Molecular dynamics simulations of spin and pure liquids with preserving all the conservation laws

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A new methodology is developed to integrate numerically the equations of motion for classical many-body systems in molecular dynamics simulations. Its distinguishable feature is the possibility to preserve, independently on the size of the time step, all the conservation laws inherent in the description without breaking the time reversibility. As a result, an implicit second-order algorithm is derived and applied to pure as well as spin liquids for which the dynamics is characterized by the conservation of total energy, linear and angular momenta as well as magnetization and individual spin lengths. It is demonstrated on the basis of Lennard-Jones and Heisenberg fluid models that when such quantities as energy and magnetization must be conserved perfectly, the new algorithm turns out to be more efficient than popular decomposition integrators and standard predictor-corrector schemes.

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I. INTRODUCTION

During the last years, considerable attention has been focussed on computer experiment studies of relaxation properties and critical phenomena in classical spin systems [1–9]. These studies dealt mainly with lattice models such as the Ising, the XY, and the Heisenberg. Of notable current interest is the investigation of continuum spin liquid models [10–15] in which additional dynamical effects are possible because of the coupling between spin and liquid subsystems.

Quite recently [16,17], a set of symplectic algorithms of different orders in the time step has been constructed for numerical integration of motion at the presence of both translational and spin degrees of freedom. As a consequence, the molecular dynamics simulations (MD) of a Heisenberg spin fluid have been carried out for the first time. The symplectic integrators were derived by developing the Suzuki-Trotter technique [18] for decompositions of exponential operators. Their main advantages over standard predictor-corrector schemes are explicitness, time reversibility and exact conservation of spin lengths. It was also shown that the decomposition algorithms permit significantly larger time steps and lead to a substantial speedup of the calculations. In a particular case when the spin degrees of freedom are frozen, these algorithms can be reduced to the well-known velocity Verlet integrator [19], widely used for simulating of pure liquid dynamics.

However, the decomposition approach (like predictor-corrector and other existing traditional numerical schemes [20], such as Runge-Kutta, etc.) do not preserve the total energy and magnetization of the system. In most MD applications the accuracy achieved for the energy-magnetization conservation by the decomposition algorithms is high enough in order to obtain reliable results. Moreover, this accuracy can be improved using higher-order versions [16] of the decomposition approach or decreasing the step size. But if the integrals of motion have to be conserved perfectly, the non-conservative algorithms may not be an optimal choice for the solution of the problem. The reason is that then the time step should be divided into a lot of subintervals, reducing considerably the efficiency of the computations.

The exact conservation of integrals of motion is especially important for simulations of spin liquids near phase transitions, when the phase diagrams, dynamical scaling, long-time correlated behavior or derivatives of the thermodynamic functions are investigated. This is so because in these cases, the presence of artificial fluctuations in energy and magnetization may have a significant influence on the results. Therefore, it is desirable to look for an algorithm which conserves the fundamental physical invariants exactly or at least within machine accuracy.

In the present paper, a novel approach to numerical integration of the equations of motion for spin and pure liquids is introduced. The main feature of this approach is its intrinsic preservation of all the conservation laws inherent in the system without violating the time reversibility property. The paper is organized as follows. The basic equations and their integrals of motion are described in Section 2. Section 3 is devoted to a consequent derivation of the desired second-order algorithm. Its possibility to conserve exactly the integrals of motion is demonstrated there also. In Sections 4 and 5, the algorithm is tested in actual MD simulations on Heisenberg and Lennard-Jones fluid models, respectively, and compared with previous numerical schemes. The discussion and concluding remarks are given in Section 6.
II. BASIC EQUATIONS OF MOTION AND CONSERVATION LAWS

Let us consider a classical $N$-body system described by the Hamiltonian

$$H = \sum_{i=1}^{N} \frac{m_i v_i^2}{2} + \frac{1}{2} \sum_{i \neq j} \left( \varphi(r_{ij}) - J(r_{ij}) s_i \cdot s_j \right), \quad (1)$$

where $r_i$ and $v_i$ are the translational position and velocity, respectively, of particle $i$ with mass $m_i$ carrying spin $s_i$. The fluid part of the potential is denoted by $\varphi(r_{ij})$, whereas $J(r_{ij})$ is the exchange integral corresponding to a pair of spins with the interparticle separation $r_{ij} = |r_i - r_j|$. Note that within the classical approach, each spin $s_i$ is treated as a continuous three-component vector with fixed length (putting for convenience $|s_i| = 1$, so that $J$ will be measured in energy units). Although the results which will be obtained below can easily be adapted to a larger class of Hamiltonians (to multi-component systems, for instance), we restrict ourselves for the sake of simplicity to the basic model (1) which represents a typical isotropic Heisenberg spin fluid [12,14]. For $J \equiv 0$, Eq. (1) reduces to a pure liquid model.

In MD simulations it is necessary to solve numerically the equations of motion, $d\rho/dt = \{\rho, H\}$, where $\{,\}$ denotes the Poisson bracket and $\rho \equiv \{r_i, v_i, s_i\}$ is the full set of microscopic phase variables. For the system under consideration, the dynamical equations can be written more explicitly [12,17],

$$\frac{dr_i}{dt} = v_i,$$

$$\frac{dv_i}{dt} = m_i \frac{s_i}{h} = -\frac{1}{m_i} \sum_{j(i \neq j)} \left( \frac{\partial \varphi_{ij}}{\partial r_{ij}} - \frac{\partial J_{ij}}{\partial r_{ij}} s_i \cdot s_j \right) \frac{r_{ij}}{r_{ij}}, \quad (2)$$

$$\frac{ds_i}{dt} = \frac{s_i}{h} \times \vec{g}_i \equiv \frac{s_i}{h} \times \sum_{j(j \neq i)} J_{ij} s_j.$$

Here, $f_i = \sum_{j(j \neq i)} f_{ij}$ and $g_i = \sum_{j(j \neq i)} g_{ij}$ are the force and internal magnetic field, respectively, acting on particle $i$ due to the interactions $f_{ij} = -\left( \varphi'_{ij} - J'_{ij} s_i \cdot s_j \right) r_{ij}/r_{ij}$ and $g_{ij} = J_{ij} s_j$ with all the rest of bodies, where $\varphi'_{ij} = \partial \varphi(r_{ij})$ and $J'_{ij} = J(r_{ij})$. Note that the quantum Poisson bracket was applied [12] to derive the equations for spin subdynamics. If an initial state $\rho(0)$ is specified, the time evolution $\rho(t)$ can be uniquely obtained by integrating Eq. (2).

