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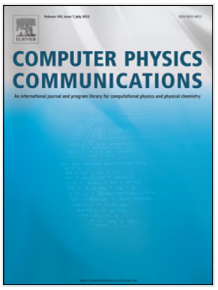

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An efficient algorithm for time propagation as applied to linearized augmented plane wave method

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An algorithm for time propagation of the time-dependent Kohn-Sham equations is presented. The algorithm is based on dividing the Hamiltonian into small time steps and assuming that it is constant over these steps. This allows for the time-propagating Kohn-Sham wave function to be expanded in the instantaneous eigenstates of the Hamiltonian. The method is particularly efficient for basis sets which allow for a full diagonalization of the Hamiltonian matrix. One such basis is the linearized augmented plane waves. In this case we find it is sufficient to perform the evolution as a second-variational step alone, so long as sufficient number of first variational states are used. The algorithm is tested not just for non-magnetic but also for fully non-collinear magnetic systems. We show that even for delicate properties, like the magnetization density, fairly large time-step sizes can be used demonstrating the stability and efficiency of the algorithm.

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In the 1990s, light-induced demagnetization in Ni was experimentally demonstrated[1]. The field of femtomagnetism[2] has since grown[3–13] and there is a lot of research to achieve light-induced control of spins. However, the physics of light-spin interactions remains highly debated[14–16] largely because of lack of *ab-initio* calculations. Time-dependent density functional theory (TDDFT)[17], which extends density functional theory into the time domain, is a formally exact *ab-initio* method for describing the real-time dynamics of interacting electrons under the influence of a laser field. In order to study femtomegnetism a fully non-collinear extension of TDDFT is required. Within this extension Kohn-Sham (KS) states need to be treated as Pauli spinors and the magnetization density as an unconstrained vector field. Since such a vector field requires very little energy to alter its direction, a highly precise time-evolution algorithm is required for reliable results.

Such an algorithm is required to propagate the time-dependent Schrödinger equation:

$$i\frac{\partial}{\partial t}\left|\Phi_i(t)\right\rangle = \hat{H}(t)\left|\Phi_i(t)\right\rangle, \tag{1}$$

where $\hat{H}$ is the Hamiltonian and Φ the wave function of interacting electrons. By the virtue of the Runge-Gross theorem[17], one can obtain the exact time-propagation of the density of this fully interacting system by solving single particle time-dependent KS equations. In our particular case, where the orbitals are Pauli spinors, these are

$$i\frac{\partial\psi_j(\mathbf{r},t)}{\partial t} = \Big[\tfrac{1}{2}\Big(-i\overrightarrow{\nabla} + \tfrac{1}{c}\overrightarrow{A}_{\rm ext}(t)\Big)^2 + v_s(\mathbf{r},t) \tag{2}$$
$$+ \tfrac{1}{2c}\overrightarrow{\sigma}\cdot\overrightarrow{B}_s(\mathbf{r},t) + \tfrac{1}{4c^2}\overrightarrow{\sigma}\cdot(\overrightarrow{\nabla}v_s(\mathbf{r},t)\times -i\overrightarrow{\nabla})\Big]\psi_j(\mathbf{r},t),$$

where $\overrightarrow{A}_{\rm ext}(t)$ is a external vector potential, $\overrightarrow{\sigma}$ are the Pauli matrices and ψ_j are the KS orbitals. The KS effective potential $v_s(\mathbf{r},t) = v_{\rm ext}(\mathbf{r},t) + v_{\rm H}(\mathbf{r},t) + v_{\rm xc}(\mathbf{r},t)$ is decomposed into the external potential $v_{\rm ext}$, the classical electrostatic Hartree potential $v_{\rm H}$ and the exchange-correlation (XC) potential $v_{\rm xc}$. Similarly the KS magnetic field is written as $\mathbf{B}_s(\mathbf{r},t) = \mathbf{B}_{\rm ext}(t) + \mathbf{B}_{\rm xc}(\mathbf{r},t)$ where $\mathbf{B}_{\rm ext}(t)$ is an external magnetic field and $\mathbf{B}_{\rm xc}(\mathbf{r},t)$ is the XC magnetic field. The final term of Eq. (2) is the spin-orbit coupling term.

Some of the important requirements for accurate[18, 19] time-propagation algorithm for solving Eq. 2 are (a) stability: the errors do not build up as the system is propagated for longer times, (b) efficiency: time propagation is performed by dividing the the total time interval into steps and it is essential for an efficient algorithm to allow for large time steps and (c) unitarity: which is required for maintaining the normalization of the wave function at each time-step. In the following we outline one such algorithm which satisfies all the above criteria and is particularly suited for computer codes that deal with dual basis like linearized augmented plane waves (LAPW) and perform exact diagonalization of the Hamiltonian.

The solution of the KS equations can be represented by means of the time evolution operator:

$$\left|\psi_i(T)\right\rangle = \hat{U}(T,0)\left|\psi_i(0)\right\rangle, \tag{3}$$

where $\hat{U}(T,0)$ is the time evolution operator that propagates all TD-KS states from time $t=0$ to the final time $t=T$ and satisfies the decomposition law:

$$\hat{U}(T,0) = \hat{U}(T,T-\Delta t)\ldots\hat{U}(2\Delta t,\Delta t)\,\hat{U}(\Delta t,0) \tag{4}$$

which allows for division of the total time propagation into small steps of step length Δt. In the limit $\Delta t\to 0$ this time propagation operator can be approximated as:

$$\hat{U}(t+\Delta t,t) = e^{-i\hat{h}_s(t)\Delta t}. \tag{5}$$