory, thereby reducing its efficiency.

The minimisation of the energy of a spin system is challenging because it involves moving on a curved manifold. The energy of a system of \(N\) spins is defined as a function of the orientation of the spins

\[
E = F(\hat{s}_1, \hat{s}_2, ..., \hat{s}_N),
\]

where \(\hat{s}_i = (s_{ix}, s_{iy}, s_{iZ})\) is a normalized row-vector \(|\hat{s}_i|^2 = 1\) for \(i = 1, ..., N\). Either the length of the spin vectors is fixed, as in a Heisenberg-type Hamiltonian, or it is obtained from self-consistency calculations, as in DFT or NCAA calculations, by invoking the adiabatic approximation where the length of the spin vector is treated as a...
FIG. 1. Energy surface of a single spin with easy-axis anisotropy in an external magnetic field: $E = -B \sin(\phi)\sin(\theta) - K \cos^2(\theta)$, $B = K = 1$ meV. The angles $\theta$ and $\phi$ give the orientation of the spin. Several algorithms for finding the energy minimum starting from a point near the maximum are compared. The ‘damp-LL’ trajectory is obtained from the damped Landau-Lifshitz equation \(1\). The ‘LL-dis’ trajectory is obtained by including only the dissipative term, Eq. \(2\), and corresponds to a steepest descent algorithm. The damping parameter is 0.19. The OSO-LBFGS and OSO-VPO paths correspond to orthogonal spin optimisation employed with the limited-memory Broyden-Fletcher-Goldfarb-Shanno algorithm with inexact line search and velocity projection optimisation algorithms, respectively. The OSO-LBFGS clearly outperforms the other approaches as it requires significantly fewer iterations. Stars on the paths correspond to iteration steps of the OSO algorithms.

fast variable. Well-established minimisation algorithms that are widely used in atomic simulations to determine the location of atoms at an energy minimum cannot be applied to spin systems without some modifications. It is possible to use Cartesian coordinates for the spin vectors, but then a normalization constraint needs to be added, introducing Lagrange multipliers and thereby increasing the dimensionality of the problem. Alternatively, one can use spherical polar coordinates, but then a special treatment is required near the poles, $\theta = 0$ and $\pi/2$.

Another approach is to use an orthogonal optimisation algorithm. This approach has been successfully applied to a variety of problems, including signal processing and electronic structure calculations. Here, we extend this approach to spin systems and show that the orthogonal optimisation employed with the limited-memory Broyden-Fletcher-Goldfarb-Shanno (LBFGS) minimisation algorithm with inexact line search provides fast convergence for large systems involving magnetic Skyrmions. The algorithm significantly outperforms the damped dynamics and dissipative dynamics approaches.

The article is organised as follows. In section II, the orthogonal minimisation algorithm is presented. In section III, results of numerical tests are presented where the orthogonal minimisation is carried out in combination with the LBFGS, conjugate gradient (CG) or velocity projection optimisation (VPO) algorithms, and performance compared also with dissipative dynamics approaches. Finally, conclusions are presented in section IV. Details of the implementation of the orthogonal optimisation algorithm are presented in the Appendices. The code and initial and final spin-configurations are available in a GitLab repository.

II. METHODOLOGY

Consider a system consisting of $N$ interacting spins and let $\{\vec{s}_i\}_{i=1}^N$ be a reference orientation of the spins. Then, any spin configuration, $\{\hat{s}_i\}_{i=1}^N$, can be obtained from this reference by applying some orthogonal transformation

$$\hat{s}_i = s_i U^i,$$ (3)

The objective is to find the set of orthogonal matrices that transform the reference spin configuration to the minimum energy configuration. Since orthogonal matrices must satisfy orthonormality constraints, $U^i U^i = I$, it is convenient to parametrise them by exponentials of skew-symmetric matrices, $A_i$:

$$U^i = e^{A_i},$$ (4)

where

$$A_i = \begin{pmatrix} 0 & a_{12}^i & a_{13}^i \\ -a_{12}^i & 0 & a_{23}^i \\ -a_{13}^i & -a_{23}^i & 0 \end{pmatrix}.$$ (5)

Since the reference spin vectors satisfy the normality constraints $|\hat{s}_i|^2 = 1$, the transformed spin vectors $\{\hat{s}_i\}_{i=1}^N$ also satisfy the normality constraints for any set of $\{A_i\}_{i=1}^N$.

