

# Fast and stable method for simulating quantum electron dynamics

Naoki Watanabe and Masaru Tsukada  
Department of Physics, University of Tokyo

A fast and stable method is formulated to compute the time evolution of wave functions under scalar and vector potentials by directly computing the time dependent Schrödinger equation. This method is a real space time evolution method implemented by several techniques of the computational physics to realize high accuracy, perfect stability and a good suitability for vector- and parallel-type supercomputers. Applying this method to some simple electron dynamics, we confirmed the efficiency and the accuracy of the method.

## Introduction

Owing to the rapid progress in computers and electronic state theories, it becomes possible to simulate dynamics of many body quantum systems with the first principles approaches. To carry out the computation of the time-dependent Kohn-Sham equation or the time dependent Schrödinger equation, efficient numerical methods have been desired and many methods are proposed.<sup>1-4)</sup> The important points of those methods are, a unitary time evolution operator, a small amount of the computational effort, and suitability for vector and parallel computers.

In the present work, we formulate a new method to compute the time dependent Schrödinger equation, which is more improved in those points mentioned above by using a real space time evolution approach based on Suzuki's "fractal decomposition"<sup>5)</sup> with Cayley form.

In the method we propose, wave functions are represented by values at spatial grids. The numerical stability is perfect. Probability and ortho-normality are preserved perfectly. The computational effort required is proportional to the number of grids in the system. Moreover, the algorithm is quite simple and suitable for vector and parallel computers by introducing "adhesive operator".

## Formulations of the real space time evolution method

The time dependent Schrödinger equation with scalar and vector potentials takes the following form;

$$i\frac{\partial\psi(\mathbf{r},t)}{\partial t} = \left[-\frac{1}{2}(\nabla - i\mathbf{A}(\mathbf{r}))^2 + V(\mathbf{r})\right] \psi(\mathbf{r},t).$$

Here  $\hbar$  and  $m$  are taken as unit. The strict, analytical solution of this differential equation is given by,

$$\psi(\mathbf{r},t + \Delta t) = \exp\left[\frac{i\Delta t}{2}(\nabla - i\mathbf{A})^2 - i\Delta tV\right] \psi(\mathbf{r},t).$$

Some approximations are needed to compute this exponential operator. For example, in the case of a two-dimensional system with no vector potential;  $\mathbf{A} = 0$ , the exponential operator is approximated as a product of exponential operators

as below,

$$\begin{aligned} \psi(\mathbf{r},t + \Delta t) \simeq & \exp\left[-i\frac{\Delta t}{2}V\right] \exp\left[i\Delta t\frac{\partial_x^2}{2}\right] \\ & \times \exp\left[i\Delta t\frac{\partial_y^2}{2}\right] \exp\left[-i\frac{\Delta t}{2}V\right] \psi(\mathbf{r},t). \end{aligned}$$

In another case, with only a uniform magnetic field;  $\mathbf{A} = (-By, 0, 0)$ ,  $V = 0$ , exponentials of the magnetic field appear in the product as below,

$$\begin{aligned} \psi(\mathbf{r},t + \Delta t) \simeq & \exp\left[i\Delta t\frac{\partial_y^2}{4}\right] \exp[-iBxy] \\ & \times \exp\left[i\Delta t\frac{\partial_x^2}{2}\right] \exp[iBxy] \exp\left[i\Delta t\frac{\partial_y^2}{4}\right] \psi(\mathbf{r},t). \end{aligned}$$

We treat the wave functions  $\psi(\mathbf{r})$  as a set of values at spatial grids  $\psi(\mathbf{r}_i)$ . Exponentials of a scalar potential and a magnetic field turn the phase of the wave function at each grids. Exponential of a spatial second derivative is computed by using another approximation called Cayley form;

$$\exp\left[i\Delta t\frac{\partial_x^2}{2}\right] \psi(\mathbf{r}_i) \simeq \frac{1 + i\Delta t\partial_x^2/4}{1 - i\Delta t\partial_x^2/4} \psi(\mathbf{r}_i).$$

This is equivalent with the following Crank-Nicholson scheme,

$$\left[1 - i\Delta t\frac{\partial_x^2}{4}\right] \phi(x, \Delta t) = \left[1 + i\Delta t\frac{\partial_x^2}{4}\right] \phi(x, 0)$$