Taking into account the symmetry $\varphi_{ij} = \varphi_{ji}$ and $J_{ij} = J_{ji}$ of interaction potentials, it follows from Eq. (2) that the total energy $E \equiv H$, the total magnetization $M = \sum_i s_i$, the total linear momentum $P = \sum_i m_i v_i$ as well as angular momentum $L = \sum_i m_i v_i \times s_i$, are integrals of motion, i.e., $dE/dt = dM/dt = dP/dt = dL/dt = 0$. The structure of spin equations of motion (the last line of Eq. (2)) imposes in addition the conservation of individual spin lengths, $|s_i| = s_i = const$. Indeed, $ds_i/dt = d(s_i \cdot s_i)^{1/2}/dt = (s_i \cdot ds_i/dt)/s_i \equiv s_i [s_i \times \vec{g}_i]/h s_i = 0$, because the equality $a_i [a \times b] = 0$ is valid for arbitrary vectors $a$ and $b$. Besides, the exact solutions are time reversible, since the equations of motion are invariant with respect to the time inversion transformation $t \to -t$, $\{v_i, s_i\} \to \{-v_i, -s_i\}$.

No existing numerical scheme can obey perfectly all the just mentioned properties. The exact conservation during the integration can be achieved only for some of the integrals of motion, such as linear momentum, for example. Usually, it is required that the deviations of conservative quantities from their original values to be within an acceptable level of precision. This results, however, in limitations on the size of the time steps which actually can be used for MD simulating.

III. THE METHOD OF INTEGRATION

We will show in this section that it is possible to generate time-reversible microscopic trajectories along of which all the integrals of motion are preserved at arbitrary finite time steps. Our derivation of the desired algorithm is started by considering a mid-point scheme of the second order. According to this scheme, the dynamical variables can be propagated as

$$\rho(t + \tau) = \rho(t) + \tau [d\rho/dt]_{t+\tau/2} + O(\tau^3)$$

with $\tau$ being the step size and $O(\tau^3)$ denoting the truncation terms. In view of Eq. (2), the explicit expressions for such a propagation read

$$r_i(t + \tau) = r_i(t) + \tau v_i(t + \tau/2),$$

$$v_i(t + \tau) = v_i(t) + \tau f_i(t + \tau/2)/m_i,$$

$$s_i(t + \tau) = s_i(t) + \tau [s_i \times g_i]/m_i + \tau/2 h,$$

where the mid-step values of $v_i$, $f_i$ and $g_i$ should be specified additionally.

The only way to construct mid-point translational velocities maintaining the time reversibility property is

$$\tau v_i(t + \tau/2) = \frac{\tau}{2} [v_i(t) + v_i(t + \tau)] + O(\tau^3). \quad (4)$$

The terms $O(\tau^3)$ of third- and higher-orders can be ignored because the corresponding terms of the same orders have been truncated already within the mid-point approach. Eq. (4) represents, in fact, an implicit interpolation formula in which past (at time $t$), and future (at $t + \tau$) values of dynamical quantities enter symmetrically, assuring automatically the reversibility of the solutions.

In the case of translational forces, there are several possibilities to build the mid-point values. The reason is that the interparticle function $f_{ij} \equiv f(r_{ij}, s_i, s_j)$ depends explicitly on relative position $r_{ij}$ and orientation $s_i, s_j$ which in turn vary with time. Thus, we can apply
the mid-point interpolation either to the function \( f_{ij} \) as a whole, or directly to the dynamical variables \( r_{ij} \) and \( s_i s_j \). As a result, two different approaches to the force evaluation may be introduced, namely,

\[
f_{ij}(t + \frac{\tau}{2}) = \frac{1}{2} \left[ f(r_{ij}(t), [s_i s_j]_t) + f(r_{ij}(t + \tau), [s_i s_j]_{t + \tau}) \right]
\]

and

\[
f_{ij}(t + \frac{\tau}{2}) = f(r_{ij}(t + \frac{\tau}{2}), [s_i s_j]_{t + \frac{\tau}{2}}). \quad (5)
\]

The last approach requires the knowledge of mid-point values for \( r_{ij} \) and \( s_i s_j \). The obvious choice for the relative positions is

\[
r_{ij}(t + \frac{\tau}{2}) = \frac{1}{2} \left[ r_{ij}(t) + r_{ij}(t + \tau) \right]. \quad (6)
\]

The interpolation of the scalar product \([s_i s_j]_{t + \tau/2}\) will be described latter. None of the above approaches for evaluating \( f_{ij}(t + \tau/2) \) can lead to a scheme with exact preservation of the total energy. The energy will only be conserved approximately with the precision within which the microscopic solutions are calculated, i.e., \( E(t + \tau) = E(t) + \mathcal{O}(\tau^2) \).

We will show now that the second approach (Eq. (5)) can be modified in such a way as to compensate the loss of precision in the total energy. The idea lies in the following. Since, according to Eq. (1), the energy difference \( E(t + \tau) - E(t) \) is a function of the quantities \( \phi(r_{ij}(t + \tau)) \) and \( \phi(r_{ij}(t)) \) as well as \( J(r_{ij}(t + \tau)) \) and \( J(r_{ij}(t)) \), it is natural to try to evaluate numerically the partial derivatives \( \phi'(r_{ij}) = \partial \phi / \partial r_{ij} \) and \( J'(r_{ij}) = \partial J / \partial r_{ij} \) (which appear in Eq. (5) at \( t + \tau/2 \)) in terms of the same quantities, rather than to calculate the derivatives analytically. This is possible because for any function \( \xi(r_{ij}) \) depending only on the interparticle distance \( r_{ij} \) we can write the following two expressions

\[
\frac{d\xi(r_{ij})}{dt} = \frac{\partial \xi}{\partial r_{ij}} \frac{dr_{ij}}{dt} = \xi'(r_{ij}) r_{ij} \cdot v_{ij}
\]

and

\[
\frac{d\xi(r_{ij})}{dt} \bigg|_{t + \frac{\tau}{2}} = \xi'(r_{ij}(t + \tau)) - \xi'(r_{ij}(t)) + \mathcal{O}(\tau^2),
\]

combining of which gives

\[
\xi'(r_{ij}(t + \tau) = \xi'(r_{ij}(t)) + \frac{\xi(r_{ij}(t + \tau)) - \xi(r_{ij}(t))}{\tau r_{ij}(t + \tau) \cdot v_{ij}(t + \tau/2)} + \mathcal{O}(\tau^2), \quad (7)
\]

where the mid-point values of relative velocity \( v_{ij} = v_i - v_j \) are calculated according to Eq. (4) as

\[
v_{ij}(t + \frac{\tau}{2}) = \frac{1}{2} \left[ v_{ij}(t) + v_{ij}(t + \tau) \right].
\]