Skew-symmetric matrices form a linear space and the minimum of the energy can be found as

$$E_{\text{min}} = \min_{\vec{a} \in \mathbb{R}^{3N}} E(\vec{a}),$$ (6)

where $\vec{a}$ is 3$N$-dimensional vector

$$\vec{a} = (a_{12}^1, a_{13}^1, a_{23}^1, ..., a_{12}^N, a_{13}^N, a_{23}^N),$$ (7)

and $i$ refers to the spin site.

With this formulation, the energy can be minimised as a function of a 3$N$-dimensional vector instead of a minimisation with respect to the spin orientations subject to the normality constraints. In practice, the reference spin orientation can represent a guess for the minimum energy spin configuration, chosen in such a way as to obtain convergence with as few iterations as possible, or it can be chosen at random in a search for an unknown local minimum on the energy surface.
The energy gradient with respect to \( \vec{a} \) needs to be evaluated in order to minimise the energy in an efficient way. This can be done as follows:

\[
\begin{align*}
\mathbf{g}_{\alpha\beta}^i := \frac{\partial E}{\partial \alpha_{\alpha\beta}^i} &= \left( \int_0^1 e^{tA^i}T_s e^{-tA^i} dt \right)_{\alpha\beta}, \tag{8}
\end{align*}
\]

where the matrix \( T^i \) is

\[
T^i = \begin{pmatrix}
0 & t_{iz} & -t_{iy} \\
-t_{iz} & 0 & t_{ix} \\
t_{iy} & -t_{ix} & 0
\end{pmatrix}, \quad \dot{s}_i = \dot{s}_i + \lambda \vec{s}_i \times \frac{\partial F}{\partial \vec{s}_i}. \tag{9}
\]

This equation can be obtained from the chain rule of differentiation and the definition of the first directional derivative of the matrix exponential.\(^{13}\)

This orthogonal optimization method can be shown to reduce to the steepest descent and dissipative LL dynamics in the limit of small rotations. For a small rotation, \( \| A^i \| \ll 1 \) for all \( i = 1 \ldots N \), the gradient becomes \( \mathbf{g}_{\alpha\beta}^i \approx T_{\alpha\beta}^i \) and rotation in the steepest descent direction at \( A^i = 0 \) is

\[
\dot{s}_i = \dot{s}_i^0 e^{-\lambda T^i} \approx \dot{s}_i^0 (I - \lambda T^i) = \dot{s}_i^0 + \lambda \dot{s}_i^0 \times \vec{f}_i \times \frac{\partial F}{\partial \vec{s}_i}. \tag{10}
\]

If one chooses \( \lambda = \gamma \alpha \Delta t / \mu_s \) and \( \Delta t \to 0 \), then the equation above transforms into

\[
\frac{d\dot{s}_i}{dt} = \alpha \gamma \mu_s \dot{s}_i \times \dot{s}_i \times \frac{\partial F}{\partial \vec{s}_i},
\]

which is the same as Eq. (2). Thus, in the limit of small rotations, where the orthogonal optimization becomes equivalent to a steepest descent algorithm, it corresponds to the dissipative LL trajectory. If one defines the \( t_i \) vector as \( \alpha \dot{s}_i \times \frac{\partial F}{\partial \vec{s}_i} - \vec{f}_i \), where \( \vec{f}_i \) is a random field, then the orthogonal rotations correspond to the trajectory obtained from the stochastic LL equations.

By defining the energy of a spin system as a function of variables in linear space and given the corresponding expression for the energy gradient, well-established algorithms can be used to carry out the minimization. We will refer to this approach as orthogonal spin optimisation (OSO) and will test three different minimisation algorithms for finding the coefficients in the vector \( \vec{a} \), namely the LBFGS, CG and VPO. The details of our implementation of the minimisation algorithms and the calculation of the orthogonal matrix and gradients of the energy are given in the Appendices A and D.