By utilizing a finite differential method, it turns into a simultaneous linear equation as below,

$$\begin{aligned} & \begin{bmatrix} A & -1 & 0 & 0 \\ -1 & \ddots & \ddots & 0 \\ 0 & \ddots & \ddots & -1 \\ 0 & 0 & -1 & A \end{bmatrix} \begin{bmatrix} \phi_1(\Delta t) \\ \vdots \\ \vdots \\ \phi_N(\Delta t) \end{bmatrix} \\ & = \begin{bmatrix} B & +1 & 0 & 0 \\ +1 & \ddots & \ddots & 0 \\ 0 & \ddots & \ddots & +1 \\ 0 & 0 & +1 & B \end{bmatrix} \begin{bmatrix} \phi_1(0) \\ \vdots \\ \vdots \\ \phi_N(0) \end{bmatrix}, \end{aligned}$$

where  $A \equiv -4i\Delta x^2/\Delta t + 2$ ,  $B \equiv -4i\Delta x^2/\Delta t - 2$ .



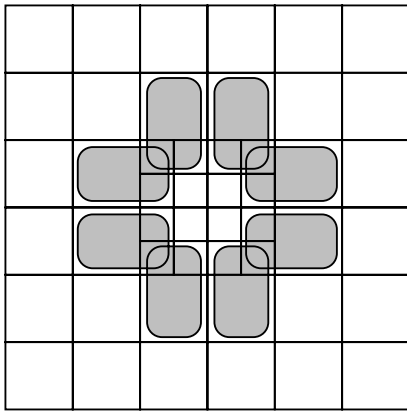


Fig. 5. The adhesion for adaptive mesh refinement.

### Test of the real space time evolution method

To confirm the accuracy, we simulated a photon excitation of a hydrogen atom. Photon field was treated as a semi-classical oscillating electric force. It was found that the electron density started from 1S state is excited in an oscillatory way and after some elapsed time it becomes to the 2Pz state as shown in Fig. 6.

Such electron oscillation generates a scattering light as shown in Fig. 7. In the spectrum of the scattering light, several peaks are found at the energies known from the theory of nonlinear optics.

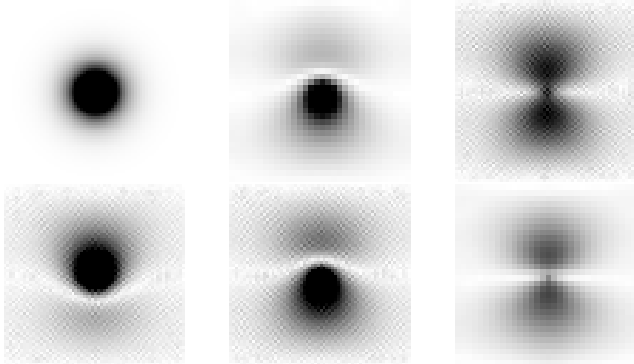


Fig. 6. The process of excitation of a hydrogen atom.

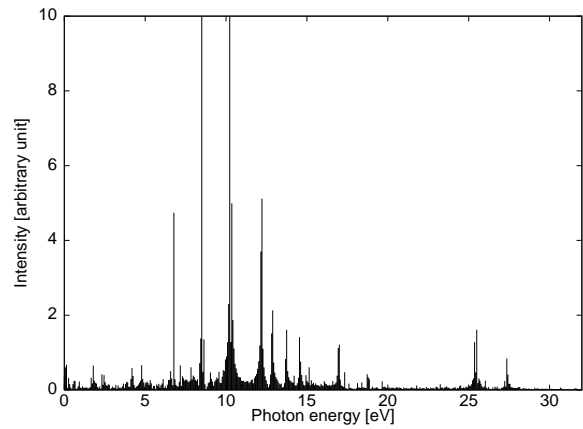


Fig. 7. The spectrum of the scattering light from hydrogen.

The transition energies can be obtained analytically by using the perturbation theory. However, with the present method, we can directly calculate the behavior without the perturbation theory and without the information about excited states of the system.

### Summary

We have formulated a new scheme for simulating one electron dynamics under scalar and vector potentials. We have found that by using Cayley form and Suzuki's fractal decomposition, the simulation is fast, stable, accurate, and suitable for vector computers. We have proposed the adhesive operator to simulate periodic and large systems on parallel computers.

These techniques will be also useful for the time-dependent Kohn Sham equation, namely, for the simulations of dynamics or chemical reactions of many body large systems. It is our future work.

### References

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