Then choosing \( \xi = \phi, J \), one finds the expression

\[
\tau f_{ij}(t + \frac{\tau}{2}) = - \sum_{j \neq i} \frac{r_{ij}(t + \frac{\tau}{2})}{r_{ij}(t + \frac{\tau}{2}) \cdot v_{ij}(t + \frac{\tau}{2})} \left( \phi(r_{ij}(t + \tau)) - \phi(r_{ij}(t)) - \frac{2}{r_{ij}(t + \tau) - r_{ij}(t)} \right).
\]

for mid-step translational forces, where the \( \mathcal{O}(\tau^3) \) terms have been neglected.

Performing scalar multiplication of Eq. (8) with the vector \( v_i(t + \tau/2) \), then taking the sum over all the particles \((i = 1, 2, \ldots, N)\), and using the fact that the double sum obtained in the right-hand side is invariant with respect to the replacements \( i \leftrightarrow j \), it can be shown that

\[
\tau \sum_{i=1}^{N} f_i(t + \frac{\tau}{2}) \cdot v_i(t + \frac{\tau}{2}) = - \sum_{i 

\]

Assuming for the moment that spin degrees of freedom are frozen (i.e., that \([s_i s_j]\) do not depend on time), the last relation can be presented in the form

\[
\tau \sum_{i=1}^{N} f_i(t + \tau/2) \cdot v_i(t + \tau/2) = U(t) - U(t + \tau),
\]

where \( U \) denotes the potential energy of the system. On the other hand, multiplying the second line of Eq. (3) by \( v_i(t + \tau/2) \) and summing over the particles yields

\[
\sum_{i} \frac{m_i}{2} (v_i(t + \tau) - v_i(t)) \cdot (v_i(t + \tau) + v_i(t)) = K(t + \tau) - K(t) = \tau \sum_{i=1}^{N} f_i(t + \frac{\tau}{2}) \cdot v_i(t + \frac{\tau}{2}),
\]

where \( K \) denotes the kinetic energy. We see, therefore, that during the time propagation given by Eqs. (3) and (8), the total energy \( E = K + U \) is conserved exactly for any \( \tau \), i.e., \( E(t + \tau) = E(t) \).

Note that Eqs. (7) and (8) are well defined when the scalar product \( r_{ij}(t + \tau/2) \cdot v_{ij}(t + \tau/2) \) tends to zero. This is so because according to the first line of Eq. (3), the mid-step relative velocity is connected with the change in position by the constraint \( v_{ij}(t + \tau/2) = (r_{ij}(t + \tau) - r_{ij}(t))/\tau \). So that the scalar product is merely equal to \( (r_{ij}(t + \tau) + r_{ij}(t)) \cdot (r_{ij}(t + \tau) - r_{ij}(t))/(2\tau) \approx (r_{ij}(t + \tau) + r_{ij}(t)) / (2\tau) = (r_{ij}(t + \tau)) / (2\tau) \). As a result, the right-hand side of Eq. (7) can be rewritten in the following mathematically equivalent form

\[
\xi(r_{ij}(t + \tau)) - \xi(r_{ij}(t)) \frac{2}{r_{ij}(t + \tau) - r_{ij}(t)} + \mathcal{O}(\tau^2) \quad (9)
\]

which reduces to \( \xi'(r_{ij}(t + \tau)) / r_{ij}(t + \tau) + c^2 \mathcal{O}(\tau^2) \) when the value \(|r_{ij}(t + \tau) - r_{ij}(t)| < \epsilon \) is small enough,
where \( r_{ij}(t+\tau/2) = (r_{ij}(t)+r_{ij}(t+\tau))/2 \) and \( c^2 \) denotes a machine zero.

The exact energy conservation can also be achieved in the presence of spin subdynamics. In order to show this, unfreeze now the spin variables, and consider first the question of how to interpolate the vector product \( s_i \times g_i \) arising in the third line of Eq. (3). Again, since this product depends on time implicitly via dynamical variables \( s_i \) and \( g_i \), we will have here a lot of possibilities. The first of them

\[
[s_i \times g_i]_{t+\tau/2} = \frac{1}{2} \left[ s_i(t) \times g_i(t) + s_i(t+\tau) \times g_i(t+\tau) \right]
\]

is not suitable because it does not lead to the conservation of individual spin lengths, i.e., \( s_i(t+\tau) = s_i(t) + O(\tau^3) \). At the same time, the second interpolation

\[
[s_i \times g_i]_{t+\tau/2} = \frac{1}{2} \left[ s_i(t) + s_i(t+\tau) \right] \times g_i(t+\tau/2) \quad (10)
\]

does conserve spin lengths exactly, \( s_i(t+\tau) = s_i(t), \) for arbitrary choice of \( g_i(t+\tau/2) \). Indeed, substituting Eq. (10) into the propagation equation \( s_i(t+\tau) = s_i(t) + \tau s_i \times g_i(t+\tau/2)/\hbar \) and solving analytically the obtained expression with respect to \( s_i(t+\tau), \) one obtains

\[
s_i(t+\tau) = \frac{1}{1 + \frac{\tau^2}{4\hbar^2}|g_i|_{t+\tau/2}^2} \left[ s_i(t) + \frac{\tau}{\hbar} g_i(t) \times [g_i]_{t+\tau/2} + \frac{\tau^2}{4\hbar^2} \left( 2[g_i]_{t+\tau/2} \cdot s_i(t) - (|g_i|_{t+\tau/2}^2 s_i(t)) \right) \right]. \quad (11)
\]

As can be verified readily, Eq. (11) represents a unitary transformation, \( s_i(t+\tau) = \Theta_i(t,\tau) s_i(t), \) where \( \Theta_i(t,\tau) \) is a rotation matrix which, of course, does not change the norm of vectors.