### III. NUMERICAL TESTS

A system described by an extended Heisenberg-type Hamiltonian is chosen as a test problem. The energy is given by

\[
F = - \sum_{<i,j>} \left[ J \dot{s}_i \cdot \dot{s}_j + \vec{D}_{ij} \cdot \dot{s}_i \times \dot{s}_j \right] - \sum_i \mu_i \dot{s}_i \cdot \vec{H}. \tag{11}
\]

where the first and second contributions are exchange and Dzyaloshinskii-Moriya interactions, respectively, including only the first neighbour interaction and the third term is the Zeeman energy describing the interaction of the spins with an external magnetic field \( \vec{H} \). The Dzyaloshinskii-Moriya (DM) vector, \( \vec{D}_{ij} \), is chosen to lie along the vector pointing from site \( i \) towards site \( j \).

In order to benchmark the performance of the various minimisation algorithms, a square lattice of spins is simulated. This can represent a monolayer of magnetic atoms commensurate with a (100) surface of a FCC or BCC crystal. The supercell consists of either 20x20 or 40x40 spins subject to periodic boundary conditions. The parameters in the Hamiltonian are \( J = 10 \) meV, \( D = J/2 \), where \( D \) is the length of the DM vector. All magnetic moments have the same length \( \mu_s \), magnetic field is perpendicular to the monolayer and \( \mu_s \vec{H} = J/5 \). While the energy surface of the system has multiple local minima, corresponding to Skyrmions of varying density, the ground state is ferromagnetic. The energy of the ferromagnetic state is -22 meV per spin. The energy of 20x20 and 40x40 skyrmionic states is -21.94 and -21.98 meV per spin, respectively.

The performance of four algorithms is compared. The OSO is combined with either VPO, CG or LBFGS algorithms using the gradient expression given in section II. The latter pair of algorithms are used with inexact line search. The fourth algorithm is dissipative LL dynamics (LL-dis), Eq. (2), where the SIB algorithm is used as integrator since it preserves the length of the spins even for a large time step. There, the damping parameter is taken to be \( \alpha = 0.1 \) and the time step set to 0.7 ps. For a larger time step of 0.8 ps the SIB algorithm does not converge. The initial spin orientations are chosen from a random distribution on a unit sphere. Convergence is considered to be achieved when the average torque is less than \( 10^{-5} \) meV, that is \( \sum i |\vec{t}_i| / N < 10^{-5} \) meV.

The rate of convergence of the energy is shown in Fig. 3. The OSO-LBFGS algorithm shows the best performance, requiring ca. 300 energy and gradient evaluations to converge on the Skyrmion state. The OSO-CG requires about 50% more evaluations to reach the same level of convergence. The OSO-VPO is much slower requiring more than 3000 evaluations. The LL-dis algorithm has the worst performance requiring more than 4000 evaluations. Large savings in computer time can be achieved by using the OSO-LBFGS in energy minimisations for magnetic systems.

The number of energy and gradient evaluations is not the same as the number of iterations. In the SIB dynamics algorithm, two evaluations of the effective fields (first in order to calculate the predictor and then in order to calculate the corrector) are required for each iteration. While LBFGS and CG at principle requires calculation of the effective fields once at each iterations as well, the energy must be estimated in order to test the strong Wolfe conditions and if these conditions are violated the line search procedure needs to be applied (see Appendix A).
FIG. 2. A 20×20 lattice of spins subject to periodic boundary conditions (the simulation cell is marked by solid black lines, the figure additionally shows three periodic images of the system). Coordinates are given as a multiple of the lattice constant. (a): Initial state where the spins are randomly oriented. (b): Local minimum obtained with OSO-LBFGS. The three other algorithms tested converge to an equivalent state with the same energy corresponding to two magnetic Skyrmions in the simulation cell.

In general, it is necessary to perform the line search only in the beginning phase of the LBFGS minimisation procedure. After some minimisation steps have been carried out, the unity step length along the LBFGS search direction is guaranteed to satisfy the strong Wolfe conditions if earlier steps have satisfied the conditions. Therefore, most iterations in the LBFGS algorithm require only one estimation of energy and the gradient.