Three different time-reversible interpolations can be introduced for the factor \( |g_i|_{t+\tau/2} \equiv g_i(t+\tau/2) = \sum_{j(j\neq i)} |g_{ij}|_{t+\tau/2}. \) They are:

\[
[g_{ij}]_{t+\tau/2} = \frac{J(r_{ij}(t)) s_j(t) + J(r_{ij}(t+\tau)) s_j(t+\tau)}{2},
\]

\[
[g_{ij}]_{t+\tau/2} = J \left( \frac{r_{ij}(t) + r_{ij}(t+\tau)}{2} \right) \frac{s_i(t) + s_j(t+\tau)}{2},
\]

and

\[
[g_{ij}]_{t+\tau/2} = J \left( \frac{r_{ij}(t) + r_{ij}(t+\tau)}{2} \right) \frac{s_j(t) + s_i(t+\tau)}{2}.
\]

The first approximation cannot be chosen for our purpose because it destroys the total magnetization of the system, i.e., \( M(t+\tau) = M(t) + O(\tau^3) \). The last two interpolations do reproduce the magnetization vector perfectly. Indeed, summing up the individual spin propagation (third of Eqs. (3)) over all the particles and taking into account Eq. (10) gives \( M(t+\tau) = M(t) + \Delta M, \) where

\[
\Delta M = \frac{\tau}{4\hbar} \sum_{i \neq j} J_{ij}(t+\tau/2) \left[ s_i(t) + s_i(t+\tau) \right] \times \left[ s_j(t) + s_j(t+\tau) \right]
\]

and \( J_{ij}(t+\tau/2) \) may be equal either to \( J((r_{ij}(t)+r_{ij}(t+\tau))/2) \) or \( J(r_{ij}(t)) + J(r_{ij}(t+\tau))/2. \) The term \( \Delta M \) is canceled because of the invariance of the double sum with respect to the transformation \( i \leftrightarrow j, \) and of the obvious equality \( a \times b + b \times a = 0 \) which fulfills for any vectors \( a \) and \( b. \) Thus, \( M(t+\tau) = M(t) \) in both the cases. However, in the first of them when \( J_{ij}(t+\tau/2) = J(r_{ij}(t) + r_{ij}(t+\tau))/2, \) the energy difference \( E(t+\tau) - E(t), \) being a function of two quantities \( J(r_{ij}(t+\tau)) \) and \( J(r_{ij}(t)), \) cannot be reduced to zero exactly using only one midpoint value \( J((r_{ij}(t)+r_{ij}(t+\tau))/2). \)

At the same time, within the last third interpolation given by Eq. (12) we are able to perform such a reduction. To demonstrate this, let us consider finally the interpolation of the spin scalar product \( |s_i| s_j|_{t+\tau/2} \) appearing in Eq. (8). Similarly to Eq. (6), one defines this interpolation in the form

\[
|s_i \cdot s_j|_{t+\tau/2} = \frac{1}{2} \left[ s_i(t) \cdot s_j(t) + s_i(t+\tau) \cdot s_j(t+\tau) \right]. \quad (13)
\]

Then, using Eqs. (3), (8), (10) and (12), it can be shown that the following equality holds

\[
\sum_{i \neq j} \left[ J(r_{ij}(t+\tau)) - J(r_{ij}(t)) \right] |s_i \cdot s_j|_{t+\tau/2} = \sum_{i \neq j} \left( J(r_{ij}(t+\tau)) s_i(t+\tau) \cdot s_j(t+\tau) - J(r_{ij}(t)) s_i(t) \cdot s_j(t) \right)
\]

\[
\equiv U^{(s)}(t) - U^{(s)}(t+\tau),
\]

where \( U^{(s)} \) denotes the spin part of the potential energy. So that, as in the case of frozen spin subdynamics, the sum \( \tau \sum_{i=1}^{N} E(t+\tau/2) \cdot v_i(t+\tau/2) \) is reduced to the potential energy difference \( U(t) - U(t+\tau). \) But as was shown earlier using Eq. (3), this sum can be expressed also as the difference \( K(t+\tau) - K(t) \) in the kinetic energy. This indicates again that the total energy \( E = K + U \) is conserved exactly, i.e., \( E(t+\tau) = E(t), \) despite the microscopic solutions \( r_i(t+\tau), v_i(t+\tau) \) as well as \( s_i(t+\tau) \) are obtained with a limited \( O(\tau^3) \) accuracy. It is worth mentioning that the interpolation

\[
|s_i \cdot s_j|_{t+\tau/2} = \frac{s_i(t) + s_i(t+\tau)}{2}, \frac{s_j(t) + s_j(t+\tau)}{2}
\]

instead of Eq. (13) is possible, in principle, too but it will not lead to the energy conservation.

The approach considered conserves also the total linear and angular momenta, i.e., \( P(t+\tau) = P(t) \) and \( L(t+\tau) = L(t). \) The first follows directly from the
structure of mid-point translational forces (8) for which \( \sum_i f_i(t + \tau/2) = 0 \), so that

\[
\sum_i m_i v_i(t + \tau) = \sum_i m_i v_i(t).
\]

Further, taking into account Eqs. (3) and (4), the position propagation can be cast as

\[
r_i(t + \tau) = r_i(t) + v_i(t)\tau + f_i(t + \frac{\tau}{2})\tau^2/2m_i.
\]

Then the sum \( \sum_i m_i r_i(t + \tau) \times v_i(t + \tau) \) reduces to \( \sum_i m_i r_i(t) \times v_i(t + \tau) + \sum_i r_i(t + \tau/2) \times f_i(t + \tau/2) \). The last term is canceled since, according to Eq. (8), the interparticle forces are parallel to mid-step vectors \( r_{ij}(t + \tau/2) \), and, thus, the second property

\[
\sum_i m_i r_i(t + \tau) \times v_i(t + \tau) = \sum_i m_i r_i(t) \times v_i(t)
\]

is also satisfied.

Thus, the desired algorithm of the second order has been constructed. In view of Eqs. (3), (4), (6)–(10), (12) and (13), the algorithm can be presented in the following compact form

\[
r_i(t + \tau) = r_i(t) + \frac{\tau}{2} \left[ v_i(t) + v_i(t + \tau) \right],
\]

\[
v_i(t + \tau) = v_i(t) - \frac{\tau}{m_i} \sum_{j(j \neq i)} r_{ij}(t) + r_{ij}(t + \tau) \times \left( \frac{\varphi(r_{ij}(t + \tau)) - \varphi(r_{ij}(t))}{r_{ij}(t + \tau) - r_{ij}(t)} - \frac{J(r_{ij}(t + \tau)) - J(r_{ij}(t))}{r_{ij}(t + \tau) - r_{ij}(t)} \right) \times \left( s_i(t) \cdot s_i(t + \tau) \right),
\]

\[
s_i(t + \tau) = s_i(t) + \frac{\tau}{\hbar} \left[ s_i(t) + s_i(t + \tau) \right] \times \frac{J(r_{ij}(t + \tau)) + J(r_{ij}(t + \tau))}{2} \frac{s_j(t) + s_j(t + \tau)}{2}.
\]

Equation (14) constitutes, in fact, a coupled system of three nonlinear vector equations for each particle with respect to the same number of unknowns \( r_i(t + \tau), v_i(t + \tau) \) and \( s_i(t + \tau) \). The system can be solved in a quite efficient way by iteration, letting initially \( v_i^{(0)}(t + \tau) = v_i(t) \) and \( s_i^{(0)}(t + \tau) = s_i(t) \) in the right-hand sides of Eq. (14). Then the current values for \( r_i(t + \tau), v_i(t + \tau) \) and \( s_i(t + \tau) \) obtained in the left-hand sides of Eq. (14) are treated as initial guesses for the next iteration. Already two iterations are sufficient to reach the \( O(\tau^3) \) accuracy for the microscopic solutions and energy conservation, i.e., \( E(t + \tau) - E(t) = O(\tau^3) \). The goal of carrying out further several updates of Eq. (14) is to reduce the uncertainty \( \varepsilon = E(t + \tau) - E(t) \) in energy deviation to a negligibly small value (by adjusting the number \( l \geq 2 \) of iterations for a given \( \tau \)). The rapid convergence \( \varepsilon \to 0 \) is guaranteed by the relative smallness of the step size \( \tau \) and an exponential decaying of \( \varepsilon \) with increasing \( l \) (see the next section).

It is interesting to remark that the total linear and angular momenta are conserved exactly within each iteration of Eq. (14). The reason is that the interparticle forces are evaluated exploiting Newton’s third law and the velocities \( v_i(t + \tau) \) are updated before all \( (i = 1, 2, \ldots, N) \) the advanced positions \( r_i(t + \tau) \) were calculated. For similar reasons, the magnetization conservation is also fulfilled for each iteration, when the spins are updated according to the third line of Eq. (14). In this case, however, the individual spin lengths will only be preserved like energy in an iterative sense, i.e., \( s_i(t + \tau) - s_i(t) = O(\varepsilon) \). Nevertheless, the spin lengths can be maintained exactly within each iteration by replacing this third line by its mathematically equivalent counterpart (11) (were \( \{g_{ij}\}_{i = 0}^{i + \tau/2} \) being evaluated with the help of Eq. (12)).

IV. NUMERICAL TESTS. COMPARISON WITH OTHER METHODS

In our MD simulations of the Heisenberg spin fluid (Eq. (1)), the Yukawa function

\[
Y(r) = u \frac{\sigma}{r} \exp \left( \frac{\sigma - r}{\sigma} \right)
\]

was used to describe the spin-spin interactions. The liquid subsystem was modelled by a soft-core potential \( \varphi(r) \),

\[
\varphi(r) = 4u \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} \right] + u
\]

which accepts nonzero values at \( r < 2^{1/6}\sigma \) and \( \varphi(r) = 0 \) at \( r \geq 2^{1/6}\sigma \). Here \( \sigma \) is the diameter of particles, and \( u \) as well as \( w \) denote the intensities of core-core and spin-spin interactions, respectively. The simulations were carried out in a microcanonical ensemble for \( N = 1000 \) identical \( (m = n, s = 1) \) particles in a cubic box of volume \( V = L^3 \) employing periodic boundary conditions. The Yukawa function was truncated at \( R_c = 2.5\sigma < L/2 \) and shifted to be zero at the truncation point to avoid the force singularities, i.e., \( J(r) = Y(r) - Y(R_c) \) at \( r < R_c \) and \( J(r) = 0 \) otherwise. We have chosen the same thermodynamic point as considered in previous papers [16, 17], namely, a reduced density of \( n^* = N\sigma^3/V = 0.6 \), a reduced temperature of \( T^* = k_B T/w = 1.5 < T_c^* \) (where \( k_B \) is the Boltzmann’s constant and \( T_c \approx 2.055 \) the critical temperature of the system [18]), a non-zero magnetization per particle of \( |M|/N \approx 0.6356 \) as well as the same values for the reduced core intensity \( u/w = 1 \) and the dynamical coupling parameter \( d = \sigma(mw)^{1/2}/\hbar = 2 \).

The equations of motion were solved using a well established Adams-Bashforth-Moulton (ABM) predictor-corrector integrator of the fourth order [20]. The explicit
where \( \dot{\rho} \) only at very small time steps (namely, at other existing predictor-corrector schemes can be used of an algorithm). For this reason, the ABM as well as numerical stability follows directly from the reversibility of reversible solutions (as is now rigorously proved \([23]\), the procedure.

The situation is completely different in the case of our new approach, because the CSFD algorithm preserves the integrals of motion for arbitrary time steps. Of course, we cannot apply too large step sizes (\( \tau^* \sim 1 \)) since then the microscopic solutions will deviate considerably from their exact counterparts and because of too large number of iterations needed to achieve the convergence. Choosing, for instance, \( \tau^* = \tau_{\text{max}} = 0.01 \) we have determined the following levels \( \mathcal{E} \) in the averaged total energy fluctuations \( \mathcal{E} = \langle (E^*(t) - E^*(0))^2 \rangle^{1/2}/N \) at the end of the 100 000 time step runs: \( 9.2 \times 10^{-4}, 2.3 \times 10^{-5}, 3.1 \times 10^{-6}, 2.2 \times 10^{-7}, \) and \( 2.8 \times 10^{-8} \) corresponding to the numbers \( l \) of iterations: 2, 3, 4, 6, and 8, respectively. We see, therefore, that the iterations converge rapidly with increasing \( l \) and the uncertainties can be approximately described by the exponential dependence \( \mathcal{E} \sim 3 \times 10^{-4} \exp(-1.2l) \) at \( l \geq 4 \). Of course, the iterative solutions require additional computational efforts, but they are justified when a high level of the energy conservation is necessary. In order to demonstrate this, we have tried to reduce the.