When the number of degrees of freedom is increased by adding more spins to the simulated system, the number of energy and gradient evaluations needed to reach convergence also increases. This is illustrated by simulating a 40×40 lattice where a 20×20 subsection of the lattice is started with random spin orientations, the same as in the calculation of the system shown in Fig. 2(a), while the rest of the spins are oriented as in the ferromagnetic phase, see Fig. 4(a). The OSO-LBFGS calculation now requires 1330 evaluations to converge to a tolerance of 10^{-5} meV, while OSO-CG and OSO-VPO require 4600 and 40000, respectively. The dissipative LL minimization requires ca. 50000 evaluations. Interestingly, all four methods converge on the same local minimum energy spin configuration where the density of Skyrmions is ca. half as large as in the 20×20 calculation. This calculation illustrates how the addition of degrees of freedom, without addition of complexity (in that the number of randomly oriented spins is the same as in the calculation illustrated in Fig. 2), increases the number of evaluations required to reach convergence.

FIG. 3. Energy as a function of the number of energy and gradient evaluations using the dissipative LL dynamics equation integrated using the SIB algorithm, and using three different implementations of the orthogonal spin optimisation algorithm, either LBFGS, CG or VOP minimisation. Convergence is considered to be achieved when ∑_{i=1}^{N} |s_i × ∂E/∂s_i|/N < 10^{-5} meV. (a): Calculations for the 20×20 lattice depicted in Fig. 2. (b): Calculations for the 40×40 lattice depicted in Fig. 4.
FIG. 4. A 40×40 lattice of spins subject to periodic boundary conditions. Coordinates are given as a multiple the units of the lattice constant. (a): Initial state. (b): Local minimum obtained with OSO-LBFGS. The three other algorithms tested converge to an equivalent state with the same energy corresponding to five magnetic Skyrmions in the simulation cell.

The computational time spent on minimisation of the energy consists of time spent on evaluation of the energy value and effective fields as well as algebra operations of the minimisation algorithm. The CPU time spent on the algebra operations of the OSO algorithm can be reduced by approximating the expression for the gradient in Eq. (8). Namely, if the rotation matrix is small then the first term from Taylor expansion of Eq. (8) can be used to estimate the gradient:

$$g_{\alpha\beta}^i \approx T_{\alpha\beta}^i,$$  \hspace{1cm} (12)

When the norm of the matrix $A^i$ becomes large the reference orbitals must be updated $\hat{s}_i \leftarrow \hat{s}_i e^{A_i}$, and the minimisation can be continued starting from $A_i = 0$ (see details in Appendix (A)). With this approach, Eq. (12) gives a good estimate of the gradient. The approximate gradients increases number of energy and gradient evaluations for OSO-LBFGS from 1300 to 1800 but reduces the elapsed time of the calculations from 2.1 s to 1.4 s. The OSO-LBFGS with the exact gradient can be relatively more efficient in the problems when the energy and effective fields evaluations are the most time consuming operations.

**IV. CONCLUSION.**

An orthogonal spin optimisation algorithm is presented for the minimisation of the energy of a magnetic system with respect to the orientation of the spins. The relation between this algorithm and a dynamics simulation including only the dissipative term in the LL equation is demonstrated. The orthogonal optimisation in combination with the LBFGS algorithm and inexact line search procedure for the minimum energy, OSO-LBFGS, greatly outperforms dissipative dynamics as well as other algorithms tested.

The algorithm should also be useful in other spin configuration optimisations, such as minimum energy path calculations using the geodesic nudged elastic band method, which has previously been implemented with a VPO algorithm mimicking particle dynamics. Also, the algorithm could be used in searches for first order saddle points on energy surfaces for spin systems starting only from the initial state and without knowledge of final states. From the saddle points, the rate of magnetic transitions can be estimated using harmonic transition state theory as has, for example, been done to estimate the lifetime of Skyrmions as a function of temperature.

The performance of the OSO-LBFGS algorithm could likely be improved further by developing a preconditioner for the orthogonal optimisation.