### FIG. 1

The total energy \( E^*/N \) (subset (a)) and magnetization \( M/N \) (subset (b)) per particle as functions of the length \( t/\tau \) of the simulations performed for a Heisenberg spin fluid using the predictor-corrector (dashed curve in (a), marked as ABM, decomposition (solid curves, ED/ED4), and our new (bold solid horizontal lines, CSFD) algorithms. Note that the ABM and ED4 curves are indistinguishable in (b) from the CSFD line.
energy fluctuations within the ED/ED4 algorithms by decreasing the time step. The corresponding result for $\mathcal{E}$ at the time steps $0.01$, $0.005$, $0.0025$, and $0.00125$, i.e., at $\tau^* \equiv \tau_{\text{max}}^*/l$ with $l = 1, 2, 4$, and $8$ is presented in Fig. 2 in comparison with the above CSFD data.

![Graph showing energy fluctuations](image)

**Fig. 2.** The averaged total energy fluctuations $\mathcal{E}$ as a function of the number $l$ of iterations, obtained in the Heisenberg fluid simulations within the CSFD algorithm at the reduced time step $\tau_{\text{max}} = 0.01$ (bold solid curve). The levels of $\mathcal{E}$ corresponding to the decomposition integrators at the time steps $\tau^* \equiv \tau_{\text{max}}^*/l$ are connected by solid (ED) and dashed (ED4) curves.

As one can see, such a reduction of the ED/ED4 energy fluctuations is not efficient, since the deviations $\mathcal{E}$ behave like $\sim l^{-2}$, i.e., decrease with increasing $l$ much more slowly than the exponential dependence obtained within the CSFD algorithm. In view of the results of Fig. 2 and taking into account that at the same value of $l$ one needs approximately the same processor time with both the ED and CSFD algorithms (the ED integrator needs the time larger by factor 5 and is less economical) in order to investigate the system over an identical time interval, we come to the following conclusion: When the total energy must be conserved up to a precision of $\mathcal{E}_0 \sim 10^{-4}$ (the intersection point of the ED and CSFD curves, see the horizontal dashed line in Fig. 2) or better, the preference should be done to the CSFD algorithm. For example, a level of $\mathcal{E} \sim 10^{-8}$ in the conservation is achieved at $l \sim 5$ within the CSFD algorithm, while at $l \sim 50$ for the ED scheme (the last value was obtained by extrapolating the $\sim l^{-2}$ dependence to larger $l$). Thus the CSFD algorithm appears to be approximately in 10 times faster than the ED integrator at this level of $\mathcal{E}$. For $\mathcal{E} > \mathcal{E}_0$, we can restrict ourselves to the usual explicit decomposition integrators.

Note that despite the uncertainties $\mathcal{E}_0 \sim 10^{-4}$ look quite small, they can influence considerably on some observable macroscopic quantities. The influence can be estimated quantitatively in terms of the ratio $\Gamma = ((E(t) - E(0))^2)/(U(t) - U(0))^2)^{1/2}$ of total and potential energy fluctuations. For our system $U = (U^*(t) - U^*(0))^2/\mathcal{N} \sim 10^{-2}$, where $U^* = U/w$, so that $\Gamma_0 = \mathcal{E}_0/\Gamma \sim 1\%$. Usual investigated quantities, such as thermodynamic functions, structure factors, etc., will be calculated approximately with the same relative precision $\Gamma_0$ (provided the averaging over the produced trajectories is performed during a sufficiently large time interval to be entitled to ignore the statistical noise). But when long-time correlation functions or derivatives of the thermodynamic functions are involved into the computations, the impact of the artificial energy fluctuations on the results will be much greater. For instance, the relative uncertainty in the measurements of the specific heat (which are based on a microcanonical ensemble fluctuation formula) is estimated to be already $(\Gamma_0)^{1/2} \sim 10\%$. This uncertainty may appear to be too large to determine correctly a phase diagram of the system.

A similar pattern to that shown in Fig. 2 was observed within the CSFD approach at greater time steps $\tau > 0.01$. The energy as well as magnetization fluctuations continued to damp exponentially with increasing $l$, although a greater number of the iterations was necessary to reach the same level of the conservation. Note that a rotational matrix version of the CSFD algorithm (when the third line of Eq. (14) is replaced by Eq. (11)), in which individual spin lengths are maintained exactly (when the third line of Eq. (14) is replaced by Eq. (11)), leads to a somewhat better energy preservation at a given $l$ (but then the total magnetization like energy will be conserved in the iterative sense, i.e., at sufficiently large $l$). The CSFD results presented above for $\mathcal{E}$ have been obtained using this version for spin subdynamics propagations.

Further improvements in the efficiency of the CSFD algorithm can be reached applying the following computational trick. It can occur that after a some period of time during the integration process the energy difference $E(t) - E(0)$ corresponding to the last $l$-th iteration (within a current time step $t/\tau$) exceeds the difference obtained for the previous $(l-1)$-th iteration. Such a situation is possible because of round-off errors and an accumulation of other numerical uncertainties, especially at relatively small values of $l$, where the lack in the time-reversibility can lead to an instability of the solutions (note the CSFD algorithm is time-reversible in the iterative sense, i.e., at large enough values of $l$). Then to avoid the accumulation, we should take merely the values for microscopic phase variables corresponding to this previous $(l-1)$-th iteration. The trick with a flexible number of the iterations will guarantee a good stability for small $l \sim 2-4$ as well.