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**Appendix A: Orthogonal Spin Optimisation**

The task is to minimise the energy as a function of spin orientations, $F(\hat{s}_1, \hat{s}_2, \ldots \hat{s}_N)$. The $\hat{s}_i$ are parametrised using skew-symmetric matrices $\hat{s}_i = \hat{s}_i' \exp(A_i)$

$$A_i = \begin{pmatrix} 0 & a_{12}^i & a_{13}^i \\ -a_{12}^i & 0 & a_{23}^i \\ -a_{13}^i & -a_{23}^i & 0 \end{pmatrix}.$$  \hspace{1cm} (A1)

It is enough to consider only the upper diagonal part of $A_i$, and the energy is therefore a function of a 3N-dimensional vector...
\[ \vec{a} = (a_{12}, a_{13}, a_{23}, \ldots, a_{12}, a_{13}, a_{23}, \ldots, a_{12}, a_{13}, a_{23}) \]

where the upper index, \( i \), refers to the spin site.

**Algorithm:**

1. Choose reference spin orientations \( \{ \hat{s}_i \} \) for \( i = 1, \ldots, N \), set initial skew-symmetric matrices to zero, \( \vec{a}^{(0)} = 0 \), \( \hat{s}_i^{(0)} = \hat{s}_i \) and calculate initial gradient \( \vec{g}^{(0)} = \partial E / \partial \vec{a}^{(0)} \).

2. Set \( k = 0 \) and calculate initial search direction \( \vec{p}^{(0)} \) according to the particular minimisation algorithm chosen, see Appendix [B] for VPO example.

3. Set integer \( u \) that counts the number of steps before updating the reference spins. For example, \( u = 20 \).

4. Calculate the magnitude of the average torque:

\[
\Delta^{(0)} = \frac{1}{N} \sum_{i=1}^{N} \| \hat{s}_i^{(0)} \times \frac{\partial E}{\partial \hat{s}_i^{(0)}} \| \quad (A2)
\]

and set tolerance \( \epsilon \) (for example, \( \epsilon = 10^{-5} \text{ meV} \)).

5. While \( \Delta^{(k)} > \epsilon \):

   (a) If \( k \) mod \( u = 0 \) then update reference spins:

   \[
   \hat{s}_i' = \hat{s}_i e^{A_i^{(k)}}, \forall i = 1, 2, \ldots, N, \quad (A3)
   \]

   \[
   \vec{a}^{(k)} = 0, \quad \vec{g}^{(k)} = \partial E / \partial \vec{a}^{(k)}, \quad (A4)
   \]

   calculate new \( \vec{p}^{(k)} \). \quad (A5)

(b) Compute \( \lambda^{(k)} \) using a line search (for VPO algorithm \( \lambda^{(k)} = 1 \)) and calculate:

\[
\vec{a}^{(k+1)} = \vec{a}^{(k)} + \lambda^{(k)} \vec{p}^{(k)}, \quad (A6)
\]

\[
\hat{s}_i^{(k+1)} = \hat{s}_i e^{A_i^{(k+1)}}, \forall i = 1, 2, \ldots, N \quad (A7)
\]

(c) Calculate new gradients \( \vec{g}^{(k+1)} \) and \( \Delta^{(k+1)} \). Calculate new search direction, \( \vec{p}^{(k+1)} \), according to the particular minimisation algorithm chosen, see Appendix [B] for VPO example. Set \( k \leftarrow k + 1 \)

6. End

In this article, the Limited-Memory Broyden-Fletcher-Goldfarb-Shanno (LBFGS) and Fletcher-Reeves Conjugate-Gradient algorithms have been used in order to calculate the search direction \( \vec{p}^{(k)} \). The step length parameter \( \lambda^{(k)} \) is chosen in such a way that the strong Wolfe conditions\(^{[23]}\) and/or approximate Wolfe conditions\(^{[30]}\) are satisfied:

\[
E(\vec{a}^{(k)} + \lambda^{(k)} \vec{p}^{(k)}) \leq E(\vec{a}^{(k)}) + c_1 \lambda^{(k)} \nabla \vec{a} E(\vec{a}^{(k)}) \cdot \vec{p}^{(k)} \quad (A8)
\]

\[
|\nabla E(\vec{a}^{(k)} + \lambda^{(k)} \vec{p}^{(k)}) \cdot \vec{p}^{(k)}| \leq c_2 |\nabla \vec{a} E(\vec{a}^{(k)}) \cdot \vec{p}^{(k)}| \quad (A9)
\]

and/or

\[
(2\delta - 1) \nabla \vec{a} E(\vec{a}^{(k)}) \cdot \vec{p}^{(k)} \geq \nabla \vec{a} E(\vec{a}^{(k)} + \lambda^{(k)} \vec{p}^{(k)}) \cdot \vec{p}^{(k)} \geq \sigma \nabla \vec{a} E(\vec{a}^{(k)}) \cdot \vec{p}^{(k)},
\]

\[
(2\delta - 1) \nabla \vec{a} E(\vec{a}^{(k)}) \cdot \vec{p}^{(k)} \geq \nabla \vec{a} E(\vec{a}^{(k)} + \lambda^{(k)} \vec{p}^{(k)}) \cdot \vec{p}^{(k)} \geq \sigma \nabla \vec{a} E(\vec{a}^{(k)}) \cdot \vec{p}^{(k)},
\]

with \( 0 < c_1 < c_2 < 1, \epsilon > 0, \delta < \min(5, \sigma, \sigma < 1) \) The parameters were chosen according to Ref.\(^{[14]}\) and\(^{[20]}\)

\[
c_1 = 10^{-4}, c_2 = 0.9, \delta = 0.1, \sigma = 0.9, \epsilon = 10^{-6} \quad (A12)
\]

A trial step of \( \lambda^{(k)} = 1 \) is always used first to test these conditions. If they are not satisfied, an inexact line search procedure based on the cubic interpolation is used\(^{[13]}\). Approximate Wolfe conditions are always examined at the minimum of the cubic interpolation [see Appendix [C]].

**Appendix B: VPO algorithm**

Values of two parameters need to be chosen, \( \Delta t \) and \( m \), for example \( \Delta t = 0.01 \) and \( m = 0.01 \). Let \( \vec{a}_0 \) be the initial vector in \( \mathbb{R}^{3N} \) and \( \vec{g}_0 = \partial E / \partial \vec{a}_0 \). At the \( k \)-th iteration the search direction is chosen according to the following:

\[
\text{if } k = 0:
\]

\[
\vec{p}^{(k)} = -\vec{g}^{(k)} \Delta t / m \quad (B1)
\]

\[
\vec{p}^{(k)} = \vec{p}^{(k)} \Delta t. \quad (B2)
\]

\[
\text{return } \vec{p}^{(k)}. \quad (B3)
\]
else:
\[ \beta^{(k)} = \tilde{g}^{(k)} \cdot \tilde{g}^{(k-1)} / \tilde{g}^{(k)} \cdot \tilde{g}^{(k)} \]  
if \( \beta^{(k)} > 0 \) then set \( \beta^{(k)} = 0 \).
\[ \gamma^{(k)} = \Delta t / m - \beta^{(k)} \]  
\[ \tilde{v}^{(k)} = -\gamma^{(k)} \tilde{g}^{(k)} \]  
\[ \tilde{p}^{(k)} = \tilde{v}^{(k)} \Delta t \]  
return \( \tilde{p}^{(k)} \).  
\[ (B4) \]
\[ (B5) \]
\[ (B6) \]
\[ (B7) \]
\[ (B8) \]
\[ (B9) \]

**Appendix C: Cubic interpolation**

Let \( f \) be defined on \([0,r]\) and \( f(0), f(r), f'(0), f'(r) \) be known. Then the cubic interpolation of \( f \) is:
\[ f(\alpha)^{\text{approx}} = c_1 \alpha^3 + c_2 \alpha^2 + c_3 \alpha + c_4 \]
\[ c_1 = -\frac{2f(r) - 2f(0)}{r^2} + \frac{f'(r) + f'(0)}{r} \]
\[ c_2 = \frac{3f(r) - 3f(0)}{r^2} - \frac{f'(r) + f'(0)}{r} \]
\[ c_3 = f'(0), \quad c_4 = f(0) \]
\[ (C1) \]
\[ (C2) \]
\[ (C3) \]
\[ (C4) \]
and the minimum of this cubic function is at
\[ \alpha_0 = (-c_2 + \sqrt{c_2^2 - 3c_1c_3}) / 3c_1 \]
\[ (C5) \]
or at the boundaries of the interval \([0,r]\).