Another technical detail concerns the way in which the expression $[\xi(r(t + \tau)) - \xi(r(t))]/[r(t + \tau) - r(t)]$ (appearing in Eq. (14) for $\xi \equiv \varphi, J$) should be treated in the
limit $r(t + \tau) \to r(t)$. As was pointed out earlier, this expression must be computed using its limiting representation $|r(t) + r(t + \tau)|/2 + c^2\mathcal{O}(\tau^2)$ when the difference $|r(t + \tau) − r(t)| < \epsilon$ is small enough. Then letting $c^2$ being equal to zero, the truncated term $c^2\mathcal{O}(\tau^2)$ can be ignored completely. In our program code we have used a double precision throughout with 16 significant digits, $c^2 = 10^{-16}$, so that the value $\epsilon$ was set to be equal to $10^{-8}$. It is interesting to remark that the condition $|r_{ij}(t + \tau) − r_{ij}(t)| < \epsilon$ was never achieved for any pair $ij$ of particles during the simulations and, thus, the limiting expression was never used. This can be explained by the fact that the probability of finding the system in such a state is prohibitively small and is proportional to $C\epsilon$. The coefficient $C$ increases with increasing the length of the simulations and the number of particles as $C \sim t N^2$. Thus, the limiting expression is expected to be applied for systems with a greater size or when extra long simulations are performed.

Finally, some words about the angular momentum conservation. As is well known, the periodic boundary conditions, which are commonly used in MD simulations to reduce the finite-size effects, destroy the angular momentum vector. Nevertheless, it has been established that this vector is conserved in our simulations in mean, namely, $\langle \mathbf{L}(t) \rangle \approx \mathbf{L}(0)$. Note that initial values for the total angular as well linear momenta were put to be equal to zero, $\mathbf{L}(0) = 0$ and $\mathbf{P}(0) = 0$, i.e., the system was considered at the very beginning as one which does not move as a whole translationally and rotationally.

V. APPLICATIONS TO OTHER SYSTEMS: PURE LIQUIDS AND HARMONIC OSCILLATOR

The algorithm derived in the preceding section can also be applied with equal successes to dynamics simulations of other liquid models. For instance, letting formally $J \equiv 0$, we come to the usual equations of motion corresponding to a pure liquid system. These equations can be integrated using the first two lines of the same propagation equation (14), where the terms with $J$ in the right-hand side of the second line must be omitted (the third line describes spin subdynamics and is not relevant in this case).

Our simulations of pure liquid dynamics were based on a system composed of $N = 256$ particles interacting through a cut-off Lennard-Jones (LJ) potential, $\varphi(r) = \Phi(r) − \Phi(R_c)$ at $r < R_c = 3.25\sigma$ with $\varphi(r) = 0$ otherwise, where

$$\Phi = 4u \left[ \left( \frac{\sigma}{r} \right)^{12} − \left( \frac{\sigma}{r} \right)^{6} \right].$$

(17)

The MD test runs have been performed at a reduced density of $n^* = 0.845$ and a reduced temperature of $T^* = k_B T^*/u = 1.7$. For the purpose of comparison, the equations of motion were integrated applying also a well-established velocity Verlet (VV) algorithm [19] of the second order and its forth-order (VV4) counterpart [13]. Our algorithm we will call now as CPFD (conservative pure fluid dynamics). A typical maximal value for the reduced time step in simulating such a system is $\tau_{max}^* = \tau (u/m\sigma^2)^{1/2} \approx 0.005$ [23]. All the runs started from a well equilibrated configuration and covered an identical time interval of $t^* = t (u/m\sigma^2)^{1/2} = 50$ (corresponding to 10 000 time steps at $\tau^* = 0.005$).

It is worth mentioning that the explicit VV integrator propagates the phase variables according to the relations

$$r_i(t + \tau) = r_i(t) + v_i(t) \tau + f_i(t) \frac{\tau^2}{2m} + \mathcal{O}(\tau^3),$$

$$v_i(t + \tau) = v_i(t) + \left[ f_i(t) + f_i(t + \tau) \right] \frac{\tau}{2m} + \mathcal{O}(\tau^3).$$

This propagation can be presented as

$$\{ r_i(t + \tau), v_i(t + \tau) \} = \mathbf{D}(t, \tau) \{ r_i(t), v_i(t) \} + \mathcal{O}(\tau^3),$$

where $\mathbf{D}(t, \tau)$ denotes the evolutionary operator. The VV4 algorithm deals (similarly to the ED4 scheme) with the five stages propagation

$$\{ r_i(t + \tau), v_i(t + \tau) \} = \prod_{k=1}^{5} \mathbf{D}(t, \xi_k \tau) \{ r_i(t), v_i(t) \} + \mathcal{O}(\tau^5),$$

where the coefficients $\xi_k$ are: $\xi_1 = \xi_2 = \xi_4 = \xi_5 \equiv \xi = 1/(4 - 4^{1/3})$, and $\xi_3 = 1 - 4\xi$. The VV approach needs only in one force evaluation (the most time-consuming part of the calculations) per time step, $p_{VV} = 1$, while $p_{VV4} = 5$. The CPFD algorithm requires two force evaluation per iteration within the time step, $p_{CPFD} = 2$.

The averaged total energy fluctuations $\mathcal{E} = \langle (E(t) − E(0))^2 \rangle^{1/2} / (u N)$ obtained within the CPFD integration at the time step $\tau_{max}^* = 0.005$ and the numbers of iterations of $l = 2, 3, 4, 8$ are plotted in Fig. 3 as a function of the reduced processor time $l p$ (where in this case $p = p_{CPFD}$) needed to perform the run of the mentioned above length $t^* = 50$. The fluctuations identified during the integration at the time steps $\tau^* = \tau_{max}^*/l$ using the VV algorithm with $l = 1, 2, 4, 8$ and 16 as well as the VV4 algorithm with $l = 0.5, 1, 2, 4$ are also included in this figure. The processor time spent to carry out the VV run of the length $t^* = 50$ at $\tau^* = 0.005$ is assumed to be equal to unity in our dimensionless presentation $l p$ (where $p = 1, 2$ and 5 for the VV, CPFD, and VV4 integrators).

The LJ energy fluctuations damp with increasing $l$ like $\sim l^{-2}$, $\sim l^{-4}$, and $\sim \exp(-2.4l)$ within the VV, VV4, and CPFD integrations, respectively. Up to three intersection points corresponding to the VV–VV4, VV4–CPFD, and VV4–CPFD curves with the energy conservation levels of $\xi_1 \sim 6 \cdot 10^{-5}$, $\xi_2 \sim 10^{-5}$, and $\xi_3 \sim 3 \cdot 10^{-7}$ can be observed in Fig. 3. So that the usual VV algorithm is recommended to be used when the precision $\mathcal{E}$ of energy conservation plays not so important role in the computations, namely, when $\mathcal{E} \geq \xi_1$. The calculation with the
help of the VV4 integrator appears to be most computationally efficient in the intermediate regime $\mathcal{E}_3 < \mathcal{E} < \mathcal{E}_1$. Finally, when a very accurate conservation, $\mathcal{E}_1 < \mathcal{E}_2$, is required, the best choice is to apply the CPFD algorithm because then it becomes to be most economical.

Choosing the initial conditions $x(0) = 0$ and $\dot{x}(0) \equiv v(0) = 1$, the above two types of numerical trajectories can be compared between themselves and with respect to the exact solution $x(t) = \sin(t)$ and $\dot{x}(t) \equiv v(t) = \cos(t)$. The result of comparison for $x(t)$ is presented in Fig. 4 at a typical time step of $\tau = 0.05T$, where $T = 2\pi$ denotes the period of the oscillations. As can be seen easily, the conservative solution leads to a better reproduction of the original dependence than the VV trajectory, despite the both CPFD and VV approaches are valid to the same $O(\tau^3)$ order in truncation errors. Therefore, additional cancellations of the truncation uncertainties are possible due to the exact preservation of the integral of motion $2E = \dot{x}^2 + x^2 \equiv 1$ along the CPFD trajectory (note that maximal VV deviations in $E$ consist about 20% at $\tau = 0.05T$). Similar cancellations of the truncation uncertainties in microscopic solutions within our conservative approach should be expected for other systems of differential equations, in particular, for spin and pure liquid dynamics.

![Graph showing energy fluctuations](image1)

**FIG. 3.** The averaged total energy fluctuations $\mathcal{E}$ as a function of the reduced processor time $lp$ needed for the simulations of a Lennard-Jones liquid within the CPFD (bold solid curve), usual velocity Verlet (solid curve, VV), and fourth-order velocity Verlet (dashed curve, VV4) algorithms.

The CPFD approach can also be used for the prediction of dynamical phenomena in other many-body collections (such as the solar system, for instance) and treated as an efficient numerical solver of first-order differential equations. The most notorious example (which allows to be analyzed analytically) is the equation $d^2x/dt^2 \equiv \ddot{x} = -x$ describing dynamics of a simple harmonic oscillator. This equation reduces to a system of two first-order differential equations, $\dot{x} = v$ and $\dot{v} = -x$, which in turn can be reproduced from the first two lines of general equation (2) putting formally $r_i \equiv x$, $v_i \equiv v$, $\varphi = x^2/2$, $m_i \equiv 1$ and $J = 0$. Then in view of Eq. (14), the time propagation reads $x(t + \tau) = x(t) + \tau [v(t) + v(t + \tau)]/2$ and $v(t + \tau) = v(t) - \tau [x(t) + v(t + \tau)]$. The last two relations can be solved explicitly, and the result for the conservative numerical trajectories is

$$x(t + \tau) = \frac{x(t)(1 - \tau^2/4) + v(t)\tau}{1 + \tau^2/4},$$

$$v(t + \tau) = v(t) - \tau \left[ x(t) + \frac{v(t)\tau/2}{1 + \tau^2/4} \right],$$

whereas the VV solutions are

$$x(t + \tau) = x(t)(1 - \tau^2/2) + v(t)\tau,$$

$$v(t + \tau) = v(t) - \tau \left[ x(t)(1 - \tau^2/4) + v(t)\tau/2 \right].$$

**VI. CONCLUDING REMARKS**

One of the most fundamental characteristics in physics are the conservation laws. Therefore, it is desirable that the numerical methods in computational physics obey these laws. Unfortunately, the most popular algorithms,
such as predictor-corrector, Runge-Kutta, decomposition Suzuki-Trotter, etc., being applied to the nonlinear many-body problem, do not preserve fundamental physical invariants, like energy and angular momentum, when these are inherent in the description.

In the present paper we have tried to remedy such a situation and formulated a novel completely conservative approach for numerical integration of the equations of motion in classical systems. The approach is general enough to be used for a wide class of systems such as spin and pure liquids, collections of charged particles, etc. It can also be considered for the prediction of other phenomena in physics, astrophysics, chemistry and biology, whenever the numerical solutions to systems of differential equations are necessary.

Our main attention in this study was concentrated on dynamics of spin liquid models in which additional effects with respect to pure liquids are possible because of the energy exchange between spin and liquid subsystems [15,17]. As a result, a new second-order MD algorithm (called as CSFD) has been consequently derived within the above presented approach. Its greatest advantage is that all the integrals of motion existing in the system, namely, the total energy, linear and angular momenta, individual spin lengths and total magnetization are conserved independently on the size of the time step. It is worth emphasizing that such a complete conservation has been achieved intrinsically, i.e., without the introduction of any artificial external forces or numerical constraints. Moreover, the resulting algorithm maintains the time reversibility property inherent in the basic equations. This is also important for long-duration MD observations because the stability of an algorithm is closely connected with its time reversibility [23].

The presented algorithm is implicit, i.e., it requires iterative solutions. Thus, when a high precision in conservation is not needed, the CSFD scheme may be less efficient in practice than explicit decomposition methods [16,17]. We have shown, however, on the basis of actual simulation of a Heisenberg fluid model that when the total energy and magnetization must be reproduced precisely, the CSFD algorithm may be in order or even more faster than the decomposition integrators. Another important feature of the conservative method is that additional cancellations of the truncation uncertainties are possible in microscopic solutions due to the exact preservation of the macroscopically observable integrals of motion, as was demonstrated analytically on a simple example of the harmonic oscillator.

For a particular case when the spin subsystem is absent, the CSFD algorithm reduces to a so-called CPFD integrator. While this work has been done we have learned that this integrator is equivalent, in fact, to that developed independently by Greenspan [25] as well as Gonzalez and Simo [24]. These authors, however, considered the integration in respect of applying it to mechanical systems when the number of particles is not very large. Here we have shown within the LJ model that the CPFD integrator can be used with equal success in MD simulations of pure liquids.

The approach presented can be adapted to many-component systems, optimized to a multiple time stepping integration and extended to higher-order versions. These and other related problems will be the subject of a separate investigation.

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