**Appendix D: Orthogonal matrix and gradient of the energy**

Let \( A \) be
\[ A = \begin{pmatrix} 0 & a & b \\ -a & 0 & c \\ -b & -c & 0 \end{pmatrix} \]
\[ (D1) \]

Let \( x = \sqrt{a^2 + b^2 + c^2} \) Then eigenvalues are:
\[ \lambda_1 = 0, \]  
\[ (D2) \]
\[ \lambda_2 = -ix, \]  
\[ (D3) \]
\[ \lambda_3 = ix. \]  
\[ (D4) \]

The eigenvectors are:
\[ \tilde{v}_1 = \frac{1}{x} \begin{pmatrix} c \\ -b \\ a \end{pmatrix}, \quad \tilde{v}_2 = \frac{1}{x \sqrt{2(a^2 + c^2)}} \begin{pmatrix} bc + iax \\ a^2 + c^2 \\ ab - icx \end{pmatrix}, \quad \tilde{v}_3 = \tilde{v}_2^* \]
\[ (D5) \]

if \( a = 0 \) and \( c = 0 \) then
\[ \tilde{v}_2 = \begin{pmatrix} \text{sign}(b)i \\ 0.0 \\ 1.0 \end{pmatrix} \]
\[ (D6) \]

The matrix exponential can then be calculated as:
\[ e^A = VLV^\dagger, \]
\[ (D7) \]
1 L. D. Landau and E. M. Lifshitz, Phys. Z. Sowjetunion 8, 153 (1935).
2 P. Hohenberg and W. Kohn, Phys. Rev. 136, B864 (1964).
3 W. Kohn and L. J. Sham, Phys. Rev. 140, A1133 (1965).
4 P. F. Bessarab, V. M. Uzdin, and H. Jónsson, Phys. Rev. B 89, 1 (2014).
5 V. Antropov, M. Katsnelson, B. Harmon, and M. van Schilfgaarde, Phys. Rev. B 54, 1019 (1996).
6 A. Edelman, T. A. Arias, and S. T. Smith, SIAM Journal on Matrix Analysis and Applications 20, 303 (1998).
7 T. Abrudan, J. Eriksson, and V. Kövári, Signal Processing 89, 1704 (2009).
8 J. Hutter, M. Parrinello, and S. Vogel, J. Chem. Phys. 101, 3862 (1994).
9 T. Van Voorhis and M. Head-Gordon, Molecular Physics 100, 1713 (2002).
10 S. Lehtola and H. Jónsson, J. Chem. Theory Comput. 10, 3324 (2014).
11 H. Jónsson, G. Mills, and K. Jacobsen, B.J. Berne, G. Ciccotti, D.F. Coker (Eds.), Classical and Quantum Dynamics in Condensed Phase Simulations, World Scientific (1998), 385 (1998).
12 https://gitlab.com/alxvov/spinmin.
13 I. Najfeld and T. Havel, Advances in Applied Mathematics 16, 321 (1995).
14 J. Nocedal and S. J. Wright, Numerical Optimization 2nd ed. (Springer, New York, NY, USA, 2006).
15 J. H. Mentink, M. V. Tretyakov, A. Fasolino, M. I. Katsnelson, and T. Rasing, J. Phys. Condens. Matter 22, 176001 (2010).
16 P. F. Bessarab, V. M. Uzdin, and H. Jónsson, Comput. Phys. Commun. 196, 335 (2015).
17 G. P. Müller, P. F. Bessarab, S. M. Vlasov, F. Lux, N. S. Kiselev, S. Blügel, V. M. Uzdin, and H. Jónsson, Phys. Rev. Lett. 121, 197202 (2018).
18 P. F. Bessarab, V. M. Uzdin, and H. Jónsson, Phys. Rev. B 85, 184409 (2012).
19 V. M. Uzdin, M. N. Potkina, I. S. Lobanov, P. F. Bessarab, and H. Jónsson, J. Magn. Magn. Mater. 459, 236 (2018).
20 W. W. Hager and H. Zhang, SIAM Journal on Optimization 16, 170 (2006).